THESIS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY

Fuel Gas Technology for Biomass and Waste Environmental and Techno-Economic Assessments

Fabrizio Di Gregorio

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Caserta (Italy), 2012







Energy generation from biomass and waste is one of the most concern of the society at this time.

On one hand, despite increasing attention for waste prevention and sustainability, total municipal solid waste has increased in the last two decades and actual value is projected to double by 2030. Today, modern incinerators provide one of

the most robust and energy-efficient options for energy recovery from waste, and due to the increasing of energy demand and environmental concerns, especially related with the use of fossil fuels, new solutions are continuously sought, also in the order to limit the greenhouse gas effect.

On the other hand, biomass, and in general renewable energy, can have a significant positive impact for industrialized societies, and even more for developing countries. In rural areas, particularly in remote locations, transmission and distribution of energy generated from fossil fuels can be difficult and expensive, a challenge that renewable energy can attempt to correct by facilitating economic and social development in communities.

Fuel gas technologies, including for final application grinding of fuel, drying, gas cooling, gas cleaning and final utilization of the gas, could be proposed as a techno-economical viable alternative solution for different biomass and waste valorisation processes.

The research activity carried out in my PhD project was evaluating the technical feasibility of the gasification process and comparing the environmental, energetic and economic performances of the most promising design configurations for industrial applications of gasification-based waste- and biomass-to-energy generators. To this end, experimental tests with selected fuels of interest (wood biomass, mixed plastic waste and food packaging) were carried out in a pre-pilot and in a pilot scale bubbling fluidized bed gasifiers. Moreover, innovative gasification concepts and technologies were investigated, and applied in innovative scheme of waste management system.



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Fuel gas technology for biomass and waste - Environmental and techno-economic assessments

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What is essential is invisible to the eye...

...Only children know what they are looking for.

Abstract

Despite increasing attention for waste prevention and recycling, total municipal solid waste (MSW) generation has increased in EU25 from about 150 million tonnes in 1980 to more than 250 million tonnes in 2005 and is forecasted to reach 300 million tonnes by 2015. In addition, United States MSW generation has increased from about 1.1 tons per capita in 1990 to 1.3 tons in 2002, and worldwide it is projected to double by 2030.

Today, modern thermochemical plants provide a robust and efficient option for the treatment of wastes, ensuring a considerable reduction in mass and volume (as process residues), thereby preserving landfill spaces, and at the same time, allowing energy recovery. Moreover, due to the increasing of energy demand and environmental concerns, especially related with the use of fossil fuels, new solutions are continuously sought, also in the order to limit the greenhouse gas effect.

Energy is a vital input for social and economic development of a nation. Global population and energy needs are increasing day by day. It has been found that total world population rose from 6.08 billion in 2000 to 6.85 billion in 2010 and projected to reach 9.34 billion in 2050. Currently, fossil fuel based energy such as oil (40%), coal (23%) and natural gas (23%) are the major sources of energy for domestic, commercial and industrial activities, followed by nuclear (8%) and renewable (6%). This latter involves solar, wind, hydroelectric, biomass and geothermal power. World renewable sources of energy consumption are projected to increase from 3.577E+10 GJ in 1990 to 4.389E+10 GJ in 2000 to 5.492E+10GJ in 2010 and to 1.053E+11 GJ in 2035, at an average annual growth of 2.4%. On these bases, the European Union adopted a "20-20-20" Renewable Energy Directive, setting climate change reduction goals for the next decade. The targets call for a 20 percent reduction in greenhouse gas (GHG) emissions by 2020 compared with 1990 levels, a 20 percent cut in energy consumption through improved energy efficiency by 2020 and a 20 percent increase in the use of renewable energy by 2020, 50% of which by the use of biomass. This latter aspect, along with substantial economic incentives that member Countries established for the production of energy from biomass, have greatly stimulated the attention of the market and scientific community.

Biomass is a general term for material derived from growing plants or from animal manure, including algae, trees and crops. Although a lot of studies on biomass potential already available, their results vary widely, from close to zero to potential satisfying multiple times the world energy demand, because they are based on theoretical, technical, economic or implementation potential concepts. Wood biomass has great potential as relatively clean feedstock for producing modern

energy carriers, such as electricity and transportation fuels, because the energy in biomass from plant matter originally comes from solar energy through the photosynthesis process. This energy can be recovered by burning biomass as a fuel, releasing heat and carbon dioxide that was absorbed while the plant was growing. In order to compete with fossil energy sources an highly efficient utilization of biomass resources is desired. Therefore, in the last decade great R&D efforts were made to efficient conversion technologies to convert biomass into gaseous products, subsequently used for the production of heat and power energy and bio-fuels, such as ethanol, dimethyl ether, diesel. Currently, biomass gasification, is not more considered as one of the most promising thermochemical technologies but it could be proposed as a technological and economical viable alternative solution for different biomass valorisation processes. In this sense, biomass and biomass waste have been widely recognized as (a clean and) renewable energy sources because energy conversion systems based on their use reduce global CO2 emissions. Gasification, and in particular fluidized bed gasification, has been attracting worldwide attention due to its varied uses and benefits. Renewable energy can have a significant positive impact for developing countries. In rural areas, particularly in remote locations, transmission and distribution of energy generated from fossil fuels can be difficult and expensive, a challenge that renewable energy can attempt to correct by facilitating economic and social development in communities.

The aim of the research activity carried out in my PhD project was evaluating the technical feasibility of the gasification process and comparing the environmental, energetic and economic performances of the most promising design configurations for industrial applications of gasification-based waste- and biomass-to-energy generators. To this end, experiments with selected fuels of interest (wood- and agro-biomass, mixed plastic waste and food packaging) were carried out in a pre-pilot and in a pilot scale bubbling fluidized bed gasifiers. The experimental data were processed by mass and energy balances and an innovative and environmental friendly assessment tool such as the material and substance flow analyses. The aim was to assess and compare: 1) the fate of the main and trace elements such as carbon, cadmium, mercury, lead, zinc, etc. along different stages of the process under different values of operating parameters, and 2) some alternative design solutions. Particular attention was focused on the cleaning section to obtain a syngas with an energetically valuable heating value and a small amount of pollutants. It is infact recognized that removal of tar, fly ash, alkali compounds, ammonia is the true obstacle to the commercialization of the fluidized bed process. The environmental, energetic and economic

performances have been then estimated on the basis of the experimental data and manufacturer's specifications for small-scale plant capacity (100kWe – 6MWe).

Gasification-based plants, also in thermal configuration (i.e. first burning the fuel gas and then cleaning the flue gas), involve reduced environmental loads compared to those related to the combustion-based process because of the reducing reaction atmosphere. This aspect also implies lower exhaust gas rates compared to those from combustion plants, which must be operated with an air excess between 40 and 70%. Moreover, the substoichiometric oxygen flow rates fed in the gasification reactors promotes the partial oxidation of the carbon content of the fuel and, therefore, a low CO₂ emission. A further advantage is the operating reactor temperature lower than that typical of the combustion process, which in turn implies a lower concentration of trace elements such as cadmium, mercury and lead in the gas phase. The element partitioning of trace elements, and in a specific gasification case (direct melting system) seems to predict the direct recyclability of almost all the process solid residues. This innovative aspect is a turning point for the thermal treatments of waste as it allows for the first time to look at the thermochemical technologies mainly as material recovery process (and not only as energy recovery).

Finally, on the basis of the estimation of standard accounting items such as total plant costs, operating costs, taxation and direct revenues from the sale of the generated energy, an economic assessment compared for power production, combined heat and power and district heating solutions was carried out by a homemade economic model and a sensitivity analysis.

Keywords: gasification, fluidized bed, biomass, waste, waste management, element partitioning, techno-economic assessment

This PhD thesis is based on the results reported in the following scientific papers:

- Fluidized bed gasification of biomass: a substance flow analysis, Arena U., Di Gregorio F., Zaccariello L., Mastellone M.L., in Fluidization XIII-New Paradigm in Fluidization Engineering, S.D. Kim, Y. Kang, J.K. Lee, Y.C. Seo (Eds.). Engineering Foundation Conference International (ISBN 978-0-918902-57-3), pp. 805–812, 2010.
- Fluidized Bed Gasification of a Chicken Manure, U. Arena, F. Di Gregorio, L. Zaccariello, in Proceedings of 21th International Conference on Fluidized Bed Combustion, U. Arena, R. Chirone, M. Miccio, P. Salatino (Eds), pp. 821-828, ISBN 978-88-89677-83-4, Napoli, June 2012.
- A Comparison Between Fluidized Bed Combustion and Gasification of a Mixed Plastic Waste, U. Arena, R. Chirone, F. Di Gregorio, R. Solimene, M. Urciuolo, L. Zaccariello, in Proceedings of 21th International Conference on Fluidized Bed Combustion), U. Arena, R. Chirone, M. Miccio, P. Salatino (Eds), pp. 752-759, ISBN 978-88-89677-83-4, Napoli, June 2012.
- A Techno-Economic Comparison Between Two Design Configurations for a Small Scale Biomass-to-Energy Gasification Based System, U. Arena, F. Di Gregorio, M. Santonastasi, 2010; Chemical Engineering Journal, 142:180-190.
- 5. A Techno-Economic Comparison of Fluidized Bed Gasification of Two Mixed Plastic Wastes, U. Arena, F. Di Gregorio, C. Amorese, M.L. Mastellone, 2011; Waste Management, 31:1494-1504.
- Fluidized bed gasification of a packaging derived Fuel: Energetic, Environmental and Economic Performances Comparison for Waste-to-Energy Plants, F. Di Gregorio and L. Zaccariello, 2012; Energy, 42 (1):331-341.
- Element partitioning in combustion- and gasification-based waste-to-energy units, U. Arena and F. Di Gregorio, 2013; Waste Management, in press and available at http://dx.doi.org/10.1016/j.wasman.2013.01.035
- A waste management planning based on substance flow analysis, U. Arena and F. Di Gregorio, published in Proceedings of 1st International EIMPack Congress, Lisbona, 29-30 November 2012 and submitted for publication on Resource, Conservation & Recycling.

The following papers have been published but are not included in this thesis. Their content overlap

with the included papers or are out of the scope of this thesis.

- Fluidized bed gasification of a natural biomass: a process performance comparison of two design configurations, U. Arena, F. Di Gregorio, M.L. Mastellone, L. Zaccariello; published in Proceedings of XXXIII Meeting of the Italian Section of Combustion Institute, Ischia 2010.
- A techno-economic comparison of fluidized bed gasification of two mixed plastic wastes, U. Arena, F. Di Gregorio, C. Amorese, M.L. Mastellone, in Proceedings of Third International Symposium on Energy from Biomass and Waste, Venezia, November 2010.
- Bubbling Fluidized Bed Gasification of a Poultry Farm Waste, U. Arena, F. Di Gregorio, M. L. Mastellone, D. Santoro, L. Zaccariello; published in Proceedings of XXXIV Meeting of the Italian Section of Combustion Institute, Roma 2011.
- 4. A Substance Flow Analysis of a Combustion- and a Gasification-based Waste-to-Energy Units, F. Di Gregorio, N. Arena and U. Arena, published in Proceedings of SIDISA 2012 9° International symposium of Environmental Engineering, Milano, 26-29 June 2012.
- A comparison between combustion and gasification-based waste-to-energy systems, U. Arena and F. Di Gregorio, in Proceedings of Fourth International Symposium on Energy from Biomass and Waste, Venezia, November 2012.

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1. Fluidized bed gasification of biomass and waste

In recent years public and political sensitivities to environmental issues and necessity of reliable energy supply have led to the promotion of indigenous energy resources. In this perspective, renewable energy and energy from waste can play a key role, able to meet the future needs of more intense energy requirements by growing regional and world populations.

Development of gasification technology dates back to the end of the 18th century (Buekens et al., 1990). The first reported use of gasification happened in 1812 for the lighting of London. A gasification method designed by Bishoff in 1839, and modified by Siemens in 1857, was used for one century. Then, during both world wars, when oil was scarce in most common applications, thermal conversion processes utilizing mainly coal and biomass were stimulated. With the new expected of the depletion of oil wells and natural gas, along with the growth in costs of fossil fuels and the concerns with the emission of pollutant gases, in last decades research activities in gasification have been stimulated, in particular for an efficient exploitation of renewable sources and waste.

Gasification process is the thermochemical conversion of carbonaceous material into fuel gas through partial oxidation at elevated temperatures. The resulting energetic gas, called syngas or producer gas, is a mixture intentionally rich in carbon monoxide (CO) and hydrogen (H₂), and affected by varying amounts of methane (CH₄), carbon dioxide (CO₂), steam (H₂O), small quantities of light hydrocarbons (C_nH_m), besides nitrogen (N₂) if air is supplied as oxidizing medium. The syngas composition is related to operating conditions, such as temperature, equivalent ratio and pressure.

Ideally the process produces only a non-condensable fuel gas and ash residues. In the practice industrial plants produce a gas containing varying levels of contaminants, such as solid particulate, condensable tars, alkali metals, chlorine, nitrogen and sulfur compounds and an ash residue containing some char. Syngas composition and its contamination level vary depending on feedstock, and reactor type as well as values of the key operating parameters.

Gasifiers have been designed in various configurations, typically classified in entrained bed, fluidized bed, and fixed or moving bed according to how the gas and fuel contact each other. Although many gasification have been developed commercially, fluidized bed has several advantages over that in fixed/moving bed or entrained flow. In a fluidized bed gasifier, granular solid such as sand, fuel and gasifying medium are mixed in a hot bed that can be bubbling (BFBG)

or circulated (CFBG). Granular bed works as a heat exchanger so the heat and the mass transfer from the fuel is improved and throughput and syngas heating value are higher. Fluidized bed provides high mixing and reaction rates, and then accommodates variation in fuel quality and allows scaling-up of the process (Higman and van der Burgt, 2008). Fluidized bed gasifiers are more tolerant with the fuel particle quality and function at lower temperature (800-900°C) respect to other reactors, making it ideal for the processing of biomass and waste (Gomez-Barea, 2012). Moreover, with reference to biomass gasification only fluidized bed configurations are being considered in applications that generate over 1MWe (Overend et al., 1993; Palonen et al., 1995) both for operation at atmospheric or elevated pressures.

	Gas	compos	ition, %	%v∕v dr	HHV, MJ/Nm³	Gas quality		
	H_2	CO	CO_2	CH_4	N_2		Tars	Dust
Fluid bed air-blown	9	14	20	7	50	5.4	Fair	Poor
Updraft, air-blown	11	24	9	3	53	5.5	Poor	Good
Downdraft, air-blown	17	21	13	1	48	5.7	Good	Fair
Downdraft, oxygen-blown	32	48	15	2	3	10.4	Good	Good
Multi-solid fluid bed	15	47	15	23	0	16.1	Fair	Poor
Twin fluidised bed gasification	31	48	0	21	0	17.4	Fair	Poor



Table 1. Typical syngas composition and quality for different gasification technologies (Bridgwater, 2002)

Figure 1. The main gasifier configurations for biomass gasification process

Based on new concepts, in recent years innovative process and reactor designs were developed, such as dual (Kern et al., 2012) and three stage (Gomez-Barea, 2012) FBG, with the aim to improve the process efficiency and to obtain a higher syngas purity. Although they have generated

promising results in pilot scale reactors, at the time still expects the respective validation on commercial-scale plants.



Figure 2. Conceptual performance diagrams of the three stage gasifier (left) and dual fluidized bed gasifier (right)

Gasification enables the use of cheap, potentially CO₂-neutral (biomass) or low grade fuels (waste), that are usually available locally, and long distance transportation can be avoided. By converting them into fuel gas, these fuels can be exploited with much better efficiency than with direct combustion.

Energy	Net Electrical	Required level of syngas cleaning									
Conversion Device	Efficiency of Gasification Plant	Tar	Dust	Alkali (Na, K)	Heavy Metals	H₂S					
Steam Turbine	15-24%	not limited	not limited	not limited	not limited	not limited					
Organic Rankine Cycle	15-18%	not limited	not limited	not limited	not limited	not limited					
Gas Engine	13-28%	< 50 mg/m³ _N	< 50 mg/m³ _N	< 0.025-0.1 ppm,wb	< 0.1 ppm,wb	< 20 ppm,vb					
Gas Turbine	20-30%	< 10 mg/m ³ _N	< 10 mg/m ³ _N	< 0.025-0.1 mg/m ³ _N	< 0.1 ppm,wb	< 20 ppm,vb					

Table 2. Required level of syngas cleaning for the main energy generation devices. wb: weight basis; vb: volume basis.

Electricity generation is considered the most lucrative opportunity for commercial exploitation of biomass and waste, by virtue of the high value of electricity and incentive schemes, but

gasification application offers more opportunities. There are ongoing projects seeking sustainable gasification systems for the generation of several forms of energy. Modern day gasification units are mostly based on integrated gasification combined cycle (IGCC) and produce electricity along with hydrogen/methanol/Fischer-Tropsch liquid fuels/chemicals/synthetic natural gas or a combination of these. This concept, called polygeneration, is infusing high research interest as it reduces the emissions as well as improves the plant economy (Mondal et al., 2011). However, the requirements of syngas purity differ widely for different downstream applications. In this perspective, power generation requires a lower syngas quality than other applications.

1.1 Experimental activity on pre-pilot and pilot scale FBGs

Fluidization: art or knowledge?

When a new commercial-scale physical or chemical process is planned, proper selection of a contacting mode is crucial. Fluidization is an operation by which solid particles behave as a fluid through contact with a gas (Basu, 2006; Grace et al., 1997). If a fluid is passed upward through a bed of fine particles, when the frictional forces between particle and fluid just counterbalances the weight of the bed particles, the vertical component of the compressive force between adjacent particles disappears and the pressure drop through any section of the bed about equals the weight of fluid and particles in this section. At this moment, the bed is considered to be just fluidized and is referred to as a bed at minimum fluidization. With an increase in flow rate large instabilities with bubbling and channeling of gas are observed but the bed does not expand much beyond its volume at minimum fluidization. It become a bubbling fluidized bed and is considered a densephase as long as there is a fairly clearly defined upper limit or surface to the bed. In gas-solid systems, gas bubbles coalesce and grow as the rise and they may eventually become large enough to spread across the vessel and flow smoothly down by the wall around the rising void of the gas or the portion of the bed above the bubbles is pushed upward and particles rain down from the slug, which finally disintegrates. When fine particles are fluidized at a sufficiently high gas flow rate, the terminal velocity of the solids is exceeded, the upper surface of the bed disappears, entrainment becomes appreciable, and, instead of bubbles, a solid turbulent motion of solid clusters and void of gas of various size and shape can be observed. Compared to other methods of gas-solid contacting, such as fixed bed, moving bed and rotary cylinders, fluidized beds have some

rather unusual and useful properties. A dense-phase gas fluidized bed, infact looks very much like a boiling liquid and many ways exhibits liquidlike behavior.



Figure 3. Fluidization regimes increasing gas velocity (redrawn from PSRI, 2010).

The main advantages for industrial applications of fluidized beds are related to: i) the isothermal conditions throughout the reactor, guaranteed by the rapid mixing of solids, ii) the excellent heat and mass transfer rates between gas and particles and between the bed particles and an immersed material, iii) the suitability for both small and large-scale operations. Anyway, it should be taken into account possible operating troubles related to the erosion of pipes and vessel from abrasion by particle, agglomeration and sintering phenomena of the bed, elutriation of pulverized solid bed particles, and nonuniform residence time of solids in the reactor.

Businessmen and process engineers feel that fluidization is an interesting operation but is a choice affected by high risk because it is still too much of an art requiring practical experience and knowhow, and because too much uncertainty is involved, particularly in scale-up, at which stage the cost of failure is serious. Infact, the history of fluidized bed plants is done of a series of commercial disasters, and only large companies can absorb the cost of possible failure. For sure the design of fluidized bed processes is often more complex than other modes of contacting, but not always. In any case, when technical and economic considerations both point strongly to the fluidized bed, then one must put up with possible difficulties and complications, although in the last 30 years significantly progress was done and know-how grown constantly and pervasively to reduce uncertainties and design fluidized beds successfully. Interest in renewable sources as biomass and low grade fuels as waste, together with the world need to aim to high energy efficiencies process, appear to be at the moment the renewed drive to the invest in fluidization.

Description of pilot and pre-pilot BFB gasifiers



Figure 4. Sketch of the pre-pilot bubbling fluidized bed gasifier and syngas conditioning lines.

The pre-pilot reactor is an atmospheric bubbling fluidized bed gasifier (BFBG) with a feeding capacity of approximately 3kg/h. The BFBG is a 102mmID cylindrical column, made of AISI 316L and electrically heated by five shell furnaces, each capable of a maximum power of 3.5kW. All the heating elements are controlled by a data acquisition system connected to five thermocouples, located in the reactor internal wall, which allow to independently set the temperature of each reactor section (blast feeding, air pre-heater, bed and freeboard). The air utilized as fluidizing agent was injected at the bed bottom through a distributor plate composed of three nozzles. These have a truncate pyramidal shape and were specifically designed in order to ensure an homogeneous distributor plate to the syngas outlet is equal to 2.5 meters. The feedstock was continuously over-bed fed by means of a screw-feeder device and a nitrogen flow was used to help the fuel feeding and, at the same time, to avoid the back flow of the produced gas. At the

reactor outlet, the syngas is addressed to an high efficiency cyclone for dust removal. Downstream of it there are two alternative symmetric syngas conditioning lines, each one consisting of a bubbler and filters for tar, residual fly ashes, and acid and basic gases. Syngas composition, in terms of CO, O₂ and CO₂, was on-line measured by IR analyzers to monitor the process evolution. The main syngas compounds (CO, H₂, CH₄, CO₂, N₂, O₂, as well as C_nH_m and BTX) were measured by using an Agilent Micro-GC 3000 located downstream of the tar sampling line. The syngas is further sampled by means of tedlar bags in other two points along the reactor height (0.9m and 1.8m) and then off-line analyzed.



Figure 5. Flow-sheet of the pilot scale bubbling fluidized bed gasifier and syngas cleaning section.

The pilot scale gasifier is a bubbling fluidized bed with an ID of 381 mm and a nominal capacity of 500 kW, related to a maximum feeding capacity of 100kg/h depending on the fuel heating value . The experimental plant runs was carried out injecting air at the bed bottom as reducing agent while an over-bed feeding system was used for fuel. A couple of electric heaters heated up to the desired temperature the fluidizing agent stream before entering the reactor. Fixing the fluidizing velocity and adjusting both fuel and blast flow rates, the desired equivalence ratio (ER) was obtained (i.e. the ratio between the oxygen amount of air supply and that required for the stoichiometric fuel complete combustion). The BFB gasifier was heated up to the reaction temperature by the sensible heat of pre-heated blast gases and by a set of three external electrical furnaces, then turned off to allow operating autothermic conditions. The synthesis gas produced in the reactor was first sent to a high efficiency cyclone to remove fly ashes and then to a wet scrubber to segregate tars, acid gases and residuals. Finally the syngas was incinerated by a safety

flare. Syngas composition (in terms of CO, CO_2 , H_2 and CH_4) upstream and downstream conditioning section was on-line measured by IR analyzers and by micro-gas-chromatographs equipped with different columns for the detection of light hydrocarbons, up to BTX, as well as of carbon monoxide and dioxide, hydrogen and nitrogen.

To optimize the gasification process and produce a considerably clean raw gas, not eliminating the need of downstream hot gas cleaning, for all the experimental runs on both reactors, olivine, an iron and magnesium silicate, was selected as material for the fluidized bed on the basis of results of scientific investigations reported in literature. Considering the demonstrated activity and selectivity in terms of tar reduction, the economical availability and the excellent attrition resistance in the fluidized bed reactor, olivine seems to be a promising candidate to act as a bed catalyst for the tar cracking reactions (i.e. as "primary method") in waste derived fuel gasification. Gas and solids sampling procedures were activated just when the values of pressure, temperature and gas composition were at steady state conditions for not less than 1h. For the sampling of condensable species, a system consisted of four in-series cooling coils plugged in an ice bath, a suction pump and a flow meter, was installed and operated with a syngas flow rate of about $3 dm_N^3/min$ for 30min to obtain tar and water phase. Water was separated from tar in order to evaluate its content in the syngas and the condensed hydrocarbons were off-line analyzed, with a specific pre-treatment, in a Perkin-Elmer Clarus 500 gas chromatograph coupled with a mass spectrometer (GC-MS). This procedure allows to recognize tar belonging to the classes between 2 and 5 of the classification system proposed by ECN (Van Paasen and Kiel, 2004). Hydrogen chloride, hydrogen sulfide and ammonia were collected by bubbling the product gas through a pair of gas stripping bubblers, connected in series and containing basic and acid solutions respectively, and subsequently analysed by means of a Dionex DX-120 ion chromatograph.

Feedstock

Recognizing the limitations of many current power production technologies, in terms of resource potential, greenhouse gas savings and economic viability, there is considerable interest in fuel gas technologies. These offer the potential for a wider range of feedstocks to be used, lower greenhouse gas impacts, and lower costs.

There are a large number of different feedstock types for use in a gasifier, especially in the case of heterogeneous fuels as biomass and wastes, each having different characteristics, in terms of size, shape, bulk density, moisture, energy content, chemical composition, ash melting and

composition, and homogeneity of all these properties. Feedstock properties often form the basis for the technology chosen for an identified process. Depending on these properties, a fuel can be excluded for specific process, partially for technical or environmental reasons. For these reasons, it is necessary a preliminary knowledge of proximate and ultimate analysis, heating value and ash melting point.

Feedstock moisture contents above 30% result in lower gasification thermal efficiency, as energy is needed to evaporate the water, with the resulting steam also affecting the gas composition. Higher moisture contents (30-70%) also reduce the temperatures that are achieved, increasing the proportion of syngas tars in the syngas due to the incomplete cracking reactions. However, drying feedstocks to less than 10% requires ever increasing energy inputs, and hence moisture contents in the 10-20% range are preferred. This heat can be provided externally, or extracted from the gasifier syngas or other plant process steps (i.e. partial recycling of exhaust gases in a thermal configuration).

Ash is the feedstock mineral content which cannot be gasified. It ranges from less than 1% in wood to above 30-40% in some animal manures and herbaceous crops. Low ash content feedstocks are usually preferred to minimize disposal streams and to ensure high heating value of the fuel. Ash composition is a crucial aspect, since feedstocks with low ash melting point can be difficult to gasify in some reactors. In particular in fluidized beds, ash alkali species can play a critical role during thermal conversion (Khan et al., 2009; Giuntoli et al., 2009; Gatternig et al., 2011; Yrjas et al., 2012). These elements appear to be responsible of undesired phenomena in a bubbling fluidized bed such as coating formation and the consequent sintering and bridging between bed particles (Font-Palma, 2012). These in turn could imply agglomerate and deposit formation and, in some cases, worsening of the fluidization quality until to a definitive defluidization and unscheduled shutdown of the plant. Typically eutectics of low boiling point are created by absorption and chemical interaction between silica and potassium, dispersed in biomass in ionic and organometallic forms, which tends to stay in the bed by recapture of vapors by mineral components in the bottom ash under fluidized bed conditions. In general, this constraint induces to run the gasifier at lower temperature (typical lower than 700°C), reducing kinetic reactions and tar cracking. Whilst woody biomass feedstocks usually meet the ash requirements, farm manure and agricultural residues may have to be first treated for their ash melting characteristics. Catalytic bed additives, such as kaolin, dolomite limestone or alumina, can be used to prevent sand bed

agglomeration. Moreover, an efficient dust removal system is always necessary to handle particulate emissions (for example leaching).

The size of the feedstocks into the gasifier can have a large influence on the gasification performance. The requires sizing is mainly a function of feeding rate, residence time, tar production, temperature and gasifier efficiency, which need evaluation for each feedstock. For example, smaller particles have a larger surface area to volume ratio, and the gasification reactions occur faster. Crude sizing operations include chipping, cutting, and chopping, but in order to get small ground particles, specific equipments is needed, and this is generally an energy intensive process. Moreover a screening process is often used to ensure any remaining larger particles and extraneous materials are removed. In general, it is desirable feedstocks fairly uniform in size, shape and density to prevent non steady state chemical conditions and troubles to the feeding system. Biomass preparation, such as drying and/or sizing is needed to some extent for most combinations of feedstock and gasifier type. Some gasifier type and feedstock require more pre-treatment, in the form of additional biomass conversion step, to make biomass suitable for use.

Of all the physical properties, the bulk density of the fuel is of major importance because of economical and technical reasons. Most biofuels, especially from agricultural origin are characterized by a low bulk densities (50-150kg/m³). A number of disadvantages are attributed to the lower bulk density of biomass including relatively low heating value per unit volume, process control difficulties, feeding control, requirement of huge storage, expensive transportation, and limitation of technologies applied. Densification is a well known process utilized to overcome these disadvantages, removing the inter and intra-particles voids. Briquetting and pelletization (up to 450-650kg/m³) are the most commonly used densification techniques, but require energy and costs. In general, woody biomass are typically collected and stored directly as chips and not need pelletization or briquetting to provide the desirable bulk density (> 300kg/m³) and avoid feeding problems.

Definitively, plant economics can be greatly improved through the use of lower cost feedstock, however pre-treatments add to costs and energy requirements, which must be compared with those of using alternative feedstocks.

Scope

Thermal processing of biomass and waste has the potential to offer a major contribution in meeting the increasing demand of the bio-energy and renewable energy sectors as well as the

targets set by the EC and member Countries for global warming mitigation. Biomass and waste fluidized bed gasification are considered one of the most promising routes for syngas or combined heat and power production because of the potential for higher efficiency cycles. Instead of utilizing biomass and waste in traditional low-efficient steam cycles, high-efficient gas engines or gas turbines can be applied.

Theory

During fluidized bed gasification generally air or steam, respectively for heat and power or hydrogen production, but also oxygen, carbon dioxide or a mixture of two of more of these, can be used as fluidizing agent through a bed generally made of quartz sand, dolomite or olivine. In the partial oxidation process, the gasifying agent is chosen and its ratio to carbonaceous feedstock (called equivalence ratio, ER) is adjusted accordingly to meet the desired chemical composition of syngas and efficiency of the process. ER is important in designing the parameters of a gasifier because a higher equivalence ratio provides lower yields of hydrogen, syngas and gas low heating value. This is because reactions with higher ER favor a more complete conversion (CO₂ and H₂O instead of CO and H₂, respectively). Typical values of ER for biomass and waste fluidized bed gasification vary between 0.2 and 0.5. The wide variation in syngas yield and composition is directly correlated to the variation in the feedstock composition, as well as on the type of gasifier and the operating conditions used (temperature, equivalence ratio, residence time, fuel size, etc). Describing the whole process of gasification is very complex and cannot be generalized for different feedstocks. However the phenomenon of pyrolisis followed by volatilization of the remaining carbon is predominant. Major reactions (reported in Table 3) involved in the gasification process are combustion (reactions 1-5, with O_2), steam gasification (reactions 6-9, with H_2O), hydrogenation (reactions 10-11, with H_2), Boudouard reaction (reaction 13, with CO_2). Moreover, after entering into the gasifier, a feedstock gets devolatilized and produces hydrocarbons and char. The hydrocarbons thus react to give carbon monoxide and hydrogen as per the following generalized reaction (reactions 14-15, PAH and tar decomposition):

1	$C + \frac{1}{2}O_2 \rightarrow CO$	-111 kJ/mol
2	$CO + \frac{1}{2}O_2 \rightarrow CO_2$	-283 kJ/mol
2	$C + O_2 \rightarrow CO_2$	-394 kJ/mol

4	$H_2 + \frac{1}{2} O_2 \rightarrow H_2 O$	-242 kJ/mol
5	$C_nH_m + n/2 O_2 \leftrightarrow n CO + n/2 H_2$	exothermic
6	$C + H_2O \leftrightarrow CO + H_2$	+ 131 kJ/mol
7	$CO + H_2O \leftrightarrow CO_2 + H_2$	- 41 kJ/mol
8	$CH_4 + H_2O \leftrightarrow CO + 3H_2$	+206 kJ/mol
9	$C_nH_m + n H_2O \leftrightarrow nCO + (n + m/2) H_2$	endothermic
10	$C + 2H_2 \leftrightarrow CH_4$	- 75 kJ/mol
11	$CO + 3H_2 \leftrightarrow CH_4 + H_2O$	- 227 kJ/mol
12	$C + CO_2 \leftrightarrow 2CO$	+172 kJ/mol
13	$C_nH_m + nCO_2 \leftrightarrow 2nCO + m/2 H_2$	endothermic
14	$pC_{x}H_{y} \rightarrow qC_{n}H_{m} + rH_{2}$	endothermic
15	$C_nH_m \rightarrow nC + m/2 H_2$	endothermic

Table 3. Reactions involved in the gasification process

The gasification reactions are mainly endothermic (reactions 6 and 12) wherefore heat has to be supplied to the reactor. This can be done by partial combustion of the feedstock (autothermic gasification) or by an external source (allothermic gasification) by means of a heat exchanger or a heat carrier (dual fluidized bed gasifier). Autothermic gasification reactors mostly use air as gasification medium because pure oxygen is economic feasible only in large scale installations. The gained product gas is therefore diluted with nitrogen and for biomass because of the low heating value (3–6.5 MJ/Nm³) it is called lean gas. Biomass and plastic wastes contain considerable amounts of carbon and hydrogen (in a low C/H ratio) and a large amount of volatile matter (>70%). Then, they are highly reactive and their gasification takes place at low temperature (800-900°C).

Raw syngas also contains different pollutants as tar, solid particulate, alkali metals and nitrogen/chlorine/sulfur compounds. Tars are undesired heavy condensable organic compounds and represent the real obstacle to the diffuse commercialization of the fluidized bed technology. If tars condense on cool surfaces of the cleanup section or into the end-of-use device, severe operating problems can result. On the other hand, tars constitute such an important energy component of the fuel gas, then removing them from the gas could result in a loss of system energetic efficiency. Therefore is preferable use tar cracking technology (thermal or catalytic) to

broke it into smaller hydrocarbon molecules that are lighter and stable. Tar yield in the syngas is above all function of combination of equivalence ratio and temperature. Higher gasifier temperature provides a high gas flow and increases the yield of hydrogen and syngas, gives high cold gas efficiency and high carbon conversion. But if the temperature is above the ash melting temperature, clinker or ash agglomerate takes place and as a result gasifier life reduces. Whilst particulate and alkali rates are directly correlated to the natural presence in the feedstock, nitrogen and sulfur compounds are also related to the reactor environment. In particular, due to the gasification substoichiometric conditions, S is converted principally in H₂S, along with methanation and shift reaction, a small amount of COS is also formed, and N, also due to the low temperature, is generally converted in NH₃ instead of environmental hazardous NO_x.



Figure 6. Summary of the temperature effect on the key output parameters during gasification.

It was seen that raising the reactor temperature increases tar and char conversion. However, the risk for sintering of ash and bed material also increases and sets the maximum temperature. Considering the balance between benefits and drawbacks associated with the thermal level, the temperature range of a waste and biomass gasifier is between 800 to 900 °C. Even at the highest acceptable temperature, it is difficult to convert PAH into gas via steam reforming by non-catalytic reactions or by contact with bed material. Infact, mass transport resistances provoked by fluid-dynamic effects may limit the effective contact of tar and catalyst.

Experimental runs

The pre-pilot and pilot BFBGs were fired with the following commercially available waste and biomass fuels:

<u>Wood Biomass (WB)</u>, a natural wood pellets mainly made of cellulose, hemicelluloses and lignin, chosen as the traditionally biomass utilized for domestic heating;

<u>Chicken Manure (CM)</u>, typical low grade bio-fuel, chosen because produced in large amount worldwide due to the increase of meat demand and that, by the introduction of specific legislation, cannot be yet dispose off directly on agricultural soil;

<u>Mixed Plastic Waste (MPW)</u>, mainly made of polyolefin (polypropylene and polystyrene), therefore characterized by high heating value, chosen because is a process residues of the sorting process of the household separate collection of plastic solid waste (about 35–40% of the total plastic waste collection);

<u>Packaging Derived Fuel (PDF)</u>, mainly made of plastic polymers such as polyethylene, polypropylene, polystyrene, polycarbonate, polyamide and polyethylene terephthalate, chosen because pervasive supplied most of common food packaging materials.

For all the experimental tests with these different feedstocks, quartz sand and olivine were selected as materials for the fluidized bed on the basis of results of previous investigations carried out on the same pilot-scale BFBG and those reported on the scientific literature. All indicated olivine as an interesting candidate to act as a bed catalyst for the tar cracking reactions during gasification process, even taking into account its low cost and excellent resistance to attrition in the fluidized bed reactor. Moreover in all the reported experiments, air was used as fluidizing/reducing agent and always injected at the bed particles bottom while the fuels were always fed by means of an over-bed system. The fluidizing air stream was heated up to the desired temperature by a couple of electric heaters before entering the reactor. The fuel and blast flow rates were mutually adjusted so that, at the fixed fluidizing velocity, the desired equivalence ratio ER was obtained. In the start up phase the cylindrical BFBG was heated up to the reaction temperature by the sensible heat of pre-heated blast gases and by a set of three external electrical furnaces, then turned off to attend to an autothermic process.

Material and Substance Flow Analysis

The proposed studies combine a series of experimental results obtained from the pre-pilot and pilot scale bubbling fluidized bed gasifiers fed with plastic wastes and natural biomass, together with a recently defined environmental assessment tool, the Material Flow Analysis (MFA), which is named Substance Flow Analysis (SFA) when it is referred to a specific chemical species. MFA/SFA is a systematic assessment of the flows and stocks of materials and elements within a system

defined in space and time. It connects the sources, the pathways, and the intermediate and final sinks of each species in a specific process. These characteristics make MFA/SFA attractive as a decision support tool, as showed by its utilization in process evaluation of waste treatments and recycling options and in waste management planning (Rotter et al., 2004). In these studies MFA/SFA was applied to the bubbling fluidized bed gasification process to deeper understand the performance of the whole pilot plant as well as that of its specific components (reactor, cyclone, wet scrubber) and to define and quantitatively assess some design solutions and operating criteria of the biomass and waste gasification systems. Moreover, SFA is a prerequisite for environmental impact statements and forms the data base for every Life Cycle Assessment (LCA) and Risk Assessment (RA). The Material and Substance Flow Analysis was developed by an idea conceived by Prof. Paul Helmut Brunner of Technology University of Vienna (TUV) for environmental assessments on waste treatment and management. He thought that a comprehensive and systemic approach was needed to design sustainable waste management systems because of the complexity of the matter, requiring knowledge on system behavior. A goal-oriented approach helps to resolve the issue and because goals (environmental and health protection, resource conservation) are substance-oriented, evaluation methods must be based on analysis of substance flows and stocks, too. Today in Austria there is a Standard for the application of MFA in waste management, including terms and definitions for SFA. On the basis of this standard, the freeware STAN was developed at TUV, which allows balancing and presentation of material and substance flows through waste management systems. Companies applying for certification are now required to use STAN to display their material turnover.

Results and discussion

This section presents and summaries a number of typical experimental results with the aim of providing an overview of the most promising findings of the studies. The presentation is not limited to the original content of the related papers. If additional data and explanations exist that were not available at the time of publication, they have been added.

Feedstocks used in bubbling fluidized bed reactors were biomass and wastes. Here, for the sake of comparison, proximate and ultimate analyses, correlated with heating values (Table 4), and chemical composition of inorganic fractions (Table 5) for all the fuel types are reported.

Biomass and waste feedstocks differ in many important ways each other, including the organic, inorganic and energy content, besides physical properties. Relatively to wastes and waste-derived-

fuels, biomass has generally less carbon and hydrogen content, more oxygen, and lower but anyway high, volatile component, especially in the case of agricultural residues. Due to the high volatile content, feedstocks are easier to ignite even at low temperature and gasification is expected to occur at a rapid rate and therefore has to be controller accordingly. The amount of fuel devolatilized during the pyrolisis stage of gasification increases with increasing hydrogen to carbon ratio and, to a lesser extent, with increasing oxygen to carbon ratio. The quick release of a larger fraction of fuels as volatilizes makes it necessary to have longer high temperature zones in order to achieve complete gasification at high process efficiency and to ensure low pollutant emissions.

	WB	CM1	CM2	MPW	PDF
Ultimate analysis, % on weight basis					
С	45,9	33,0	30,7	79,5	53,9
н	5,6	4,4	4,2	13,1	7,7
N	0,3	5,6	3,2	0,3	0,5
S	0,01	0,3	0,2	0,2	0,1
CI	0,04	0,5	0,4	0,5	0,3
Moisture	7,0	9,9	11,0	0,7	5,6
Ash	1,3	17,2	25,1	1,9	5,9
0	39,9	29,1	25,2	3,8	26,0
C:H ratio	8,2	7,5	7,3	6,1	7,0
Proximate analysis, % on weight basis					
Moisture	7,0	9,9	11,0	0,7	5,6
Volatile matter	72,0	57,2	50,2	97,2	87,9
Fixed carbon	19,7	15,7	13,7	0,2	0,6
Ash	1,3	17,2	25,1	1,9	5,9
Heating value ^a					
HHV, kJ/kg	18.600	14.590	13.670	43.400	26.500
LHV, kJ/kg	15.900	11.940	10.980	40.200	23.200

^a calculated by the relationship of Sheng and Azevedo for WB and CM and by the relationship of Channiwala and Parikh for MPW and PDF.

Table 4. Proximate and ultimate analyses and heating values of biomass and wastes used in the gasification tests.

These implication could affect fluidized bed gasifier design and operation principles. For example for plastic waste feedstocks, the distance between top of the bed and the fuel feeding point must be reduced respect to typical value to avoid that a large part of the fuel vaporize directly into the freeboard. Since the biomass moisture content can reach very high value, influencing the gasification efficiency and the volume of syngas produced for energy unit and decreasing the

syngas heating value, all bio-feedstocks utilized in the experimental activity have values of about 10%. On the other side, plastic wastes, and in particular MPW, show very low moisture value (less than 1%). Moreover, biomass and plastic waste greatly differ in heating value, being the latter characterized by a valuable energy content, similar to that of utilized polymers and RDFs, and not so less to that of common oils. Some biomass, as chicken manure, show very high ash content values (17-25%). Inorganic material of biomass can be divided into two fractions, one inherent to the fuel and the other added to the fuel through processing steps. The latter, adventitious material, originates from skidding and other operations whereby soil is incorporated into the fuel. This dirt often makes up major fraction of the ash content of bio-fuels. It is typically different from that of inherent materials, for both composition and mode of occurrence of the elements. For example for chicken manure, calcium and sodium from the incorporation of sands, clays and other soil particles, and potassium incorporated in feldspars with relatively little contribution to alkali reactions, lead to fouling other than by inertial impaction and sticking of particles. Table 4 shows the ash analysis of the selected biomass and waste feedstocks.

mg/kg _{db}	WB	CM1	CM2	MPW	PDF
Al	150	211	345	1.040	2.620
Ar	<0,1	0,1	0,2	<0,1	0,1
Ca	1.750	57.930	93.200	10.070	22.875
Cd	0,05	0,5	0,2	0,5	0,25
Со	0,2	1,4	1,4	1,1	0,7
Cr	1,5	7,2	9,1	10,2	6,1
Cu	3,0	37,3	33,5	17,3	18,7
Fe	290	418	436	305	140
Hg	<0,1	0,2	0,1	<0,1	0,1
Mn	15	213	207	6,5	7,1
Na	110	3.372	4.350	529	910
Ni	0,5	2,2	3,3	1,1	6,7
Pb	1,2	0,1	0,2	33,8	82
К	330	16.780	14.740	1.480	2.074
Sb	2	0,1	0,04	<0,1	2
Sn	150	0,1	0,05	16,8	7,1
V	<0,1	2,1	2,9	0,8	2

Table 5. Chemical composition of inorganic fraction of biomass and waste feedstocks used in the gasification tests

The ash content varies from one fuel to another (from 1% to 25%), as well as its composition. With high ash containing fuel, an efficient dust removal system becomes a must to handle particulate

emission and to perform better heating value of the fuel. Moreover, high alkali (potassium) and silica content typically gives low ash melting temperatures. Although a major fraction of low melting point alkali is released into the gas phase, the part left in the ash may deliver detrimental effects during fluidized bed gasification, as bed agglomeration or even bed defluidization. As cited above (see "Experimental Runs" paragraph) all the tests were carried out on pre-pilot and pilot airblown gasifiers, utilizing a quartz sand or olivine as bed material, with the aim to obtain an energetic syngas, particularly enriched in hydrogen and carbon monoxide, and more free as possible of pollutants, particularly of tars. Table 6 provides a synthesis of the range of the main operating parameters, especially bed temperature, equivalence ratio (ER), air to fuel ratio (AF), and fluidization velocity (v_f), utilized for all the experimental runs with biomass and waste feedstocks.

	WB	CM	MPW	PDF
ER, -	0,17 - 0,30	0,25 - 0,40	0,20 - 0,26	0,24 - 0,31
AF ratio, -	0,9 – 1,5	1,2 – 1,5	2,8 – 3,2	2,0 – 2,4
v _{f,} m/s	0,6	0,4	0,7	0,7
T _{air} , °C	545	300 – 575	450	550
T _{bed,} °C	810 - 880	700 - 800	810 - 860	845 – 850
bed material	olivine/ guartz sand	quartz sand	Olivine	olivine

Table 6. Operating	conditions	of	the	pre-pilot	and	pilot	scale
gasifier feeding diffe	erent biomas	s a	nd w	aste feeds	tocks	5.	

For each test in the pre-pilot and pilot plants a new batch of bed material was used. Typically, the pre-pilot scale reactor was utilized to obtain preliminary indications on fuel thermal behavior in gasification environment and to select a more narrow range of the main operating parameters. The intense utilization of calcined olivine as an active bed material was suggested by its good performance in fluidized bed gasifiers of biomass (Devi et al., 2005) and plastic wastes (Arena et al., 2009), and because is perceived as a natural catalyst. Specific studies about the catalytic role of olivine during the gasification tests indicated that magnesium and iron, both largely present in olivine particles, activate the endothermic decomposition reactions of hydrocarbon fragments that are the precursors in tar formation, at the same time improving the syngas energetic content. In particular, magnesium catalytically enhances the dehydrogenation and isomerization reactions of fragments produced by thermal cracking ($pC_xH_y = qC_nH_m + rH_2$), whereas iron catalytically assists the dehydrogenation and carbonization reactions ($C_nH_m = nC + m/2H_2$). Temperature and

equivalence ratio, for each specific gasifier, are major parameters to obtain acceptable syngas quality level. A high reaction temperature provides high gas flow and increases the yield of syngas. Gasification process involves raising the syngas temperature typically above 1000°C, to crack tars into lighter species. Morf et al. in 2002 observed that CO and CH₄ concentrations increase linearly between 700°C and 1000°C, whereas H_2 concentration increase exponentially, equaling CO concentration at 1000°C. Thermal tar destruction increases the yields of PAH and soot (Milne et al., 1998). Sooting incurs a reduction in producer gas carbon content, hence in heating value. In addition, thermal tar cracking reactors operate at high temperatures, thus have to be constructed with expensive materials that resist at high temperature. Lastly, thermal tar conversion is an energy penalty to processes utilizing fluidized bed gasifiers operated at lower temperature, typically between 800°C and 900°C. In addition, feedstocks characterized by a low ash melting temperature require further low bed temperature to prevent agglomeration phenomenon or defluidization. A higher ER provides lower yields of hydrogen, syngas, and low heating value. This is because a reaction with a higher equivalence ratio favors complete combustion. Typical values of ER for biomass and waste gasification vary between 0.2 and 0.4. An intensive experimental activity on the matter is present in literature both for biomass and waste gasification, but different trends in syngas composition and pollution control were observed for specific feedstock and gasifier design. Another parameter that affects the quality of syngas is superficial velocity (v_s) , defined as gas flow rate (v_f) divided by the internal cross section of the gasifier. Low value of v_s lead to a slow pyrolysis process with high yields of char and significant amounts of unburned tars. Although high values of v_s promote low char formation, such high values result in lower residence time inside the gasifier, reducing the efficiency in the tar cracking processes. Table 7 reports a synthesis of the best performance results of gasification tests for different bio- and waste-fuel utilized, as function of gasifier operating conditions.

	ER	т	Y_{syngas}	LHV _{syngas}	E _{syngas}	CGE	CCE	CO %	H ₂	CH4 %	CO ₂	C ₂ H _m %	C _n H _m %	N ₂
			III N/ NBfuel	NJ/III N	K VVII/ KBtuel			70	70	70	70	70	70	70
WB	0,28	880	2,1	5900	3,4	0,77	0,96	17,9	12,3	3,9	14,0	0,92	0,28	50,7
СМ	0,34	750	1,7	4360	2,1	0,63	0,92	9,6	9,7	2,8	18,22	1,37	0,18	58,13
MPW	0,24	890	3,4	9400	8,8	0,79	0,97	5,21	8,38	7,13	9,57	3,66	2,2	63,85
PDF	0,26	880	2,5	6050	4,1	0,66	0,89	11,57	9,86	6,33	14,3	2,48	-	55,46

Table 7. Synthesis of the best performance parameters and syngas compositions of the gasification tests carried out with different bio- and waste-feedstocks.

Reported data indicate better biomass gasification performances for higher ER values respect to waste-fuels. This could be related to difference in structures and chemical analysis of utilized feedstocks. In particular wood biomass is mainly made of cellulose, hemicelluloses and lignin, while utilized wastes have a prevalent fraction of plastic polymers, therefore higher heating value, amount of volatile matter and reactivity. This implies for MPW and PDF a very rapid achievement of the desired bed temperature utilizing reduced amount of fuel and oxidizing agent. Further, influence of higher moisture content in biomass is also present, since energy issued from exothermic reactions is used in the evaporation of the water contained. Typically, CO content in the syngas is higher in the case of dry fuels, while CO_2 content increases with the moisture in the feedstock, as a consequence of water gas shift reaction (n° 7 in Table 3). Additionally, higher moisture content in biomass reduces the molar fractions of the combustible components and the efficiency of the process (lower heating value of the producer gas). However, all utilized feedstocks show good energetic efficiencies in terms of cold gas (0.63-0.79) and carbon conversion (0.89-0.97) and syngas heating values suitable for energy generation. In particular, the best gasification performances are related to MPW, with highest syngas yield (3,4 m³_N/kg_{fuel}), lower heating value (9400 kJ/m³) and specific energy (8,8 kWh/kg_{fuel}), due to the extremely high fuel LHV (about 40 MJ/kg), while chicken manure appears the worst feedstock in terms of fuel gas energy and yield. The dirty syngas obtained by the gasification is then addressed to cyclone for dust abatement and then to a wet scrubber (a couple of bubblers in the pre-pilot plant) for tar and nitrogen, chlorine and sulfur compounds removals, as well as for the residual fraction of inorganic compounds. Table 8 summarizes the syngas pollutant concentrations, downstream of the reactor, for the gasification tests that provided the best performance parameters in terms of CGE and CCE for each feedstock utilized.

	FR T		FR T		Е	Ec	Tar	HCI	H_2S	NH₃
	EK	I	g/kg _{fuel}	g _C /kg _{C-fuel}	mg/m ³ _N	mg/m ³ _N	mg/m ³ _N	mg/m ³ _N		
WB	0,28	880	20,9	31,2	2300	13	1	16		
СМ	0,34	750	118	2,89	1340	41	4	3675		
MPW	0,24	890	0,9	0,5	1100	3	4	37		
PDF	0,26	880	23,2	13,8	23	1	0,4	2		

Table 8. Synthesis of the syngas pollutant concentrations as function of the best performance parameters for different feedstocks

Biomass gasification leads to relatively high emission of particulates. Particles can derive from incomplete conversion (soot, tar, char) or from the inorganic material in the fuel ash. Mass

concentrations of particles upstream of dust separation device widely vary for different biomass (i.e. $10 \text{ g/m}^3_{\text{N}}$ for wood and $69 \text{ g/m}^3_{\text{N}}$ for manure) and fuel composition is strongly related to the composition of particles emitted. Supermicron particles typically contain refractory species such as calcium, magnesium, silicon, phosphorous and aluminium, while submicron particles are mainly composed of alkali salts like potassium chloride and potassium sulfates. Further, wood biomass fine particles result very rich in carbon (up to 68%) affecting the carbon conversion efficiency of the whole process in not negligible way.

Tar formation is one of the biggest problems faced during gasification. The tars formed consist of a variable mixture of condensable hydrocarbons, with or without other oxygen-containing hydrocarbons and more complex polycyclic aromatic hydrocarbons and causes catalyst deactivation, operation interruption and the production of carcinogenic elements. Internal treatments in the gasifier, indicated as primary methods for tar removal, optimize gasification to produce a syngas with a minimum concentration of tar as possible, including selection of specific operating parameters, use of bed catalyst, and modification in gasifier design. Well operated fluidized bed gasification produce a syngas with a relatively low amount of tar for different biomass and waste feedstocks (from 0,02 to 2,3 g/m³_N). Moreover, all feedstocks produce negligible amounts of ammonia, hydrogen chlorides and sulfide, except to NH₃ concentration for chicken manure gasification $(3.7g/m^3_N)$, due to the very high nitrogen content in the feedstock (3,2% - 5,6%). On the basis of gasification test results and laboratory analyses of liquid, solid and gas residues for all the utilized bio- and waste-feedstocks, material and substance flow analyses were performed in order to deeper understand the performance of the gasifiers and to define and quantitatively assess some design solutions and operating criteria of the whole gasification system. Figure 7 (A-D) reports, for example, the results of carbon flow analysis applied to the best performance gasification tests for each feedstock utilized in the experimental activity. In some cases result have suggested the possibility to recycle fines collected by the cyclone into the reactor with the fuel or to provide a tar recycling downstream of the wet scrubber unit directly at the bed bottom of the gasifier. These options allow to recover a large quantities of carbon, and therefore of energy, both to obtain better energetic performance in the fuel gas produced (as in CCE and CGE) and to avoid, respectively, a solid (fly ash) or a liquid (water contaminated with tar) output stream and it disposal or successive treatment. In particular, for wood biomass, recycling of carbon fines increase CCE to 0,994 (from 0,963) and CGE to 0,769 (from 0,765) while for packaging derived fuel tar recycle increase CCE to 0,986 (from 0,887) and CGE to 0,684 (from 0,658). Another

advantage of fly ash recycle is the reinjection inside the gasifier of large fraction of escaped inorganic bed material, which could limit the entity of bed make-up and ensure longer life-time of bed catalytic effect.



Figure 7. Layers of carbon flow analysis throughout the pilot scale gasifier for the biomass and waste feedstock during the best performance tests. Data are reported in g/h. WSCR = Wet SCRubber; WTS = Water Treatment System.

A further interesting result of substance flow analysis is related to the feedstock energy. In Figure 8 (A-D) are reported the results of energy flow analysis applied to the best performance gasification tests for each feedstock.



Figure 8. Layers of feedstock energy flow analysis throughout the gasifier for the biomass and waste feedstock during the best performance tests. Data are reported in MJ/h. WSCR = Wet SCRubber; WTS = Water Treatment System.

The energy flow entering inside the reactor with the fuel has been determined by means of specific relationships for each feedstock. All the other flows in import or in export reported in the feedstock energy layers were evaluated on the basis of the heat of combustion of each specific substance. As expected, the feedstock energy losses are mainly concentrated inside the fluidized bed reactor. The resulting difference in feedstock energy inside the reactor is that invested at the steady-state condition to convert a solid fuel in a gaseous fuel. For example, for wood biomass fluidized bed gasification the process need of 151 MJ/h to convert 48,8 kg/h of fuel in syngas. Therefore, in the reactor there is an energy loss of 19,5% respect to the energy fed with the fuel, i.e. a specific energy of 3,1 MJ/kg is invested in solid-to-gas conversion. Similarly, gasification needs of 4,25 MJ/kg for chicken manure, 7,4 MJ/kg for mixed plastic waste and 5,4 MJ/kg for packaging derived fuel, with energy losses of 35,6%, 18,4% and 23,2% respectively. In particular very high amount of energy loss in the chicken manure gasification (more than one third) is related to the high of ash content (up to 25%) and composition (high value of alkali, especially potassium).

Infact, by evaluating experimental tests carried out at different temperature with the same manure, at same ER, and keeping fixed all the other operating parameters, difference in specific energy losses could be related to the partial ash melting that occurs at temperatures higher than 700°C. Obviously in the case of modified configuration design, such as fly ash or tar recycling in the reactor, energy feedstock result in increase of energy losses inside the reactor. In particular for packaging derived fuel energy feedstock become 311 MJ/h instead of 244 MJ/h, i.e. 30,5% of total energy feedstock entering with the fuel, due to the endothermic tar reforming reactions that runs against the exothermic tar combustion reactions.
Paper I.

Arena U., Di Gregorio F., Zaccariello L., Mastellone M.L. (2010) "Fluidized bed gasification of biomass: a substance flow analysis" published in Fluidization XIII - New Paradigm in Fluidization Engineering

FLUIDIZED BED GASIFICATION OF BIOMASS: A SUBSTANCE FLOW ANALYSIS

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ABSTRACT

A natural biomass was fed in a pilot scale bubbling fluidized bed gasifier, having a maximum feeding capacity of 100kg/h. Measurements included the syngas composition, the mass flow rate and composition of entrained fines collected at the cyclone and purge material from the wet scrubber, and the bed material characterization. The performance of the whole gasification plant and of its specific components as well as the validity of some design solutions and operating criteria have been quantitatively assessed by means of a substance flow analysis.

INTRODUCTION

Different gasification technologies are today available to convert biomass in a syngas able to provide a wide range of products, extending from clean fuel gas and electricity to bulk chemicals (<u>1</u>, <u>2</u>). Fluidization is the most promising among all biomass gasification technologies, for a series of attracting reasons, such as the possibility to utilize different fluidizing agents, reactor temperatures and gas residence times, to inject reagents along the reactor height and to operate with or without a specific catalyst (<u>3</u>, <u>4</u>).

Since the markets for biomass gasifiers without gas cleaning are rather limited, the key to achieving economically and environmentally efficient energy recovery from natural and waste biomass gasification is to overcome problems associated with the formation and release of different contaminants (tars, heavy metals, halogens and alkaline compounds) that can cause environmental and operational troubles. The syngas cleaning approaches can be divided in treatments inside the gasifier (primary methods), such as adequate selection of main operating parameters, use of a proper bed additive or catalyst, specific gasifier design modifications, and hot gas cleaning after the gasifier (secondary methods), such as thermal or catalytic tar cracking and mechanical methods (cyclones, ceramic, fabric or electrostatic filters, and wet scrubbers). It is likely that an adequate combination of primary and secondary treatments may optimize the gasifier performance and allow to produce a syngas that meets the cleaning requirements of different end-use devices (1, 2, 5).

This study combines a series of experimental results obtained from a pilot scale bubbling fluidized bed gasifier fed with a natural biomass fuel, together with a recently

defined environmental assessment tool, the Material Flow Analysis, which is named Substance Flow Analysis when it is referred to a specific chemical species. MFA/SFA is a systematic assessment of the flows and stocks of materials and elements within a system defined in space and time. It connects the sources, the pathways, and the intermediate and final sinks of each species in a specific process ($\underline{6}$). These characteristics make MFA/SFA attractive as a decision support tool, as showed by its utilization in process evaluation of waste treatments and recycling options ($\underline{7}$) and in waste management planning ($\underline{8}$). The aim of this study was to deeper understand the performance of the whole gasification plant as well as that of its specific components (reactor, cyclone, wet scrubber) and to define and quantitatively assess some design solutions and operating criteria of the biomass gasification system.

PILOT SCALE GASIFIER, MATERIALS AND EXPERIMENTAL PROCEDURES

The pilot scale bubbling fluidized bed gasifier (BFBG) has a maximum thermal output of about 500kW and the design and operating features schematically listed in Table 1. The fuel and blast flow rates were mutually adjusted so that, at the fixed fluidizing velocity, the desired equivalence ratio ER was obtained (where ER is defined as the ratio between the oxygen content of air supply and that required for the stoichiometric complete

combustion of the fuel effectively fed to the reactor). The cylindrical BFB reactor was heated up to the reaction temperature the bv sensible heat of pre-heated blast gases and by a set of
 Table 1. Main design and operating features of the BFBG.

	· · · · · · · · · · · · · · · · · · ·
Geometrical	ID: 0.381m; total height: 5.90m; reactive
parameters	zone height: 4.64m; wall thickness:12.7mm
Feedstock capacity	30-100 kg/h (depending on the type of fuel)
Thermal output	about 500kW
Typical bed amount	131-145 kg
Feeding system	over-bed screw feeder
Gasifying agents	air, oxygen, steam, carbon dioxide
Bed temperatures	700-950°C
Fluidizing velocities	0.3–1m/s
Flue gas treatments	cyclone, scrubber, flare
Safety equipments	water seal, safety valves, rupture disks,
	alarms, nitrogen line for safety inerting

three external electrical furnaces. The gas generated in the reactor is sent to the syngas treatment section composed of a high

efficiency cyclone, a simple wet scrubber (for removal of tars, residual fly ashes and acid gases) and a flare. An accurate description of the plant is provided elsewhere (5).

The biomass fuel is made of natural wood pellets for domestic heating, having the ultimate analysis and lower heating value reported in Table 2, and the chemical composition of inorganic fraction reported in Table 3. Two bed materials were used during the experimental runs: an olivine and a quartz sand (Table 4). The latter is the typical bed material of fluidized bed combustor while olivine is recognized as a good tar-removal bed additive in the gasification of biomass (1, 5, 9, 10).

Table 2. Chemical

characterization of the biomass.

Ultimate analysis, %	
С	45.3
Н	5.6
N	0.5
S	0
moisture	9
ash	1.2
O (by difference)	38.4
C/O ratio	1.2
LHV _{as received} , kJ/kg	≈15700

biomass morgan	ic traction.					
Element, mg/kg _{dry solid}						
Arsenic <0.1						
Aluminium	150					
Antimony	2					
Cadmium	0.05					
Calcium	1750					
Cobalt	0.17					
Chromo	0.95					
Iron	290					
Magnesium	465					
Manganese	15					
Mercury	0.01					
Nickel	0.35					
Lead	1.2					
Potassium	330					
Copper	1.5					
Sodium	110					
Tin	90					
Thallium	<0.25					
Vanadium	<0.1					
Clorides (ma/kaar)	3650					

Table 3. Composition ofbiomass inorganic fraction.

Experimental procedures are described in detail elsewhere (11). Here it is sufficient to highlight that in all the runs gas composition upstream and downstream of the syngas treatment section was on-line measured by IR analyzers for the main syngas components and by a couple of micro-gaschromatographs equipped with different columns for detection of lighter and heavier hydrocarbons as well as carbon monoxide and dioxide. hvdrogen. nitrogen and water. Two different methods of tar evaluation were used: the first conservatively imputes to the tar amount the whole carbon loading which, as a result of a mass balance on atomic species, cannot be attributed either to the produced gas or to the solids collected at the cyclone or present inside the bed; the second method utilizes samples taken at the reactor exit, for about 30 minutes, by means of four in-series cold traps, and then sent to a gas chromatograph coupled with a mass spectrometer. Data obtained from on-line and off-line gas measurements and those from chemical analyses of solid samples were processed to develop for each run complete mass balances on atomic species and the related energy balance. The flow rate of produced syngas was determined by the "tie component" method (12) applied to the

value of nitrogen content in the dry syngas, as obtained by (on-line and off-line) GC measurements.

RESULTS AND DISCUSSION

The operation of the pilot scale BFBG with the natural biomass at 850°C and 0.25 of ER, in a bed of olivine particles fluidized at 0.63m/s, was chosen as the base-case condition. Figure 1A is the result of the MFA applied to the whole gasification system, when operated under these conditions. This layer of total mass flow rate is the quantified flow diagram of the main process units (gasifier, cyclone, wet scrubber, water treatment system) of the pilot scale gasification system. Each flow in input to or in output from a specific unit is identified by means of a black arrow if the

	olivine	sand
	Mg-Fe	quartzite
Chemical Composition, %	silicate	
SiO ₂	39-42	96.38
MgO	48-50	-
Fe ₂ O ₃	8-10.5	0.15
CaO	<0.4	0.05
K ₂ O	-	0.75
TiO ₂	-	0.50
Al ₂ O ₃		2.3
Cr ₂ O ₃	0.8	-
Mg ₃ O ₄		-
LOI	0.20	0.27
Size range, μm	200 ÷	+ 400
Sauter mean diameter, µm	298	205
Particle density, kg/m ³	2900	2600
Minimum fluidization vel.	0.030	0.013
(850°C), m/s		
Terminal vel. (850°C), m/s	2.0	1.0

Table 4. Characteristics of the bed particles.

specific data have been measured or fixed, or by a grey arrow if the data have been obtained by means of the material or energy balances of the MFA. The input flows to the BFBG unit are the stream of biomass, that of a small flow rate of nitrogen (utilized to facilitate the fuel injection) and that of air used as reducing agent and fluidizing gas. The output flow stream is the obtained syngas, which still contains heavy hydrocarbons, inorganic pollutants and entrained fines. It is sent to the cyclone for dust abatement and then to the wet scrubber to remove tars and inorganic compounds. The final syngas stream has a volumetric flow rate of 115.8m³_N/h and the following measured composition: CO₂=14.3%: CO=19.1%: H₂=13.8%: $CH_4=4.9\%$; $C_2H_4=0.9\%$; N₂=47.0%. The specific production of syngas is equal to 2.27kg_{svndas}/kg_{fuel} (i.e. 1.94m³_{N,svngas}/kg_{fuel}) while that of elutriated fines is 28.2q_{fines}/kq_{fuel}. The stock of 145kg of olivine bed particles is progressively depleted (0.22kg/h) as a result of opposite effects of elutriation losses and fuel ash accumulation. The experimental activity provides the complete chemical composition of streams in exit from cyclone and the water treatment system. These data have been used for the substance flow analysis reported in Figs. 1B and D for carbon and iron and in Table 5 for magnesium, nickel, aluminum, ash as well as for the energy flow analysis of Fig. 1C.

Figure 1B is the result of the mass balance applied to the carbon element, i.e. the carbon layer of SFA. It gives the carbon conversion efficiency CCE, defined as the ratio between the mass flow rate of the carbon present in the syngas as CO, CO₂, CH₄ and light hydrocarbons (until C₅H_m) and the mass flow rate of the carbon that enters the reactor with the fuel. The value of 0.96 of CCE is evaluated as the ratio between the mass flow rates of the syngas carbon stream, F7, and fuel carbon stream, F1. CCE is mostly affected by the carbon losses related to the fly ash stream, F6 (for 3.9%) and, for an almost negligible fraction, to those of purge stream, F8. The specific carbon elutriation rate can also be evaluated and it is equal to $38.9g_{C-fines}/kg_{C-fuel}$. The carbon layer finally reports an important state variable of the biomass gasification process, the bed carbon loading W_C, which is the amount of carbon present in the bed as char particles at the steady-state condition (<u>13</u>). Its value of 1.71kg is a function of bed temperature and equivalence ratio.

Figure 1C is the layer of feedstock energy, i.e. the heat of combustion of each input and output streams (<u>14</u>). The energy flow entering with the biomass fuel has been determined by means of a relationships recently proposed and validated specifically for biomass fuels (<u>16</u>), while the energy flows of exit streams have been evaluated on the basis of the heats of combustion of the specific substances. The resulting difference in feedstock energy, 183MJ/h, is that "invested" at the steady-state condition to convert the solid biomass in a gaseous fuel. Reported data allow to evaluate the cold gas efficiency CGE, defined as the ratio between the chemical energy of obtained syngas and that of injected fuel: the value of 0.762 is mainly determined by the chemical energy utilized inside the gasifier (19.5%) and, for a smaller fraction, by that lost with the entrained fines (3.9%). It is negligible (0.3%) the fraction of feedstock energy lost with the heavy hydrocarbons of the purge stream from the water treatment system.

Figure 1D is the result of the mass balance applied to the iron element, i.e. the iron layer of SFA. It has an important role if olivine is used as tar removal bed material. An investigation about the role of olivine as a tar removal catalyst during the gasification



Figure 1. Layers of mass and energy balances throughout the gasification plant: A) total mass (kg/h); B) carbon element (g/h); C) feedstock energy (MJ/h); D) iron element (g/h).

of a plastic waste (<u>16</u>) indicated that magnesium and iron, both largely present in the olivine particles, activate the endothermic decomposition reactions of hydrocarbon fragments that are the first precursors of tar formation. A recent experimental study (<u>5</u>) showed that a similar behavior of iron and magnesium could be present even in the fluidized bed gasification of some waste biomass and, for a limited extent, in that of the same natural biomass utilized in this study. This conclusion was supported by the analysis of inorganic fraction of the fines collected at the cyclone. Accordingly with

conditions.	T () ()	~		-				-
Stream	Total Mass,	С,	Ash,	⊦e,	Mg,	AI,	Ni,	Energy,
	kg/h	g/h	g/h	g/h	g/h	g/h	g/h	MJ/h
F1, fuel	59.57	26,985.2	714.8	15.70	25.20	8.10	0.02	935.2
F2, transport N ₂	6.25	-	-	-	-	-	-	-
F3, air	80.41	-	-	-	-	-	-	-
F4, wet syngas								
+tar+fines	146.45	26,985.2	683.3	68.96	196.6	30.7	0.33	752.7
F5, wet syngas+tar	144.77	25,935.2	68.3	0.06	-	-	0.01	715.9
F6, fly ash	1.68	1,050	615	68.90	196.6	30.7	0.32	36.8
F7, dry syngas to end-use device	135.00	25.934.8	0.7	-	-	-	-	712.9
FO								
F8, purge	9.77	0.4	67.7	0.06	-	-	0.01	2.9
F9, tar	0.07	0.4	67.7	-	-	-	-	2.9
F10, water	9.70	-	-	-	-	-	-	-
Stock (expressed in g)	145,000	1,710	126.7	9,380	42,993	232	348	-
∆Stock	-0.22	-	31.5	-53.3	-171.4	-22.6	-0.31	182.6

Table 5. Results of material and substance flow analysis for the base case conditions.

other studies carried out with different refuse-derived fuels (<u>16</u>, <u>17</u>), the catalytic activity of olivine enhances series-parallel reactions that eventually produce molecular hydrogen and carbon coke. The latter links to the elemental iron of olivine by means of coordination complexes (<u>18</u>) and then it is entrained out of the reactor. As a consequence, when olivine is active, the fines collected at the cyclone should contain a larger quantity of iron with respect to that entering the reactor with the fuel. Data in Fig. 1D show that the ratio between the iron flow rate that escapes the reactor as fines (F6) and that of the iron that enters the reactor as inorganic fraction of the fuel (F1) is equal to 4.4, so supporting the presence of a possible even though limited



catalytic action of olivine. The same support is provided by the results of SFA for the magnesium element, reported in Table 5. Magnesium, which is one of the main components of olivine (Table 4), is present in large concentration in the fly ash stream, and the ratio between its flow rate escaping the reactor as fines (F6) and that entering the reactor as inorganic fraction of the fuel (F1) is equal to 7.8. The consequence of

Figure 2. Thermogravimetrical analysis of the fines collected at the cyclone, under the base case conditions.

these phenomena is the depletion of Fe and Mg in the bed of olivine, which is reported to be equal to -53.3g/h for Fe and -171.4g/h for Mg (Table 5).

These results suggest two possible design solutions: the make-up of bed olivine particles and the recycle of entrained fines. In particular, the latter could lead to some advantages. The first is an increase of both CCE and CGE as a consequence of the additional residence time of carbon fines inside the reactor, by taking into account that the reactivity of these fines has been demonstrated to be sufficiently high by a parallel investigation carried out by means of a thermo-gravimetrical balance (Fig. 2). The consequent advantage is that there is no necessity for a further treatment or disposal of these fines. Another, even if just potential, advantage of fly ash recycle is the reinjection inside the gasifier of large part of escaped inorganic fraction, and in particular that of magnesium and iron: if both of them would save their catalytic activity, this reinjection could limit the entity of olivine make-up.

The conditions of the base case were then changed in order to investigate the role of different operating parameters by means of the combined utilization of experimental data and substance flow analysis. The effects of the variation of the equivalence ratio, in the range between 0.17-0.26, and that of the bed material at the same ER of the base case, were investigated. The general behavior of the plant remains substantially unchanged, even though the values of the process performance parameters have in some cases not negligible variations. Table 6 provides a synthesis of the obtained results. The feedstock energy that in the BFBG is invested to convert biomass in a syngas fuel progressively reduces when ER increases, with a trend that is mainly related to the amount of solid fuel that is effectively converted in syngas. The bed carbon loading has a similar trend, i.e. it reduces as a consequence of an increase of ER. On the contrary, the CGE and the specific chemical energy production increase with ER, as a consequence of the larger syngas yield, which is, in turn, related to the reduced amount of heavy hydrocarbons in the syngas. The carbon conversion efficiency appears less affected by the equivalence ratio.

The plant performance with a bed of quartz sand is substantially similar, as it can be deduced by the values in Table 6 that appear just slightly affected by the type of bed material. The reason should be related to the high quality of the natural biomass tested (and then to the limited tar production during its gasification) that remarkably reduces the improvement that could be obtained by the utilization of olivine particles.

gasilication at valious equivalence ratios and	with an		Ju mato	lais.	
Equivalence ratio	0.172	0.212	0.247	0.254	0.260
Bed material	olivine	olivine	olivine	q.sand	olivine
Air-to-fuel ratio, kg _{air} /kg _{fuel}	0.93	1.14	1.35	1.39	1.40
Carbon conversion efficiency, -	0.976	0.982	0.961	0.967	0.978
Cold gas efficiency, -	0.746	0.742	0.762	0.764	0.780
Bed carbon loading, kg	2.93	2.43	1.71	1.90	0.84
∆Feedstock energy in the BFBG, MJ/h	322	266	183	196	178
Specific syngas yield, kg _{syngas} /kg _{fuel}	1.79	2.03	2.27	2.31	2.33
LHV of syngas, MJ/m ³ _{N,syngas}	7.67	6.81	6.16	6.11	6.19
Specific chemical energy production, MJ/kg _{fuel}	11.94	11.87	11.97	11.94	12.47

Table 6. Synthesis of results of material and substance flow analysis for gasification at various equivalence ratios and with different bed materials.

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Paper II.

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FLUIDIZED BED GASIFICATION OF A CHICKEN MANURE

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Abstract: A bubbling fluidized bed air gasifier, having an internal diameter of 102mm and a maximum feeding capacity of 3kg/h, was operated with a waste manure coming from an Italian chicken farm. The experimental runs have been carried out by keeping fixed the fluidized bed velocity (0.4m/s) and the type and size range of bed material (quartz sand, 0.2-0.4mm) and by varying the equivalence ratio between 0.27 and 0.40 and the reactor bed temperature between 700 and 800°C. The main components of the obtained syngas (CO, CO₂, H₂, CH₄, C₂H_m, C₃H_m, BTX, N₂) were measured by means of on-line analysers and a gas-chromatograph. The process performance was assessed on the basis of different parameters, such as carbon conversion efficiency (CCE), cold gas efficiency (CGE), specific energy production, syngas low heating value and yield of undesired by-products (mainly tar and fly ash). The results indicate that the air gasification process of chicken manure is technically feasible even though a preliminary careful characterization of ash composition and properties must be made, in order to avoid operating troubles and to optimize the conversion of the feedstock energy.

Keywords: chicken manure; waste; gasification; bubbling fluidized bed; biomass

INTRODUCTION

In the last years, the intensive population growth and the following increase of the meat demand, determined a proliferation of the food industry. In particular, the Italian poultry industry is one of the Europe's largest producer and exporter of poultry meat (12% of the overall market) with a turnover of 5,300 million euro, mainly due to the continuous national growing meat consumption (18kg/cap/y), with a production of a large quantity of manure by-products (UNA, 2010). Landfilling of chicken manure was the traditional disposal solution until the beginning of 2000. The necessity of an adequate treatment of pathogens and heavy metals as well as that of avoiding potential eutrophication effects have driven to restrictive environmental regulations (Boesch et al., 2001; Dagnal et al., 2000). Consequently, bio-chemical (composting and anaerobic digestion) and thermo-chemical (combustion and gasification) conversion processes were recently identified as possible better management options.

The bio-chemical units allow to obtain weight and volume reduction together with the elimination of pathogens but they have additional equipment and handling costs and do not solve the problems of nutrient losses and heavy metals contamination (Florin et al., 2009). The thermo-chemical units can instead obtain the destruction of pathogen agents together with a remarkable weight reduction and energy generation: they can then appear as an economical and environmental viable solution (Zhu and Lee, 2005). The gasification process, in particular, can be used to convert the poultry farm waste in a fuel gas (syngas), which can significantly contribute to the sustainability of chicken manure management (Zhang et al., 2009). Problems associated with the formation and release of different contaminants (tar, ashes, heavy metals, halogens and alkaline compounds) could cause environmental and operational troubles, such as clogged or blockage in fuel lines, filters, heat exchangers and energy conversion devices, material corrosion, wastewater pollution, and still are today a crucial obstacle to be overcome.

This study aims to evaluate the performances of the air-gasification process of a chicken manure by utilizing a bubbling fluidized bed reactor. Fluidization is the most promising among all biomass gasification technologies, due to its peculiar characteristics and, in particular, for the possibility to utilize different fluidizing agents, reactor temperatures and gas residence times, to inject reagents at different reactor heights and to operate with or without a specific catalyst (Basu, 2006; Arena et al., 2010a). An appropriate utilization of these features could therefore be the key to define an efficient and sustainable management of this kind of waste.

EXPERIMENTAL APPARATUS AND PROCEDURE

The experimental work was carried out in an atmospheric bubbling fluidized bed gasifier (BFBG) with a feeding capacity of approximately 3kg/h. The BFBG is a 102mmID cylindrical column, made of AISI 316L and electrically heated by five shell furnaces, each capable of a maximum power of 3.5kW. All the heating elements are controlled by a data acquisition system connected to five thermocouples, located in the reactor internal wall, which allow to independently set the temperature of each reactor section (blast feeding, pre-heater, bed and freeboard). The air utilized as fluidizing agent was injected at the bed bottom through a distributor plate composed of three nozzles. These have a truncate pyramidal shape and were specifically designed in order to ensure a homogeneous distribution of the fluidizing gas in the bed crosssection. The total height is 2.5m from the metal distributor plate to the syngas outlet. The feedstock was continuously over-bed fed by means of a screw-feeder device. A nitrogen flow was used to help the fuel feeding and to avoid the back flow of the produced gas. At the syngas outlet a high efficiency cyclone allows dust removal. Downstream of it there are two alternative symmetric syngas conditioning lines, each one consisting of a bubbler and a filter for tar, residual fly ash, acid and basic gases. The main syngas compounds (CO, H₂, CH₄, CO₂, N₂, O₂, C_nH_m and BTX) were measured by using an Agilent Micro-GC 3000 located downstream of the tar sampling line. The syngas is further sampled by means of tedlar bags in other two points along the reactor height (0.9m and 1.8m) and then off-line analyzed. More details about the experimental apparatus used can be found in Mastellone et al. (2010).

Gas and solids sampling procedures were activated when the values of pressure, temperature and gas composition were at steady state conditions for not less than 1h. For the sampling of condensable species, a system consisted of four in-series cooling coils plugged in an ice bath, a suction pump and a flow meter, was installed and operated with a syngas flow rate of about $3dm^3_N/min$ for 30min to obtain tar and water phase. Water was separated from tar in order to evaluate its content in the syngas and the condensed hydrocarbons were off-line analyzed, with a specific pre-treatment, in a Perkin-Elmer Clarus 500 gas chromatograph coupled with a mass spectrometer (GC-MS). This procedure allows to recognize tar belonging to the classes between 2 and 5 of the classification system proposed by ECN (Van Paasen and Kiel, 2004). Hydrogen chloride, hydrogen sulphide and ammonia were collected by bubbling the product gas through a pair of gas stripping bubblers, connected in series and containing basic and acid solutions respectively, and subsequently analysed by means of a Dionex DX-120 ion chromatograph.

Data obtained from on-line and off-line gas measurements and from chemical analyses of solid samples were processed to develop mass balance on atomic species and the related energy balance for each chicken manure gasification test. The bed carbon loading, i.e. the amount of carbon present in the bed as char particles at steady-state condition, was experimentally determined at the end of each test by switching from reducing to oxidizing conditions and by recording and integrating the CO₂ and CO contents produced by oxidation. Finally, the flow rate of produced syngas was determined by the "tie-component method" applied to the value of nitrogen content in the dry syngas, as obtained by (on-line and off-line) GC measurements, and adequately corrected to take into account the nitrogen fed into the gasifier with the waste and that leaving it as ammonia.

CHARACTERIZATION OF CHICKEN MANURE TESTED

The feedstocks used were two chicken manures coming from the same Italian poultry farm, as collected in two different seasons. Both wastes, indicated in the following as CM1 and CM2, were obtained by a random sampling method in the chicken farm storage area. Their ultimate analyses are reported in Table 1 together with the moisture and ash content, the heating values and a detailed composition of inorganic fraction. It is noteworthy that the data in Table 1 are very close to those reported by the main scientific literature in the field (Davalos et al., 2002; Quiroga et al., 2010).

	CM1	CM2
Ultimate analysis, % on weight basis ±SD		
С	33.0±3.1	30.7±1.3
Н	4.4±0.5	4.2±0.3
N	5.6±0.1	3.2±0.2
S	0.3±0.1	0.2±0.1
Cl	0.5±0.1	0.4±0.1
O (by difference)	29.1±2.9	25.2±2.0
Moisture	9.9±1.9	11.0±0.6
Ashes	17.2±0.4	25.1±2.1
C/N ratio	5.90	9.6
Heating value [°] , kJ/kg		
HHV	14,590	13,670
LHV	11,940	10,980
Chemical composition of inorganic fraction, mg/kg _{db}		
Aluminium	210.9	344.6
Antimony	0.1	0.04
Arsenic	0.1	0.18
Cadmium	0.5	0.23
Calcium	57930	93200
Chromium	7.2	9.09
Cobalt	1.38	1.39
Copper	37.32	33.51
Iron	418.1	435.6
Lead	0.13	0.19
Magnesium	2936	3032
Manganese	213	206.8
Mercury	0.23	0.13
Nickel	2.19	3.3
Phosphorus	11490	10400
Potassium	16780	14740
Sodium	3372	4350
Tin	0.07	0.05
Vanadium	2.07	2.9
Zinc	283.7	214.3

Table 1. Main chemical properties of the chicken manure utilized for the gasification tests.

^a evaluated by means of a specific biomass relationship proposed by Sheng and Azevedo (2005)

Fig. 1 reports the thermal analyses of the two wastes, as obtained by a Perkin-Elmer Pyris Diamond Thermogravimetric Differential Thermal Analyzer (TG/DTA), operated with nitrogen and a heating rate of 20°C/min. The curves of TG (sample weight losses) and DTA (endo- or exo-thermic nature of the reactions) seem to predict an enough similar behaviour of the two wastes with most of the materials decomposed between 280 and 710°C.



Figure 1. Thermal analyses (TG and DTA) of the two chicken manures tested

EXPERIMENTAL RESULTS AND DISCUSSION

Effect of operating conditions

A first series of experimental tests were carried out with the chicken manure CM1, by keeping fixed the fluidized bed velocity (0.4m/s), the reactor bed temperature (between 750 and 800°C) and the bed material (a quartz sand having a SiO₂ content of 96% and a size range of 0.2-0.4mm) and by varying the equivalence ratio between 0.27 and 0.40. The results reported in Fig. 2 show that, as expected on the basis of previous experiences of biomass wastes (Devi et al., 2003; Arena et al., 2010a and b), the contents of hydrogen, carbon monoxide and methane in the obtained syngas as well as those of carbon losses, evaluated as elutriated fines and tar, decrease as the equivalence ratio ER increases. In particular, the data reported in Fig. 2 indicate that, for the tests carried out at a same ER of 0.34, higher temperature promotes a carbon loss increasing, due to a large production of tar and fines, and consequently a hydrogen and carbon monoxide decreasing.



Figure 2. Concentrations of the main syngas compounds and carbon loss in the CM1 gasification tests.

Fig. 3 reports the variation of main performance parameters (Basu, 2006) as a function of ER. It appears that an increases of ER induces a reduction of syngas lower heating value (from about 5.0 to $3.4\text{MJ/m}^3_{\text{N}}$) and an increase in the specific syngas yield (from 1.5 to $1.9\text{m}^3_{\text{N}}/\text{kg}_{fuel}$). These opposite effects only partially balance to each other, since a variation in the specific energy yield from about 2 to less than 1.7kWh/kg_{fuel} was detected. The same effect is also present when the focus is on the chemical energy transferred to the syngas (CGE), which reduces from 0.63 to a very low value of 0.49, which is below the generally recognized minimum acceptable value for a gasification process (Higman and van der Burgt, 2003). The latter two effects could not be only attributed to the ER decreasing, since, as it is evident by trends reported in the diagrams of Fig. 3, an effect related to the higher temperature at which some tests were carried out (800°C instead of 750°C) is also present. The temperature increasing leads always to a worsening of all the performance parameters, mainly as a consequence of the increased carbon losses.

To deeper investigate this effect, a Material Flow Analysis (MFA) was developed following the approach proposed by Brunner and Rechberger (2004). The analysis was carried out with the criteria utilized in recent investigations (Arena et al., 2010a) to quantitatively assess some design solutions and operating criteria of a biomass gasification system.



Figure 3. Main performance parameters in the CM1 gasification tests.

Fig. 4 shows the layers of feedstock energy (expressed as MJ/h), as obtained by the MFA applied to the tests carried out with the CM1 at ER=0.34 and bed temperature equal to 750°C (Fig. 4A) and 800°C (Fig. 4B), respectively.



Figure 4. Layers of feedstock energy (MJ/h). Tests carried out with the CM1 at ER=0.34.

The flow diagrams indicate that the losses of feedstock energy are mainly concentrated inside the BFBG, i.e. they are related to the energy necessary to convert the solid waste in a gaseous fuel. These values are equal to about 12MJ/h and 16MJ/h, respectively, so indicating that in the test at lower temperature (Fig. 4A) the loss of feedstock energy was equal to 4.1MJ/kg while that in the test at 800°C (Fig. 4B) was equal to 5.7MJ/kg. Since the tests were carried out with the same waste, at the same ER, and keeping fixed all the other operating parameters, the difference should be attributed to the softening or the partial melting of manure ash that occurs at temperatures higher than 750°C.

Effect of ash content and composition

A second series of tests were carried out with a second lot of a chicken manure, named CM2, received by the same farm. This waste shows a similar thermal behavior (Fig.1) but a higher ash content (25.1% instead of 17.2%), having a greater calcium percentage (Table 1).

The first test (indicated as A in Table 2) was carried out by keeping fixed the equivalence ratio at 0.34 and reducing the reactor temperature as low as 700°C, i.e. at a value enough lower than that involving possible melting or softening phenomena (Fig. 1). The results indicated a dramatic reduction of all the process parameters: CGE reduces until 0.36 and the specific energy becomes as low as 1.1kWh/kg_{fuel}.

Table 2. Main operating parameters and experimental results of Civiz gasification tests.																
Test	Tbed	ER	A/F	CO ₂	CO	H ₂	CH_4	$C_{n}H_{m}$	BTX	Q _{syngas}	LHV _{syngas}	Specific energy	Dust	Tar	CGE	CCE
#	°C	-	$\rm kg_{\rm air}/\rm kg_{\rm fuel}$	%	%	%	%	%	%	m³ _N /h	kJ/m³ _N	kWh/kg _{fuel}	g/m ³ _N	g/m ³ _N	-	-
Α	700	0.34	1.32	19.61	5.05	4.61	2.18	1.10	0.13	4.77	2800	1.11	68	11.24	0.36	0.92
В	760	0.34	1.29	20.67	4.76	4.68	2.02	0.90	0.14	4.57	2600	1.01	240	1.09	0.33	0.95
С	770	0.32	1.23	20.33	6.86	7.17	2.20	1.15	0.16	4.64	3400	1.35	96	1.92	0.44	0.94
D	760	0.40	1.53	21.56	4.95	5.29	2.24	1.03	0.14	4.45	2800	1.34	65	0.88	0.44	0.96

Table 2. Main operating parameters and	experimental results of	CM2 gasification test
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Since this remarkable worsening of the process performance could be attributed to a too much low bed temperature, a new test (indicated as B) was carried out at the same ER but at a bed temperature of 760°C and by using the same air stream pre-heating temperature of all the tests with CM1. Results of this test again showed a poor process performance. Therefore, the main reason of this unsatisfying behavior is likely to be found in the worst quality of the chicken manure CM2, which has a reduced LHV (-1.1MJ/kg), a higher ash content (+7.9%) and a larger amount of calcium (+61%), as reported in Table 1. The MFA applied to this test B (Fig. 5) allows to quantify these effects. The feedstock energy necessary to convert the solid waste in a gaseous fuel inside the BFBG is more than 22MJ/h (i.e. about 6.8MJ/kg), then 66% larger than the value measured for the test with CM1, carried out keeping fixed all the other operating parameters (Fig. 4A).



Figure 5. Layer of feedstock energy (MJ/h). Test carried out with the CM2 at ER=0.34.

The necessity of a larger energy consumption for the conversion of CM2 to a gaseous fuel is further supported by the TG/DTA curves reported in Fig. 6, as obtained in nitrogen atmosphere and at a heating rate of 20°C/min. The different weight losses (0.3mg and 3.6mg, respectively for CM1 and CM2) and DTA peaks confirm a strong different thermal behavior.



Figure 6. Thermal analyses (TG and DTA) of the ash of the two chicken manures tested.

Tar content and composition in the obtained syngas

The organic impurities of syngas range from low molecular weight hydrocarbons to heavy PAHs (usually collectively known as tar). Tars tend to condense or polymerize into more complex structures in pipes, heat exchangers or on particulate filters, leading to high risks of plant stop and larger maintenance costs and, above all, to impede the utilization of high efficiency energy conversion devices (Arena et al., 2010a; Arena et al., 2010b). It is generally recognized that tar control is a key issue for a successful application of biomass derived producer gas (Li and Suzuki, 2009). The reliable evaluation of their concentration and composition is then a pre-requisite to define and adopt a suitable combination of primary and secondary methods (i.e. inside the reactor and downstream of it) to control its content below the limit imposed by the specific syngas end-use device (Han and Kim, 2008). Fig. 7 reports the content and composition of these heavy PAHs for two tests with CM1, carried out at different equivalence ratios and reactor temperatures, respectively.



Figure 7. Tar content and composition for the tests carried out with CM1

All the tests exhibit the largest concentration in tar class 4, i.e. that of light PAHs with 2 or 3 rings, and naphthalene is in all the tests the main detected compound. It can also be observed (Fig. 7A) a growth of undetected tars concentration with increasing ER (47.4% instead of 76.3%) while the concentrations of tar classes 3, 4 and 5 decrease. An increase of undetected tars (class 1) was also observed (Fig. 7B) with increasing gasification temperature (76.3% instead of 93.1%) while all the detected compound concentrations decrease. A possible explanation of the observed increase of undetected tar compounds

assumes that the PAH growth reactions involve compounds of tar classes 2-5 or unsaturated hydrocarbons in the formation of heavier tars (Van Paasen and Kiel, 2004).

CONCLUSIONS

The results indicate that the air-gasification process of chicken manure is technically feasible but a preliminary and careful characterization of waste properties (mainly, LHV, moisture and ash content) as well as that of ash fraction must be made, in order to avoid operating troubles and to optimize the conversion of the feedstock energy. In particular, the possible non-homogeneity of the manure properties could be related to the handling and collection procedures that must be optimized in order to avoid a too large amount of floor dirt in the waste.

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Paper III.

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A COMPARISON BETWEEN FLUIDIZED BED COMBUSTION AND GASIFICATION OF A MIXED PLASTIC WASTE

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Abstract: The bubbling fluidized bed combustors and gasifiers could both be proposed as a convenient conversion section of a plastics-to-energy plant having a medium-size throughput (8-30MW). In order to develop a preliminary comparison of their process performances, a number of tests was carried out in two pilot scale bubbling fluidized bed reactors, a combustor and a gasifier, having the same internal diameter and a feeding capacity of about 40kg/h of a commercially available mixed plastic waste.

The collected experimental data were processed by different analytical tools in order to assess and compare the performances of the two fluidized bed waste-to-energy units. In particular, mass and energy balances, together with a Material and Substance Flow Analysis, were utilized to quantitatively define the paths of some crucial elements, such as carbon and heavy metals, as well as to evaluate the hot gas efficiency in the combustion-based and gasification-based plastics-to-energy fluidized bed systems.

Keywords: plastic waste; combustion; gasification; bubbling fluidized bed, waste-to-energy

INTRODUCTION

Plastic waste is a remarkable fraction of municipal solid waste and its treatment and disposal is one of the crucial aspects of a waste management system. Source separation and collection of end-of-life plastic goods is generally recognized as a necessary step of a sustainable management, produces three waste streams (Arena et al., 2011):

- material essentially made of polyethylene (PE) and polyethylenterephthalate (PET), which allows an environmental and economic sustainable recycle process (and is estimated to be about 55-60% of the total collection);
- II. material "out of target", which does not allow any recycling or energy recovery process (about 5-10% of the total collection);
- III. material essentially made of mixed plastic waste (MPW), with predominance of polyolefins (mainly polypropylene and polystyrene), which has a very high heating value (between 27 and 40MJ/kg) and is preferentially utilized as waste-derived fuel (about 35-40% of the total collection).

The energy value of this MPW can be converted to electricity, to process heat for industrial facilities and district heating as well as to vehicle fuels, by means of the thermochemical processes of combustion and gasification. Combustion is the more utilized plastics-to-energy process even though it presents some constraints related to the very low softening temperature of the plastic fuels, with the consequent high risk of sintering in the combustion chamber (Brandrup at al., 1996). Low values of the net electric energy conversion efficiency (about 20%) are generally reported: higher values may be obtained for co-combustion in coal-fired power plants even though this option is limited by concerns related to plugging of feeding systems. Gasification converts plastics in a combustible gas mixture (called syngas), mainly made of carbon monoxide, hydrogen and lower content of methane and it is able to provide a wide range of products, extending from clean fuel gas and electricity to bulk chemicals (Arena, 2012). Recent policies aimed at tackling climate change and promoting landfill diversion provided a renewed interest on the gasification technology: it is considered an advanced and viable alternative for waste thermal treatment with energy recovery (Defra, 2007), particularly for unsorted residual waste, i.e. the waste left downstream of separate collection that cannot be conveniently recycled from an environmental and economic point of view, as well as for combustible residues from recycling chain (Arena, 2012).

Recent technical and economic studies indicated the fluidization as the most promising technology for a medium-scale industrial application of plastics-to-energy cogenerators (Yassin et al., 2009; Arena et al., 2011). Therefore, the aim of this study is to evaluate and compare the process performances of a bubbling fluidized bed (BFB) combustor and a BFB air-gasifier, which both could be proposed as a convenient conversion section of a plastic-to-energy plant having a medium-size throughput (8-30MW). To this end, experimental tests were carried out by firing a commercially available mixed plastic waste in two pilot scale bubbling fluidized bed reactors, a combustor and a gasifier, having the same size.

The collected experimental data were processed by different analytical tools. In particular, a recently defined environmental assessment tool, the Material Flow Analysis, which is named Substance Flow Analysis when it is referred to a specific chemical species, was utilized. MFA/SFA is a systematic assessment of the flows and stocks of materials and elements within a system defined in space and time, which connects the sources, the pathways, and the intermediate and final sinks of each species in a specific process. These characteristics make it attractive as a decision support tool, as showed by its utilization in process evaluation of waste treatment and recycling options and in waste management planning. In this study MFA/SFA was used to deeper understand the performance of the pilot scale reactors and to quantitatively assess some operating criteria of the combustion- and gasification-based plastics-to-energy fluidized bed systems.

EXPERIMENTAL APPARATUS AND PROCEDURES

The pilot-scale bubbling fluidized bed combustor (BFBC) is visualized on the left side of Fig. 1. The bottom of the column contains the plenum chamber, which is divided into an annulus and core sections. The distributor sustains bed material allocated in the intermediate section, which is also equipped with several access ports. The freeboard section provides disengagement of elutriated solids and is fitted with several ports for temperature, pressure, gas concentration and particulate probes. Two cyclones, having medium and high efficiency respectively, are used for flue gas de-dusting. A ceramic wool blanket thermally insulates the entire vessel in order to ensure a safe temperature at the external surface. The heat exchange is obtained by means of: i) a water-cooled external jacket for a height of about 0.3m from the distributor plate; ii) an array of horizontal bayonet-type tubes whose adjustable penetration into the bed controls the heat removal rate; iii) an air-cooled exchanger located inside the upper part of the freeboard (ID=0.700m) to prevent the operation of cyclones at high temperature. A probe is installed at the exit of the second cyclone for gas sampling. The combustor start-up is accomplished thanks to a propane-premixed burner located in the annulus section of the plenum chamber. After the bed reaches a temperature high enough to ignite fuel particles (e.g., 600°C), the feeding of fuel is started and propane is switched off (Chirone et al., 2004).

The pilot-scale bubbling fluidized bed gasifier (BFBG) is visualized on the right side of Fig. 1. It is composed of three main sections: the feeding system, the fluidized bed gasifier and the syngas treatment unit. The feeding system can be divided in the blast feeding (measuring, mixing and injection of gasification agents) and the fuel feeding (measuring and injection of solid feedstock). The blast feeding is heated up to about 550°C before entering the reactor. In the experiments carried out for this study, atmospheric air was injected at the bed bottom as blast agent while the plastic waste was fed by means of an over-bed feeding system, as in the BFBC unit. The fuel and the blast flow rates were mutually adjusted so that, at a given fluidizing velocity (U_g) , the desired equivalence ratio ER was obtained (where ER is defined as the ratio between the oxygen content supplied to the reactor and that required for the stoichiometric complete combustion of the fuel effectively fed). The gasification section is heated up to the reaction temperature by the sensible heat of pre-heated blast gases and by a set of three external electrical furnaces. The gas generated in the reactor is sent to the syngas treatment section composed of a high efficiency cyclone, a wet scrubber (consisting of a water spray tower and utilized for removal of tars, residual fly ashes and acid gases) and a flare (Arena et al., 2008).



Figure 1. Photos of the two pilot-scale BFB reactors, combustion-based (left) and gasification-based (right).

In both the units, adjustments of operating variables are required in order to achieve the desired steady-state condition, which is maintained for at least 60 min. Temperatures, pressures, and flue/fuel gas concentrations in various points of the fluidization column are on-line monitored and recorded using a data control and acquisition system. Fines collected at cyclones are weighed at fixed times and analyzed for their carbon, hydrogen and nitrogen content and for the inorganic fraction composition. Table 1 synthesizes main design and operating parameters of the two plants, while Table 2 reports the main properties of the commercial available mixed plastic waste, utilized for the experimental investigation.

	BFBC	BFBG
Geometrical parameters	ID: 0.370 m (but 0.700 m at the top)	ID: 0.381 m
	total height: 6.90 m	total height: 5.90 m
	reactive zone height: 4.42 m	reactive zone height: 4.64 m
	wall thickness: 12 mm	wall thickness: 12.7 mm
Maximum feedstock capacity	100kg/h (up to 30kg/h with MPW)	100kg/h (up to 45kg/h with MPW)
Thermal output	Up to about 300 kW	Up to about 500 kW
Typical bed amount	80 kg	145 kg
Oxidizing agent	Air	Air (but also oxygen, steam, carbon
		dioxide and their mixtures)
Feeding system	Over-bed belt feeder and/or under-bed	Over-bed water-cooled screw feeder
	screw feeder	and/or under-bed screw feeder
Range of bed temperatures	700-900°C	700-950°C
Range of fluidizing velocities	0.3-3 m/s	0.3-1 m/s
Flue/Fuel gas treatments	Dual cyclones	Cyclone, scrubber, flare
Safety equipments	Rupture disk	Water seal, safety valves, rupture
		disks, alarms, nitrogen line for safety
		inerting

Table 1. Main design and operating parameters of the two pilot-scale BFB reactors.

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Proximate analysis, % _{wt, ar}		Ash composition, mg/kg _{db}			
Moisture	0.7	Aluminum	1040		
Volatile matter	97.2	Antimony	<0.1		
Fixed carbon	0.2	Arsenic	<0.1		
Ash	1.9	Cadmium	0.48		
Ultimate analysis, % _{wt ar}		Calcium	10070		
,, <u>.</u>		Chrome	10.17		
C	79.5	Cobalt	1.07		
н	13.1	Copper	17.3		
N	0.3	Iron	305		
S	0.2	Lead	33.8		
CI	0.5	Magnesium	246		
O (by diff.)	3.8	Manganese	6.44		
Moisture	0.7	Mercury	<0.1		
Ash	1.9	Nickel	1.1		
Heating value, MJ/kg _{fuel, ar}		Potassium	1480		
		Sodium	529		
LHV (measured)	40.4	Thallium	<0.25		
LHV (estimated)*	40.2	Tin	16.8		
*by means of correlation by Channiv	vala and	Vanadium	0.8		
Parikh (2002)		Chlorides (mg/kg _{ar})	517		

Table 2. Main properties of the mixed plastic waste.

ar= as received; db=dry basis; wt= weight basis

EXPERIMENTAL RESULTS AND DISCUSSION

Table 3 reports the values of main operating parameters for a couple of the experimental tests carried out with each of the BFB reactors. It is noteworthy that the two reactors have the same internal diameter and were operated at the same fluidizing velocity: this allows to assume that the whole hydrodynamics of the two units was substantially the same. Therefore, in all the following considerations, the effect of the fluidization quality can be assumed negligible or absent. Accordingly, the flow rate of elutriated fines in the BFBG unit was evaluated, and not measured, in order to avoid the misleading effects coming from the long fuel gas duct which is present in the pilot plant gasifier.

	COMB-1	COMB-2	GAS-1	GAS-2
Operating p	arameters			
F, kg _{fuel} /h	5.6	5.7	25.1	28.4
ER, -	1.45	1.41	0.24	0.21
Ug, m/s	0.67	0.68	0.65	0.65
T _{bed} , °C	822	844	894	890
T _{preheating air} , °C	20	20	448	543
AF ratio, -	19.64	19.14	3.20	2.84
Flue/Fuel go	as compositic	n, % _{vol}		
O ₂	7.00	6.35	-	-
N ₂	82.83	83.05	63.85	64.29
CO ₂	10.12	10.49	9.57	9.74
CO	0.05	0.11	5.21	3.97
H ₂	-	-	8.38	8.56
CH ₄	-	-	7.13	7.63
C₃Hmª	-	-	3.67	3.42
BTX	-	-	2.19	2.39
Main pollut	ants, mg/m ³ _N			
Tar	-	-	1,100	2,600
C as C _n H _m ^b	137	367	149,000	157,000
NOx	68.3	73.6	-	-
NH ₃	-	-	34	16
SO ₂	5.7	31.4	-	-
H ₂ S	-	-	4	9
HCI	-	-	3	58
Flow rate of	f elutriated fi	nes		
E/F, g/kg _{fuel}	16.9	19.3	17.1 °	17.1 ^c
E _c /F _c , g _c /kg _{C,fuel}	0.2	0.4	9.2	7.4

Table 3. Main operating parameters and results of the experimental tests.

^a light hydrocarbons, with 2 or 3 carbon atoms; ^b total organic carbon, CH₄, C₃H_m, BTX and tar included; ^c calculated value F= Fuel mass flow rate; AF= Air-to-Fuel ratio; E= mass flow rate of elutriated fines

Material Flow Analysis (MFA)

A Material Flow Analysis was carried out by means of the freeware STAN (subSTance flow ANalysis) developed by the Vienna University of Technology (Cencic and Rechberger, 2008). During the last 20 years Material and Substance Flow Analysis has become a reliable instrument to describe material flows and stocks within various systems. In particular, it was recently used to understand the performance of a BFB gasifier and to define and quantitatively assess some design solutions and operating criteria of a gasification-based plastics-to-energy system (Arena et al., 2011).



Figure 2. Layers of mass flows (kg/h), as obtained by the MFA: COMB-1 (left) and GAS-1 (right).

The MFA was applied to the tests indicated in Table 3 as COMB-1 and GAS-1. The quantified flow diagrams related to the total mass flow rates (also known as "layers") are reported in Fig. 2, respectively for the BFBC and the BFBG plant. The input flows to each reactor unit are the streams of plastic fuel, of air used as oxidizing agent and fluidizing gas, and, only for the gasification test, of a small flow rate of nitrogen utilized to facilitate the fuel injection. The output flow stream is the obtained flue/fuel gas, which still contains heavy hydrocarbons, inorganic pollutants and entrained fines. The raw gas is first sent to the cyclone for dust abatement. The specific productions are equal to 20.6kg_{fluegas}/kg_{fuel} and to 4.4kg_{fuelgas}/kg_{fuel} (i.e. $16.0m^3_{N,fue gas}/kg_{fuel}$ and $3.6m^3_{N,fue gas}/kg_{fuel}$) for the combustion and gasification tests, respectively.

Inside the reactors the bed amounts are progressively incremented (0.1kg/h for BFBC and 0.9kg/h for the BFBG) as a result of the opposite effects of elutriation losses and fuel ash accumulation.

Substance Flow Analysis (SFA)

Fig. 3 reports the result of the SFA applied to the carbon element. It gives the carbon conversion efficiency CCE, defined as the ratio between the carbon flow rate present in the flue/fuel gas (as CO_2 and CO or as CO_2 , CO, CH₄ and light hydrocarbons (C_3H_m), respectively) and the carbon flow rate fed to the reactor with the fuel. The calculated values of CCE are equal to 0.9998 and 0.9908, respectively. The difference is related to the higher C content in the elutriated fines, which is 0.7% for the BFBC and 42.8% for the BFBG. This very high C percentage in the gasifier ash could be explained with the high content of heavy hydrocarbons in the fuel gas, partially collected at the cyclone after condensation on particulates and nucleation mechanisms as a result of the decreasing temperature in the fuel gas duct and the cyclone (Zevenhoven and Kilpinen, 2001). Another possible contribution to the increased elutriation of carbon fines is the higher values of the bed carbon loading W_c , i.e. the amount of carbon present in the bed as char particles at steady-state condition (Arena et al., 1995), which is an important state variable also for plastic waste thermal treatments (Arena and Mastellone, 1999; Arena et al., 2011). As it is highlighted in the layers of Fig. 3, W_c is undetectable in the BFBC, as a consequence of the strongly oxidant environment and the very high reaction rate but it is equal to 0.8kg (i.e. 0.55% of the bed amount) in the BFBG, due to the nature of the independent heterogeneous equilibrium reactions, all characterized by a slower kinetics.



Figure 3. Layers of carbon flows (g/h), as obtained by the SFA: COMB-1 (left) and GAS-1 (right).

The Substance Flow Analysis was also applied to the main inorganic compounds present in the MPW in order to determine their fate as bottom ash stock in the reactor bed material, as collected fly ash or as compounds entrained in the gas phase downstream of the cyclone (and then almost completely intercepted in the adsorbing/dedusting air-pollution-control system). Figs. 4 and 5 show the results of the balance on the atomic species applied to cadmium (Cd) and lead (Pb), respectively.







Figure 5. Layers of lead flows (mg/h), as obtained by the SFA: COMB-1 (left) and GAS-1 (right).

It is known (Zevenhoven and Kilpinen, 2001) that many factors can influence whether and in what form a trace element eventually ends up in the gaseous or particulate phase. The most important among these factors are: i) how the trace element resides in the fuel; ii) system temperature and pressure; iii) presence of halogens (in particular, of chlorine); iv) presence of sorbent compounds; and v) oxidizing or reducing conditions. The latter is the only factor that can affect the fate of these elements in the reported BFBC and BFBG tests, being all the others kept fixed. Table 4 reports the transfer coefficients for some of these trace elements (Cd, Pb, Sn and V), together with some other compounds of interest for the combustion- and the gasification-based plastics-to-energy processes, as it has been already made for other waste-to-energy technologies (Jung et al., 2005).

Inorganic	BA, %		FA, %		Flue/Fuel gas, %	
compound	COMB-1	GAS-1	COMB-1	GAS-1	COMB-1	GAS-1
Aluminum	1.0	2.1	77.0	48.4	22.0	49.5
Cadmium	0.3	0.1	23.3	39.2	76.4	60.7
Calcium	3.8	8.1	7.7	11.9	88.4	80.0
Lead	0.4	0.1	31.4	36.1	68.2	63.8
Potassium	0.1	0.7	1.7	7.7	98.2	91.6
Tin	0.1	0.1	3.8	8.3	96.1	91.7
Vanadium	2.7	23.1	35.9	53.1	61.4	23.8

Table 4. Transfer coefficients of the main inorganic compounds in bottom ash (BA), fly ash (FA) and flue/fuel gas, during COMB-1 and GAS-1 tests.

It is evident that in the BFBC test there is a small percentage of all the inorganic compounds as stock in the bed material (BA). In particular, all of them, except for the AI, show a larger fraction escaping the system in the flue gas (from 61 to 98%), with a not negligible fraction collected as dust (FA) by the cyclone for Cd, Pb and V (between 23 to 36%). In the fluidized bed gasification test the fate of some inorganic compounds seems to be different, even though also in this case (with the exception of V) their fraction as stock in the reactor is unimportant. In particular, AI, Cd and Pb reveal a balanced repartition betweens FA and fuel gas phase while the largest fractions of K, Sn and Ca are detected in the fuel gas.

Energetic Performance

The reported balances have been also used as a basis to assess and compare the energetic performances of the combustion- and gasification-based plastics-to-energy processes. For thermal applications, such as when the gas is not cooled before combustion and the sensible heat of the gas is also useful, the hot gas efficiency (HGE) is used as the best parameter to an accurate assessment of the energetic performances (Basu, 2006). The conventional definition of this parameter (Arena, 2012) has been modified to take into account the different terms which have a relevance in the two thermochemical processes. HGE has been then defined as:

Table 5 reports all the terms that contribute to the evaluation of the HGE in the four experimental tests. A comparison of the energetic performances of the two units cannot be carried out just on the basis of the HGE, even because it has a range of variation, as a consequence of the variability of different operating parameters. Nevertheless, it is possible to make some preliminary observations: i) the two processes have similar energetic performances; ii) the BFBC process is negatively affected by the utilization of an air excess greater than 40% (i.e. ER>1.4); iii) the BFBG is less advantageous when values of ER<0.24 were utilized. The latter conclusion is supported by the values of cold gas efficiency CGE, defined as the ratio between the chemical energy of the produced syngas and the chemical energy of the plastic waste,

which is equal to 0.71 for the GAS-2 test and to 0.79 for the GAS-1 test, then in agreement with the typical range of ER for medium or large units, which is between 0.25 and 0.5 (Arena, 2012).

# test	LHV _{gas} *Q _{gas} , kW	H _{flue/fuel gas} , kW	H _{water} , kW	LHV _{fuel} *Q _{fuel} , kW	H _{air} , kW	HGE, -
COMB-1	-	31.3	21.3	62.7	0.61	0.83
COMB-2	-	32.1	26.0	64.3	0.61	0.90
GAS-1	206	35.1	-	280	9.86	0.83
GAS-2	202	38.6	-	317	12.2	0.73

Table 5. Chemical energies and sensible heats that contribute to the Hot Gas Efficiency, for all the fluidized bed combustion/gasification tests.

CONCLUSIONS

The study proposes a preliminary comparison between a combustion- and a gasification-based BFB plastics-to-energy process. The results cannot be considered as exhaustive since other important aspects – such as the specific configurations of the two plants, the type and the efficiency of the energy conversion devices and those of the flue/fuel gas cleaning units – should be taken into account. The reported results can be utilized as a basis to develop accurate comparisons between different specific processes or plant solutions. The future experimental activity will investigate a wider range of operating parameters as well as the performance of the two processes when operated in co-feeding of different waste-derived fuels.

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1.2 Techno-economic assessment for FBG biomass- and waste-to-energy plants

All the data obtained from fluidized bed gasification tests for different bio- and waste-fuels were combined with relationships of fluidization engineering in order to determine main geometrical parameters of the gasification section. In particular, for fixed nominal plant capacities, the reactor diameter was determined, on the basis of the cold gas efficiency and equivalence ratio, by keeping fixed the fluidizing velocity and the type and granulometric distribution of bed materials:

$$ID_{reactor} = \sqrt{\frac{Q_{air}}{v_f} / \frac{\pi}{4}}$$

where Q_{air} is the air flow rate, evaluated for a set equivalence ratio value, by the fuel composition and v_f is the fluidizing bed velocity, kept fixed between U_{mf} and U_t to avoid or reduce carryover of particles from the fluidized bed.

In particular U_{mf} , the minimum fluidization velocity, is evaluated for small bed particles (i.e. for a $Re_{mf} < 20$) by the simplified Ergun equation:

$$U_{mf} = \frac{d_p^2 \left(\rho_p - \rho_g\right) g}{150 \,\mu} \frac{\varepsilon_{mf}^3 \,\varphi_p^2}{1 - \varepsilon_{mf}}$$

and U_t , the terminal free-fall velocity of a particle through a fluid, can be estimated by the expression:

$$U_t = \sqrt{\frac{4 d_p (\rho_p - \rho_g)}{3 \rho_g C_D}}$$

where d_p is the particle size, ρ_p and ρ_g are respectively the bed material and fluidizing agent densities, μ is the fluid viscosity, ε_{mf} is the bed voidage at U_{mf} , φ_p is the particle sphericity and C_D is an experimentally determined drag coefficient.

The distance above the bed at which the entrainment becomes constant is the transport disengaging height, TDH. This reactor height was determined by means correlation respect to the reactor internal diameter in order to minimize the entrainment of fines from the bubbling bed gasifier.

In Table 9 these gasifier geometrical parameters are summarized for each feedstock utilized in the experimental tests with the pilot plant in the relative base case, i.e. in terms of the plant capacity of interest for the related study.



Figure 9. Correlation between TDH and reactor diameter, based on Zenz and Weil relationship (redrawn from PSRI, 2010)

	ER	T, °C	v _f , m/s	CGE	Plant Capacity, kWe	Q _{fuel} , kg/h	$ID_{reactor},m$	TDH _{reactor} , m
WB	0,28	880	0,6	0,77	200	167	0,65	5,50
MPW	0,24	890	0,7	0,79	2000	750	1,98	5,50
PDF	0,26	880	0,7	0,66	500	514	1,3	5,50

Table 9. Reactor geometrical parameters for each feedstock utilized in the pilot plant for a specific nominal plant capacity.

Plant capacities for biomass and waste derived fuel are related to the feedstock availability, generally commercially produced in large amount. Therefore for MPW and PDF the selected range of plant capacities were 2-6MWe and 0,2-1MWe respectively. When using biomass as fuel an important aspect that must be taken into consideration is its low energy density. Typically power production with biomass fuel makes sense only when it is used in decentralized or small to medium scale applications, in which the logistics cost is kept at a reasonable level, due to the short transportation distances required. Furthermore, the seasonality that characterize the availability of most biomass types creates many problems in the biomass logistics, such as the increased ware-housing requirements and seasonal use of resources. Some attempts have been made to deal with this problem by considering the simultaneous use of multiple biomass sources. For this reasons, in the reported study the range plant capacity considered for wood biomass plants was related to small scale applications (0,1-1MWe).

Syngas valorization

A biomass- or waste-driven generation plant can supply electricity and/or heat. The latter is normally in the form of steam or hot exhaust gas, or it can be applied to the process, for example for drying the feedstock or for preheating some gas agents. Alternatively, heat can be utilized in the form of hot water of around 70-90°C for district heating. Biomass has been used at a wide extend for district heating in Northern Europe, and is often used at combined heat and power (CHP) plants. The simultaneous heat and power generation reduces the primary energy consumption, compared to independent generation of heat and power, therefore making it environmentally friendlier and financially more attractive. District heating coupled with CHP has been used for many years, but it has not been considered a viable option for areas with warm climate up to now, as in these areas traditional cogeneration applications tend to prove financially unviable, due to the short operational time within the year. Also if not considered in these studies, in the last years an innovative concept has been introduced as an extension of the cogeneration, called trigeneration. It is the simultaneous generation of electricity, heat and cooling, with the addition of absorption chillers that transform heat to cooling. Absorption chillers have the ability of easily integrating with cogeneration systems and have lately gained widespread acceptance, together with the significant reduction of their price.

Gasification is a very promising conversion technology for biomass and waste, as it is considered that it may prove to be more cost effective compared to other technologies. In particular for biomass, some economic studies report that gasification plants can be as economical as conventional coal fired plants, while other researchers mention that gasification is the only technology among the most cost-efficient ones that is likely to be commercially viable for combining with a spark-ignition gas engine, which is a typical power generation scheme.

Gas engine (GE) is generally defined as internal combustion engine running on natural gas, syngas or biogas. It has been in commercial operation for decades, in units ranging from a few kWe up to several MWe, and with price ranges of around 500-1000 \in /kWe. Gas engine is described as robust against fuel quality changes, however it exhaust gas temperature is rather low with around 80-100°C, which makes further usage of it difficult. Gas engine efficiency lies in the range of 30-40% with a decrease under part-load operation (similar to microturbines). One problem in using biomass or waste fuels is that H₂S, which is present as contaminant in the syngas, is highly soluble in oil lubricants. As a consequence, frequent lubrication changes are necessary and maintenance cycles (oil and filter changes) can be required every 500 hours. This effect could highly reduces the

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applicability of those engines in rural and remote areas, especially when they are intended to run as a single generation unit and no skilled personnel is at hand. Finally, reciprocating engine emission levels (especially in terms of CO and NO_x) are significantly higher than those of other generators. Difference of up to an order of magnitude were reported (Gomes et al., 2004; Bruno et al., 2004; Rabou et al., 2007), and especially when running on biomass fuels. In terms of flexibility of operation, reciprocating engines again perform better than other generation technologies. Reasonable load changes can be adopted very quickly, and the suitability for fluctuating loads is high, hence they were the standard solution for emergency gensets.

Gas turbines (GT), in particular microturbines (i.e. < 500kWe), are small, predominantly aeroderivative turbines, using a comparably simple design and a generator directly mounted on the turbines shaft. Air is compressed, heated and then expanded in the turbine to produce motion. A recuperator can be used to preheat the compressed air with the exhaust gas heat before entering the combustion chambers. This increases the turbine efficiency by around 5%. However the turbine exhaust gas temperature is lowered from around 600°C in simple cycles to around 300°C in recuperative cycles and there is a significant cost impact. A number of suppliers have designed turbines from a few kWe up to several MWe. One of the advantages over internal combustion engines is their high exhaust gas temperature and therefore the possibility to use this heat within the process. Gas turbines can also be run on fuels with varying calorific values, and special designs for compressed low-calorific syngas are available. Another major advantage is their low maintenance need. They use air bearings, and together with a smooth rotation of the turbine, very long maintenance cycles of up to 10k-15k hours of continuous operation can be achieved. Finally, due to their constant combustion, emission levels are far below those of reciprocating engines, and given their price range of around 1000-2000€/kWe they are a promising alternative.



Figure 10. Schematic of the reference cycle for the operation of a gas turbine without recovery of the exhaust gases.

In terms of fuel efficiency, gas turbines employing recuperative cycles result in efficiencies of around 25-30% which is less than reciprocating engines, and they have a comparably robust efficiency behavior under part-load.

Although most turbines employ combustion chambers and expand the combustion air to generate shaft motion, some design use heat exchangers to heat the turbine working gas. In this case the process is called externally fired gas turbine (EFGT) and the engine can be operated based on all combustion fuels, similar to the stirling engine technology. In EFGT gasification application, syngas combustion provides the heat to be transmitted to compressed air used in the turbine. A high temperature heat exchanger, fired by a syngas combustion furnace, is employed to heat precompressed air. This hot air is then continuously expanded in the turbine and a generator, mounted on the turbine shaft, generates power. The expanded hot air at the turbine outlet can be used in CHP applications or within the process. Due to material constraints and limits, the heat exchanger temperature limit is around 900-1100°C. Therefore the temperature limit of the compressed hot air to be expanded in the turbine is around 800-900°C. Compared to a common gas turbine which directly uses the combustion flue gas of 900-1100°C, a lower level of work and thus efficiency can be achieved. Levels of 20-25% were reported for the comparably low number of EFGT plants in operation. When considering load flexibility, although fast load changes can be applied by using a heat exchanger air bypass valve to rapidly lower the temperature of the working air volume, this results in very poor part-load efficiencies. Since very long maintenance cycles of up to 10k hours of continuous operation can be achieved, price range of around 2000-2200€/kWe are justified by the high cost of special materials required for the heat exchanger, due to the high temperature differences on both sides of the exchanger surface as well as corrosive combustion flue gases.



Figure 11. Schematic of the reference cycle for the operation of a externally fired gas turbine.

Steam Rankine cycle is the common application. It has its main positive feature in insuring that the expanding fluid is completely isolated from the syngas combustion fumes, therefore avoiding

corrosion, fouling and plugging of the rotating parts. Moreover, due to the change of phase in the working fluid, the specific power of the machinery is extremely high. Rankine cycle power plants have a net electrical efficiency between 15% and 24% for small to medium plant capacities, reaching 32-36% for larger plants. They run at lower superheated steam temperature and pressure because of the possible presence of contaminants in the flue gas which can cause high temperature corrosion. These plants, for economic reasons, are not equipped with a reheater section and turbines are simpler and less efficient and condenser pressure is higher. Since very long maintenance cycles for continuous operation can be achieved, Rankine cycles are expensive for the cost of the heat recovery steam generator (HRSG), composed of economizer to warm up the water output the feed pump, evaporator to produce steam, and overheat system, a shell-and-tube exchangers containing the flowing steam, run over by the hot flue gases from to allow the heat transfer. As consequence, Rankine cycle plants are extremely large due to high space requirements for the boiler and the condenser and related to low electrical efficiency for small sizes, are justified only for MW-scale plants.

In thermodynamic point of view, the organic Rankine cycle (ORC) turbine is identical to the steam turbine in each and every aspect. In fact, the same name (Rankine) identifies the ideal reference cycle for both machines. The ORC differs in that it utilizes organic compounds as the working fluid, thus extending the choice of the possible temperatures in the boiler or heat exchanger. This, in consequence, allows for the use of "low" temperature heat sources, such as waste process heat. The technology is therefore considered opportunistic in that it is engineered to extract useful work from whatever heat streams might be available on site. This technology is widely used nowdays, but special attention must be drawn to the selection of the appropriate organic fluid and to the adjustment of the optimal operation parameter of the cycle, since these two factors play a significant role in the economic feasibility of the system. Most of the success of an ORC application lies on the ability to engineer the working fluid (single molecule or azeotropic mix) so that its phase transition curve encapsulates the temperature limits of the application. It is a technology in commercial operation in units ranging from a few kWe up to 2-3 MWe, with a low power-to-heat ratio resulting in limited electricity generation efficiency (15-18%), and with wide price ranges of around 1500-3000€/kWe, but highly standardized, reliable and utilized.


Figure 12. Schematic of the reference cycle for the operation of an organic Rankine cycle turbine

Syngas cleaning

The raw syngas obtained from gasification of biomass and waste contains tar, char, inorganic solid particulate, ammonia and other impurities. These contaminants must be removed before its utilization in a power generation equipment. Tars can foul heat exchange surfaces and engine valves or they may not burn adequately in combustion chambers of gas turbines. Alkali forms deposits on cold heat exchange surfaces and turbine blades where it promotes corrosion. Removal of these gas pollutants is essential to the long-term success of biomass or waste gasification process. Thereby, syngas cleaning is the part of the process where most of concerns are focused on.

Two gas treatment methods have been proposed in these studies: hot gas filtration and wet gas scrubbing. The basic idea is that in the first case the syngas is first burned and then cleaned while in the second case it is first cleaned and then burned. In hot gas filtration the gases are partially cooled down to about 400-500°C to condense alkali metal vapours onto particulate in the gas. Gas cooling is followed by a hot gas filter that removes both the particulate and the condensed alkali metals. Typically, the gas is then released to the gas turbine at relatively high temperatures of around 450°C that allow tars in the gas to be retained as vapours. In some cases, a sorbent addition occurs just after the cooling and the syngas is then sent to a ceramic filter to remove chlorine salts at a temperature higher than 350°C: this allows a high efficiency combustion in a boiler, where the tar heating content is also recovered. In the wet scrubbing approach the syngas is cooled under 150°C upstream of the wet scrubber inlet. This removes particulate, alkali metals, tars and soluble nitrogen compounds such as ammonia. Wet gas scrubbing is considered an

established gas cleaning technology, and when it is used is usual to incorporate thermal or catalytic cracking of the tars before gas clean-up to produce non-condensable hydrocarbon gases and so retain the chemical energy of the fuel gas.

Hot gas filters are currently subject of a great deal of research and development activity and are perceived to be the better solution if their technical problems can be overcome because the tars and sensible heat in the syngas are retained and the effluent stream that would be produced in wet scrubbing is avoided. Furthermore, hot gas cleaning is typically considered for gas turbine applications. Indeed, considering the lower tolerance of gas turbines to tar, and pressure losses incurred in scrubbers, cold gas cleaning is not necessarily attractive for GT. Internal combustion engines need to be fuelled with cold gas, therefore wet scrubbers are generally used to clean producer gas for this type of prime mover. Although gas cooling can simplify gas cleaning, it reduces the overall efficiency of the power cycle. This solution also implies the uneconomical disposal of tar contaminated liquid effluent.

Particulate control can be obtained by means of cyclones, wet scrubbers, ceramic filters, and electrostatic precipitators, with different levels of removal efficiency. Cyclone collectors are very effective for removing all but the finest particles and are frequently arranged in series, with the first designed to capture the largest particles and the second those progressively finer. Unfortunately, particles smaller than 5µm, which escape capture in cyclones, can degrade performance of many kinds of power systems. Ceramic filters consist of arrays of candle-shaped elements, which are effective in removing fine particles not captured by cyclones. Filter arrays are employed in series to ensure complete particulate removal. The ceramic candle filters that are currently available are made of clay-bonded silicon carbide, while the main body is composed of course-ground silicon carbide or aluminosilicate surrounds the main body. It is this outer layer that performs the filtering duties. Ceramic candles have collection efficiencies approaching 100%. However, there is a not excellent reliability of the ceramic filters when operated at extremely high temperatures (>800°C), as well as the possibility of chemical degradation, creep, and static fatigue failure over time. These concerns, and some negative experience in demonstration plants, led to the development of sintered metal filters, which, like ceramic filters, are barrier capable of high collection efficiencies for micron-sized particles. Metal filters are produced by filling molds with powdered metal and heating them to slightly below the melting point of the metal. Under these conditions, the metal particles sinter together to form a porous metal matrix. The advantage of ceramic filters is predominantly related to the material qualities of porous metals, which have

demonstrated good mechanical strength under constant and transient loads. In addition to these physical characteristics, they are resistant to the corrosion under typical gasification conditions. Granular bed filters, i.e. large bins containing a quantity of a granular material, typically made of limestone or alumina, could be an interesting alternative. They can be employed either as static or moving beds of granular material. In the first case, a dirty syngas stream flows down through the bed where particles impact and adhere to the granules: as particles accumulate in the bed, the void spaces become clogged, pressure drop increases and the bed must be cleaned, requiring an interruption in the filtering process. In the second case, this interruption can be achieved. In these filters the media flows downward through the filter, while dirty syngas flows upward through the bed, allowing a collection efficiency greater than 99% for particles greater than 4µm in size and more than 90% of smaller particles.

Tar control is among the greatest technical challenges to be overcome for a wider application of gasification systems. The types of tars produced (phenolic ethers, alkyl phenolics, heterocyclic ethers, polynucleic aromatic hydrocarbons (PAH), and larger PAH) is mainly a function of time and temperature over which reaction occurs. Methods to remove tars from producer gases fall in physical removal (separation), thermal conversion (cracking) and catalytic degradation. Physical processes for tar removal include wet scrubbers, demisters, wet granular bed filters and wet electrostatic precipitators, all only effective when the producer gas has been cooled to less than 150°C, which is thermodynamically inefficient for power systems. A second problem, as it has been mentioned above, is that the water used must be treated to remove tars before it can be disposed, and the cost of treatment may make really expensive these methods. Thermal conversion is achieved by passing the syngas through a second, high-temperature reactor where tars decompose or reform CO, H₂ and other light gases. Milne et al. (1998) concluded that temperatures in excess of 1000°C and reasonable residence times are necessary to destroy aromatic tars without the use of a catalyst, but this temperature presents material problems, requiring expensive alloys. Moreover, generally high-temperature reaction conditions produce soot, which may be even more problematic that tars. Addition of steam and/or oxygen is effective in increasing cracking efficiencies. Oxygen inhibits the formation of aromatics; steam produces fewer refractory tars, enhances phenol formation, reduces concentration of other oxygenates, affects for a limited extent the conversion of the aromatics, produces tars that are easier to reform catalytically, facilitates the water/gas shift reaction (greater amounts of steam increase H_2 and CO_2 and decrease CO and H_2O). Catalytic destruction can be accomplished by passing the

syngas in a bed of metallic, such as nickel or alumina, or non-metallic, such as dolomite or limestone, catalyst. In particular, metallic catalysts have been more successful, reducing tar levels by more than 90%, but are typically very expensive.

Alkali, especially potassium contained in biomass feedstocks, readily vaporizes during gasification and then condenses at a temperature below about 600°C. The resulting alkali aerosol has several detrimental effects. Deposited on metal surface is forms a sticky film that causes impacting particulate matter to adhere to the surface forming ash deposits that interfere with heat transfer through boiler tubes or with aerodynamics of turbine blades. Moreover, alkali metals are erosive to turbomachinery and corrosive to metal surfaces. If alkali exceeds limits recommended by turbine manufacturers (typically total alkali concentration at the turbine inlet must be under 24 ppb), then some method for controlling alkali emission is required: adsorption or leaching. Adsorption can occur in the bed itself if sorbents are mixed into the fluidized bed or it can take place in a separate, post-gasifier sorbent bed. The ideal adsorbent has a high-temperature capacity, a rapid rate of adsorption, and a high loading capacity. A number of materials have been tested including diatomaceous earth, kaolinite, and a variety of clays, but bauxite showed the most promise alkali removal efficiency (99%) and its regeneration is a relatively simple process, due to the physical nature of adsorption. Leaching is an alternative approach for alkali control, reducing its concentration in the feedstock before the injection into the gasifier. For example, in biomass alkali are typically present as water-soluble compounds, and washing the feedstock readily dissolves most of the alkali (more than 80% of potassium and sodium, as well as more than 90% of the chlorine).

On the basis of the considerations above, downstream of the results of the experimental pilot plant gasification tests, cleaning sections have been specifically designed for each fuel tested, taking into account the most promising plant configurations in order to obtain a syngas that could meet the requirements of each specific end-use device chosen for energy generation. In the case of wood biomass power gasification with a gas engine and a thermal gasification with an externally fired gas turbine processes were designed and quantified developing material and energy balances for the specific feedstock capacities selected. For the mixed plastic waste feedstock a designed process with a steam Rankine cycle was proposed as unique viable configuration for the plant capacities considered (4MWe). The configuration applies the solution with two twin gasifiers operated in parallel, providing a rather large plant capacity even when one of the two reactors is on maintenance, and was quantified for two different wastes as obtained with or without a pre-

selection process. For the packaging derived fuel for technical reliability and economical sustainability a power gasification configuration was designed for a plant capacity of 500kWe and then quantified developing mass and energy balances for three different solutions, power generation, heat and power cogeneration and district heating. In this work the various technological options examined apply to the base-load device, not considering peak-load. The heating is transferred via a district energy network to the final consumers. This district heating system consists of two main parts. Firstly, the long-distance heat transfer pipeline, which connects the CHP gasification plant with the terminal station and transfers only heat. The terminal station is located close to the district heating consumption and contains heat exchangers and the absorption chillers. The second part is the district energy distribution network, which connects the terminal station with the final consumers, supplying them with heat.

Economic assessments

The choice of appropriate conversion process is influenced by many factors, such as type and quantity of feedstock availability, end-use applications, environmental standards, and economic aspects. In order to assess plants feasibility and the impact of main variables, a comprehensive cost-estimating procedure has been preliminary established, and then coupled with an overall economic evaluation model able to indicate the effects of varying parameters values on plant costs and revenues. The homemade economic model used in these studies is based on the estimation of standard accounting items such as capital costs, taxation and direct revenues from the sale of the generated energy. All monetary values have been subject to time-value of money adjustments, i.e. future costs and revenues have been discounted to their present worth based on a fixed discount rate of 5% per year, even thought a range of variation of 2,5-7% has been taken into account in the sensitivity analysis. This is needed to compare investment options that might generate costs and revenues in different time points along their expected life.

In these studies capital costs represent the total plant costs (TPC), which cover main equipment costs, direct costs and indirect plant costs. The main equipment costs include biomass and waste pretreatment and storage, gasification system with heat exchangers, gas cleaning system and energy generation device. Direct costs cover costing for piping, auxiliary systems and services, electrical, instrumentation and control. Indirect cost include engineering and supervision, contingency and contractor fee. Annual amortization of total plant costs has been calculated as a constant rate of 6,7% that corresponds to an expected plant life on 15 years. This value of the working life of the plant has been assumed on the basis of the life of the end-use device, a proper

maintenance program, and the incentivized period. The scaling factor utilized for the equipment costs is based on a power law applied to estimates for the reference installation size obtained directly from manufacturers. A scale exponent equal to 0,6 was used, as it is derived from basic literature and recent works in the field.

Operating costs include maintenance, consumables and utility, waste streams disposal, labor and insurance. Maintenance costs cover both running and extraordinary repairs, and was calculated as a percentage of equipment costs. The plants would operate for 320-330 days a year, depending on the plant capacities and the end-use device applied. Consumables and utility are system capacity dependent, therefore their costs was evaluated for the installation size, while the electrical consumption by the auxiliary units was subtracted from the gross electrical output of the systems. Labor costs was determined at the recurring wage for different number of shifts of different workers, depending on the gasification plant size. For instance, kW-scale plants are capable to operating unmanned, therefore a single shift of a single worker was considered, MW-scale plants with Rankine cycle need of almost three shift of different worker. An average salary for simple worker (35k \in) and for skilled steam driver (50k \in) was assumed. The disposal cost of the waste streams amounts to the product of the mass flow rate of solid waste by a fixed disposal fee on 120€/t and of liquid waste by a disposal of 70€/t. These costs cover transport, treatment and landfill tax. Moreover, some plant configurations for biomass and waste present additional operating costs for residual stream disposal and energy demand produced by the in situ feedstock pre-treatment section. All costs have been calculated in today's money and then discounted according to the year in which they occur.

Revenues from different biomass and waste gasification plants depend on gate fees, sales of electricity and/or heat, renewable obligation certificates, "white certificates", and, in the case of waste feedstocks, avoided disposal costs. For these studies the Italian incentive scheme was adopted as the basis for the energy compensation estimation. For biomass power plants, an all-inclusive feed-in tariff of 0,28€/kWhe delivered to the grid was used even though a range of variation 0,21–0,35€/kWhe was then taken in consideration. The all-inclusive tariff encompasses compensation for the electrical energy sold and all the incentives associated with production of electricity from renewable resources and is valid for a period of 15 years. Access to the all-inclusive feed-in tariff, therefore excludes the attribution of renewable obligation certificates (green credits or green certificates) or other incentives under current Italian legislation. For waste power plants, it was assumed that revenues come from the sale of the electrical energy produced

and from the gate fee of MPW utilized as fuel. Tariff of 0,062€ per kWhe delivered to the grid has been used. The tariff encompasses compensation for the electrical energy sold, but not yet incentives associated with production of electricity from waste. A value of 0,065€/kWhe for the energy from waste recovery was taken in consideration for a valid period of 15 years. The fuel delivery revenues was assumed respectively equal to 0 and 95€/t for the two different mixed plastic waste utilized (indicated as SFA and EBR), even though ranges of variation of these values was taken into account. For the EBR solution a fuel feed rate in entrance to the plant 30% larger than that for SRA solution has been taken in consideration, as a consequence of the material losses from the pre-treatment section, corresponding to a moisture reduction of about 15% and an ash reduction of another 15%. Moreover, in the study applied to packaging derived fuel, "white certificates" calculated on the basis of the electrical and thermal MWh produced, are guaranteed for a period of 10 years and 15 years respectively for combined heat and power (CHP) plants and, in case of demand, for district heating (DH). A conservative tariff of 0,08€/kWht has been used for the sale of saturated or superheated steam production in CHP plant, and for the sale of hot water in case of demand of district heating occurs. Compatibly with the Italian waste market and the tariff applied by the Italian National Consortium for Packaging (CONAI) revenues as avoided food packaging disposal has been assumed equal to 30€/t, even though ranges of variation of these values have been taken into account.

Standardized financial tools, such as the net present value (NPV), the internal rate of return (IRR) and the payback time (PT), were employed to assess the profitability of different plant solutions. An option is economically attractive if it has the highest IRR and the NPV is higher than zero. The NPV refers to the difference between the present values of all costs and associated revenues, therefore the IRR was calculated as the discount rate that makes the NPV equal to zero.

For a 200kWe biomass power plant, gas engine solution offers higher global efficiency than gas turbine (about 27% vs 23%) and lower capital cost $(6000 \in /kWe vs 7600 \in /kWe)$, but has very higher maintenance costs $(449(\in/y)/kWe vs 115(\in/y)/kWe)$ due to the wastewater treatment unit downstream of the wet scrubber and a light lower availability (7680 h/y vs 7920h/y). The EFGT solution must dispose a solid waste stream downstream of the air pollution control system instead of a liquid one $(0,11kg/kg_{fuel})$ but present the disadvantage of a very larger mass of flue gases to be treated at the stack (32,3kg/kg_{fuel}vs 6,6kg/kg_{fuel}).

Moreover this solution is more affected by the biomass cost due to the lower specific biomass conversion rate $(1,02kWhe/kg_{fuel} vs 1,20kWhe/kg_{fuel})$. For all investigated plant sizes (100-

600kWe), also if both installations benefit of a power scale effect in the cost function, the GE solution has always lower total plant costs than the EFGT alternative, but decrease in cost is expected for future EFGT installations due to economy attainable by the "nth plant effect". The gap in the operating costs between the two alternatives is mainly due to the different maintenance costs of the rotating equipment. Moreover, despite having a 17,8% higher annual electricity yield, GE's cash flow is always lower than the EFGT one due to the operating costs. Both plants become financially attractive for a nominal plant capacity larger than 200 kWe (IRR >12%). Anyway, the EFGT's lower operating costs cannot compensate for the higher capital costs and the internal rate of return is always favorable to the GE alternative for nominal plant capacities equal or larger than 200 kWe (32% vs 26% for a 600kWe). This is indicated even by the payback time values, equal to 3 years and 4 years at a nominal plant capacity of 600 kWe.

	200kWe WB		200kWe PDF			4 MWe MPW	
	GE	EFGT	PP	CHP	DH	EBR	SRA
Energy Performance							
Total energy conversion efficiency, %	27,1	23	23,8	78,2	78,2	23,7	23,7
Specific fuel conversion rate, kWhe/kg _{fuel}	1,2	1,02	0,97	3,2	3,2	2,09	2,65
Environmental Performance							
Liquid export, kg/kg _{fuel}	0,11	-	0,035	0,035	0,035	-	-
Solid export, kg/kg _{fuel}	-	0,01	0,033	0,033	0,033	0,44	0,25
Gas export, kg/kg _{fuel}	6,64	32,3	7,96	7,96	7,96	18,9	23,1
Economic Performance							
Total plant costs, €/kWe	6000	7600	4860	5040	7440	4790	4360
Operating costs, (k€/y)/kWe	0,94	0,69	0,53	0,54	0,63	0,74	0,62
Average cash flow, (k€/y)/kWe	0,96	1,21	0,35	1,55	1,56	0,65	0,36
Internal rate of return, %	13,2	13	0,5	29,8	18,9	8,3	n.a.

Table 10. Summary of energetic, environmental and economic performances for biomass- and waste-toenergy gasification plants.

For a 4MWe mixed plastic waste power plant, steam turbine solution offers a global efficiency of 23,7%, but higher capital cost is needed for EBR respect to SRA feedstock ($4790 \in /kWe$ vs $4360 \in /kWe$). The solution fuelled with EBR infact is affected by the lower specific feedstock conversion rate (2,09kWhe/kg_{fuel} vs 2,65kWhe/kg_{fuel}) and by the installation of pretreatment equipment. Moreover it must dispose higher solid waste stream due to the in-situ fuel pretreatment (0,44kg/kg_{fuel} vs 0,25kg/kg_{fuel}), but treat lighter mass of flue gases at the stack (18,9kg/kg_{fuel} vs 23,1kg/kg_{fuel}). On the other hand SRA solution is negatively affected by a lower

average cash flow imputable to the lack of revenues from the fuel delivery (instead of 1818k€/y for EBR). As consequence, for all investigated plant sizes (2-6MWe), also if both installations benefit of a power scale effect in the cost function, the SRA solution show always no achievable internal rate of return. Instead EBR plants become financially attractive for a nominal plant capacity larger than 5 MWe (IRR >12%).

For a 500kWe PDF plant, gas engine solution offers interesting global electrical efficiency (about 24%) for power generation (PP) and also good thermal efficiency (about 54%) in the case of cogeneration (CHP) or district heating (DH). Power production configuration show the lowest capital cost (4860€/kWe, instead of 5040€/kWe and 7440€/kWe respectively for CHP and DH), and the lowest operating costs (530(€/y)/kWe, instead of 540(€/y)/kWe and 630(€/y)/kWe respectively for CHP and DH) because the other solutions need of more heat exchangers to recovery the heat by the hot fumes leaving the engine and of an expensive network to guarantee the transport of hot water to the users respectively. However, CHP and DH solutions show higher average discounted cash flow (1550(€/y)/kWe and 1560(€/y)/kWe) and IRR (29,8% and 18,9%) due to the greater revenues from the sold of thermal energy produced and to the incentive life. On the contrary, the PP solution is affected by an extremely low ADCF and the not achievable IRR for all the investigated plant sizes (0,2-1MWe), imputable to the lack of revenues from the thermal energy sold, undermine the plant economic feasibility.

The sensitivity analysis of the main variables on the main economical parameters (ADCF and IRR) indicates always the crucial role of the feed-in tariff for biomass and mixed plastic waste plants. This means that the absence of an adequate incentive policy may undermine the economic sustainability of the biomass- and waste-to-energy plants. For instance, a 4 MWe plastic-to-energy plant needs a feed-in tariff of 0,151€/kWhe to reach the minimum attractive rate of return (MARR). Moreover, for the MPW gasifiers the sensitivity related to the fuel delivery revenue appears not negligible since it determines a remarkable change of the IRR. As expected, the gasifier performance has a not relevant role in the assumed range of variation of cold gas efficiency for each study, since the extremes of the interval however represent very good reactor performances.

Paper IV.

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A techno-economic comparison between two design configurations for a small scale, biomass-to-energy gasification based system

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ABSTRACT

Biomass has great potential as a clean and renewable feedstock for producing modern energy carriers. This paper focuses on the process of biomass gasification, wherein the synthesis gas is subsequently used to produce electricity. A comparison between the most promising design configurations for the industrial application of gasification based, biomass-to-energy cogenerators in the 100–600 kWe range is presented. Mass and energy balances and material and substance flow analyses drawn for each design solutions are based on the experimental data obtained from a pilot scale bubbling fluidized bed air gasifier, having a feeding capacity of 100 kg/h and operated with a commercially available, natural biomass. Measurements taken during the experimental tests include the syngas complete composition as well as the characterization of the bed material, the entrained fines collected at the cyclone and the purge material from the scrubber. The techno-economic performances of two energy generation devices, a gas engine and an externally-fired gas turbine, have been estimated on the basis of the manufacturer's specifications. The study concludes that the internal combustion engine layout is the solution that currently offers the higher reliability and provides the higher internal rate of return for the investigated range of electrical energy production.

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1. Introduction and framework

Biomass is the oldest known source of energy and it is a renewable energy. The possible utilization of the biomass energy content gained a great interest in the last decade, because of its potential to displace a large part of conventional fossil fuel for electricity production. The main reasons lay in the large availability of biomass resources, the progressive depletion of conventional fossil fuels and the potential better air pollution control of the related power generation processes [1-3]. A large amount of energy is in fact potentially available from biomass, since sources that can be used for energy production cover a wide range of materials (wood and wood waste, agricultural crops and their waste by-products, organic fraction of municipal solid waste, residues from agro-industrial and food processes, aquatic plants such as algae and waterweeds). Moreover, the limitate sulphur and greenhouse gas emissions associated with the use of biomass for energy production could respond to the growing pressure for the achievement of a better environmental sustainability of power generation processes.

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Despite the widely agreed potential of bioenergy utilization, key problems regarding the use of biomass remain the unsteady availability, related to biomass seasonality and geographical distribution over the territory that often make the logistics (collection, transport and storage operations) complex and expensive [2], as well as the necessity of an energy production which should be not only environmental sustainable but also economic competitive. In other words, biomass has great potential as a renewable and relatively clean feedstock for producing energy carriers, such as electricity and transportation fuels, but in order to compete with fossil energy sources it needs to utilize efficient conversion technologies [4,5].

Biomass can be converted to a wide variety of energy forms (electricity, process heat for industrial facilities, domestic heating, vehicle fuels) by means of a number of thermochemical and biochemical processes [3]. With reference to low-value lignocellulosic biomass, biological conversion processes still faces challenges in low ecomomy and efficiency, even though fermentation and anaerobic digestion are today commercially proven technologies, suitably used to produce ethanol from biomass containing sugar [6–8] and biogas from high-moisture content biomass, such as the organic fraction of MSW [9].

Combustion, pyrolysis and gasification are the three main thermochemical process solutions. Combustion is traditionally used to convert biomass energy into heat and power in the process indus-

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try: the net conversion efficiency is generally low, even if higher values may be obtained in co-combustion in coal-fired power plants [2]. Pyrolysis is the thermal degradation of biomass in a bio-oil, a solid fraction and a high-heating value gas: a wide application is still restricted by difficulties in the efficient processing of bio-oil [3]. Gasification converts biomass in a combustible gas mixture (called producer gas or syngas), mainly made of carbon monoxide, hydrogen and lower content of methane and able to provide a wide range of products, extending from clean fuel gas and electricity to bulk chemicals [10,11].

Different gasification technologies are available today and fluidization is the most promising among all of them, for a series of reasons, among which the possibility to use different fluidizing agents, reactor temperatures and gas residence times, to inject reagents along the reactor height and to operate with or without a specific catalyst [12,13]. The key to achieving economically and environmentally efficient energy recovery from natural and waste biomass gasification is to overcome the problems associated with the formation and release of different contaminants (mainly tars, i.e. high molecular weight hydrocarbons that condensate at ambient temperature, but also heavy metals, halogens and alkaline compounds) that have an environmental and operating negative impact. The syngas cleaning approaches can be classified in treatments inside the gasifier (primary methods), such as adequate selection of main operating parameters, use of a proper bed additive or catalyst, specific gasifier design modifications, and hot gas treatments downstream of the gasifier (secondary methods), such as thermal or catalytic tar cracking and mechanical methods (ceramic, fabric or electrostatic filters, cyclones and wet scrubbers) [14,10,13]. The type and the possible combination of primary and secondary methods are strongly dependent on the nature of biomass fuel and gasification technology as well as on the level of syngas cleaning required by the specific end-use device.

The aim of this study is to evaluate and compare the technical and economic performance of the most promising design configurations for the small scale industrial application of gasification-based biomass-to-energy cogenerators. To this end, a number of tests with a selected natural biomass was carried out in a pilot scale bubbling fluidized bed gasifier (BFBG). The collected experimental data were processed by different analytical tools such as mass and energy balances and material and substance flow analyses, in order to obtain information useful to define design solutions and configurations suitable for different electricity generation devices. The energy conversion devices for the range of electric output of interest, among all those commercially available, are then analyzed and selected. The technical and economic performances of the best two plant configurations are finally described in details and compared.

2. The pilot scale fluidized bed gasifier

The utilized pilot scale bubbling fluidized bed gasifier has the characteristics schematically listed in Table 1. An olivine – a magnesium-iron silicate, $(Mg,Fe_2)SiO_4$ – was selected as material for the fluidized bed on the basis of results of previous investigations carried out on the same pilot-scale BFBG [15] and those reported on the scientific literature [16,17]. All indicated olivine as an interesting candidate to act as a bed catalyst for the tar cracking reactions in biomass gasification, even taking into account its low cost and excellent resistance to attrition in the fluidized bed reactor. The main characteristics of the utilized olivine are reported in Table 2.

In the reported experiments, air was used as reducing agent and always injected at the bed bottom while the fuel was always fed by means of an over-bed feeding system. The fluidizing air stream was

Table 1

Main design and operating features of the utilized pilot scale bubbling fluidized bed gasifier.

Geometrical parameters	ID: 0.381 m; total height: 5.90 m; reactive zone height: 4.64 m; wall thickness:12.7 mm
Feedstock capacity	100 kg/h
Thermal output	Up to about 500 kW
Typical bed amount	145 kg
Feeding system	Over-bed air-cooled screw feeder
Gasifying agents	Air, oxygen, steam, carbon dioxide
Range of bed temperatures	700–950 °C
Range of fluidizing velocities	0.3-1 m/s
Flue gas treatments	Cyclone, scrubber, flare
Safety equipments	Water seal, safety valves, rupture disks, alarms, nitrogen line for safety inerting

heated up to 545 °C by a couple of electric heaters before entering the reactor. The fuel and blast flow rates were mutually adjusted so that, at the fixed fluidizing velocity, the desired equivalence ratio ER was obtained (where ER is defined as the ratio between the oxygen content of air supply and that required for the stoichiometric complete combustion of the fuel effectively fed to the reactor). The cylindrical BFB reactor was heated up to the reaction temperature by the sensible heat of pre-heated blast gases and by a set of three external electrical furnaces. The gas generated in the reactor was sent to the syngas conditioning section composed of a high efficiency cyclone and a wet scrubber (for the removal of tars, residual fly ashes and acid gases) and finally incinerated by a safety flare. An accurate description of the plant and of experimental procedures is provided elsewhere [13,18]. Here it is sufficient to highlight that gas composition, upstream and downstream of the syngas conditioning section, was on-line measured by IR analyzers for the main syngas components (carbon monoxide and dioxide, hydrogen, methane) and by two micro-gas-chromatographs equipped with different columns for the detection of lighter and heavier hydrocarbons as well as of carbon monoxide and dioxide, hydrogen, nitrogen and water. Two different methods of tar evaluation were used: the first conservatively imputes to the tar amount the whole carbon loading which, as a result of a mass balance on atomic species, cannot be attributed either to the produced gas or to the solids collected at the cyclone or present inside the bed; the second method utilizes samples taken at the reactor exit, for about 30 min, by means of four in-series cold traps, and then sent to a gas chromatograph coupled with a mass spectrometer. Data obtained from on-line and off-line gas measurements and those from chemical analyses of solid samples were processed to develop complete mass balances on atomic species and the related energy balance for each run. The flow rate of produced syngas was determined by the "tie component" method

Table 2

Characteristics of the olivine particles utilized as bed material in the pilot scale bubbling fluidized bed gasifier.

Mineral	Mg-Fe silicate
Chemical composition, %	
SiO ₂	39-42
MgO	48-50
Fe ₂ O ₃	8-10.5
CaO	<0.4
K ₂ O	-
TiO ₂	-
Al ₂ O ₃ , Cr ₂ O ₃ , Mg ₃ O ₄	0.8
LOI (loss of ignition)	0.20
Ci	200 400
Size range, µm	200-400
Sauter mean diameter, µm	298
Particle density, kg/m ³	2900
Minimum fluidization velocity (at 850 °C), m/s	0.030
Terminal velocity (at 850 °C), m/s	2.0

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Table 3

Chemical characterization of the reference biomass
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Ultimate analysis, % on weight basis 45.9 (45.7-46.1) M (min-max) 5.63 (5.60-5.66) N (min-max) 0.33 (0.30-0.36) S (min-max) 0.33 (0.30-0.36) S (min-max) 0.01 Moisture (min-max) 7.0 (5.9-7.1) Ash (min-max) 1.3 (1.2-1.4) O (by difference) 39.83 C:O ratio 1.15 Proximate analysis, % on weight basis 7.0 (6.9-7.1) Noisture (min-max) 7.0 (6.9-7.1) Volatile matter (min-max) 7.0 (7.0-7.4) Fixed carbon (min-max) 1.3 (1.2-1.4) Chemical analysis, g/100 g 1.3 (1.2-1.4) Cellulose ⁴ 45.1 Hemicelluloses ^b 19.6 Lignin ⁶ 22.3 Heating value (by the relationship of Sheng and Azevedo [27]) Heating value (by the relationship of Sheng and Azevedo [27]) HHV, k/jkg 18,600 LHV, k/jkg 15,900		
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N (min-max) 0.33 (0.30-0.36) S (min-max) 0.01 Moisture (min-max) 7(6.9-7.1) Ash (min-max) 1.3 (1.2-1.4) O (by difference) 39.83 C:O ratio 1.15 Proximate analysis, % on weight basis 7.0 (6.9-7.1) Volatile matter (min-max) 7.0 (6.9-7.4) Fixed carbon (min-max) 1.9.7 (19-20) Ash (min-max) 1.3 (1.2-1.4) Chemical analysis, g/100 g Cellulose ⁴ Cellulose ⁴ 45.1 Hemicelluloses ^b 19.6 Lignin ⁴ 22.3 Heating value (by the relationship of Sheng and Azevedo [27]) 1HV, kJ/kg HHV, kJ/kg 18,600 LHV, kJ/kg 15,900	H (min-max)	5.63 (5.60-5.66)
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Proximate analysis, % on weight basis Moisture (min-max) 7.0 (6.9–7.1) Volatile matter (min-max) 72.0 (70–74) Fixed carbon (min-max) 19.7 (19–20) Ash (min-max) 1.3 (1.2–1.4) Chemical analysis, g/100 g Cellulose ^a 45.1 Hemicelluloses ^b 19.6 Lignin ^c 22.3 Heating value (by the relationship of Sheng and Azevedo [27]) HHV, kJ/kg 18,600 LHV, kJ/kg 15,900	C:O ratio	1.15
Moisture (min-max) 7.0 (6.9-7.1) Volatile matter (min-max) 72.0 (70-74) Fixed carbon (min-max) 19.7 (19-20) Ash (min-max) 1.3 (1.2-1.4) Chemical analysis g/100 g Cellulose ^a 45.1 Hemicelluloses ^b 19.6 Lignin ⁶ 22.3 Heating value (by the relationship of Sheng and Azevedo [27]) HHV, kJ/kg 18,600 LHV, kJ/kg 15,900	Proximate analysis, % on weight basis	
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Fixed carbon (min-max) 19.7 (19-20) Ash (min-max) 1.3 (1.2-1.4) Chemical analysis, g/100 g 6 Cellulose ^a 45.1 Hemicelluloses ^b 19.6 Lignin ⁶ 22.3 Heating value (by the relationship of Sheng and Azevedo [27]) 1HHV, kJ/kg HHV, kJ/kg 15,900	Volatile matter (min-max)	72.0 (70-74)
Ash (min-max) 1.3 (1.2-1.4) Chemical analysis, g/100 g Cellulosea 45.1 Hemicelluloses ^b 19.6 Lignin ⁶ 22.3 Heating value (by the relationship of Sheng and Azevedo [27]) HHV, kJ/kg 18,600 LHV, kJ/kg 15,900	Fixed carbon (min-max)	19.7 (19-20)
Chemical analysis, g/100 g Cellulose ^a 45.1 Hemicelluloses ^b 19.6 Lignin ^c 22.3 Heating value (by the relationship of Sheng and Azevedo [27]) HHV, kJ/kg HHV, kJ/kg 18,600 LHV, kJ/kg 15,900	Ash (min-max)	1.3 (1.2-1.4)
Cellulose ^a 45.1 Hemicelluloses ^b 19.6 Lignin ^c 22.3 Heating value (by the relationship of Sheng and Azevedo [27]) HHV, kJ/kg HHV, kJ/kg 18,600 LHV, kJ/kg 15,900	Chemical analysis g/100 g	
Hemicelluloses ^b 19.6 Lignin ^c 22.3 Heating value (by the relationship of Sheng and Azevedo [27]) HHV, kJ/kg 18.600 LHV, kJ/kg 15,900	Cellulose ^a	45.1
Lignin ^c 22.3 Heating value (by the relationship of Sheng and Azevedo [27]) HHV, kJ/kg 18,600 LHV, kJ/kg 15,900	Hemicelluloses ^b	19.6
Heating value (by the relationship of Sheng and Azevedo [27]) HHV, kJ/kg 18,600 LHV, kJ/kg 15,900	Lignin ^c	22.3
Heating value (by the relationship of Sheng and Azevedo [27]) HHV, kJ/kg 18,600 LHV, kJ/kg 15,900		
HHV, kJ/kg 18,600 LHV, kJ/kg 15,900	Heating value (by the relationship of Sheng and Aze	evedo [27])
LHV, kJ/kg 15,900	HHV, kJ/kg	18,600
	LHV, kJ/kg	15,900

^a As obtained by the value of acid detergent fiber (ADF) less that of acid detergent lignin (ADL).

 b As obtained by the value of neutral detergent fiber (NDF) less that of acid detergent fiber (ADF).

^c As obtained by the value of acid detergent lignin (ADL).

[19] applied to the value of nitrogen content in the dry syngas, as obtained by (on-line and off-line) GC measurements.

3. The configurations of the biomass-to-energy system

The configurations of the gasification based, biomass-to-energy system investigated in this study were defined on the basis of the following design specifications. The plant is designed to be fed with a natural biomass: a commercially available beechwood for domestic heating, having the chemical characteristics reported in Table 3. The process is designed to produce electricity, even though additional thermal energy is available to use in case a demand is present at the installation site. The electrical size range of interest is that together with the evidence that fluidized reactors allow a continuous operation, a sufficient flexibility on biomass feedstock and a limited tar content in the syngas [10,11] lead to individuate the atmospheric bubbling fluidized bed air gasification as the conversion process to be adopted.

The design configurations for the industrial application of gasification plants in the range of interest can be sketched as a combination of three sections: syngas production, syngas utilization and syngas or flue gas cleaning. The first defines the syngas that can be produced and then, for fixed biomass fuel and gasification technology, the quantity and quality of this syngas. The utilization section indicates the producer gas that can be utilized in a specific energy conversion device and then, for a given machinery (steam turbine, gas engine, internally or externally-fired gas turbine), its temperature, heating value and cleaning level (i.e. tar and dust content but also that of alkaly and inorganic contaminants). The relative succession of the utilization and cleaning sections depends on the two possible types of biomass-to-energy gasification system that can be adopted: the "power gasification", where the producer gas is first cleaned then burned, and the "heat gasification", where the producer gas is first burned then cleaned [11]. Then, for a "power gasifier" the cleaning section must function as an interface between the characteristics of the producer gas and those required by the specific generator set, even though the conditioning of the gas up to the specifications imposed by the generator

Table 4

Operating conditions and performance parameters of the pilot scale gasifier under two values of equivalence ratio.

Operating conditions		
ER (equivalence ratio)	0.23	0.28
AF (air/fuel ratio), kg _{air} /kg _{fuel}	1.26	1.53
Temperature of fluidizing air at gasifier entrance, °C	545	545
Output process data		
Temperature of fluidized bed at thermal steady-state, °C	810	880
Temperature of syngas at gasifier exit, °C	680	740
Q _{syngas} , m ³ _N /kg _{fuel}	1.8	2.1
LHV _{syngas} , kJ/m ³ _N	6800	5900
Specific energy, kWh/kg _{fuel}	3.4	3.4
CGE (cold gas efficiency)	0.77	0.77
Composition of syngas (downstream of cyclone and scrubbe	г), %	
N ₂	47.6	50.7
CO ₂	16.0	14.0
CO	16.9	17.9
H ₂	12.5	12.3
CH ₄	5.0	3.9
C ₂ H ₄	1.2	0.8
C ₂ H ₆	0.17	0.04
C ₂ H ₂	0.08	0.08
C ₃ H ₆	0.06	0
C ₆ H ₆	0.26	0.25
C ₇ H ₈	0.10	0.02
C ₈ H ₁₀	0.05	0.01
Syngas contaminants (upstream of cyclone and scrubber)		
Entrained fines, g/kg _{fuel}	26.2	20.9
Entrained carbon fines, g _C /kg _{C-fuel}	43.3	31.2
PAH, mg/m ³ _N	450	2300
HCl, mg/m ³ _N	17	13
H ₂ S, mg/m ³ _N	0.7	1
NH ₃ , mg/m ³ _N	14	16

set is not always economically viable [14,20]. Instead, for a "heat gasifier", it consists of a possible pre-treatment of the syngas to remove contaminants (such as hydrogen chloride) before it goes into the combustor and, above all, of an air-pollution control (APC) system for flue gas cleaning.

The following paragraph investigates the syngas characteristics that can be obtained by a BFBG fired with the design biomass fuel, mainly on the basis of the experimental activity carried out with the described pilot scale gasifier. The energy conversion devices for the range of electric output of interest, among all those commercially available, are then described and selected. The cleaning sections that complete the two most promising plant configurations are finally defined.

3.1. The gasification section

The gasification section has been designed on the basis of an experimental activity carried out on the pilot scale BFBG operated under autothermal conditions, i.e. with the only external heat addition being provided for the pre-heating of the reducing and fluidizing air stream. The reactor was operated with the natural biomass, in a bed of olivine particles fluidized at a velocity of 0.6 m/s, a bed temperature of about 850 °C, an air preheating temperature of 545 °C and with an equivalence ratio ER between 0.2 and 0.3. The performances of the BFBG were measured and recorded only when the chemical composition of the produced syngas and the temperature profile along the reactor reached stedy-state conditions. The obtained results, reported in Table 4 for two values of ER, have been combined with a recently defined environmental assessment tool, the Material Flow Analysis, which is named Substance Flow Analysis when it is referred to a specific chemical species. MFA/SFA is a systematic assessment of the flows and stocks of materials and elements within a system defined in space and time. It connects the sources, the pathways, and the intermediate



Fig. 1. Layers of mass and energy balances throughout the pilot scale gasifier in its present configuration: (A) total mass (kg/h); (B) carbon element (g/h); (C) feedstock energy (MJ/h).

and final sinks of each species in a specific process [21]. These characteristics make MFA/SFA attractive as a decision support tool, as showed by its utilization in process evaluation of waste treatments and recycling options [22] and in waste management planning [23]. In this study MFA/SFA was used to deeper understand the performance of the pilot scale gasifier and to define and quantitatively assess some design solutions and operating criteria of the biomass gasification system.

The quantified flow diagrams reported in Fig. 1 are the result of the MFA/SFA applied to the main process units (gasifier, cyclone, wet scrubber, water treatment system) of the pilot scale gasification system, when operated at an equivalence ratio of 0.28. Each flow in entrance to or in exit from a specific unit is identified by means of a black arrow if the specific data have been measured or fixed, or by a grey arrow if the data have been obtained by means of MFA/SFA. The layer of total mass flow rate is reported in Fig. 1A. The input flows to the BFBG unit are the stream of biomass fuel, that of a small flow rate of nitrogen utilized to facilitate the fuel injection and that of air used as reducing agent and fluidizing gas. The output flow stream is the obtained syngas, which still contains heavy hydrocarbons, inorganic pollutants and entrained fines. The dirty syngas is sent to the cyclone for dust abatement and then to the wet scrubber for removal of tars and inorganic compounds. The specific production of syngas is equal to 2.45 kg_{syngas}/kg_{fuel} (i.e. $2.1 \text{ m}^3_{N \cdot syngas}/kg_{fuel}$) while that of elutriated fines is $20.9 \text{ g}_{fines}/kg_{fuel}$. The stock of 145 kg of bed particles is progressively incremented (0.30 kg/h) as a result of opposite effects of elutriation losses and fuel ash accumulation. The experimental activity provides the complete chemical composition of streams leaving the cyclone and the water treatment system. These data have been used for the substance flow analysis of carbon, iron, magnesium and other elements and for the feedstock energy flow analysis, as made in a similar study [24].

Fig. 1B reports the result of the mass balance applied to the carbon element, i.e. the carbon layer of SFA. It gives the carbon conversion efficiency CCE, defined as the ratio between the mass flow rate of the carbon present in the syngas as CO, CO₂, CH₄ and light hydrocarbons (until C_5H_m) and the mass flow rate of the carbon that enters the reactor with the fuel. The value of 0.96 of CCE is evaluated as the ratio between the mass flow rates of the syn

gas carbon stream, F7, and fuel carbon stream, F1. CCE is mostly affected by the carbon losses related to the fly ash stream, F6 (for 3.1%) and, for an almost negligible fraction (0.6%), to those of purge stream, F8. The carbon layer finally reports an important state variable of the biomass gasification process, the bed carbon loading $W_{\rm C}$, which is the amount of carbon present in the bed as char particles at the steady-state condition [25]. Its value of 1.45 kg is a function of bed temperature and equivalence ratio. Fig. 1C reports the layer of feedstock energy, i.e. the heat of combustion of each input and output streams [26]. The energy flow entering with the biomass fuel has been determined by means of a relationship recently proposed and validated specifically for biomass fuels [27], while the energy flows of exit streams have been evaluated on the basis of the heats of combustion of the specific substances. The resulting difference in feedstock energy, 151 MJ/h, is that "invested" at the steady-state condition to convert the solid biomass in a gaseous fuel. Reported data allow to evaluate the cold gas efficiency CGE, defined as the ratio between the chemical energy of obtained syngas and that of injected fuel: the value of 0.765 is mainly determined by the chemical energy utilized inside the gasifier (19.5%) and, for a smaller part, by the fraction of feedstock energy lost with the entrained fines (3.2%) and with the heavy hydrocarbons of the purge stream from the water treatment system (0.8%).

These results suggest two possible design solutions: the makeup of bed olivine particles and the recycle of entrained fines. In particular, the latter could lead to some advantages. The first is an increase of both CCE and CGE as a consequence of the additional residence time of carbon fines inside the reactor, by taking into account that the reactivity of these fines has been demonstrated to be sufficiently high by a parallel investigation carried out by means of a thermo-gravimetrical balance [24]. The consequent advantage is that there is no necessity for a further treatment or disposal of these fines. Another advantage of fly ash recycle is the reinjection inside the gasifier of large part of escaped inorganic fraction, which could limit the entity of olivine make-up. Fig. 2 reports the results of material and substance flow analyses in the gasifier design solution that includes the recycling of fines into the reactor and shows the increased values of syngas yield, carbon conversion efficiency and cold gas efficiency. These data were finally combined with relationships of fluidization engineering [28] in order to determine the



Fig. 2. Layers of mass and energy balances throughout the pilot scale gasifier in the configuration with the recycling of entrained fines: (A) total mass (kg/h); (B) carbon element (g/h); (C) feedstock energy (MJ/h).

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Table 5

Advantages and disadvantages of different energy conver-	on devices for syngas	from biomass gasification.
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Energy conversion device	Net electrical efficiency of gasification plant	Main advantages	Main disadvantages
Steam turbine	10–20%	•Turbine components are isolated from combustion products •Long maintenance intervals, high availability •High specific work (kJ/kg yielded for working fluid)	•Expensive •Electrical efficiency is low at small sizes •Partial load decreases efficiency significantly •Plants is extremely large due to space requirements for the condenser and the boiler
Gas turbine	15-25%	•Electrical efficiency is good even at small sizes •Compact assembly •Long maintenance intervals, high availability •Ideal for cogeneration plants (CHP) due to high exhaust temperatures	•Turbine components are exposed to combustion products •Partial load decreases efficiency significantly •Moderately expensive
Externally fired gas turbine	10–20%	•Turbine components are isolated from combustion products •Electrical efficiency is acceptable even at small sizes •Long maintenance intervals, high availability •Ideal for cogeneration plants (CHP) due to high exhaust temperatures	•Expensive •Heat exchanger is exposed to high temperature, aggressive combustion gases •Partial load decreases efficiency
Gas engine	13–28%	•High electrical efficiency also at small sizes •Relatively inexpensive •Durable and reliable •Partial load effects efficiency only marginally	•Engine components are exposed to combustion products •Short and expensive maintenance intervals, low availability

main geometrical parameters of the gasification section. In particular, the reactor diameter was determined, for the fixed nominal plant capacity, on the basis of the cold gas efficiency and equivalence ratio, by keeping fixed the fluidizing velocity and the type and size of bed materials while the reactor height was determined by means of the Zenz and Weil relationship [29] in order to minimize the entrainment of fines from the bubbling bed gasifier.

3.2. The energy generation section

The list of possible devices that can be used to convert the syngas into electricity are schematically listed and compared in Table 5. Each of them has its advantages and disadvantages when coupled with a BFB gasifier.

The steam turbine and boiler combination has its main positive feature in insuring that the expanding fluid is completely isolated from the syngas combustion fumes, therefore avoiding the corrosion, fouling and plugging of the rotating parts. Moreover, due to the change of phase in the working fluid, the specific power of the machinery is extremely high. Commercially available steam turbines in the size range considered for this study, have an extremely low net electrical efficiency [10–20%] and additionally require a large condenser if the steam cycle is to be run in a closed loop configuration [30]. The intensive capital costs and the limited performance of the boiler and steam turbine configuration lead to the exclusion of this solution as a viable one [31].

Another combination that was not further analyzed is that with an internal combustion gas turbine. Although internal combustion gas turbines offer very good net electric efficiency across small size ranges, the direct combustion and expansion of the syngas and its fumes into the turbomachinery poses technical difficulties [31]. In fact, decontaminating the syngas of particulate, tar, alkali and acids to manufacturer's specification if often unfeasible due to incongruent costs of the equipment for the size range of the installation. Conversely, designing for costs can lead to residual contamination that fails to meet manufacturer's specifications which can cause unpredictable shortening of life or major failures of the machinery.

Recently a customization of the basic gas turbine machine has been readied for commercialization that overcomes the main problems associated with internal combustion gas turbines. This configuration is named either externally-fired gas turbine or hot-air gas turbine, since the working fluid is ambient air and the heat addition happens in a gas-gas high temperature exchanger [32]. The separation of the working fluid from the combustion fumes assures that the rotating parts are not deteriorated, fouled or plugged, as for a steam turbine, while the use of the exhaust clean hot air from the turbine outlet as the oxidizing gas in the syngas combustion, assures that high thermodynamic efficiencies are achieved.

The last solution that has been investigated is a syngas optimized high efficiency alternating engine. This type of engine is a proven technology that yields high electrical efficiency but has somewhat stringent requirements on both purity and technical conditions for the syngas supply [33,34]. In the case of the gas engine setup though, the decontamination of the syngas can be achieved with a sufficiently inexpensive equipment, an aspect that renders the solution viable and competitive. In fact, the engine based installation is usually regarded as the standard against which other alternatives have to be compared in terms of electrical and economical efficiency.

As mentioned above, the cleaning section must combine the characteristics of the produced syngas and those required by the specific generator set. On the basis of the preliminary selection process illustrated above, the cleaning section has been designed for with a gas engine and a "heat gasifier" with an externally-fired gas turbine. The following paragraphs present a detailed analysis and the quantified process flow diagrams (PFDs) of these configurations, on the basis of the mass and energy balances developed for a plant feedstock capacity of 100 kg/h (i.e., about 750 t/y) of the selected biomass fuel, which corresponds to a net electric power output of about 100 kWe.

4. The gas engine configuration

The process flow diagram devised for the gas engine solution is reported in Fig. 3. The configuration is composed of three sections, the gasification, the cleaning and conditioning and the electricity generation sections. In the following paragraphs, the role and main characteristics of the single components of each section are schematically described. It is noteworthy that, while the gasification section has been modelled by using experimental data for the gasifier and ancillary equipments, the successive unit operations (i.e., preheating exchanger, dissipator, chiller, gas engine, exhaust U. Arena et al. / Chemical Engineering Journal 162 (2010) 580-590



Fig. 3. Quantified process flow diagram for the gas engine configuration.

treatment) have been simulated on the basis of the performance data claimed by manufacturers and of standard mass and energy balances.

4.1. Gasification section

Bubbling fluidized bed reactor: the BFB reactor operates with a bed of olivine particles. Cyclone: this centrifugal collector, widely used for the separation and recovery of industrial dusts from process gases, is characterized by a high reliability and low capital and operating costs, due to the low pressure drop and the inexpensive maintenance schedule. The continuous operation mode of the cyclone allows for the devising of a recirculation circuit of the carbonaceous fines that can further increase the conversion efficiency of the fuel carbon and therefore increase the efficiency of the process.

4.2. Conditioning and cleaning section

Air preheating heat exchanger: it is a standard shell-and-tube exchanger that transfers the sensible heat from the hot syngas to the inlet gasification air so that the former is aptly cooled before being scrubbed and the latter is brought to the nominal temperature. The preheater is located downstream of the cyclone so as to reduce fouling and abrasion onto its hot side. Dissipator: it is an additional, inexpensive and low-maintenance heat exchanger required to bring the syngas temperature down to that compatible with the downstream scrubber inlet design point. Scrubber: it is the key component of the cleaning section, since it must guarantee the achievement of the final contaminants concentrations (residual dust, tar, acids and alkali compounds) required by the gas engine. Its cost is low but it necessitates a small water treatment unit or a connection to a water treatment plant. The component also separates small water particles that are entrained in the syngas stream to prevent their migration into downstream components. Chiller and demister: this component further cools the syngas below its dew point to reach the values of 25 °C and 60% of relative humidity, typically required by the engine inlet specification.

4.3. Electricity generation section

Gas engine: it is an internal combustion reciprocating piston engine, specifically optimized for syngas combustion rotating at 1500 rpm and directly coupled to an alternator. *Exhaust gas treatment section*: it is the engine exhaust stack, completed with a de-NOx catalytic system.

5. The externally-fired gas turbine design solution

The process flow diagram devised for the externally-fired gas turbine solution is reported in Fig. 4. The configuration is composed again of three sections, the gasification, the combustion and heat recovery with flue gas cleaning and the electricity generation sections. In the following, the role and main characteristics of the single components are schematically described. Also for this configuration, the gasification section has been modelled by using experimental data for the gasifier and ancillary equipments while the successive unit operations have been simulated on the basis of the performance data claimed by manufacturers and of standard mass and energy balances.

5.1. Gasification section

Bubbling fluidized bed reactor: it is the same type of BFB reactor adopted in the gas engine solution. *Cyclone:* this centrifugal collector too is identical to that of the gas engine solution and also in this case the recirculation of the fines is attainable.

5.2. Combustion and heat recovery section

Syngas combustor: it is a burner furnace where the syngas is combusted to yield hot flue gases to be sent to the high temperature gas exchanger. The placement of the combustor downstream of the cyclone avoids having to design a burner for particle laden gases which would make it more expensive and require higher maintenance. *Gasification air preheater*: it is a shell-and-tube heat exchanger that transfers heat from the flue gas stream from the



Fig. 4. Quantified process flow diagram for the externally-fired gas turbine configuration.

furnace to the inlet gasification air stream. This stage is also useful to lower the flue gases temperature in order to protect the flanged connection of the downstream high temperature heat exchanger. *High temperature heat exchanger (HTHE)*: it is the crucial and key component of the proposed configuration [32]. It is a recuperative type heat exchanger that has the furnace hot flue gases on the hot side and the compressed air coming from the compressor stage of the turbine on the cold side. *Air pollution control*: it is the stack where flue gases from the furnace are sent before being released. It must be equipped with adequate devices for air pollution control, such as a de-NOx system.

5.3. Electricity generation section

Externally-fired gas turbine: it is a custom modified gas turbine where the combustion chamber has been replaced by an external exchanger for heat addition before the compressed ambient air expands into the turbine wheel. The addition of the high temperature exchanger replaces the traditional combustion chamber. Auxiliary burner: it is an in-line burner where a small flow rate of high LHV fuel (such as methane) is utilized to raise the air temperature up to the design setpoint of the turbine expander.

6. The costs and revenues extimation model

The economic model used in this study is based on the estimation of standard accounting items such as total plant costs, operating costs, taxation and direct revenues from the sale of the generated energy. All monetary values have been subject to timevalue of money adjustment, i.e. future costs and revenues have been discounted to their present worth based on a fixed discount rate of 5% per year. This is needed to compare investment options that might generate costs and revenues in different time points along their expected life. Adopted models for total plant costs, operating costs and revenues [35] utilize manufacturer's information, average industry standard and the current incentive scheme available in Italy. Each item is detailed hereinafter.

6.1. Total plant costs

Total plant costs are the sum of equipment costs (i.e. the purchase cost of the equipment), direct costs (i.e. the costs associated with site preparation and assembly of components) and indirect costs (i.e. all costs associated with logistics and engineering). For each of the two configurations, equipment costs have been compiled on the basis of the manufacturer's quotes for the bill of materials associated with each layout and direct and indirect figures have been calculated by empirical factors applied to the cost of the equipment, based on a method first proposed by Lang [36,37]. For the size range under consideration, it was unpractical to base the estimation on available literature that has typically been accumulated for large to very large plants [38,39]; a comparison with existing installations that use equivalent technology is also difficult, because very few are operating in the investigated range. Equipment costs quotes have been gathered for a size in the middle of investigated range and then scaled to estimate the costs for the extremes of the size range. Direct and indirect costs have been calculated on the basis of the appropriate equipment costs, keeping the multiplying Lang factor constant across the range and equal to 1.5. Annual amortization of total plant costs has been calculated as a constant rate of 6.7% that corresponds to an expected plant life of 15 years. This value of the working life of the plant has been assumed on the basis of the following considerations: (i) the life of the energy generation device is the value that dictates the life of the whole plant, it being generally the most expensive piece of equipment; (ii) a proper maintenance program can reasonably extend the life of such devices to 15 years, as confirmed by manufacturers; (iii) it seems reasonable to assume a life non inferior to the available incentivized period. The scaling factor utilized for the equipment costs is based on a power law applied to estimates for the reference installation size obtained directly from manufacturers. An exponent of 0.6 was used, in accordance to basic literature [35,40] and recent works in the field [41].

6.2. Operating costs

The operating costs are the sum of the following items: maintenance, consumables and utility, waste streams disposal, labor and biomass cost. Maintenance costs (including running and extraordinary repairs) have been calculated as a percentage of equipment costs, with percentage values different for static equipment, the engine and the turbine. Consumables and utilities costs have been calculated for the reference installation size and then linearly scaled. Labor costs have been determined at the recurring wage for a single shift of a single worker (i.e. one man-year) because these plants are capable of operating unmanned. The disposal cost of the waste streams amounts to the product of the mass flow rate of waste by a fixed disposal fee of 120 €/t. The biomass fuel cost has been assumed to be equal to $20 \in /t$ even though a range of variation $0-40 \in /t$ has been then taken in consideration. All costs have been calculated in today's money and then discounted according to the year in which they occur.

6.3. Revenues

It has been conservatively assumed that revenues only come from the sale of the electrical energy produced. For this study, the Italian incentive scheme has been adopted as the basis for the energy compensation estimation. Then, an all-inclusive feed-in tariff of $0.28 \in /k$ Whe delivered to the grid has been used even though a range of variation $0.21-0.35 \in /k$ Whe has been then taken in consideration. The all-inclusive tariff encompasses compensation for the electrical energy sold and all the incentives associated with production of electricity from renewable resources and is valid for a period of 15 years. Access to the all-inclusive feed-in tariff, therefore excludes the attribution of renewable obligation certificates (green credits, or green certificates) or other incentives under current Italian legislation.

6.4. Taxes

Taxation has been set to 27.5% according to the current national fiscal imposition in Italy. No local taxation coefficient has been applied since no specific localization has been foreseen for the plant.

7. Technical and economical comparison

Although the two alternative plant configurations are based on identical gasification sections, they nonetheless differ in their energetic and environmental performance. Comparing the two plants on the basis on one aspect of their performance alone, e.g. their overall energy conversion efficiency, might be reductive since this would overlook other equally important aspects of the operation of power generation systems, such as their environmental burden, maintenance costs and ease of conduction. A broader comparison between the two biomass-to-energy configurations is traced in Table 6 while the economic comparison is visualized by Figs. 5 and 6. On one hand, the gas engine solution offers higher global efficiency (about 27%) due to the performance of the generator set and a lower capital cost (Fig. 5A), but has a generally lower availability (7680 h/y) and higher maintenance costs

Table 6

Synthesis of technical and economic performances for the two biomass-to-energy configurations, with reference to a nominal plant capacity of 200 kWe.

	Gas engine	Externally-fired gas turbine
Total energy conversion efficiency, % Specific biomass conversion rate, kWhe/kg _{fuel}	27.1 1.20	23.0 1.02
Waste export, kg/kg _{fuel}	Gas: 6.64 Liquid: 0.11 Solid: –	Gas: 32.25 Liquid: – Solidª: 0.01
Exhaust gas temperature, °C Total plant costs, \in/kWe Operating costs, $(\in/y)/kWe$ Internal rate of return (IRR), %	145 6000 940 13.2	313 7600 690 13.0

^a This value takes into account the sorbent utilized before the High Temperature Heat Exchanger but not the residues from APC unit.

(Fig. 5B). Moreover, it requires a suitable treatment unit for the waste water from the scrubber purge that is contaminated by tars, particulate and inorganics. On the other hand, the externally-fired gas turbine solution has a less efficient process (about 23%) due to the intrinsic thermodynamic limits and, for a less extent, to some losses inherent to the heat exchanger steps it embeds and has also a higher initial investment costs (Fig. 5A). The EFGT has a higher annual availability (7920 h/y), a lower maintenance costs and must dispose a solid waste stream (coming from APC unit) instead of a liquid one (coming from the wet scrubber unit), even though the advantage of the lack of an onerous water treatment system is balanced by the disadvantage of a very larger mass of flue gases to be treated at the stack (Figs. 3 and 4). Moreover, the EFGT configuration is more affected by the biomass cost due to the lower specific



Fig. 5. Comparison of total and operating costs of the two biomass-to-energy design configurations, for a biomass fuel cost equal to $20 \in /t$. Squares: gas engine. Circles: externally-fired gas turbine.

biomass conversion rate (Table 6), which results in larger fuel feed rates.

The different temperature and flow rate between the flue gas streams of the two alternative configurations are key elements in the evaluation of their cogenerative potential. The higher temperature and flow rate yielded by the EFGT configuration is apt to be used as a heat source for steam generation or bottoming Rankine cycle (Integrated Gasification Combined Cycle, IGCC), both valid ways to extract the residual available energy in the flue gases. On the other hand, the gas engine configuration has lower flow rates and lower temperatures, therefore it might not lend itself to be used viably as an energy source.

The graphs in Fig. 5A show that for all investigated sizes the GE solution has always total plant costs lower than the EFGT alternative and that both installations benefit of a power scale effect in the cost function. Anyway, a decrease in cost is expected for future EFGT installations due to economy attainable by the "nth plant effect" [39]: this aspect leaves a margin for the EFGT to become cost competitive with the GE in the near future. Fig. 5B illustrates the gap in the operating costs between the two alternatives that is mainly due to the different maintenance costs of the rotating equipment. Fig. 6A illustrates the influence of the GE's operating burden on the generated cash flow: despite having a 17.8% higher annual electricity yield (as can be deduced from Table 6), GE's cash flow is always lower than the EFGT one. Anyway, the EFGT's lower operating costs cannot compensate for the higher capital costs and the graph in Fig. 6B shows how the internal rate of return (IRR) is always favourable to the GE alternative for nominal plant capacities equal or larger than 200 kWe. This is indicated even by the payback time values, which for the GE and EFGT configurations, respectively, result equal to 4 years and 5 years at a nominal plant capacity of 400 kWe and to 3 years and 4 years at a nominal plant capacity of 600 kWe. A closer examination of the graphs in Fig. 6B



Fig. 6. Comparison of the financial performance indexes of the two biomass-toenergy configurations, for a biomass fuel cost equal to $20 \in /t$. Squares: gas engine. Circles: externally-fired gas turbine.

Table 7

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Sensitivities of out	nut variables to cha	nges of individual inn	ut variables for the two	a biomass-to-energy	configurations
beinsterrites of out	put vuriubies to thu	inges of mainfadaa mp	at fullables for the tim	s biolinuss to energy	configurations.

	Input variable	Base case	Variation	OC, k€/y	S _{OC}	ADCF, k€/y	S _{ADCF}	IRR, %	S _{IRR}
Gas engine	Nominal plant capacity, kWe	200	100 300	145 187 227	144	66 192 318	1.31	1.2 13.2 19.7	1.40
	CGE	0.77	0.74 0.80	189 187 186	-0.15	191 192 193	0.11	13.1 13.2 13.3	0.19
	Biomass cost, €/t	20	0 40	162 187 213	0.14	209 192 174	-0.09	15.0 13.2 11.3	-0.14
	Feed-in tariff, €/kWhe	0.28	0.21 0.35	187 187 187	-	118 192 265	1.53	4.6 13.2 20.5	2.41
Externally fired GT	Nominal plant capacity, kWe	200	100 300	97 138 177	0.58	107 241 375	1.11	5.6 13.0 17.5	0.92
	CGE	0.77	0.74 0.80	140 138 137	-0.24	240 241 242	0.11	12.9 13.0 13.1	0.20
	Biomass cost, €/t	20	0 40	107 138 169	0.22	263 241 220	-0.09	14.8 13.0 11.2	-0.14
	Feed-in tariff, €/kWhe	0.28	0.21 0.35	138 138 138	-	165 241 317	1.26	6.2 13.0 19.1	1.98

also shows how both plants become financially attractive only for a nominal plant capacity larger than 200 kWe.

The study was further pursued to determine sensitivities of relevant output variables to changes in plant capacity, operating and economic variables. To this end, the standard procedure for linearized sensitivity [35] has been used and applied at a "base case" assumed to be that of configurations reported in Figs. 3 and 4, for a nominal plant capacity of 200 kWe. Each input variable has been then changed in a fixed range of variation with respect to the base case. The sensitivity of generic output variable z was evaluated as:

$$S_z = \frac{(z^- - z^+)/z_b}{(v^- - v^+)/v_b}$$

where subscript b indicates the base case value. Superscripts - and + indicate, for the generic input variable v, the left and right extremes of assumed range of variation, whereas for the output variable z they indicate the values that it assumes for these extremes. The selected input variables were: the nominal plant capacity, whose range of variation has been assumed to be ± 100 kWe with respect to the base case; the cold gas efficiency (CGE) that can be utilized as a state variable that synthesizes the gasifier performance: its range of variation has been determined on the basis of present and previous investigations [24] as well as of literature data [10,14]; the biomass fuel cost, whose range of variation has been determined on the basis of information from the European market [42]; and the incentive tariff, whose range of variation has been assumed to be $\pm 25\%$ of the Italian tariff. The output variables chosen to characterize the performance of the two proposed configurations were: the operating costs (OC), the average discounted cash flow (ADCF) and the internal rate of return (IRR). Values of input and output variables are reported in Table 7: an analysis of these data indicates the crucial role of the all-inclusive feed-in tariff on the main economical parameters (ADCF, IRR), with very high values of the sensitivity. This highlights that the absence of an adequate incentivization policy may undermine the economic sustainability of the biomass-to-energy plant, in particular for small size plants. These have worse economical performances, as visualized by curves in Fig. 6 and data in Table 7. The sensitivity related to the biomass cost appears less important, even though the expected large effect on the operating costs results in estimated values of the IRR that are remarkably different in the extremes of the assumed range of variation. As expected, the gasifier performance has a not relevant role in the assumed range of variation of cold gas efficiency, since the extremes of the interval (0.74 and 0.80) however represent very good reactor performances.

8. Concluding remarks

The industrial application of gasification based, biomassto-energy cogenerators in the 100–600 kWe range has been investigated. The techno-economic performances of two promising design configurations, which implement a gas engine and an externally-fired gas turbine respectively, have been evaluated.

Mass and energy balances and material and substance flow analyses drawn for each design solutions were based on the experimental data obtained from a pilot scale bubbling fluidized bed air gasifier. The economic comparison has been carried out on the basis of the estimation of standard accounting items such as total plant costs, operating costs, taxation and direct revenues from the sale of the generated energy, all evaluated in the Italian context.

The results indicate that the internal combustion engine layout is the solution that currently offers the higher reliability and provides the higher internal rate of return for the investigated range of electrical energy production. Such conclusion does not take into account a cost decrease expected for future EFGT intallations due to economy attainable at the "nth installation", which leaves a margin for the EFGT to become cost competitive with the GE in the near future. Moreover, not one alternative is always preferable over the other: the choice has to account for site specific variables such as the presence of a heat demand and the costs of waste streams treatment and disposal.

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Paper V.

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A techno-economic comparison of fluidized bed gasification of two mixed plastic wastes

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ABSTRACT

A comparison between the most promising design configurations for the industrial application of gasification based, plastics-to-energy cogenerators in the 2–6 MWe range is presented. A pilot scale bubbling fluidized bed air gasifier, having a feeding capacity of 100 kg/h, provided experimental data: the syngas complete composition, the characterization of the bed material, the entrained fines collected at the cyclone and the purge material from the scrubber. Mass and energy balances and material and substance flow analyses have been therefore drawn to assess and compare design solutions utilizing two mixed plastic wastes (MPW) obtained from separate collection of plastic packaging, after different levels of pre-treatments. The related techno-economic performances have been finally estimated on the basis of the manufacturer's specifications. The study concludes that the MPW obtained after a very simple pretreatment and fed to a gasifier coupled with a steam turbine is the solution that currently offers the higher reliability and provides the higher internal rate of return for the investigated range of electrical energy production.

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1. Introduction

Plastics are the engineering material of our age, being used to substitute traditional materials, like wood, glass and metal, in a variety of forms and in a large number of applications of our daily life, from packaging, clothing, films, bags and containers to furniture, insulations, greenhouses, automotive parts and others. It is then reasonable that waste plastics represent a remarkable fraction of municipal solid waste (MSW) and then one of the crucial aspects of solid waste management (Brandrup et al., 1996; Al-Salem et al., 2009). The best approach to this problem must include all major categories of plastic solid waste treatment and recycling, such as re-extrusion, mechanical, chemical and feedstock recycling and energy recovery (Clift et al., 2000; Arena and Mastellone, 2006) as well as the development of an objective quantification of the environmental and economic advantages associated with each alternative over the whole life cycle (Arena et al., 2003; Perugini et al., 2005).

It is also generally recognized that a necessary step of a sustainable plastic solid waste management is the household separate collection (McDougall et al., 2001). The whole amount of this

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collection is generally made of three streams: (i) material essentially made of polyethylene (PE) and polyethylenterephthalate (PET), which allows an environmental and economic sustainable recycle process (and is estimated to be about 55-60% of the total collection); (ii) material "out of target", which does not allow any recycling or energy recovery process (about 5-10% of the total collection) and (iii) material essentially made of mixed plastic waste (MPW), with predominance of polyolefins (mainly polypropylene and polystyrene), which has a very high heating value and is preferentially utilized as waste-derived fuel (about 35-40% of the total collection). To highlight the energy content of this fuel, Table 1 compares lower heating values (LHVs) of a number of mono-polymeric plastics and MPWs to those of MSW, RDF, biomass and oil, on the basis of some literature data (Channiwala and Parikh, 2002; Consonni et al., 2005; Arena et al., 2010). The energy value of MPW can be converted to a variety of energy forms (electricity, process heat for industrial facilities and district heating, vehicle fuels) by means of the three main thermochemical processes: combustion, pyrolysis and gasification. Combustion of MPW presents severe constraints related to the very low softening temperature of the plastic fuels, with the consequent high risk of plugging in the feeding system and sintering in the combustion chamber. Problems can also arise, particularly for moving grate furnaces, due to the high calorific value of the plastics that could induce overheating of moving components. Low values of the net electric energy conversion efficiency (about 20%) are generally

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Table 1

Typical ranges of lower heating values of some plastic polymers and wastes, compared with those of common fuels.

Table 2

Main design and operating features of the pilot scale bubbling fluidized bed gasifier.

compared with those of common fuers.			Geometrical	ID: 0.381 m: total height: 5.90 m:		
Fuel Lower heating value, (MJ/kg)		Lower heating value, (MJ/kg)	parameters	Reactive zone height: 4.64 m; wall thickness:12.7 mm		
	Polyethylene	42.8-45.5	Feedstock capacity	100 kg/h		
	Polypropylene	46.5	Thermal output	Up to about 500 kW		
	Polystyrene	41.9	Typical bed amount	145 kg		
	Municipal solid waste	9.5-10.5	Feeding system	Over-bed air-cooled screw feeder		
	Refuse-derived fuel	14.9-18.0	Gasifying agents	Air, oxygen, steam, carbon dioxide		
	Household mixed plastic waste	27.0-32.0	Range of bed	700-950 °C		
	Selected mixed plastic waste	30.5-40.2	temperatures			
	Paper and cardboard	13.0-13.5	Range of fluidizing	0.3-1 m/s		
	Biomass	13.8-20.4	velocities			
	Heavy oil	42.5	Flue gas treatments	Cyclone, scrubber, flare		
	Petroleum	42.3	Safety equipments	Water seal, safety valves, rupture disks, alarms,		
	Gas oil	45.2		nitrogen line for safety inerting		
	Kerosene	46.5				

reported: higher values may be obtained for co-combustion in coal-fired power plants even though this option is limited by the above cited concerns related to plugging of feeding systems (Brandrup et al., 1996). Pyrolysis is the thermal degradation of plastic wastes, i.e. a thermal cracking in an inert atmosphere, carried out between 400 and 800 °C. It converts the hydrocarbon content of the waste in a high-heating value gas (having a LHV of 22-30 MJ/m³) together with condensable (waxes and oils) and solid fractions (Brandrup et al., 1996; Al-Salem et al., 2009). The experiences carried out in different countries, and especially in Japan, show the economic sustainability of the pyrolysis process when it is applied to very clean and pure plastic waste stream (Brandrup et al., 1996; Arena and Mastellone, 2006; Malkow, 2004). Gasification converts plastics in a combustible gas mixture (producer gas or syngas), mainly made of carbon monoxide, hydrogen and lower content of methane and it is able to provide a wide range of products, extending from clean fuel gas and electricity to bulk chemicals (Malkow, 2004; Arena et al., 2009, 2010). Diminishing landfill volume and high costs associated with traditional incineration technologies strongly increase the interest on the application of the gasification process to MSW: it is considered by some authors the advanced thermal treatment of the near future, particularly for restwastes, i.e. the residual dry fractions from separate collection, and for wastes coming from mechanical treatment of MSW (Heermann et al., 1979; DEFRA and Advanced Thermal Treatment of Municipal Solid Waste, 2007; Yassin et al., 2009).

The aim of this study is to evaluate and compare the technical and economic performance of the most promising design configurations for a medium scale industrial application of gasificationbased plastics-to-energy cogenerators. To this end, a number of tests with selected mixed plastic waste was carried out in a pilot scale bubbling fluidized bed gasifier (BFBG). The collected experimental data were processed by different analytical tools such as mass and energy balances and material and substance flow analyses, in order to assess and compare design solutions utilizing two mixed plastic wastes obtained from separate collection of plastic packagings, after different levels of pre-treatments. The related techno-economic performances have been finally estimated on the basis of the manufacturer's specifications.

2. The pilot scale fluidized bed gasifier

The utilized pilot scale BFB gasifier has the characteristics schematically listed in Table 2.

A special type of olivine – magnesium-iron silicate, (Mg,Fe_2) -SiO₄ – was selected as material for the fluidized bed on the basis of results of previous investigations carried out on the same pilot-scale BFBG (Arena et al., 2010) that indicated olivine as an interesting candidate to act as a bed catalyst for the tar cracking reactions in waste-derived fuel gasification, even taking into account its low cost and excellent resistance to attrition in the fluidized bed reactor. The main characteristics of the utilized olivine are reported in Table 3.

In the reported experiments, air was used as oxidizing agent and always injected at the bed bottom while the fuel was fed by means of an over-bed feeding system. The fluidizing air stream was heated up to 450 °C by a couple of electric heaters before entering the reactor. The fuel and blast flow rates were mutually adjusted so that, at the fixed fluidizing velocity, the desired equivalence ratio ER was obtained (where ER is defined as the ratio between the oxygen content of air supply and that required for the stoichiometric complete combustion of the fuel effectively fed to the reactor). The cylindrical BFB reactor was heated up to the reaction temperature by the sensible heat of pre-heated blast gases and by a set of three external electrical furnaces. The gas generated in the reactor was sent to a high efficiency cyclone and then to a wet scrubber (for the removal of tars, residual fly ashes and acid gases) and finally incinerated by a safety flare. An accurate description of the plant and of experimental procedures is provided elsewhere (Arena et al., 2009). Here it is sufficient to highlight that gas composition, upstream and downstream of the syngas conditioning section, was on-line measured by IR analyzers for the main syngas components (carbon monoxide and dioxide, hydrogen, methane) and by two micro-gas-chromatographs equipped with different columns for the detection of light and heavy hydrocarbons as well as of carbon monoxide and dioxide, hydrogen, nitrogen and water.

3. The configurations of the plastics-to-energy system

The configurations of the gasification based, plastics-to-energy system investigated in this study were defined on the basis of the following design specifications. The plant is designed to be fed with one of two mixed plastic wastes obtained as by-products of the sorting process of end-of-use plastic packaging from separate collection (Table 4). The first, named EBR (end-belt refuse), is obtained by a limited treatment (essentially mechanical shredding and air classification) made just before the feeding system of the gasification plant in order to reduce moisture and ash content. The second, named SRA (secondary reducing agent), is the result of an intensive treatment aimed to produce a fuel that can meet even high quality standards, as those of metallurgical industry. The plastic-to-energy process is designed to produce electricity, even though additional thermal energy is available to use in case a demand is present at the installation site. The electrical size range of interest is that of medium scale plants, between 2 and 6 MWe. These input data, together with the evidence that circulating fluidized bed reactors are advantageous for plant size larger

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Table 3

Characteristics of the olivine particles utilized as bed material in the pilot scale bubbling fluidized bed gasifier.

Mineral	Mg-Fe silicate
Chemical composition, (%)	
SiO ₂	39-42
MgO	48-50
Fe ₂ O ₃	8-10.5
CaO	<0.4
K ₂ O	-
TiO ₂	-
Al ₂ O ₃	0.8
Cr ₂ O ₃	
Mg ₃ O ₄	
LOI (loss of ignition)	0.20
Size range, (µm)	$200 \div 400$
Sauter mean diameter, (µm)	298
Particle density, (kg/m ³)	2900

Table 4

Chemical characterization of the two mixed plastic wastes utilized in the study.

Mixed plastic wastes	EBR	SRA
Ultimate analysis, % on weight basis		
C (min-max)	65.0 (62.1-67.9)	79.5 (75.9-83.1)
H (min-max)	9.7 (9.5-9.9)	13.1 (12.8-13.4)
N (min-max)	0.5 (0.4-0.6)	0.2 (0.15-0.25)
S (min-max)	0.3 (0.2-0.32)	0.1 (0.08-0.12)
Moisture (min-max)	2.1 (1.8-2.4)	0.7 (0.6-0.8)
Ash (min-max)	3.2 (2.4-4.0)	1.9 (1.4-2.4)
O (by difference)	19.2	4.5
C:O ratio	3.4	17.7
HHV, MJ/kg (by the relationship of Channiwala e Parikh)	34.6 (33.4-35.8)	43.4 (41.8-45.0)
LHV, MJ/kg	31.7 (30.4-33.0)	40.2 (38.6-41.8)

than 20MWth (Heermann et al., 1979; Basu, 2006) lead to individuate the atmospheric bubbling fluidized bed air gasification as the conversion process to be adopted. The design configurations for the plastics-to-energy gasification-based industrial plants can be sketched as a combination of three sections: syngas production, syngas utilization and syngas or flue gas cleaning. The first defines the syngas that can be produced and then, for fixed plastic fuel and gasification technology, the quantity and quality of this syngas. The utilization section indicates the producer gas that can be fed in a specific energy conversion device and then, for a given machinery (steam turbine, gas engine, internally or externally fired gas turbine), its temperature, heating value and cleaning level (i.e. tar and dust content but also that of alkali and inorganic contaminants). The relative succession of the utilization and cleaning sections depends on two possible configurations of the wasteto-energy gasification system that can be adopted: the "power gasification", where the producer gas is first cleaned then burned, and the "heat gasification", where the producer gas is first burned then cleaned (Fig. 1).

Then, for a "power gasifier" the cleaning section must function as the crucial interface between the characteristics of the producer gas and those required by the specific generator set whereas for a "heat gasifier" it consists of a possible pre-treatment of the syngas to remove contaminants (such as hydrogen chloride) before it goes into the combustor and, above all, of an air-pollution control (APC) system for flue gas cleaning (Arena et al., 2010; Heermann et al., 1979).

The following paragraph investigates the syngas characteristics that can be obtained by a BFBG fired with the design MPW fuels, mainly on the basis of the experimental activity carried out with the described pilot scale gasifier. The energy conversion devices for the range of electric output of interest, among all those commercially available, are then described and selected. The cleaning section that completes the most promising plant configurations is finally defined.

3.1. The gasification section

The gasification section has been designed on the basis of an experimental activity carried out on the pilot scale BFBG operated under autothermal conditions, i.e. with the only external heat addition being provided for the pre-heating of the fluidizing air stream. The reactor was operated with the mixed plastic waste, in a bed of olivine particles fluidized at a velocity of 0.7 m/s, a bed temperature of about 850 °C, an air pre heating between 450 and 550 °C and with an equivalence ratio ER between 0.21 and 0.24. The performances of the BFBG were measured and recorded only when the chemical composition of the produced syngas and the temperature profile along the reactor reached steady-state conditions.

The obtained results, reported in Table 5 for tests with SRA fuel, have been combined with a recently defined environmental assessment tool, the Material Flow Analysis, which is named Substance Flow Analysis when it is referred to a specific chemical species (Brunner and Rechberger, 2004). MFA/SFA is a systematic assessment of the flows and stocks of materials and elements within a system defined in space and time. It connects the sources, the pathways, and the intermediate and final sinks of each species in a specific process. These characteristics make MFA/SFA attractive as a decision support tool, as showed by its utilization in process evaluation of waste treatment and recycling options (Rotter et al., 2004) and in waste management planning (Mastellone et al., 2009). In this study MFA/SFA was used to deeper understand the performance of the pilot scale gasifier and to define and quantitatively assess some design solutions and operating criteria of the plastic waste gasification system.

Fig. 2 reports the quantified flow diagrams resulting by the MFA/SFA applied to the main process units (gasifier, cyclone, wet scrubber, water treatment system) of the pilot scale gasification system, when operated with SRA at an equivalence ratio of 0.24. Similar diagrams are available also for EBR fuel. Each flow in entrance to or in exit from a specific unit is identified by means of a black arrow if the specific data have been measured or fixed, or by a grey arrow if the data have been obtained by means of MFA/SFA.

The quantified flow diagram (also known as "layer") related to the total mass flow rate is reported in Fig. 2A. The input flows to the BFBG unit are the stream of plastic fuel, that of a small flow rate of nitrogen utilized to facilitate the fuel injection and that of air used as reducing agent and fluidizing gas. The output flow stream is the obtained syngas, which still contains heavy hydrocarbons, inorganic pollutants and entrained fines. The dirty syngas is sent to the cyclone for dust abatement and then to the wet scrubber for removal of tars and inorganic compounds. The specific production of syngas is equal to 4.19 kg_{syngas}/kg_{fuel} (i.e. 3.4 m³_{N,syngas}/ kg_{fuel}) while that of elutriated fines is 0.92 g_{fines}/kg_{fuel}. The stock of 145 kg of bed particles is progressively incremented (0.72 kg/h) as a result of opposite effects of elutriation losses and fuel ash accumulation. The experimental activity provides the complete chemical composition of streams leaving the cyclone and the water treatment system. These data have been used for the substance flow analysis of carbon, iron, magnesium and other elements and for the feedstock energy flow analysis (Arena et al., 2010).

Fig. 2B reports the result of the mass balance applied to the carbon element, i.e. the carbon layer of SFA. It gives the carbon conversion efficiency CCE, defined as the ratio between the mass flow rate of the carbon present in the syngas as CO, CO₂, CH₄ and



Fig. 1. Schematic representation of a heat and a power gasifier for waste processing, as elaborated from Heermann et al. (1979).

light hydrocarbons (until C_5H_m) and the mass flow rate of the carbon that enters the reactor with the fuel. The value of 0.97 of CCE is evaluated as the ratio between the mass flow rates of the syngas carbon stream, F7, and fuel carbon stream, F1. CCE is affected for 2.3% by the carbon losses related to the purge stream, F8, and for a negligible fraction (0.05%) to those of fly ash stream, F6. The carbon layer finally reports an important state variable of the plastic gasification process, the bed carbon loading W_c, which is the amount of carbon present in the bed as char particles at steady-state condition. Its value of 0.8 kg is a function of bed temperature and equivalence ratio.

Fig. 2C reports the layer of feedstock energy, i.e. the heat of combustion of each input and output stream (McDougall et al., 2001). The energy flow entering with the plastic fuel has been determined by means of a recently proposed relationship (Channiwala and Parikh, 2002), while the energy flows of exit streams have been evaluated on the basis of the heats of combustion of the specific substances. The resulting difference in feedstock energy, 186 MJ/h, is that "invested" at steady-state condition to convert the solid plastic into a gaseous fuel. Reported data allow to evaluate the cold gas efficiency CGE, defined as the ratio between the chemical energy of obtained syngas and that of injected fuel: the value of 0.792 is mainly determined by the chemical energy utilized inside the gasifier (18.4%), with the heavy hydrocarbons

of the purge stream from the water treatment system (2.3%) and, almost negligible, by the fraction of feedstock energy lost with the entrained fines (0.04%).

These results suggest two possible design solutions: the overflow of bed particles to avoid the increase of bed height and to refresh the olivine catalytic effect, and the disposal of the entrained fines collected by the cyclone, since their negligible production rate runs against the recycle opportunity. These data were finally combined with relationships of fluidization engineering (Kunii and Levenspiel, 1991) in order to determine the main geometrical parameters of the gasification section for a nominal plant capacity of 4 MWe. In particular, the reactor diameter was determined, for the fixed nominal plant capacity, on the basis of the cold gas efficiency and equivalence ratio, by keeping the fluidizing velocity and the type and size of bed materials constant. For both the MPW fuels, a configuration with two twin cylindrical BFBGs was chosen rather than that with a single larger reactor, as reported in Table 6. This solution reduces the reactor ID by about 30% so strongly limiting the potential transversal solid mixing concerns and then allowing a simpler fuel feeding system. Moreover, it makes the planned maintenance program easier: thanks to the rather wide range of operating fluidizing velocity (that can be as high as 1 m/s, then giving the possibility to increase the feedstock capacity of each reactor), the solution with two twin gasifiers pro-

Table 5

Operating conditions and performance parameters of the pilot scale bubbling fluidized bed gasifier operated with the SRA fuel under two values of equivalence ratio.

Operating conditions ER (equivalence ratio) AF (air/fuel ratio), kg _{sir} /kg _{fuel} Temperature of fluidizing air at gasifier entrance. °C	0.21 2.84 448	0.24 3.21 448
Output process data		
Temperature of fluidized bed at thermal steady-state, °C Temperature of sugas at gasifier exit, °C Q _{syngas} , M ₃ /M ₅ fuet HV _{syngas} , kJ/m ³ _N Specific energy, kVM/kg _{fuet} CGE (cold gas efficiency)	890 860 3.0 9580 7.9 0.71	890 810 3.4 9400 8.8 0.79
Summas composition (downstraam of cyclone and scrubbar)		
Syngas composition (downstream of cyclone and scrubber) N ₂ , % CO ₂ , % CO ₃ , % CO, % H ₂ , % CH ₄ , % C ₂ H ₄ , % C ₂ H ₄ , % C ₂ H ₄ , % C ₄ H ₆ , % C ₄ H ₆ , %	64.23 9.74 3.97 8.56 7.63 3.24 0.14 0 0.04 2.39	63.74 9.57 5.21 8.38 7.13 3.24 0.04 0.38 0.01 2.19
Syngas contaminants (upstream of cyclone and scrubber)		
Entrained fines, g/kg _{fuel}	0.3	0.9
PAH. mg/m_{N}^{3}	2600	1100

vides a rather large plant capacity even when one of the two reactors is on maintenance.

3.2. The energy generation section

The list of possible devices that can be used to convert the syngas into electricity are schematically listed and compared in Table 7. Each of them has its advantages and disadvantages when coupled with a BFB gasifier.

The steam turbine and boiler combination has its main positive feature in insuring that the expanding fluid is completely isolated from the syngas combustion fumes, therefore avoiding corrosion, fouling and plugging of the rotating parts. Moreover, due to the change of phase in the working fluid, the specific power of the machinery is extremely high. Rankine cycle power plants in the size range considered for this study have a net electrical efficiency between 15% and 24%, i.e. lower than those of larger plants (32–36%). Infact they are run at lower superheated steam temperature and pressure because of the possible presence of contaminants in the flue gas which can cause high temperature corrosion. These plants, for economic reasons, are not equipped with a reheater section and the turbines are simpler and less efficient and condenser pressure is higher (DEFRA and Advanced Thermal Treatment of Municipal Solid Waste, 2007; Saravanamuttoo et al., 2001).

A combination that was not further analyzed is that with an internal combustion gas turbine. Although internal combustion gas turbines offer very good net electric efficiency across the size ranges considered, the direct combustion and expansion of the syngas and its fumes into the turbo machinery poses technical difficulties. In fact, decontaminating the syngas of particulate, tar, al-kali and acids to manufacturer's specification if often unfeasible due to incongruent costs of the cleaning unit and, for scrubber devices, as a consequence of energy losses connected to tar abatement (Arena et al., 2010). Conversely, designing for costs can lead to residual contamination that fails to meet manufacturer's specifications which can cause unpredictable shortening of life or major failures of the machinery.

A recent customization of the basic gas turbine machine has been readied for commercialization, overcoming the main problems associated with internal combustion gas turbines. This configuration is called either externally-fired gas turbine (EFGT) or hot-air gas turbine, since the working fluid is ambient air and the heat addition happens in a gas-gas high temperature heat exchanger (Cocco et al., 2006). The separation of the working fluid from the combustion fumes assures that the rotating parts are not deteriorated, fouled or plugged, while the use of the exhaust clean hot air from the turbine outlet as the oxidizing gas in the syngas combustion, assures that high thermodynamic efficiencies are achieved. Unfortunately, this end-use device is not a proven technology and it is then affected by high economic risk. Moreover, EFGT needs a high compression ratio so that an extremely large and expensive gas cleaning section is necessary. For these reasons, the plant configuration with an EFGT was not considered for further analysis.

Another solution that has been investigated is a syngas optimized high efficiency reciprocating internal combustion engine. This type of engine is a proven technology that yields high electrical efficiency but has somewhat stringent requirements on both purity and technical conditions for the syngas supply (Boehman and Le Corre, 2008). In case of the gas engine setup though, the decontamination of the syngas can only be achieved by extremely expensive equipment, an aspect that makes this solution not viable and not competitive for the waste-derived fuel to be utilized and the plant capacity range analyzed.

In conclusion, the reported analysis of the possible end-use devices indicated that for the range of plant capacity of interest and for MPWs considered in this study (and, then, for the quality of the syngas obtainable by a BFB air gasification), the best solution for technical reliability and economic sustainability is that of a "heat gasification" configuration, having a steam turbine and boiler combination as energy conversion device. The heat gasification can be seen as an indirect combustion or, better, as a staged combustion. It is useful to highlight that a process that "first gasify and then burn the gas" has a number of advantages over direct combustion of wastes: (i) an efficient and clean combustion, since the exact required air can be mixed for optimum combustion; (ii) the consequent lower thermal losses at the stack; (iii) a clean combustion of producer gas since impurities can be removed from the producer gas, the volume of which is much smaller compared to that of flue gas and (iv) ease of control and continuous operation.

3.2.1. The steam turbine design solution

A detailed analysis and the quantified process flow diagrams (PFDs) of steam turbine configurations for the two MPWs are reported (Fig. 3), on the basis of the mass and energy balances developed for a net electric power output of about 4 MWe, which corresponds to a plant feedstock capacity of about 1,500 kg/h (i.e., about 11,500 t/y) for the SRA fuel and of about 1,900 kg/h (i.e., about 15,000 t/y) for the EBR one. It is noteworthy that, while the gasification section has been modeled by using experimental data for the gasifier and ancillary equipments, the successive unit operations (i.e., syngas combustor, pre-heating exchangers, heat recovery steam generator, steam turbine, condenser, exhaust treatment) have been simulated on the basis of the performance data claimed by manufacturers and of standard mass and energy balances (Saravanamuttoo et al., 2001; McBride and B.J., 1993; Reynolds, 1979).

Both the configurations are composed by three identical sections (gasification, cleaning and energy generation) but that for EBR fuel needs a preliminary fuel treatment section, composed by a *thermal drier* to reduce the moisture content, a *shredding system* to reduce the fuel size and an *air classifier* to eliminate materi-



Fig. 2. Quantified flow diagrams ("layers") obtained by MFA/SFA analysis. (A) Total mass (kg/h), (B) carbon element (g/h) and (C) feedstock energy (MJ/h) balances throughout the pilot scale gasifier in its present configuration, when operated with SRA fuel at ER = 0.24.

Table 6

Fuel and syngas rates and geometrical parameters for each of the two twin bubbling fluidized bed gasfifers fed with the EBR or the SRA plastic waste fuels in a 4 MWe nominal plant capacity.

	EBR	SRA
Air-to-fuel ratio	2.38	3.21
Fuel feed rate, t/h	0.95	0.75
Air flow rate, t/h	2.30	2.42
Annual fuel feed rate (320 days/y), t/y	7350	5800
Reactor diameter, m	1.92	1.98
Syngas rate, m ³ _N /h	2660	2100
Syngas LHV, kJ/m ³ N	7600	9400

ER = 0.24; T = 850 °C; U = 0.7 m/s; CGE = 0.79.

als of different weights and shapes, in order to reduce the ash content. After this section, the EBR fuel should have chemical characteristics similar to those reported in Table 4.

The gasification section is composed by two twin cylindrical bubbling fluidized bed reactors operated with a bed of olivine particles and a high efficiency cyclone, a centrifugal collector widely used for the separation and recovery of industrial dusts from process gases.

The combustion and heat recovery section is assembled with a *syngas combustor*, a burner furnace where the syngas is combusted to yield hot flue gases to be sent to the high temperature gas-gas heat exchangers; two gasification air preheaters, that are a couple of shell-and-tube heat exchangers that transfer heat from the flue gas stream from the furnace to the inlet gasification and combustion air streams, respectively. This stage is also useful to lower the flue gas temperature in order to protect the flanged connection of the downstream heat recovery steam generator. The heat recovery steam generator (HRSG) is a crucial and key component of the proposed configuration, since it is the unit where the heat transfer between the turbine output gases and the water of bottoming loop happens. It is a recuperative type heat boiler composed of an economizer to warm up the water output from the feed pump, an evaporator to produce steam and an overheat system, a shell-and-tube exchangers containing the flowing steam, run over by the hot flue gases to allow the heat transfer. The stack where flue gases from the HRSG are sent before being released into the atmosphere is composed by an absorbing/adsorbing zone to allow a sufficiently high contact time between the flue gases and the reagents put inside the reactor to remove acids and heavy metals, a bag filter to remove the residual dust and finally a de-NOx system.

The electricity generation section is composed by a *steam tur*bine, a machine insuring that the expanding fluid (i.e. water) is completely isolated from the syngas combustion fumes, therefore avoiding corrosion, fouling and plugging of the rotating parts, coupled with a large *condenser*, a heat exchanger that condenses the steam and allows to restart the Rankine cycle. 1500

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Comparison between the possible energy generation devices.

Energy conversion device	Net electrical efficiency of gasification plant	Main advantages	Main disadvantages
Steam turbine	15–24%	High electrical efficiency Turbine components are isolated from combustion products Long maintenance. intervals, high availability High specific work (kg/kg yielded for working fluid)	Expensive Partial load decreases efficiency significantly Plants is extremely large due to space requirements for the condenser and the HRSG
Gas turbine	20–30%	Compact assembly Long maintenance intervals, high availability Ideal for cogeneration plants (CHP) due to high exhaust temperatures	Turbine components are exposed to combustion products Partial load decreases efficiency significantly Moderately expensive
Externally fired gas turbine	15–20%	temperatures Turbine components are isolated from combustion products Long maintenance intervals, high availability Ideal for cogeneration plants (CHP) due to high exhaust temperatures	Not reliable technology Expensive Heat exchanger is exposed to high temperature, aggressive combustion gases Partial load decreases efficiency
Gas engine	20-30%	High electrical efficiency Relatively inexpensive Durable and reliable Partial load effects efficiency only marginally	Engine components are exposed to combustion products Short and expensive maintenance intervals, low availability

4. The costs and revenues estimation model

The economic model used in this study is based on the estimation of standard accounting items such as total plant costs, operating costs, taxation and direct revenues from the sale of the generated energy, following the procedure recently utilized by similar studies (Arena et al., 2010; Yassin et al., 2009). All monetary values have been subject to time-value of money adjustment, i.e. future costs and revenues have been discounted to their present worth based on a fixed discount rate of 5% per vear, even though a range of variation of 2.5-7.5% has been taken into consideration. This is needed to compare investment options that might generate costs and revenues in different time points along their expected life. Adopted models for total plant costs, operating costs and revenues (Rudd and Watson, 1968) utilize manufacturer's information, average industry standard and the current incentive scheme available in Italy. Each item is detailed hereinafter.

4.1. Total plant costs

Total plant costs are the sum of equipment costs (i.e. the purchase cost of the equipment), direct costs (i.e. the costs associated with site preparation and assembly of components) and indirect costs (i.e. all costs associated with logistics and engineering).

For each of the two configurations, equipment costs have been compiled on the basis of the manufacturer's quotes for the bill of materials associated with each layout and direct and indirect figures have been calculated by empirical factors applied to the cost of the equipment, based on a method first proposed by Lang (Lang, 1947; Lang, 1948). Equipment cost quotes have been gathered for a size in the middle of investigated range and then scaled to estimate the costs for the extremes of the size range. Direct and indirect costs have been calculated on the basis of the appropriate equipment costs, taking into account the multiplying Lang factor constant across the range. Annual amortization of total plant costs has been calculated as a constant rate of 8.3% that corresponds to an expected plant life of 12 years. This value of the working life of the plant has been assumed on the basis of the following considerations: (i) the life of the energy generation device is the value that dictates the life of the whole plant, it being generally the most expensive piece of equipment; (ii) a proper maintenance program can reasonably extend the life of such devices to 12 years, as confirmed by manufacturers; (iii) it seems reasonable to assume a life non inferior to the available incentivized period. The scaling factor utilized for the equipment costs is based on a power law applied to estimates for the reference installation size obtained directly from manufacturers. An exponent of 0.6 was used, in accordance to basic literature (Rudd and Watson, 1968; Park, 1984) and recent works in the field (Arena et al., 2010; Yassin et al., 2009).

4.2. Operating costs

The operating costs are the sum of the following items: maintenance, consumables and utility, waste streams disposal, labor, and insurance. Maintenance costs (including running and extraordinary repairs) have been calculated as a percentage of equipment costs. Consumables and utilities costs have been calculated for the reference installation size and then linearly scaled. Labor costs have been determined at the recurring wage for three shifts of two different workers, a skilled steam driver and a simple worker. The disposal cost of the waste streams amounts to the product of the mass flow rate of solid waste by a fixed disposal fee of 120 c/t. Moreover, the configuration for EBR fuel presents an additional solid waste stream (about 15% of the entrance material) produced by the in situ fuel pre-treatment section. All costs have been calculated in today's money and then discounted according to the year in which they occur.

4.3. Revenues

It has been assumed that revenues come from the sale of the electrical energy produced and from the gate fee of MPW utilized as fuel. For this study, the Italian incentive scheme has been adopted as the basis for the energy compensation estimation. A tariff of 0.062 € per kWhe delivered to the grid has been used. The tariff encompasses compensation for the electrical energy sold, but not yet incentives associated with production of electricity from waste. A value of 0.065 €/kWhe for the energy from waste recovery was taken in consideration for a valid period of 12 years. The fuel delivery revenues has been assumed, respectively equal to 0 and $95 \in t$ for the SFA and EBR configurations, even though ranges of variation of these values have been taken into account. For the EBR solution a fuel feed rate in entrance to the plant 30% larger than that for SRA solution has been taken in consideration, as a consequence of the material losses from the pre-treatment section, corresponding to a moisture reduction of about 15% and an ash reduction of another 15%.





Table 8

Synthesis of technical and economic performances for the two plastics-to-energy configurations, with reference to a nominal plant capacity of 4 MWe.

EBR	SRA
23.7	23.7
2.09	2.65
Gas: 18.9	Gas: 23.1
solid: 0.44	solid: 0.25
4.79	4.36
0.74	0.62
1818	0
8.3	No achievable return
	EBR 23.7 2.09 Gas: 18.9 solid: 0.44 4.79 0.74 1818 8.3

^a Both configurations have been evaluated for an identical generator set and on the basis of the same CGE of the gasification process.

4.4. Taxes

Taxation has been set to 27.5% according to the current national fiscal imposition in Italy. Moreover, an in force base coefficient of 3.9% concerning to productive activities local taxation has been applied.

5. Technical and economical comparison

Although the two alternative plants are based on identical gasification, cleaning and electricity generation sections, they nonetheless diverge for different pre-treatment level and economic and environmental performances. A broader comparison between the two configurations is traced in Table 8 while the economic comparison is visualized in Figs. 4 and 5.

Both solutions offer a high global efficiency (about 24%) due to the performance of the generator set coupled with the gasifier, a high annual availability (7680 h/y), about the same maintenance costs but different capital costs (Fig. 4A), due to the in situ instal-



Fig. 4. Comparison of total and operating costs as a function of nominal plant capacity, for the two plastics-to-energy design configurations. Circles: EBR fuel. Squares: SRA fuel.



Fig. 5. Comparison of the financial performance indexes as a function of nominal plant capacity, for the two plastics-to-energy design configurations. Circles: EBR fuel. Squares: SRA fuel.

lation of a pre-treatment equipment for the EBR configuration. The latter solution shows higher operating costs (Fig. 4B) since it must dispose of a larger solid waste stream but it shows also a higher average cash flow due to the greater revenues from the waste fuel delivery (Fig. 5A). On the other hand, the SRA solution is affected by a lower specific fuel conversion rate (Table 8) and by a lower average cash flow imputable to the lack of revenues from the fuel delivery.

The study was further pursued to determine sensitivities of relevant output variables to changes in plant capacity, operating and economic variables. To this end, the standard procedure for linearized sensitivity (Rudd and Watson, 1968) has been used and applied at the nominal plant capacity of configurations reported in Figs. 3 and 4. Each input variable has been changed in a fixed range of variation with respect to the base case. The sensitivity of generic output variable z was evaluated as:

$$S_{z} = \frac{(z^{-} - z^{+})/z_{b}}{(v^{-} - v^{+})/v_{1}}$$

where subscript b indicates the base case value. Superscripts – and + indicate, for the generic input variable "v", the left and right extremes of assumed range of variation, whereas for the output variable "z" they indicate the values that it assumes for these extremes.

The selected input variables were: the nominal plant capacity, whose range of variation has been assumed to be +/-2 MWe with respect to the base case; the cold gas efficiency CGE that can be utilized as a state variable that synthesizes the gasifier performance: its range of variation has been determined on the basis of present and previous investigations (Arena et al., 2010); the fuel delivery revenues, whose range of variation has been determined on the basis of information from the Italian plastics market (Petriglieri, 2010); the feed-in tariff, whose range of variation has been as sumed to be from 0.062 to 0.192 ϵ /kWhe in order to take into account the possibility of the absence of the incentive (0.065)

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Table 9

Sensitivities of output variables to changes of individual input variables for the configuration for EBR gasification.

Input variable	Base case	Variation	OC, k€/y	Soc	ADCF, k€/y	SADCF	IRR, %	S _{IRR}
		2000	1898		962		-2.2	
Nominal plant capacity, kWe	4000		2944	0.69	2582	1.19	8.3	1.21
		6000	3916		4043		13.1	
		0.74	3008		2623		8.6	
CGE,-	0.79		2944	-0.32	2582	-0.24	8.3	-0.57
		0.84	2889		2546		8.0	
		80	2944		2386		6.8	
Fuel revenue, €/t	95		2944	-	2582	-0.48	8.3	1.14
		110	2944		2779		9.8	
		0.062	2944		778		-9,6	
Feed-in tariff, €/kWhe	0.127		2944	-	2582	1.20	8.3	3.21
		0.192	2944		3952		17.7	
		2.5	2944		2490		7.6	
Discount rate, %	5		2944	-	2582	0.07	8.3	0.17
		7.5	2944		2681		9.0	
		0.15	2752		2570		8.2	
Waste stream from fuel pre-treatment, kg _{waste} /kg _{fuel in}	0.30		2944	0.08	2582	0.05	8.3	-0.12
· · · · · · · · · · · · · · · · · · ·		0.45	2984		2699		9.2	

€/kWhe) associated with production of electricity from waste; the discount rate, whose range of variation has been assumed to be +/−50%; and the disposal waste rate from the pre-treatment step for the EBR configuration, whose range of variation has been assumed to be +/−50%. The output variables chosen to characterize the performance of the two proposed configurations were: the operating costs (OC), the average discounted cash flow (ADCF) and the internal rate of return (IRR). Values of input and output variables are reported in Table 9 only for the EBR gasification plant, due to the evidence of the absence of an economic sustainability of the SRA solution.

An analysis of the data in Table 9 indicates the crucial role of the feed-in tariff on the main economical parameters (ADCF, IRR), with very high values of the sensitivity. This highlights that the absence of an adequate incentive policy may undermine the economic sustainability of the plastic-to-energy plant for the size range analyzed. As an investment is considered acceptable if its internal rate of return is greater than an established minimum attractive rate of return (MARR), an analysis has been done to set up the minimum necessary incentive to obtain a 12% IRR (Lang and Merino, 2001), i.e. the return rate assumed by most companies and based on the fact that the Standard & Poor's index (S&P 500) typically yields returns between 8% and 11%. The result is that a 4 MWe plastic-to-energy plant needs an incentive value of 0.089 €/kWhe to reach the MARR, i.e. a value of the feed-in tariff of 0.151 €/kWhe. The sensitivity related to the fuel delivery revenue appears not negligible since it determines a remarkable change of the IRR. As expected, the gasifier performance has a not relevant role in the assumed range of variation of cold gas efficiency, since the extremes of the interval (0.74 and 0.84) however represent very good reactor performances.

6. Conclusions

The industrial application of gasification based, plastics-to-energy cogenerators in the 2–6 MWe range has been investigated. The techno-economic performances of two "heat gasification" configurations, both equipped with a steam turbine, but fed with mixed plastic wastes of different quality, have been evaluated.

Mass and energy balances and material and substance flow analyses drawn for each design solution were based on the experimental data obtained from a pilot scale bubbling fluidized bed air gasifier. The economic comparison has been carried out on the basis of the estimation of standard accounting items such as total plant costs, operating costs, taxation and direct revenues from the sale of the generated energy, all evaluated in the Italian context.

The results indicate the configuration that currently offers the higher reliability and provides the higher internal rate of return for the investigated range of electrical energy production: a BFB gasifier coupled with a steam turbine and fed with the MPW generated by the sorting process of household plastic packaging separate collection after a very simple in situ pre-treatment. The utilization of a high-quality MPW, as obtained by an intense treatment process, which is presently designed to be utilized in the metallurgic industry, appears not convenient for a gasification based, plastics-to-energy plant.

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Paper VI.

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Fluidized bed gasification of a packaging derived fuel: energetic, environmental and economic performances comparison for waste-to-energy plants

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ABSTRACT

A comparison of possible solutions for small scale (0.2–1 MWe) waster-to-energy gasification-based industrial application is presented. A pilot scale bubbling fluidized bed air blown gasifier, having a capacity of 500 kWe, provided experimental data: the syngas complete composition as well as the characterization of the bed material and contaminants downstream of the cyclone and wet scrubber. Mass and energy balances and material and substance flow analyses have been drawn to assess and compare design solutions utilizing a packaging derived fuel (PDF) obtained as scrap by food industrial processes. The related environmental, energetic and economic performances have been estimated on the basis of the experimental data and manufacturer's specifications. In the scale range analyzed, the best solution is that of a power gasification coupled with an internal combustion engine, which provides high reliability and high internal rate of return.

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1. Introduction

Despite increasing attention for waste prevention and sustainability, total municipal solid waste (MSW) generation in the EU25 has increased from about 150 million tonnes in 1980 to more than 250 million tonnes in 2005 and is forecasted to reach 300 million tonnes by 2015 [1]. In addition, US MSW generation has increased from about 1.1 tons per capita in 1990 to 1.3 tons in 2002, and worldwide, MSW generation is projected to double by 2030 [2].

Integrated solid waste management is typically governed by the 'ladder of Lansink', which specifies a generally accepted hierarchy of preferred methods to treat waste (Fig. 1). Although reuse and recycling are preferred, energy recovery (the process of converting energy to heat and/or electricity starting from waste, also known as the Waste-to-Energy (WtE) concept) and landfilling are still key aspects of waste management [3,4]. In particular, in the States with rapid economic growth and massive urbanization, many cities face the problem of MSW disposal. With the lack of space for new landfills, waste-to-energy plants is playing an increasingly important role in waste management [5].

MSW is a heterogeneous feedstock containing materials with widely varying sizes, shapes and composition. If as received MSW is

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fed to WtE processes, this can lead to variable (and even unstable) operating conditions, resulting in a fluctuating product quality. Refuse derived fuel (RDF) usually referred to the segregated high calorific fraction of processed MSW is often used as input to WtE processes. This pretreatment usually consists of size reduction, screening and sorting in order to improve the handling characteristics and homogeneity of the material. The main benefits of converting MSW to RDF are a higher heating value, more homogeneous physical and chemical compositions, lower pollutant emissions, reduced excess air requirement during combustion and finally, easier storage. handling and transportation [6].

In the modern age a pervasive use of polymers have supplied most of common food packaging materials. Designed and engineered for very specific needs exploiting its almost limitless adaptability, plastic packaging is essential for processing, storing, transporting, protecting and preserving products. This materials present several desired features: i) the lightest packaging material, ii) protect and preserve perishable food for longer, iii) clear identification and labeling and easy to open and use, iiii) protection against contamination in foods and prevention of the spread of germs during manufacture, distribution and display [7]. The primary pre-product for food packaging processing is a foil, which is most commonly manufactured from plastics, such as polyethylene (PE), polypropylene (PP), polystyrene (PS), polycarbonate (PC), polyamide (PA) or polyethylene terephthalate (PET) [8,9,11]. The majority of produced films applied directly for the package or

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Fig. 1. The transition from old (1970s) to new (2002) "ladder of Lansink": P as Prevention, Rc as Recycling, C as Combustion (without energy generation), Lf as Landfill, QnP as Quantitative Prevention (prevention on production of waste, QIP as Qualitative Prevention (during production materials are used that have the least negative environmental impact when the product becomes waste), RcP as Recycling of Materials, CWE as Combustion with Energy generation.

as a coating material for the manufacture of multilayer packaging are polyethylene and polypropylene films (60-65%) of the total quantity produced film) [10]. However, increased use of synthetic packaging films has led to a serious ecological problems due to their total non-biodegradability. The use of bioplastics should be the future [12] but now plastics are an ideal packaging materials for all sorts of commercial and industrial users.

To point out the PDF valuable energy content, Table 1 compares low heating values (LHVs) of common utilized polymers and RDFs to those of MSW and oil on the basis of some literature data [14,15].

A wide variety of technologies is deployed for energy production from RDF (. Production of heat (domestic and industrial), electricity (or combined heat and power—CHP) and transport fuels is possible through a portfolio of technologies as showed in Fig. 2 [13]. The main available thermochemical conversion technologies for calorific waste (RDF) treatment deployed in Europe – direct combustion, pyrolysis, gasification – are discussed hereinafter with respect to their status and generic performance levels. Besides the individual methods, combinations of these processes, possibly combined with other treatments (i.e. plasma, melting, distillation, etc.), are also applied.

For combustion in grate furnaces, generally called mass burn facilities, raw waste has not been shredded, sized, or separated before combustion, although large items such as appliances and hazardous waste materials and batteries are removed before combustion. In mass burn systems, untreated MSW is simply burned, with the heat produced converted into steam, which can then be passed through a steam turbine to generate electricity or used directly to supply heat to nearby industries or buildings. Boilers in modern waste incineration plants of any kind reach a primary efficiency of 85%, some even slightly higher. However,

Table 1

Lower heating values of some polymers and wastes, compared with common oil fuels.

Fuel	Lower heating value, MJ/kg
Heavy oil	42.5
Petroleum	42.3
Gas oil	45.2
Kerosene	46.5
Polyethylene	42.8-45.5
Polypropylene	46.5
Polystyrene	41.9
Packaging derived fuel	20.1-24.6
Household mixed plastic waste	27.0-32.0
Selected mixed plastic waste	30.5-40.2
Refuse-derived fuel	14.9-18.0
Municipal solid waste	9.5-10.5
Paper and cardboard	13.0-13.5



Fig. 2. Termochemical processes in Waste-to-Energy conversion technologies (based on [13]).

they are operated at lower steam conditions than those in power plants (typically 400 °C and 40 bar to avoid corrosion caused by the high Cl inventory of most waste fuels and the resulting high chloride concentration in the ash layers on the boiler tubes). The consequence is a lower power efficiency of about 22–25% [17,19]. PDF combustion involves a high risk of plugging in the feeding system and sintering in the combustion chamber or induces overheating of moving components of the grate, respectively due to the low softening temperature and to the high calorific value of the polymers [14].

Pyrolysis is the thermal decomposition or fragmentation of PDF in a strictly inert atmosphere, generally in an externally heated rotary drum. The reaction starts at 200-250 °C and is, in this region, often called degassing. The highest temperature is in the order of 700 °C. Products of pyrolysis of PDF are: i) gases, predominantly CO, H₂ and short chain hydrocarbons, ii) so-called pyrolysis oil, comprising low volatile hydrocarbons up to tars, iii) solids, which are a mixture of coke and inert ashes [20,21]. Pyrolysis gas has a rather complex composition and direct use requires extensive gas cleaning, which is especially difficult for the removal of sulphur compounds and sticky dust particles or tar [22,23]. This is the reason why pyrolysis application in waste treatment is often coupled with a successive direct combustion of the gas phase without major prior cleaning. Boiler efficiency can be as high as in conventional combustion plants, while power efficiency can reach 15% [19].

Gasification is a partial oxidation at elevated temperature (500–1800 °C) that converts polymers in a combustible synthesis gas (or syngas) consisting mainly of CO and H₂ with small amount of CH₄ and other short chain hydrocarbons. This syngas can be used for efficient production of electricity (34%) and/or heat (40%), or second generation liquid fuels [16–18]. Declining landfill space and high incineration costs increase the interest on the application of the gasification as process of converting waste to energy: it is considered the advanced thermal treatment of the near future, particularly for the residual dry fractions from separate collection and for waste coming from mechanical treatments of MSW [14,24].

The aim of this study is to evaluate and compare the environmental, energetic and economic (EEE) performances of the most promising design configurations for an industrial application of gasification-based PDF-to-energy generators. To this end, a number of runs with a selected food packaging derived fuel (PDF) was carried out in a pilot scale bubbling fluidized bed gasifier (BFBG). The collected experimental data were processed by different analytical tools such as mass and energy balances and material and substance flow analyses, in order to assess and compare design solutions. The related EEE performances have been finally estimated on the basis of the experimental data and manufacturer's specifications.

2. The pilot scale BFB gasifier

The pilot scale gasifier PDF-fired is a bubbling fluidized bed (BFB) with an ID of 381 mm and a nominal capacity of 500 kWe (Fig. 3), which characteristics are schematically listed in Table 2. To optimize the gasification process and produce a considerably clean raw gas, not eliminating the need of downstream hot gas cleaning, an olivine, a calcium and magnesium silicate, was selected as material for the fluidized bed on the basis of results of previous investigations carried out on literature data [25] and on the same pilot-scale BFBG [26,28]. Considering the demonstrated activity and selectivity in terms of tar reduction, the economical availability and the excellent attrition resistance in the fluidized bed reactor,



Fig. 3. Pilot-scale Bubbling Fluidized Bed Gasifier (BFBG), consists of the reactor having an ID of 0.381 m and a total height of 5.90 m, a high efficiency cyclone, a wet scrubber and a flare.

Table 2

Main design and operating	features of the	pilot scale bubbli	ng fluidized bed gasifier.
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Geometrical parameters	ID: 0.381 m; total height: 5.90 m; reactive zone height: 4.64 m; wall thickness: 12.7 mm
Feedstock capacity	100 kg/h
Thermal output	Up to about 500 kW
Typical bed amount	145 kg
Feeding system	Over-bed air-cooled screw feeder
Gasifying agents	Air, oxygen, steam, carbon dioxide
Range of bed temperatures	700–950 °C
Range of fluidizing velocities	0.3-1 m/s
Flue gas treatments	Cyclone, scrubber, flare
Safety equipments	Water seal, safety valves, rupture
	disks, alarms, nitrogen line for safety inerting

the tar cracking reactions (i.e. as "primary method") in wastederived fuel gasification.

The experimental plant runs was carried out injecting air at the bed bottom as reducing agent while an over-bed feeding system was used for fuel. A couple of electric heaters heated up to 550 °C the fluidizing air stream before entering the reactor. Fixing the fluidizing velocity and adjusting both fuel and blast flow rates, the desired equivalence ratio (ER) was obtained (i.e. the ratio between the oxygen amount of air supply and that required for the stoichiometric fuel complete combustion). The BFB gasifier was heated up to the reaction temperature by the sensible heat of pre-heated blast gases and by a set of three external electrical furnaces. The synthesis gas produced in the reactor was sent to a high efficiency cyclone to remove fly ashes and then to a wet scrubber to segregate tars, acid gases and residual. Finally the syngas was incinerated by a safety flare. An accurate description of the plant and of experimental procedures is provided elsewhere [26]. Syngas composition (in terms of CO, CO₂, H₂ and CH₄) upstream and downstream conditioning section was on-line measured by IR analyzers and by micro-gas-chromatographs equipped with different columns for the detection of light and heavy hydrocarbons (tar) as well as of carbon monoxide and dioxide, hydrogen, nitrogen and water.

3. PDF-to-energy system: the selection process of gasification-based design configurations

The PDF-to-energy gasification-based system configurations investigated in this study was defined on the basis of: i) the fuel, ii) the energetic syngas valorization device, iii) the plant size, iiii) the available technologies. The plant is designed to be fed with food packaging material (hereinafter termed PDF), a scrap obtained by the industrial chain of food recovery, which characteristics are reported in Table 3. The PDF-to-energy process is designed to produce electricity, even though additional thermal energy is

Table 3	3
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Chemical characterization of the PDF utilized in the study.

Packaging derived fuel	PDF
Ultimate analysis, % on weight basis	
С	53.9 (50.9-56.9)
Н	7.7 (7.1-8.3)
Ν	0.5 (0.3-0.7)
S	0.1 (0.08-0.13)
Cl	0.3 (0.1-0.5)
Moisture	5.6 (5.4-5.9)
Ash	5.9 (3.9-8.0)
O (by difference)	26.0
C:O ratio	2.1
HHV, MJ/kg (by the relationship of Channiwala e Parikh)	26.5 (24.5-28.6)
LHV, MJ/kg	23.2 (21.2-25.3)

available to use in case a demand is present at the installation site. Based on the amount of feedstock availability that a typical italian company must dispose, the electrical size range of interest is between 200 kWe and 1 MWe. The evidence that circulating fluidized bed reactors are advantageous for plant size larger than 20 MWth [27] lead to individuate the atmospheric bubbling fluidized bed air gasification as the conversion process to be adopted. The design configurations for the PDF-to-energy gasification-based industrial plants can be sketched as a combination of three syngasbased sections: 1) generation, to qualitatively and quantitatively define the syngas produced, 2) valorization, to set the level of contaminants in syngas before to fed it in the energy genset (gas engine, gas turbine, steam turbine) and then, for the selected enduse device, set the inlet temperature and heating value, 3a) conditioning, a multiple-unit process as interface between generation and valorization sections ("power gasifier") or 3b) cleaning, a gasification-combustion combination as syngas pretreatment before its valorization, followed by an APC system ("thermal gasifier"). The choice between the conditioning and the cleaning section depends if the producer gas must be first cleaned and then burned or first burned and then cleaned [14,29].

First, the syngas characteristics and others output process data were investigated and assessed by operating the BFBG with the design fuel under different conditions. Then, energy conversion devices for the scale range of interest, among all those commercially available, are described and selected. At last, the cleaning/ conditioning section that complete the most promising plant configuration is defined.

3.1. The syngas generation section

The gasification section has been designed on the basis of an experimental activity carried out on the pilot scale BFBG operated under autothermal conditions, i.e. with the only external heat addition being provided for the pre-heating of the reducing and fluidizing air stream. The reactor was fired with PDF, in a bed of olivine particles fluidized with air preheating at 550 °C at a velocity of 0.7 m/s. The bed reached a temperature of about 850 °C and ER between 0.26 and 0.31 was selected. When the raw syngas composition and the temperature reactor profile reached steady-state conditions the performances of the BFBG were measured and recorded.

Table 4 reports the obtained results then processed with the Material and Substance Flow Analyses (MFA/SFA) [30], together with the inorganic compound concentrations of the PDF and of the fines collected at the cyclone reported in Table 5, using the software STAN [31]. It is a new systematic assessment of flows and stocks of materials, elements and energy within system or process defined in space and time, attractive as a decision support tool in process evaluation of waste treatments and recycling options [32] and in waste management planning [33]. In this study MFA/SFA was used to deeper understand the performance of the pilot scale gasifier and to define and quantitatively assess some design solutions and operating criteria of PDF gasification-based system.

Fig. 4 reports the quantified flow diagrams (termed "layer") applied to the main process units (gasifier, cyclone, wet scrubber, water treatment system) of the pilot scale gasification system, operating at an equivalence ratio of 0.26. Data measured or fixed are represented by black arrows (feeding fuel, nitrogen to facilitate the fuel injection, air used as fluidizing reducing agent, fly ashes collected by cyclone and produced syngas downstream wet scrubber) while grey arrows identify data obtained by means of MFA/SFA (raw syngas, water content, tar amount).

Mass flow rate layer is reported in Fig. 4A. The dirty syngas is sent to the cyclone for dust abatement and then to the wet scrubber

Table 4

Operating conditions and performance parameters of the pilot scale BFBG operated with the PDF fuel under two values of equivalence ratio.

Operating conditions		
ER (equivalence ratio), -	0.26	0.31
Fluidizing air velocity, m/s	0.72	0.74
AF (air/fuel ratio), kgair/kgfuel	1.99	2.40
Temperature of fluidizing air at gasifier entrance, °C	547	540
Output process data		
Temperature of fluidized bed at thermal steady-state. °C	879	915
Temperature of syngas at gasifier exit. °C	850	845
Osympase, m ³ N/kgfuel	2.45	2.72
LHV _{sympas} kl/m ³ N	6053	5035
Specific energy, kWh/kg _{fuel}	4.12	3.80
CGE (cold gas efficiency), -	0.657	0.588
CCE (carbon conversion efficiency), -	0.886	0.888
Syngas composition (downstream of scrubber), %		
N2	55.46	60.44
C0 ₂	14.30	14.10
со	11.57	10.87
H ₂	9.86	7.92
CH ₄	6.33	4.86
C ₂ H ₄	2.14	1.53
C ₂ H ₂	0.34	0.28
Syngas contaminants (upstream of cyclone and scrubber)		
Entrained fines, g/kgfuel	23.2	27.6
Entrained carbon fines, g _C /kg _{C-fuel}	13.8	7.9
Tar, mg/m ³ _N	22.93	5.1
NH ₃ , g/m ³ _N	2.00	1.81
H ₂ S, g/m ³ _N	0.40	0.36
HCl, g/m ³ _N	1.04	0.94

for removal of tars and inorganic compounds. The specific production of syngas is equal to 2.95 kg_{syngas}/kg_{fuel} (i.e. 2.45 km³_{syngas}/kg_{fuel}) while that of elutriated fines is 23.2 g_{fines}/kg_{fuel}. The stock of 145 kg of bed particles is progressively incremented (2 kg/h) as a result of opposite effects of elutriation losses and fuel ash accumulation. The experimental activity provides the complete chemical composition of streams leaving the cyclone and the water treatment system. These data have been used for the SFA of carbon, iron, magnesium and other elements and for the feed-stock energy flow analysis [34].

Fig. 4B reports the result of the mass balance applied to the carbon element. It gives the carbon conversion efficiency (CCE), defined as the ratio between the mass flow rate of the carbon present in the syngas and the mass flow rate of the carbon content in the fuel. The CCE value of 0.886 is evaluated as the ratio between

Table 5

Concentration of main inorganic compounds in fuel and fines collected at the cyclone.

Inorganic compounds	C _{fuel} , mg/kg	C _{fines} , mg/kg
Al	2620	43,594
Ar	0.1	0.1
Ca	22,875	166
Cd	0.25	0.76
Co	0.7	17.7
Cr	6.1	111.3
Cu	18.7	241
Fe	140	18.3
Hg	0.1	0.1
Mn	7.1	173
Na	910	4769
Ni	6.7	352
Pb	82	73.1
К	2074	1788
Sb	2	12.7
Sn	7.1	216
TI	0.25	0.25
V	2	83.9



Fig. 4. Layers of: A) total mass (kg/h), B) carbon element (g/h) and C) feedstock energy (MJ/h) balances throughout the pilot scale gasifier in its present configuration, when burned with PDF at ER = 0.26.

F7 and F1 streams. The value is affected for 9.9% by the carbon losses related to the purge stream (imputable essentially to the tar), F8, and for a negligible fraction (1.4%) to those of fly ash stream, F6. The layer also reported the bed carbon loading W_C (0.32 kg), which value is the amount of carbon present in the bed as char particles at the steady-state condition, function of bed temperature and ER.

Fig. 4C reports the layer of feedstock energy, i.e. the heat of combustion of each input and output streams [35]. The energy flow entering with the PDF has been determined by means of a specific relationships for solid, liquid and gaseous fuel [36], while the energy flows of exit streams have been evaluated on the basis of the heats of combustion of the specific substances. The resulting difference in feedstock energy, 244 MJ/h, is that "invested" at the steady-state condition to convert the PDF in a gaseous fuel. Reported data allow to evaluate the cold gas efficiency CGE, defined as the ratio between the chemical energy of the cleaned syngas and that of injected fuel: the value of 0.658 is mainly determined by the chemical energy with the heavy

hydrocarbons of the purge stream from the water treatment system (9.8%) and, for a negligible, by the fraction of feedstock energy lost with the entrained fines (1.2%).

The results obtained by MFA/SFA suggest some design solutions: the overflow of bed particles to avoid the increase of bed height and to refresh the olivine catalytic effect, the disposal of the negligible rate of entrained fines collected by the cyclone and the energy recovery of the tar removed from the syngas (around 10% of the total energy injected with the fuel) in a dedicated boiler or recycling it in the BFB reactor. Tar recycling within the gasification process may solve the associated waste problem and increase the system efficiency, provided tar is broken down under gasification conditions. Tar recycling experiments with a 500 kWth circulating fluidized bed (CFB) gasifier conducted at ECN laboratories at 830 °C report that 70–80% of tar compounds are broken down. Clearly, the presence of oxygen at the tar feed point is the essential condition to increase the tar destruction rate [37]. Fig. 5 reports MFA/SFA mass (A), carbon (B) and energy (C) layers when tar is fed to the gasifier:

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Fig. 5. Layers of: A) total mass (kg/h), B) carbon element (g/h) and C) feedstock energy (MJ/h) balances throughout the pilot scale gasifier in its present configuration with tar recycling, when burned with PDF at ER = 0.26.

the fuel feed rate is reduced to keep the heating value of the total fuel input approximately constant. CCE and CGE values grow to 0.986 and to 0.684 respectively, while energy feedstock become 311 MJ/h (30.5%) due to the endothermic tar reforming reactions that run against the esothermic tar combustion reactions [38].

Finally, the data obtained were combined with relationships of fluidization engineering [39] in order to determine the main geometrical parameters of the gasification section for a nominal plant capacity of 500 kWe. In particular, the reactor internal diameter was determined, for the fixed nominal plant capacity, on the basis of the CGE and ER, by keeping fixed the fluidizing velocity and the type and size of bed materials.

3.2. The syngas valorization section

The list of possible devices that can be used to convert the syngas into electricity are compared, considering for each of them its advantages and disadvantages when coupled with a BFB gasifier [14]. The steam turbine and boiler combination has its main positive feature in insuring that the expanding fluid is completely isolated from the syngas combustion fumes, therefore avoiding the corrosion, fouling and plugging of the rotating parts. Moreover, due to the change of phase in the working fluid, the specific power of the machinery is extremely high. Steam power cycles in the size range between 1 to 10 MW have a net electrical efficiency between 15 and 24%, but very poor steam turbine efficiencies (lower than 0.75) in the scale range considered for this study and additionally require an expensive condenser if the steam cycle is to be run in a closed loop configuration [19,40,41].

Although internal combustion gas turbines offer very good net electric efficiency across the size ranges considered, the direct combustion and expansion of the syngas and its fumes into the turbo machinery poses technical difficulties. In fact, decontaminating the syngas of particulate, tar, alkali and acids to manufacturer's specification if often unfeasible due to incongruent costs of the cleaning section [42]. Conversely, designing for costs can lead to residual contamination that fails to meet manufacturer's specifications which can cause unpredictable shortening of life or major failures of the machinery.

A recent customization of the basic gas turbine machine has been readied for commercialization, overcoming the main problems associated with internal combustion gas turbines. This configuration is called externally-fired gas turbine (EFGT) or hot-air gas turbine, since the working fluid is ambient air and the heat addition happens in a gas-gas high temperature exchanger [43-45]. The separation of the working fluid from the combustion fumes assures that the rotating parts are not deteriorated, fouled or plugged, while the use of the exhaust clean hot air from the turbine outlet as the oxidizing gas in the syngas combustion, assures that high thermodynamic efficiencies are achieved [46]. Unfortunately, this end-use device is not a proven technology and it is then affected by a high economic risk. Moreover, in order to increase the EFGT efficiency by reducing the temperature difference in the heat exchanger it increases the size of the heat exchanger, and consequently the air compression ratio, the pressure drop and the costs of the turbine. Although the plant configuration equipped with an EFGT was analyzed, to achieve efficiencies comparable to those of other devices, these reasons make this solution not economically advantageous

The last solution investigated is a syngas optimized high efficiency reciprocating internal combustion engine. This type of engine is a proven technology that yields high electrical efficiency but has somewhat stringent requirements on both purity and technical conditions for the syngas supply [47,48]. In the case of the gas engine setup though, the decontamination of the syngas can be achieved with a sufficiently inexpensive equipment, an aspect that renders the solution viable and competitive. In fact, the engine based installation is usually regarded as the standard against which other alternatives have to be compared in terms of electrical an economical efficiency in the scale range analyzed.

In conclusion, the reported analysis indicated that the best solution for technical reliability and economic sustainability is that of a "power gasification" configuration, having an internal combustion syngas optimized engine as energy conversion device (rpm: 1500, compression ratio: 9,2:1).

3.2.1. The syngas optimized engine design solution

On the basis of the mass and energy balances developed for a net electric power output of about 500 kWe, which corresponds to a plant feedstock capacity of about 500 kg/h (i.e., about 4000 t/y), an analysis and the quantified process flow diagram of syngas engine configuration for the PDF is reported in Fig. 6. It is noteworthy that, while the gasification section has been modeled by



using experimental data for the gasifier and ancillary equipments, the successive unit operations have been simulated on the basis of the performance data claimed by manufacturers and of standard mass and energy balances [49,51].

The PDF-to-energy plant configuration is composed by three syngas-based sections (generation, conditioning and valorization). The gasification section is composed by a cylindrical BFB reactor operated with a bed of olivine particles and a *high efficiency cyclone*, a centrifugal collector widely used for the separation and recovery of industrial dusts from process gases.

The conditioning section is assembled with: an air preheating heat exchanger, located downstream of the cyclone so as to reduce fouling and abrasion onto its hot side, that transfers the sensible heat from the hot raw syngas to the inlet gasification air; a dissipater, an additional, inexpensive and low-maintenance heat exchanger required to bring the syngas temperature down to that compatible with the downstream scrubber inlet design point; a wet scrubber, the key component of this section, since it must guarantee the achievement of the final contaminants concentrations required by the gas engine (in particular tar and acids gaseous but also alkali compounds and ash residual) connected with a water treatment system; chiller and demister, to further cools the syngas below its dew point to reach the values of 25 °C and 60% of relative humidity, typically required by the engine inlet specification.

The syngas energetic valorization section is composed by a syngas optimized engine, an internal combustion reciprocating piston directly coupled to an alternator and the exhaust gas treatment assembled with a de-NOx catalytic system.

4. The cost and revenues estimation model

The homemade economic model used in this study is based on the estimation of standard accounting items such as total plant costs, operating costs, taxation and direct revenues from the sale of the generated energy, following the procedure recently utilized by similar studies [14,24,50]. All monetary values have been subject to time-value of money adjustment, i.e. future costs and revenues have been discounted to their present worth based on a fixed discount rate of 5% per year, even thought a range of variation of 2.5–7% has been then taken in consideration. This is needed to compare investment options that might generate costs and revenues in different time points along their expected life. Adopted models for total plant costs, operating costs and revenues [54] utilize manufacturer's information, average industry standard and the current incentive energy scheme available in Italy. Each item is detailed hereinafter.

Total Plant Costs are the sum of equipment costs (i.e. the purchase cost of the equipment), direct costs (i.e. the costs associated with site preparation and assembly of components) and indirect costs (i.e. all costs associated with logistics and engineering).

Equipment costs have been compiled on the basis of the manufacturer's quotes for the bill of materials associated with each layout and direct and indirect figures have been calculated by empirical factors applied to the cost of the equipment, based on a method first proposed by Lang [52,53]. Equipment costs quotes have been gathered for a 500 kWe PDF-to-energy gasification-based plant and then scaled on the whole range to estimate the costs for the extremes of the size range. Direct and indirect costs have been calculated on the basis of the appropriate equipment costs, taking into account the multiplying Lang factor constant across the range. Annual amortization of total plant costs has been calculated as a constant rate of 6.7% that corresponds to an expected plant life of 15 years. This value of the working life of the plant has been assumed on the basis of the following

considerations: i) the life of the energy generation device is the value that dictates the life of the whole plant, being generally the most expensive piece of equipment; ii) a proper maintenance program can reasonably extend the life of such devices to 15 years, as confirmed by manufacturers; iii) it seems reasonable to assume a life non inferior to the available incentivized period. The scaling factor utilized for the equipment costs is based on a power law applied to estimates for the reference installation size obtained directly from manufacturers. An exponent of 0.6 was used, in accordance to basic literature [54] and recent works in the field [14,24,50].

Operating Costs are the sum of maintenance, consumables and utility, wate streams disposal, labor and insurance. Maintenance costs (including running and extraordinary repairs) have been calculated as a percentage of equipment costs. Consumables and utilities costs have been calculated for the reference installation size and then linearly scaled. Labor costs have been determined at the recurring wage for a single shift of a single worker (i.e. one man-year) because these plants are capable of operating unmanned. The disposal cost of the waste streams amounts to the product of the mass flow rate of solid waste by a fixed disposal fee of $120\varepsilon/t$ and of liquid waste by a disposal fee of $70\in/t$. All costs have been calculated in today's money and then discounted according to the year in which they occur.

Revenues has been assumed come from the sale of the power and heat energy produced and from the avoided disposal costs of PDF utilized. For this study, the Italian scheme adopted as the basis for the energy compensation estimation, not offer incentives in case of power plants (PP), while "white certificates", calculated on the basis of the electrical and thermal MWh produced, are guaranteed for a period of 10 years and 15 years respectively for combined heat and power (CHP) plants and, in case of demand, for district heating (DH). A tariff of 0.062€/kWhe delivered to the grid has been used, which encompasses compensation for the electrical energy sold and a conservative tariff of 0.08€/kWht has been used for the sale of saturated or superheated steam production in CHP plant, and for the sale of hot water in case of demand of district heating occurs. Compatibly with the Italian waste market and the tariff applied by the Italian National Consortium for Packaging (CONAI) revenues as avoided food packaging disposal has been assumed equal to 30€/t, even though ranges of variation of these values have been taken into account.

Taxes has been set to 27.5% according to the current national fiscal imposition in Italy. Moreover, an in force base coefficient of 3.9% concerning to productive activities local taxation has been applied.

5. Technical and economical comparison

An evaluation of environmental, energetic end economic (EEE) performance of the PDF-to-energy gasification-based plant for a nominal capacity of 500 kWe is reported in Table 6 for the three different opportunities proposed: power production (PP), combined heat and power (CHP) and district heating (DH).

The selected configuration offers a good global electrical efficiency (about 24%) due to the performance of the generator set coupled with the gasifier, an high annual availability (7680 h/y), and in the case of CHP and DH solutions, also a good thermal efficiency (about 54%). Figs. 7 and 8 visualize the more important economic items for the three different solutions. On one hand, the PP plant offers the lower capital cost (Fig. 7A) because the other solutions need of more heat exchanger units to recovery the heat by the hot fumes leaving the engine (CHP) and of an expensive network to guarantee the transport of hot water to the users (DH). For this reasons, the latter solution shows higher operating costs to

Table 6

Synthesis of Energetic, Environmental and Economic performances for the PDF-toenergy gasification-based plant, with reference to a nominal plant capacity of 500 kWe.

	PP	CHP	DH
Energetic performance			
Total energy conversion efficiency, %	23.8	78.2	78.2
Specific PDF conversion rate, kWh/kg _{fuel}	0.97	3.20	3.20
Environmental performance			
Waste export, kg/kg _{fuel}			
Liquid	0.035	0.035	0.035
Solid	0.033	0.033	0.033
Gas	7.96	7.96	7.96
Economic performance			
Total plant costs, k€/kWe	4.86	5.04	7.44
Operating costs, (k€/y)/kWe	0.53	0.54	0.63
Average cash flow, (k€/y)/kWe	0.35	1.55	1.56
Internal rate of return (IRR), %	0.5	29.8	18.9

the network maintenance (running and extraordinary repairs) (Fig. 7B) but, as the CHP solution, offers high average discounted cash flow (ADCF) and internal rate of return (IRR) due to the greater revenues from the sold of thermal energy produced and incentive life (Fig. 8A and B, respectively). On the other hand, the PP solution is affected by an extremely lower ADCF and a not achievable IRR, imputable to the lack of revenues from the thermal energy sold, undermine the plant economic feasibility.

The study was further pursued to determine sensitivities of relevant output variables to changes in plant capacity, operating and economic variables. To this end, the standard procedure for linearized sensitivity [54] has been used and applied at a 500 kWe nominal plant capacity on the basis of the configuration reported in Fig. 6. Each input variable has been changed in a fixed range of



Fig. 7. Comparison of total and operating costs as a function of nominal plant capacity, for the three solutions of PDF-to-energy gasification-based design configurations. Circles: PP. Squares: CHP. Rhombs: DH.



Fig. 8. Comparison of the economic performance indexes, average discounted cash flow (A) and internal rate of return (B), as function of nominal plant capacity, for the three solutions of PDFs-to-energy gasification-based design configurations. Circles: PP*. Squares: CHP. Rhombs: DH. The economic model results for the PP solutions for nominal plant capacities smaller than 500kWe return not achievable IRR values.

variation with respect to the base case. The sensitivity of generic output variable *z* was evaluated as:

$$S_z = \frac{(z^- - z^+)/z_b}{(v^- - v^+)/v_b}$$

where subscript b indicates the base case value. Superscripts – and + indicate, for the generic input variable "v", the left and right extremes of assumed range of variation, whereas for the output variable "z" they indicate the values that it assumes for these extremes.

The selected input variables were: the nominal plant capacity, whose range of variation has been assumed to be ± 300 kWe with respect to the base case; the cold gas efficiency CGE that can be utilized as a state variable that synthesizes the gasifier performance: its range of variation (0.60-0.76) has been determined on the basis of present and previous investigations [14]; the avoided PDF disposal costs, whose range of variation has been determined on the basis of information from the Italian waste market; the power energy tariff and the heat energy tariff, whose ranges of variation has been assumed to be $\pm 25\%$ of the Italian tariff; the thermal yield, whose range of variation has been assumed between 70 and 90%; the discount rate, whose range of variation has been assumed to be ±50% The output variables chosen to characterize the performance of the two proposed configurations were: the operating costs (OC), the average discounted cash flow (ADCF) and the internal rate of return (IRR). Values of input and output variables are reported in Table 7 for the CHP gasification plant, due to the evidence of the greater techno-economic feasibility than the other solutions (DH and PP). The sensitivity analysis results indicate the higher values for the thermal yield and the heat tariff on the main economical parameters (ADCF, IRR), as well as for the nominal

Table 7
Sensitivities of output variables to changes of individual input variables for the PDF-
to-energy gasification-based design configuration for the CHP solution.

Input variable	Base case	Variation	OC, k€/y	S _{OC}	ADCF, k€/y	S _{ADCF}	IRR, %	S _{IRR}
Nominal plant capacity, kWe	500	200	204 269	0.37	279 774	1.08	13.0 29.8	0.77
		800	322		1286		40.7	
CGE, -	0.68	0.60	271	-0.06	789	-0.14	30.4	-0.16
			269		774		29.8	
		0.76	267		764		29.3	
Power tariff,	0.062	0.046	269	-	732	0.21	27.9	0.23
€/kWhe			269		774		29.8	
		0.078	269		817		31.5	
Heat tariff, €/kWht	0.08	0.06	269	-	655	0.62	24.5	0.69
			269		774		29.8	
		0.10	269		895		34.8	
Discounted rate, %	5	2.5	269	-	762	0.03	29.2	0.04
			269		774		29.8	
		7.5	269		788		30.3	
Thermal yield, %	80	70	269	-	706	0.70	26.8	0.78
			269		774		29.8	
		90	269		842		32.6	
Avoided PDF	30	10	269	-	695	0.15	26.3	0.17
disposal, €/t			269		774		29.8	
		50	269		853		33.1	

plant capacity. As expected, the gasifier performance has a not relevant role in the assumed range of variation of cold gas efficiency, since the extremes of the interval however represent good reactor performances.

6. Conclusions

The industrial application of PDF-to-energy gasification-based between 200 kWe and 1 MWe has been investigated, analyzing the environmental, energetic and economical performances of a power configuration equipped with a syngas optimized engine for three different solutions (PP, CHP and DH). On the basis of experimental data obtained from a 500 kWe capacity bubbling fluidized bed air blown gasifier, mass and energy balances and MFA/SFA have been drawn for the PDF-to-energy design solution, and combined with manufacturer's specifications, allowed to deep understand environmental and energetic plant performances. Finally, on the basis of the estimation of standard accounting items such as total plant costs, operating costs, taxation and direct revenues from the sale of the generated energy, the economic performances comparison for PP, CHP and DH solutions has been carried out by a homemade economic model and a sensitivity analysis. Gasification-based plants in the power configuration (i.e. first cleaning and then burning the syngas) involve reduced environmental loads compared to those combustion-based because of the reducing reaction atmosphere. This aspect implies very low exhaust gas rates compared to those from combustion plants which must be operated with an air excess between 50 and 70%. Moreover, the substoichiometric oxygen flow rates fed in the gasification reactors promotes the partial oxidation of the carbon content of the fuel and, therefore, a low CO2 emission. Finally, utilizing the fluidized bed reactor and applying the tar recycling solution, the only solid waste stream to be disposed is that of ash residues collected at the cyclone, representing only the 2.3% of the original waste (PDF). A further advantage is that operating the reactor at temperature lower than that typical of the combustion process a very low concentration of trace elements such as cadmium, mercury and lead is present in the gas phase. Compared to equal environmental loads, the CHP is the solution that shows the best energetic and economic performances, with very high value of IRR in the scale range analyzed.

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2. Comparison between combustion- and gasification-based technologies for Waste-to-Energy: an eternal debate

Economical constrains pose a major dilemma in industry, especially with recovery methods of processes scrap and heterogeneous waste streams. Production of heat (domestic and industrial), electricity (or combined heat and power-CHP) and transport fuels from waste fuel is possible through a portfolio of technologies. Today direct combustion and gasification represent, with different status and different generic performance levels, the main available thermochemical conversion technologies for waste treatment deployed not only in Europe, but also in Asia and America. Besides the individual methods, combinations of these processes, possibly combined with other treatments (e. g. plasma, melting, distillation, etc.), are also applied. Many countries within the EU cover the electrical demand to hundreds of communities by direct incineration, e.g. Denmark, Sweden, and Germany. Japan and Korea, but also some European Country, such as Spain and Finland, adopt gasification technologies for the same purpose. Depending on difference in waste fuel composition, plant capacity and environmental aspect, either combustion- and gasification-based conversion technologies can be employed, the first being more suitable for large plant capacities and steady feedstock, whereas the latter is better for small or medium plant capacities, flexible energy supply demand, low-grade residual waste and direct material recovery. In particular, for combustion in grate furnaces, generally called mass burn facilities, waste has not been shredded, sized, or separated before combustion, although large items such as appliances and hazardous waste materials and batteries are removed before combustion. In mass burn systems, untreated waste is simply burned, with the heat produced converted into steam, which can then be passed through a steam turbine to generate electricity or used directly to supply heat to nearby industries or buildings. Boilers in modern waste incineration plants of any kind reach a primary efficiency of 85%, some even slightly higher. However, they are operated at lower steam conditions than those in power plants (typically 400°C and 40 bar to avoid corrosion caused by the high Cl inventory of most waste fuels and the resulting high chloride concentration in the ash layers on the boiler tubes). The consequence is a lower power efficiency of about 22-25%. For gasification technologies, the produced syngas can be used for efficient production of electricity (34%) and/or heat (40%), or second generation liquid fuels. Declining landfill space and high incineration costs increase the interest on the application of the gasification both as process of converting waste-to-energy and waste-to-material, particularly for the residual dry fractions from separate collection and for waste produced downstream of mechanical treatments of MSW, but also for unsorted residual waste (URW).

2.1 FBC vs FBG of Mixed Plastic Waste

In the modern age a pervasive use of polymers have supplied most of common packaging materials. Designed and engineered for very specific needs exploiting its almost limitless adaptability, especially for drink and food chain, plastic packaging is essential for processing, storing, transporting, protecting and preserving products. This materials present several desired features: i) the lightest packaging material, ii) protect and preserve perishable food for longer, iii) clear identification and labeling and easy to open and use, iiii) protect against contamination in foods and prevent the spread of germs during manufacture, distribution and display. The primary pre-product for packaging processing is a foil, which is most commonly manufactured from plastics, such as polyethylene (PE), polypropylene (PP), polystyrene (PS), polycarbonate (PC), polyamide (PA) or polyethylene terephthalate (PET). However, increased use of synthetic packaging has led to a serious ecological problems due to their total non-biodegradability. Mixed plastic waste posses' high calorific value when compared to other materials due to its crude oil origins, as Table 11 illustrates by comparison to gas oil, heavy oil and other crude oil derivatives. Since the heating value of plastics is high, they make a convenient energy source. Producing water and carbon dioxide upon combustion makes MPW similar to other petroleum based fuels. In general, it is considered that incineration of MPW results in a volume reduction of 90%, which reduces the reliability on landfilling. In the process of energy recovery, the destruction of foams and granules resulting from MPW also destroys CFCs and other harmful blowing agents present. Applications of better waste management system are vast, in cement and lime kilns and circulating and bubbling fluidized beds. In terms of improving the image of plastics, chemical recycling methods have contributed to that and proven very successful in recent years. Advanced thermochemical treatment, first of all gasification methods, have also proven very successful, especially when considering the range of valuable petrochemicals they produce. In particular fluidized bed technologies (FB) are increasing in popularity amongst many mass burn incineration lines due to: i) less complex emissions control systems, ii) high combustion efficiency with simple operation and fast response, iii) reduction in boiler size and iv) low corrosion with easier ash removal. In Europe,

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for example, the revolving fluid bed developed by Ebara Co. to produce energy is stated to be emerging very rapidly. The main principle for this technology is the mechanism of the internal furnace with no moving parts, equipped with a slanted bed floor to produce a revolving sand motion. These advantages are recognized in particular for waste fuel containing high percentages of plastic because their combustion involves a high risk of plugging in the feeding system and sintering in the combustion chamber or induces overheating of moving components of the grate, respectively due to the low softening temperature and to the high calorific value of the polymers. Fuelling twin pilot-scale air-blown bubbling fluidized bed combustor and gasifier with mixed plastic waste, a comparison of the main performance parameters was developed on experimental data and element partitioning, also applying a material and substance flow analysis.

	COMB-1 COMB-2 GAS-1		GAS-2			
Operating parameters						
F, kg _{fuel} /h	5,6	28,4				
ER, -	1,45	1,41	0,24	0,21		
U _g , m/s	0,67	0,68	0,65	0,65		
T _{bed} , °C	822	844	894	890		
T _{preheating air} , °C	20	20	448	543		
AF ratio, -	19,64	19,14	3,20	2,84		
Flue/Fuel gas compo	sition, % _{vol}					
O ₂	7,00	6,35	-	-		
N ₂	82,83	83,05	63,85	64,29		
CO ₂	10,12	10,49	9,57	9,74		
CO	0,05	0,11	5,21	3,97		
H ₂	-	-	8,38	8,56		
CH ₄	-	-	7,13	7,63		
$C_3H_m^a$	-	-	3,67	3,42		
BTX	2,19		2,19	2,39		
Main pollutants, mg/m_{N}^{3}						
Tar	-	-	1100	2600		
C as C _n H _m ^b	137	367	149.000	157.000		
NO _x	68,3	73,6	-	-		
NH₃	-	-	34	16		
SO ₂	5,7	31,4	-	-		
H ₂ S	-	-	4	9		
HCI	-	-	3	58		
Flow rate of elutriated fines						
E/F, g/kg _{fuel}	16,9	19,3	17,1	17,1		
E _c /F _c , g _c /kg _{c,fuel}	0,2	0,4	9,2	7,4		

Table 11. Operating parameters and process performances of FBC and FBG

The obtained flue/fuel gas, which still contains heavy hydrocarbons, inorganic pollutants and entrained fines, was completely analyzed. The raw flue/fuel gas is first sent to cyclone for dust

abatement. Typical specific gas productions are equal to 20,6kg_{fluegas}/kg_{fuel} and to 4,4kg_{fuelgas}/kg_{fuel} (i.e. $16,0m^{3}_{N,flue gas}/kg_{fuel}$ and $3,6m^{3}_{N,fuel gas}/kg_{fuel}$) for the combustion and gasification tests, respectively. As result by the material flow analysis, inside the reactors the bed amounts are progressively incremented (0,1kg/h for BFBC and 0,9kg/h for the BFBG) as a result of the opposite effects of elutriation losses and fuel ash accumulation.

The Substance Flow Analysis was applied to the main inorganic compounds present in the MPW in order to determine their fate as bottom ash, fly ash or compounds entrained in the gas phase downstream of cyclone (and then almost completely intercepted in the adsorbing/dedusting airpollution-control system). Following figures are examples of the balance on the atomic species applied in particular to low-boiling-heavy metals, cadmium (Cd) and lead (Pb), respectively.



Figure 13 . Cadmium flow analysis (mg/h): COMB-1 (left); GAS-1 (right).



Figure 14 . Lead flow analysis (mg/h): COMB-1 (left); GAS-1 (right).

Many factors can influence whether and in what form a trace element eventually ends up in the particulate or gaseous phase. The most important among these factors are: i) how the trace element resides in the fuel; ii) system temperature and pressure; iii) presence of halogens (in particular, of chlorine); iv) presence of sorbent compounds; and v) oxidizing or reducing conditions. The latter is the only factor that can affect the fate of these elements in the reported BFBC and BFBG tests, being all the others kept fixed. All inorganic compounds, except for the Al, show a larger fraction escaping the system in the flue gas (from 61 to 98%), with a not negligible fraction collected as dust (FA) by the cyclone for Cd, Pb and V (between 23 to 36%). In the fluidized bed gasification test the fate of some inorganic compounds seems to be different, even though also in this case (with the exception of V) their fraction as stock in the reactor is unimportant. In

particular AI, Cd and Pb reveal a balanced repartition betweens FA and fuel gas phase while the largest fractions of K, Sn and Ca are detected in the fuel gas.

The material and substance balance have been also used as a basis to assess and compare the energetic performances of the combustion- and gasification-based plastics-to-energy processes. For thermal applications, such as when the gas is not cooled before combustion and the sensible heat of the gas is also useful, the hot gas efficiency (HGE) is used as the best parameter to an accurate assessment of the energetic performances (Basu, 2006). The conventional definition of this parameter has been modified to take into account the different terms which have a relevance in the two thermochemical processes. HGE has been then defined as:

$HGE = \frac{gas chemical energy (LHV_{gas} + Q_{gas}) + gas sensible heat (H_{gas}) + cooling water sensible heat (H_{water}) fuel chemical energy (LHV_{fuel} + Q_{fuel}) + fluidizing gas sensible heat (H_{air})$

All the terms that contribute to the evaluation of the HGE in the four experimental tests was evaluated. A comparison of the energetic performances of the two units cannot be carried out just on the basis of the HGE, even because it has a range of variation, as a consequence of the variability of different operating parameters. Nevertheless, it is possible to make some preliminary observations: i) the two processes have similar energetic performances (HGE values between 0,73 and 0,90); ii) the BFBC process is negatively affected by the utilization of an air excess greater than 40% (i.e. ER>1,4); iii) the BFBG is less advantageous when values of ER<0,24 were utilized. The latter conclusion is supported by the values of cold gas efficiency CGE, defined as the ratio between the chemical energy of the produced syngas and the chemical energy of the plastic waste, which is equal to 0,71 and 0,79 for the gasification tests, then in agreement with the typical range of ER for medium or large units, which is between 0,25 and 0,5.

2.2 Combustion vs Gasification of Municipal Solid Waste

Despite increasing attention for waste prevention and sustainability, total municipal solid waste (MSW) generation in the EU25 has increased from about 150 million tonnes in 1980 to more than 250 million tonnes in 2005 and is forecasted to reach 300 million tonnes by 2015. In addition, US MSW generation has increased from about 1,1 tons per capita in 1990 to 1,3 tons in 2002, and worldwide, MSW generation is projected to double by 2030. In particular, in the States with rapid

economic growth and massive urbanization, many cities face the problem of MSW disposal. With the lack of space for new landfills, WtE plants is playing an increasingly important role in waste management because, in addition to allow energy recovery, can reduce mass and volume of waste up to 90%, and also recover materials by residuals. MSW is a heterogeneous feedstock containing materials with widely varying sizes, shapes and composition. If as received MSW is fed to WtE processes, this can lead to variable (and even unstable) operating conditions, resulting in a fluctuating product quality. However, today the environmental impact of modern, adequately constructed and operated WtE units is assessed as comparable to that of a medium industry and anyway less than almost any other source of electricity. Nevertheless, fear of pollution still brings WtE plants to the center of emotional public debate, much of it based on perception rather than on objective scientific evidence. This public perception forces the manufacturers of WtE plants to continuously improve the performance of the chemical conversion process and to develop advanced technologies for pollution control systems.



Figure 15. Conceptual overview of a modern single-stage mass burn incinerator.

Conventional combustion is a well-established technology developed over 100 years ago for energy generation from municipal solid waste (the first attempts took place in England in the 1870s). Since that time, vast technology improvements have been made making conventional combustion the most common WtE technology currently being used to treat MSW. The most common conventional combustion approach is called single-stage combustion or mass burn incineration (sometimes referred to as grate-fired technology). Over 90% of WTE facilities in Europe utilize mass burn incineration technology with the largest facility treating approximately 750kt/y. Several stages of combustion occur in mass burn incinerators. High temperature gasification system, called Direct Melting System (DMS), is employed by Nippon Steel and JFE and represent the most utilized gasification technology diffuse worldwide. The process produces a syngas that is combusted in a steam boiler, driving a steam turbine to produce electricity. The heating process begins by feeding waste into a gasification vertical shaft reactor. The high temperature causes organic material in the MSW to dissociate into syngas. The syngas is transferred to a combustion chamber which heats a boiler which in turn powers a turbine and produces electricity.



Figure 16. Overview of the high temperature vertical shaft gasifier, called Direct Melting System.

In order to reduce the environmental impacts associated with WTE facilities air pollution control (APC) systems have been developed. In general, APC systems are used to cool flue gases, scrub

acidic gases and capture particulate matter and various contaminants such as heavy metals and trace organics. Significant improvements have been made in APC systems of WtE plants over the past few decades and advancements continue to be made to the types of APC systems used for MSW incinerators. The APC system are similar for both the combustion- and gasification-based plant. The produced flue gas is typical cleaned using a bag filter (to remove solid particulate), limestone injection (to remove inorganic compounds), activated carbon (to remove acid gases) and SCR (to reduce NO_x) before it is released into the atmosphere.



Figure 17. Detail of moving grate with heat recovery boiler (left) and vertical shaft gasifier (right).

For mass burn incinerator plants, the first step reduces the water content of the waste in preparation for burning (drying and degassing). The next step involves primary burning which oxidizes the more readily combustible material while the subsequent burning step oxidizes the fixed carbon. In single-stage combustion, waste is burned in sub-stoichiometric conditions, where sufficient oxygen is not available for complete combustion. The oxygen available is approximately 30 to 80% of the required amount for complete combustion which results in the formation of pyrolysis gases. These gases are combined with excess air and combusted in the upper portions of the combustion chamber which allows complete oxidation to occur. The direct melting system utilize O₂-enriched air injection in the melting section and the solid waste charged from the top of the vertical shaft furnace, together with coke and limestone. From the top to the bottom of the

gasifier it is possible to individuate a drying and pre-heating region (which operates at about 400°C), thermal decomposition region (between 400 and 1000°C), a combustion zone (1000-1600°C) and melting region (between 1600 and 1800°C), the latter provided by the coke bed layer formed in the lower part of the vertical shaft furnace. It prevent cool-down of slag and accelerate low-boiling-point heavy metals devolatilization. Limestone is added to provide some pH buffering of the melt and to form fluid slag that can be easily discharged from the furnace bottom. In some plants, a natural gas or LPG injection system is utilized to improve the carbon conversion ratio of the injected char. The produced syngas is transferred to a swirling combustor that transfers the generated thermal energy to a boiler, which in turn powers a steam turbine that produces electricity.

Mass burn technology applications provide long residence times on the grate which in turn results in good ash quality (i.e. less non-combusted carbon). Mass burn facilities have energy efficiencies of 14% to 27% for electricity generation, depending on the plant capacities (higher energy recovery efficiencies are achieved through the cogeneration of heat). Direct melting system pay less energy efficiencies (13-15%) but produce a significant amount of directly recyclable vitreous slag and a metal stream, therefore represent a thermal treatment balanced between material and energy recovery.

A comparison between these selected combustion- and gasification-based technologies was carried out on the basis of published data on scientific journals and information directly obtained by several commercial WtE units operating worldwide. The analysis was focused on the partitioning of key elements by using substance flow analysis. In particular the ferrous and non-ferrous metal partitioning was analyzed because the recovery of these metals from WtE solid residues is of great interest for the known environmental and economic advantage of metal scraps recycling. Moreover, it is recognized that metal recovery increases the technical feasibility of ash utilization for road construction and concrete production. For instance, the presence of Al can result in swelling and expansion phenomena that create troubles to these applications. Moreover, the fate of low-boiling-point heavy metals was analyzed since it determines their emission potential and the related environmental hazards, mainly with reference to the reuse or disposal of solid residues. The results indicate that slag from direct melting system units could be assumed as immediately and completely recyclable, because the low-boiling-point metals, such as Pb and Zn, are almost completely transferred to the gas phase (98% and 99% respectively) and then caught by the APC system, while the high-boiling-point metals, such as Cu and Fe, are mainly

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concentrated in the metal stream (86% and 89% respectively). These results, coupled with those of slag leaching and acid-extraction tests, indicate that slag could be directly recyclable. Moreover, XRD analysis carried out on these slag after different types and times of exposure showed no changes in chemical composition and negligible effects on enhancing constituent release. This implies that the investigated gasification-based technology allows a reduction up to about 70% of the amount of waste to be sent to final landfill, when compared to the conventional combustion-based WtE units.

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Element partitioning in combustion- and gasification-based waste-to-energy units

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ABSTRACT

A critical comparison between combustion- and gasification-based waste-to-energy systems needs a deep knowledge of the mass flows of materials and elements inside and throughout the units. The study collected and processed data from several moving grate conventional incinerators and high-temperature shaft gasifiers with direct melting, which are in operation worldwide. A material and substance flow analysis was then developed to systematically assess the flows and stocks of materials and elements within each waste-to-energy unit, by connecting the sources, pathways, and intermediate and final sinks of each species. The patterns of key elements, such as carbon, chloride and heavy metals, in the different solid and gaseous output streams of the two compared processes have been then defined. The combination of partitioning coefficients with the mass balances on atomic species and results of mineralogical characterization from recent literatures was used to estimate a composition of bottom ashes and slags from the two types of waste-to-energy technologies. The results also allow to quantify some of the performance parameters of the units and, in particular, the potential reduction of the amount of solid residues to be sent to final disposal.

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1. Introduction

The municipal solid waste (MSW) management systems that operate successfully worldwide demonstrate the key role of thermal treatment, which appears essential to obtain the environmental and economic sustainability of the whole system (Psomopoulos et al., 2009; Brunner, 2012). The environmental impact of modern, adequately constructed and operated, waste-to-energy (WtE) units is today assessed as comparable to that of a medium industry (Rechberger and Schöller, 2006), and anyway less "than almost any other source of electricity" (US-EPA, 2003). Nevertheless, fear of pollution still brings WtE plants to the center of emotional public debate, much of it based on perception rather than on objective scientific evidence. This public perception forces the manufacturers of WtE plants to continuously improve the performance of the chemical conversion process and to develop advanced technologies for pollution control systems (Arena et al., 2012; ESA, 2012).

With specific reference to the chemical conversion process, the whole range of technologies can be grouped into two main categories: combustion- and gasification-based thermal treatment. The first is a well established and sustainable technology that results in considerable waste volume reduction with the added ability to

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reclaim a significant amount of energy. There are more than 900 plants in operation, having a capacity that ranges from 50 to 1000 kt/y: the most common type of combustion-based WtE technology used worldwide is the mass burn moving grate incinerator (Stantec, 2011; ESA, 2012). The second involves more complex processes and is less proven on a commercial scale, even though about 100 gasification-based WtE plants, having a capacity that ranges from 10 to 250 kt/y, are today in continuous operation, mainly in Japan but also in Korea and Europe. This suggests that gasification could today be proposed as a viable alternative for a WtE treatment, particularly if a dramatic reduction of the amount of residues to be disposed in landfills is required (Arena, 2012).

The paper aims to provide data for a critical comparison between a combustion- and a gasification-based process (in the following indicated as CB-WtE and GB-WtE), on the basis of a detailed analysis of the mass flows of materials and elements inside and throughout the units. The study focuses on the thermal treatment of the same municipal solid waste in a moving grate combustor and in a high-temperature vertical shaft gasifier with melting system, respectively. To this end, a large collection of data from several units in operation were processed by means of different analytical tools. In particular, a Material Flow Analysis (MFA) and a Substance Flow Analysis (SFA) have been carried out by means of the freeware STAN (subSTance flow ANalysis) developed by the Vienna University of Technology (Cencic and Rechberger, 2008). The MFA is a systematic assessment of the flows and stocks of materials and elements within a system defined in space and

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time (Brunner and Rechberger, 2004; Brunner, 2004), which is named SFA when it is referred to a specific chemical element. SFA is today largely utilized to link inputs and outputs of treatment process and management systems, so supplying data that are often relevant for the design, operation, and control of waste treatment systems. In a framework of increasing complexity of solid waste composition (Brunner, 2012), its ability to connect the sources, pathways, and intermediate and final sinks of each species in a specific process appears greatly attractive, as demonstrated by its utilization in the assessment of recycling options (Rotter et al., 2004), waste management scenarios (Morf and Brunner, 1998; Mastellone et al., 2009; Arena and Di Gregorio, 2012) and specific thermal treatments (Brunner and Mönch, 1986; Arena et al., 2011).

A combined MFA and SFA has been utilized in this study to define and compare the patterns of some key elements of municipal solid waste throughout the different sections of the two compared WtE plants. A particular attention was dedicated to the partitioning of low-boiling-point heavy metals and to their concentration in output solid streams with reference to reuse or disposal scenarios. It is in fact recognized that the recovery of metals and inert materials from waste-to-energy solid residues can become a crucial issue, from both environmental and economic point of view. It defines the possible recovery of valuable materials such as copper, iron and aluminum (Meawad et al., 2010; Rocca et al., 2012) and, above all, the reduction of the amount of residues to be sent to final disposal. This aspect is becoming crucial since the shortage of traditional disposal sites, together with stricter requirements for location and more severe environmental controls, have resulted in a strong reduction of the number of adequate sites for safe landfills, especially in areas at high density of population (Heller and Catapreta, 2003; UNEP, 2012).

2. Waste composition and WtE configurations

The fraction of MSW which is typically treated in a WtE unit is that residual from the operations of source separation and collection of dry recyclable and wet organic fractions. It is typically called unsorted residual waste (URW), and its composition varies widely between countries and within each country, depending on some factors such as the local standard of living and (quantitative and qualitative) levels of household separation and collection. This variation may affect the set of optimal operating parameters of the unit as well as the amount and characteristics of produced solid residues; rarely it can also affect the emission of pollutant species. Taking in mind these considerations, the waste composition utilized in this investigation is that reported in Table 1, as obtained by the Confederation of European Waste-to-Energy Plants as average data from 29 European States (Kreißig and Stoffregen, 2008; CEWEP, 2009).

The comparative analysis of combustion- and gasificationbased waste-to-energy processes was then developed with reference to the most utilized technologies for each WtE category, i.e. the moving grate furnace and the high temperature shaft reactor with direct melting (Arena, 2012). The first is the predominant mass burning technology: only in Europe it is utilized by 420 of the total 450 incinerators in operation. It is a well known and reliable type of furnace, with defined design and operating criteria (Stantec, 2011), even though new advanced solutions are continuously developed to improve the already high energetic and environmental performances (Gartner, 2011). The selected gasification-based technology is that most utilized in MSW gasification processes, with more than 40 units in operation. It is a high temperature gasification and melting reactor, with O2-enriched air injection in the melting section and the solid waste charged from the top of the vertical shaft furnace, together with coke and limestone (Tanigaki et al., 2012; Suzuki and Nagayama, 2011). A coke bed layer is formed in the lower part of the direct melting furnace and it is burned and kept at high temperatures in order to melt ash stably, to prevent cool-down of slag and accelerating waste thermal devolatilization and gasification. Limestone is added to provide some pH buffering of the melt and to form fluid slag that can be easily discharged from the furnace bottom. In some plants, a natural gas or LPG injection system is utilized to improve the carbon conversion ratio of the injected char (Tanigaki et al., 2008). From the top to the bottom of the gasifier it is possible to individuate a drying and pre-heating region (which operates at about 400 °C), a thermal decomposition region (between 600 and 800 °C) and a combustion and melting region (between 1000 and 1800 °C). The produced syngas is transferred to a swirling combustor that transfers the generated thermal energy to a boiler, which in turn powers a steam turbine that produces electricity (Arena, 2012).

The schematic configurations of the two WtE plants are reported in Figs. 1 and 2: both of them include similar heat recovery and air pollution control (APC) systems, so that the potential different performances could be mainly ascribed to the different conversion technologies. An accurate analysis has been carried out to define the set of values of the main operating parameters and that of reagent consumptions. The most important of them are listed in Table 2: for the CB-WtE unit the main sources were the Best Reference Document of European Community on Best Available Technologies for Waste Incineration (EC-IPPC, 2006), together with reports from several plants in operations, particularly in Italy and United Kingdom (C-Tech, 2003; CEWEP, 2009; Federambiente and Enea, 2012); for the GB-WtE unit the main sources were the scientific literature about large-scale plant operating experience (Tanigaki et al., 2008; Tanigaki et al., 2012), together with the results of a recent investigation about combustion- and gasification-based WtE units in Japan that also analyzed 15 gasification and melting systems (Matsuto, 2012). Some of these sources (Kreißig and Stoffregen, 2008; CEWEP, 2009; Tanigaki et al., 2008) contain also data of performances of the different sections

Table 1

Ultimate analysis and LHV of the unsorted residual	waste assumed as reference. Source of data: CEWEP	(2009).
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Element	Concentration (kg/t _{wet waste})	Element	Concentration (kg/t _{wet waste})	Element	Concentration (kg/t _{wet waste})	Element	Concentration (kg/tweet waste)
Ag	0.00017	Со	0.002	Mn	0.33	SiO ₂	99
AĪ	10	Cr	0.19	Ν	8.4	Sn	0.012
As	0.01	Cu	1.1	Na	4.4	Sr	0.000024
Ba	0.0000072	F	0.064	Ni	0.11	Te	0.00047
Be	0.00047	Fe	24	0	180	Ti	0.39
Br	0.00011	Н	40	Р	0.76	Tl	0.000046
С	250	Hg	0.0004	Pb	0.2	V	0.012
Ca	20	K	3.3	S	1.3	Zn	0.72
Cd	0.014	Mg	2.9	Sb	0.0071	H_20	340
Cl	3.6	Mo	0.00047	Se	0.000094	Ash	240

Low Heating Value = 9.8 MJ/kg.

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Fig. 1. Quantified flow sheet of the combustion-based WtE unit assumed as reference, as obtained by the Material Flow Analysis. Input data for MFA derived from CEWEP (2009) and EC-IPPC (2006).



Fig. 2. Quantified flow sheet of the gasification-based WtE unit assumed as reference, as obtained by the Material Flow Analysis. Input data for MFA derived from Tanigaki et al. (2008) and Matsuto (2012).

of the WtE units, which have been adequately processed in this study in order to obtain the partitioning coefficients related to the various gaseous and solid streams. These coefficients were then utilized to develop the material and substance flow analyses. It is here noteworthy to observe that the typical composition of URW from Japan (where are located most of the GB-WtE plants in operation) is different from that reported in Table 1, in particular for a higher percentage of wet organic fraction and a reduced content of ash. In order to take into account this aspect, the analysis mainly utilized operating data coming from a Korean GB-WtE plant (Tanigaki et al., 2008), which is fed with a waste more similar to that used as input data (Table 1). The performances of each section of the GB-WtE unit (and then, the values of their partitioning coefficients) were estimated on the basis of operating results reported in the study of Tanigaki et al. (2008). The MFA/SFA was anyway repeated by using the Korean waste composition as input data: the results confirm the expected limited variations in the element partitioning.

3. The fate of carbon, chlorine and sulfur

The MFA and SFA have been utilized to systematically assess the flows and stocks of materials and elements within and throughout the two WtE units taken in consideration, with the aim of quanti-

Table 2

Specific technologies and operating parameters of the combustion and gasificationbased WtE units. Source of data: EC-IPPC (2006), Gartner (2011), Matsuto (2012), Tanigaki et al. (2008).

	Combustion-based WtE	Gasification-based WtE
Waste conversion reactor	Moving grate	Vertical shaft with direct melting
Equivalence ratio	1.7	0.26
Oxidant medium	Air	Air and O ₂ -enriched air
		(O ₂ = 36%)
SNCR	Urea: 4.6 kg/t _{waste}	-
Additional fuel	-	Coke: 90.0 kg/twaste
		LPG: 9.0 kg/t _{waste}
Ash melting additive	-	Limestone: 70.2 kg/t _{waste}
Syngas conversion reactor	-	Swirling furnace
Equivalence ratio	-	1.2
Oxidant medium	-	Air
SNCR	-	Urea: 4.6 kg/t _{waste}
Air Pollution Control system	Dry scrubber	Dry scrubber
Absorption	Hydrated lime: 10 kg/	Hydrated lime: 6.5 kg/t _{waste}
-	t _{waste}	
Adsorption	Activated carbon:	Activated carbon: 0.5 kg/
-	1 kg/t _{waste}	twaste
	Bag filter	Bag filter
	0.	

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Table 3

Overall partitioning of main waste elements in the output streams from the combustion and gasification-based WtE units, as obtained by the substance flow analysis. The percentage values refer to the total input amount of each element.

Combustion-based WtE				Gasificatio	Gasification-based WtE			
Element content (%)	Bottom ash	APC residues	Flue gas	Slag	Metal	APC residues	Flue gas	
Carbon	1.0	0.9	98.1	0.1	0.0	0.6	99.3	
Chlorine	12.0	87.4	6.5×10^{-1}	0.4	0.01	98.9	$7.2 imes 10^{-1}$	
Sulfur	50.0	48.0	2.0	19.3	1.1	76.5	3.1	
Aluminum	90.0	10.0	2.6×10^{-3}	81.9	2.4	15.7	$4.0 imes 10^{-3}$	
Copper	95.0	5.0	2.1×10^{-3}	3.9	85.7	10.4	$1.0 imes 10^{-3}$	
Iron	98.0	2.0	$4.9 imes 10^{-4}$	7.6	89.2	3.2	$3.6 imes 10^{-4}$	
Silicon	95.0	5.0	9.0×10^{-4}	86.6	5.9	7.5	9.1×10^{-4}	
Calcium	70.9	29.1	$1.3 imes 10^{-4}$	21.4	0.7	77.8	$2.4 imes 10^{-2}$	
Sodium	95.0	5.0	2.2×10^{-3}	49.1	0.9	50.0	5.0×10^{-3}	
Potassium	50.0	50.0	2.2×10^{-2}	19.7	0.2	80.0	8.3×10^{-3}	
Magnesium	90.0	10.0	2.7×10^{-3}	74.3	2.0	23.7	$2.7 imes 10^{-3}$	
Lead	55.0	44.9	5.9×10^{-2}	1.1	0.8	98.0	$1.2 imes 10^{-1}$	
Zinc	45.0	55.0	2.5×10^{-2}	0.6	0.2	99.1	4.6×10^{-2}	



Fig. 3. (A and B) Substance flow analysis of the combustion-based and gasification-based WtE units, with reference to carbon.

fying and comparing the partitioning of different mass flow rates of solid and gaseous streams as well as that of some crucial elements. Table 3 summarizes the partitioning coefficients of most of the key elements: for some of them the mass flow rates in input to and output from all the sections of the units are also reported by the sequence of quantified flow diagrams in Figures from 1 to 6. An analysis of those in Figs. 1 and 2 confirms the major complexity of the GB-WtE: this is mainly related to the choice of coupling the melting furnace to the waste conversion reactor, in order to obtain slags and metals that can be immediately reused or recycled. It is also confirmed that gasification units produce a lower amount of exhaust gas (with a reduction of about 26% on mass basis and 29% on volume basis) mainly as a consequence of the lower excess air required for the syngas combustion.

The flow diagrams of Fig. 3 show the patterns of carbon element in the two units, even though the addition of coke and LPG makes difficult an immediate visualization of the fraction of carbon chemical energy that is "invested" in the GB-reactor for the ash melting



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Fig. 4. (A and B) Substance flow analysis of the combustion-based and gasification-based WtE units, with reference to chlorine.

and thermo-chemical conversion of the waste in a fuel gas. The carbon conversion efficiencies of the two units are both very high, about 99%, with a very limited amounts of unburned carbon in APC residues and bottom ash or slags.

The flow diagrams of Fig. 4, together with data reported in Table 3, quantify the patterns of chlorine, showing the very good performance of the APC system in both the WtE units, able to collect 87.4% and 98.9% of total chlorine input, respectively. It is noteworthy that the chlorine content inside slag from the gasification reactor is very low (0.4% on mass basis) and then that contained in the syngas burned into the swirling combustor is higher (Fig. 4B). This could imply higher corrosion in the heat recovery unit. On the contrary, the operating data (as those reported by Suzuki and Nagayama, 2011) indicate that these GB-WtE plants show lower losses of material from superheater surfaces (in the order of two or three times less) when compared to CB-WtE units that treat similar wastes. This behavior can be explained by the results of thermodynamic calculations and experimental tests (Osada et al., 2009) that indicated that most of Na, K, Pb, and Zn volatilize in the form of metal chlorides (NaCl, KCl, PbCl₂, ZnCl₂) at a furnace temperature of about 900 °C: this means that a not negligible fraction of chlorine is taken away from the formation of HCl. Moreover, the limestone addition further contributes to this effect by catching chlorine as calcium chloride. An analysis of data in Table 3 suggests that similar observations could be also made for the sulfur patterns in the two WtE processes: in the combustion system the partitioning is about 50% between bottom ash and APC residues while the higher temperatures of the gasification system induce a higher sulfur volatilization and then a higher content (about 77%) in APC residues. Finally, for both the WtE units, it is recognized that the APC system is able to intercept more than 90% of hazardous atmophilic elements, such as cadmium and mercury, and about 99% of dioxins (Kreißig and Stoffregen, 2008).

4. The fate of ferrous and non-ferrous metals

Table 3 also reports ferrous and non-ferrous metal partitioning in the two WtE proposed technologies. The recovery of these metals from WtE solid residues is of great interest for a series of reasons. First of all there is the known environmental and economic advantage of metal scraps recycling. Then it is recognized that metal recovery increases the technical feasibility of ash utilization for road construction and concrete production. For instance, the presence of Al can result in swelling and expansion phenomena that create troubles to these applications (Muller and Rubner, 2006). The flow diagrams reported in Fig. 5 visualize the patterns of aluminum, showing that it is largely present (90% of total Al input) in bottom ash of CB-WtE unit as well as in the slag of gasification-based one, even though to a lesser extent (about 82% of total Al input). The residual part volatilizes and subsequently concentrates on the surface of fly ash, mainly as aluminum oxide (Al2O3) and as metallic aluminum, respectively in the combustionand gasification-based process. It should also be noted that the Al inside slags coming from a high-temperature gasifier is present as amorphous phase and then is not recoverable. On the other hand,

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Fig. 5. (A and B) Substance flow analysis of the combustion-based and gasification-based WtE units, with reference to aluminum.

the amount of non-ferrous scraps, which is recoverable from the bottom ash of a combustor is remarkable and may represent a significant source of income (Grosso et al., 2011). However, this potential advantage is strongly related to the forms under which the Al-materials are present in the URW (mainly cans, trays and foils). It is known that small thickness (and then high exposed surface area) and low mechanical strength properties of some of these materials (trays and foils, in particular) are directly related to higher oxidation levels and to a higher production of fine particles, which are both conditions that do not allow any aluminum recovery by means of the actual technologies (Pruvost, 2012; Biganzoli et al., 2012). With reference to iron and copper, data reported in Table 3 indicate high percentages of both these metals in the bottom ash stream of CB-WtE (98% and 95%) and in the metal stream of GB-WtE (89% and 86%), respectively. However, an almost complete recovery is possible only in the gasification plants, where the Fe and Cu are collected under metallic forms. In the combustion units, iron and copper are instead mainly collected under oxidized forms, so reducing the actual recovery efficiency. The oxidation is further enhanced by water quenching of bottom ash after their discharge from the grate (Lopez-Delgado et al., 2003).

5. The fate of low-boiling-point heavy metals

The partitioning of low-boiling-point heavy metals among all the output streams is of great interest since it determines their emission potential and the related environmental hazards, mainly with reference to the reuse or disposal of solid residues (Jung et al., 2004, 2005; Takamiya et al., 2007). It is known that many factors can influence whether and in what form a trace element eventually ends up in the gaseous or a specific particulate phase. The most important among these factors are (Zevenhoven and Kilpinen, 2001): (i) how the trace element resides in the fuel; (ii) presence of halogens (in particular, of chlorine); (iii) presence of sorbent compounds; (iv) system temperature and pressure; and (v) oxidizing or reducing conditions. The latter two are the factors that can affect the fate of these elements in the reported analyses, since the same MSW was utilized as input fuel.

Table 3 reports the distribution of some of these trace elements. The attention was focused on Pb and Zn, due to their role in the leachability of solid residues from CB-WtE and GB-WtE, i.e. bottom ash and slag, respectively. It has been demonstrated (Osada et al., 2009, 2010) that in the gasification melting process the volatilization rate of Pb and Zn compounds was almost 100%, regardless of the molar ratio of Cl/Pb and Cl/Zn, so that these heavy metals volatilized as metals in the melting furnace and then were condensed as sulfides as the gas temperature decreases. Fig. 6 reports the fate of Pb along the different sections of the combustion- and gasification-based WtE units. Similar patterns have been evaluated for zinc, whose partitioning is strongly affected by the reactor environment: in oxidizing atmosphere, zinc orthosilicate (Zn₂SiO₄) and zinc spinell (ZnAl₂O₄) are formed, which are thermally stable; on the other hand, zinc chlorides (ZnCl2) formed in a reducing condition are favored for volatilization, as mentioned above (Jung et al., 2005). These observations together with data reported in Table 3



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Fig. 6. (A and B) Substance flow analysis of the combustion-based and gasification-based WtE units, with reference to lead.

suggest that in a GB-WtE unit the low-boiling-point heavy metals, such as lead and zinc, are substantially present only in APC residues, at rates of 98% and 99%, respectively. These results, coupled with those of slag leaching and acid-extraction tests (Suzuki and Nagayama, 2011; Tanigaki et al., 2012), indicate that slags from a GB-WtE unit could be generally assumed as recyclable. Moreover, XRD analysis carried out on these slags after different types and times of exposure showed no changes in chemical composition and negligible effects on enhancing constituent release (Sakanakura et al., 2009). The framework is different for bottom ashes from CB-WtE units, where low-boiling-point heavy metals, such as Pb and Zn, are largely present (about 50%). Moreover, studies on beneficiation of bottom ashes, as well as information related to their utilization together with cement-based materials, indicate that exposure conditions can change the mineralogical properties of the materials and, then, modify the leaching behavior of critical elements (Bayuseno and Schmahl, 2010; Wei et al., 2011). This implies that bottom ash from CB-WtE units cannot be considered as immediately recyclable. As a consequence, they are preferably sent to specific external post-treatment and recovery processes (ISWA, 2006; Meawad et al., 2010; Grosso et al., 2011) or to a melting treatment aimed to obtain a reusable product and then a reduction of volumes sent to landfill disposal (Jung et al., 2005; Gomez et al., 2009).

The material and substance flow analyses carried out in this study have been extended to all the main components of solid residues coming from the two thermal conversion processes. The related results were combined with some recent mineralogical characterizations carried out on this kind of residues from both moving grate combustors and high-temperature gasifiers (Baciocchi et al., 2010; Gori et al., 2011; Rocca, 2012; Rocca et al., 2012). These studies, and in particular that by Rocca et al. (2012), support a series of considerations: (i) bottom ashes from CB-WtE consist of an assemblage of a variety of crystalline phases while slags from GB-WtE are mainly made of amorphous glassy phases; (ii) no LOI (loss of ignition) or TOC (total organic carbon) are typically detected in slags from GB-WtE while values of about 5% of LOI and 0.35% of TOC have been detected in bottom ashes from CB-WtE: this is probably due to the ash melting temperatures (1200-1400 °C) of the GB-WtE, which are clearly higher than those (850-1000 °C) utilized in CB-WtE; (iii) the most intense diffraction peaks found by analyzing bottom ash from CB-WtE correspond to gehlenite (Ca2Al2SiO2) and calcite (CaCO3), but there is also evidence of significant contents of silica-containing phases, quartz (SiO₂), di-calcium silicate (Ca₂SiO₄) and akermanite (Ca₂MgSi₂O₇), as well as hematite (Fe₂O₃) and forsterite (Mg₂SiO₄); and (iv) for the slags, the XRD analysis reveals typically diffractograms with a wide hump, which is index of a consistent amorphous glassy matrix: therefore, only a few crystalline phases were identified, mainly akermanite (Ca2MgSi2O7) and magnetite (Fe3O4). Taking in mind these conclusions, a series of mass balances on atomic species has been carried out to determine the compositions of bottom ash and slag. The results are reported in Table 4, and appear in substantial agreement with those reported by the recalled mineralogical studies. The whole set of results obtained by the substance flow analysis allows to schematically summarize the overall fate

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Table 4

Bottom ash and slag compositions as derived by means of mass balances on atomic species, on the basis of some mineralogical characterizations (Rocca et al., 2012; Gori et al., 2011; Baciocchi et al., 2010; Saffarzadeh et al., 2009).

	Bottom ash from CB-WtE				Slag from GB-	Slag from GB-WtE		
	x (w/w)		<i>x</i> (w/w)		x (w/w)		x (w/w)	
Ca	0.096	CaCO ₃	0.012	Ca	0.209	CaO	0.193	
Si	0.234	SiO ₂	0.408	Si	0.227	SiO ₂	0.364	
Al	0.048	Ca ₂ Al ₂ SiO ₇	0.238	Al	0.073	Al(OH) ₃	0.186	
Mg	0.014	Mg ₂ SiO ₄	0.030	Mg	0.016	Ca ₂ MgSi ₂ O ₇	0.186	
Pb	0.001	Pb	0.001	Pb	0.0001	Pb	0.0000	
Zn	0.002	ZnMO ₄ ^a	0.005	Zn	0.0001	Zn	0.0001	
Na	0.022	NaOH	0.036	Na	0.029	NaOH	0.045	
К	0.009	K ₂ O	0.010	K	0.004	К	0.004	
Cl	0.002	NaCl	0.004	Cl	0.0004	NaCl	0.001	
S	0.003	CaSO ₄	0.014	S	0.002	CaSO ₄	0.008	
Cu	0.006	CuO	0.007	Cu	0.0003	Cu	0.0003	
Fe	0.125	Fe ₂ O ₃	0.175	Fe	0.009	Fe ₃ O ₄	0.011	
С	0.438	Ca ₂ MgSi ₂ O ₇	0.038	С	0.002	Cunburned	0.002	
0	0.001	Ca ₂ SiO ₄	0.010	0	0.426			
Н	0.013	Cunburned	0.012	Н	0.001			

^a Zinc is mainly present as Zn₂SiO₄ (zinc orthosilicate), ZnAl₂O₄ (zinc spinel) but also ZnFe₂O₄ and ZnCr₂O₄.

Table 5 Fate of elements in the output solid streams of the investigated combustion and gasification processes.

Class	Fate	CB-WtE	GB-WtE
1	Mainly in bottom ashes/slags	Al, Cu, Fe, Si, Ca, Na	Al, Si, Mg
2	Mainly in metals	-	Cu, Fe
3	Equally distributed between bottom ashes/slags and APC residues	S, K, Mg, Pb, Zn	Ca, Na
4	Mainly in APC solid residues	Cl, Cd*, Hg*	Cl, K, S, Pb, Zn, Cd ^a , Hg ^a

^a On the basis of data from Jung et al. (2004, 2005).

of some key elements in the output solid streams, as it is done in Table 5. The latter could be proposed as a possible classification of elements according to their behavior during the investigated combustion and gasification processes.

6. Conclusions

The study developed a substance flow analysis for the two most common technologies of combustion- and gasification-based WtE units, i.e. a moving grate combustor and a vertical shaft gasifier coupled with direct melting. The partitioning of mass flows of materials and elements has been consequently obtained on the basis of data coming from commercial units. A classification of key elements in the output solid streams according to their behavior during the investigated combustion and gasification processes has been proposed.

Moving grate combustion process is today recognized as a sustainable waste management option that allows to obtain a significant amount of energy and a considerable waste volume reduction. The output solid streams are bottom ash and APC residues (which include fly ash), for a total of about 25% of the mass input to the plant. Bottom ashes, in particular, are about 22% of the waste input and consist primarily of coarse, non-combustible materials and unburned organic matter collected in a quenching-cooling tank at the outlet of the combustion chamber. They have a heterogeneous composition and physical character: the concentrations of some heavy metals, such as cadmium and mercury, are significantly lower than that in APC residues since these are volatilized during combustion and combine with solid particulate residues; on the other hand, low-boiling-point heavy metals, in particular Pb and Zn, are equally distributed between bottom ash and APC residues. This implies that bottom ash from CB-WtE units cannot be considered as immediately recyclable.

High temperature gasification process coupled with direct melting system concentrates the original non-volatile inorganic elements into the slag, with a composition dominated by Si, Ca, Mg and Al. The low-boiling-point heavy metals, in particular Pb and Zn, are almost completely transferred to the gas phase and then caught by the APC system, while the high-boiling-point metals, such as Cu and Fe, are mainly concentrated in the metal stream and then completely reused. The results of substance partitioning indicate that slags from GB-WtE units could be assumed as immediately and completely recyclable. This implies that the investigated gasification-based technology allows a reduction up to about 65% of the amount of waste to be sent to final landfill, when compared to the conventional combustion-based WtE units.

This is a crucial aspect. The shortage or exhaustion of traditional disposal sites, stricter requirements for location, more severe environmental controls, and greater quantities of wastes have resulted in a strong reduction of adequate sites for safe landfills together with high disposal costs, especially in medium and large cities of industrialized countries and, in general, in areas at high density of population. And it is likely that it could become worldwide a major concern. On the other hand, this advantage is generally coupled with a significantly lower power production. An optimum must be therefore defined for each specific management framework only on the basis of an accurate assessment of local needs.

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3. Integrated and sustainable waste management system

In the last ten year regulation, planning and management of municipal solid waste have undergone dramatic changes. The difficulty of shifting from an oversimplified system – based on the collection of unsorted municipal wastes and transport to landfills located in quarries sites chosen independently from any rational planning – to an integrated one, interweaving waste reduction, material recycling and energy recovery, have become well apparent. A wealth of documents have been published; actions have been defined and developed by national, regional and local governments, collection and disposal waste authorities and private operators; an ample participation by citizens has been required and often obtained. These actions resulted in impressive outcomes: in Europe recycling and biological treatments reached respectively 24% and 18%, thermal treatments with energy recovery reached 20%, even though landfill utilization is still too high (38%).



Figura 18. Municipal solid waste management in Europe in 2009, source Eurostat.

Integrated solid waste management is typically governed by the 'ladder of Lansink', which specifies a generally accepted hierarchy of preferred methods to treat waste. Although reuse and recycling are preferred, energy recovery and landfilling are still key aspects of waste management. It could be argued that the issue concerning the optimal ranking of alternative treatments and solutions is still debated and that, at all events, hierarchies of whatever consistency do not always lead to the most effective waste management system and are not sufficient to develop complete, fully integrated and sustainable WM planning. The human health and environmental protection, resource conservation and aftercare-free management are widely recognized as the main goals of a sustainable waste management, particularly taking into account that there is a continuous increase of waste production and complexity of its composition. Since these goals are all substance-oriented, the substance flow analysis of specific elements have to be investigated, controlled, and addressed to appropriate processes. A series of Life Cycle Assessment studies were utilized to assess impacts for different sub-units composing a the MSW management system designed, based on the fundamental and recognized crucial role of the source separation and collection. The MFA/SFA permitting to quantify the mass flow rates of wastes and their main chemical elements, can provide scientific support to the decision-making process and ensure that the technical inputs to this process are transparent and rigorous.



Figure 19. Waste composition utilized for the study, evaluated on the basis of averaged waste fractions from 27 European Countries.

The composition of the municipal solid waste assumed as input data was evaluated and averaged in terms of different waste fractions, on the basis of different analyses carried out for Italian areas.
Then, for each specific waste fraction an ultimate analysis was developed in order to obtain a complete database for both main and trace elements, such as cadmium, chromium, lead and mercury. Different scenarios of source separation level, assumed to be equal to 35%, 50% and 65%, were developed continuously increasing the interception efficiencies of each waste fractions, in particular for organic matter, but also that of paper, glass and plastics. Collected dry fractions are addressed to the sorting and recycling chain, wet organic fraction processed into an anaerobic digestion plant coupled with a digestate post-composting, and unsorted residual waste, together with wood, paper and plastic residuals of the sorting and recycling processes, fuelled to a mass burn WtE incinerator. Results permit to identify the plant capacities for each specific treatment and decide the best practice in waste management on the basis of the landfill use, energy recovery and material recovery. In particular, the results of the substance flow analysis also indicate that too high source separation and collection levels, on the basis of the available sorting and recycling technologies, mean higher (specific) quantities of residuals and, above all, could address high amounts of toxic substances in the recycled products.

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A waste management planning based on substance flow analysis

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ABSTRACT

The paper describes the results of a municipal solid waste management planning based on an extensive utilization of material and substance flow analysis, combined with the results of specific life cycle assessment studies. The mass flow rates of wastes and their main chemical elements were quantified with a view to providing scientific support to the decision-making process and to ensure that the technical inputs to this process are transparent and rigorous. The role of each waste management option (recycling chains, biological and thermal treatments), as well as that of different levels of household source separation and collection (SSC), was quantitatively determined. The plant requirements were consequently evaluated, by assessing the benefits afforded by the application of high quality SSC, biological treatment of the wet organic fraction, and thermal treatment of unsorted residual waste. Landfill volumes and greenhouse gas emissions are minimized, toxic organic materials are mineralized, heavy metals are concentrated in a small fraction of the total former solid waste volume, and the accumulation of atmophilic metals in the air pollution control residues allows new recycling schemes to be designed for metals. The results also highlight that the sustainability of very high levels of SSC is reduced by the large quantities of sorting and recycling residues, amounts of toxic substances in the recycled products, as well as logistic and economic difficulties of obtaining very high interception levels. The combination of material and substance flow analysis with an environmental assessment method such as life cycle assessment appears an attractive tool-box for comparing alternative waste management technologies and scenarios, and then to support waste management decisions on both strategic and operating levels.

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1. Introduction

The decision making process over waste management (WM) policy is a complex issue, which has to evaluate and suitably take into account the environmental impacts, technical aspects, implementation and operating costs (preferably in a welfare economic perspective) of each specific treatment and disposal option as well as the social implications (Kinnaman, 2009; Massarutto et al., 2011; Ferreira da Cruz et al., 2012). The process often involves accurate as well as inaccurate or missing data, expert evaluation as well as il-defined and changing public opinion, and sometimes it is guided by preconceptions for or against specific waste management solutions, generally based on perception rather than on objective scientific evidence (Brunner and Ma, 2008). In the last decades, this framework has become increasingly complicated due to the growing generation and complexity of municipal solid wastes (MSWs) and the far-reaching changes that consequently occurred in their management. The latter have shifted from oversimplified procedures, such as the collection of unsorted wastes and their disposal in landfills, to integrated and sustainable systems, which have to work as a filter between human activities and the environment, providing a suitable balance between waste reduction practices, material recycling techniques, biological and thermal processes, and engineered landfill disposal (Arena et al., 2012). On the other hand, the decision making within this complicated framework does not appear adequately supported by existing regulations, such as those laid down by European Community Waste Framework Directive 2008/98. Such regulations are generally inspired by a precise ranking of solutions (the "waste hierarchy"), with material recovery to be preferred to energy recovery, and landfill to be considered as a last resort (EC, 2008). It could be argued that the issue concerning the optimal ranking of alternative treatments and solutions is still debated and that, at all events, hierarchies of whatever consistency do not always lead to the most effective waste management system and are not sufficient to develop complete, fully integrated and sustainable WM planning (Kinnaman, 2009). The same Waste Framework Directive is open to potential deviations from the hierarchy "where this is justified by life cycle thinking on the overall impacts of the generation and management of such waste [...]" (article 4(2)).

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Nomenclature

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AD	anaerobic digestion
APC	air pollution control
LCA	life cycle assessment
LHV	lower heating value
MBT	mechanical biological treatment
MFA	material flow analysis
MSW	municinal solid waste
OFMON	organic fraction of MSW
PCRs	polychlorinated binbenyls
PRDFc	polybrominated diphenyl ethers
SFA	substance flow analysis
SSC	source separation and collection
SOE	stabilized organic fraction
SOF	selection residues
SK	selection residues
22F	source separation level
RK	recycling residues
URW	unsorted residual waste
WEEE	waste from electrical and electronic equipments
WM	waste management
WtE	waste-to-energy

The considerations reported above indicate the need to adopt a comprehensive, systemic, goal-oriented approach based on indepth knowledge of the system behavior and able to provide reliable information about how environmental hazards can be minimized and potential resources maximized (Brunner and Ma, 2008; Mastellone et al., 2009). Since there is a general consensus about the main goals (protection of human health and environment; conservation of resources; and after-care-free management), and since these are all substance-oriented, the assessment tools cannot refer just to bulk flows of wastes and residues. The flows of individual substances also have to be investigated, controlled, and directed to appropriate treatments and sinks. In other words, given that individual substances are responsible for environmental loadings and resource potentials, it is necessary to observe the system even at the substance level.

The aim of the study is to describe the results of a WM planning that, in accordance with the observations reported above, is based on a substance-oriented approach. The final goal is to quantify the mass flow rates of wastes and their main chemical elements in order to provide scientific support to the decision-making process and ensure that the technical inputs to this process are transparent and rigorous. In this way, the stakeholders, i.e. any individual or organization with a legitimate interest, may be effectively involved in the decisional process (Clift, 2012). The approach was recently applied to three Italian areas, having different extension (from 2600 km² to 13,600 km²), population densities (from 72 inh/km² to 428 inh/km²) and per-capita waste generation rates (from 426 kg/(inh y) to 467 kg/(inh y)) (Provincia Caserta, 2011; Regione Campania, 2011; Arean and Di Gregorio, 2013a).

2. Methods and input data

The utilized approach is based on the extensive utilization of two valuable tools, the material flow analysis (MFA) and substance flow analysis (SFA), which can be efficiently used to support waste management decisions on both strategic and operating levels. MFA is a systemic assessment of the flows and stocks of materials and elements within a system defined in space and time (Brunner, 2004), which is called SFA when referring to a specific chemical species. Today, SFA is largely used to link the inputs and outputs of treatment processes and management systems, thereby supplying data that are crucial for the design, operation, and control of waste treatment systems. Due to the increasing complexity of solid waste composition (Bilitewsky, 2009; Brunner, 2009) what is highly attractive is SFA's ability to connect the sources, pathways, and intermediate and final sinks of each species in a specific process, as demonstrated by its use in the assessment of thermal treatments (Arena et al., 2011; Arena and Di Gregorio, 2013b), recycling options (Rotter et al., 2004) and waste management scenarios (Mastellone et al., 2009).

Following this approach, the methodology adopted for the desired substance-oriented waste management planning is made of a sequence of three steps. First, a series of life cycle analysis (LCA) studies is utilized to define the overall WM scheme and then to identify specific technical solutions to be included in the scheme. Only fully tested technologies, with proven technical reliability and environmental sustainability and with known total costs for treatment and aftercare were selected. In the second step, a specific MFA/SFA is developed for each of the recycling, biological, and thermo-chemical technologies of the defined management scheme, with the support of the freeware STAN (subSTance flow ANalysis) implemented by the Vienna University of Technology (Cencic and Rechberger, 2008). The final step applies the MFA/SFA to a series of alternative management scenarios, which are finally compared to each other. It is noteworthy that all the material and substance flow analyses have been developed on the basis of the transfer coefficients of the selected waste treatment processes, as obtained by mass balances extended to some crucial atomic species.

The composition of the municipal solid waste assumed as input data, i.e. the waste produced upstream of any form of separation and collection, was evaluated and averaged, in terms of different waste fractions, on the basis of different analyses carried out for Italian areas (Giugliano et al., 2011). Table 1 reports this composition, together with the ultimate analysis of each waste fraction, as obtained by different sources: Consonni and Viganò (2011) for the main elements, and Rotter et al. (2004), CEWEP (2009) and Zhang et al. (2011) for the trace elements. It should be noted that a certain variation in the value of cadmium, chromium and lead was found, in particular for the wet organic fraction, as may also be expected on the basis of different dietary habits. That said, the variation is always in the range of a few mg/kg (0.5-2 mg/kg for Cd, 3-12 mg/kg for Cr, 4-11 mg/kg for Pb) and was thus assumed to be negligible for the purposes of this study. The ultimate analyses were extended to these trace elements since a substance-oriented approach was adopted for WM planning. As mentioned above, a WM system cannot focus on the amount of total waste alone; it must also address the amounts of constituent substances (i.e., chemical elements and chemical compounds) since these determine whether waste has a resource potential or constitutes hazardous material. For instance, it is the content of heavy metals in the bottom ash of a wasteto-energy unit that determines whether this ash can be re-used, can be landfilled directly, or requires treatment before landfilling (Rocca et al., 2012; Arena and Di Gregorio, 2013b); similarly, it is the content of hazardous substances, such as persistent organic molecules and heavy metals, in waste of durable and non-durable goods or packaging that determines whether or not it can be safely recycled (Döberl et al., 2002; Brunner, 2009; Mastellone et al., 2009).

3. Results and discussion

3.1. Definition of the overall WM scheme

An integrated and sustainable waste management system should be defined and developed according to the following

LHV (MI/kg)

8

Ash (

(mg/kg)

£

Hg (mg/kg

6

0.057

4.85 0.84 0.02

> 2.00 1.00 1.00 1.00

5.63 0.02 0.02 4.92 8.06 8.06

8.00

06.43 2.74 3.63 7.0

0.047 0.007 0.072 0.072 0.072 0.027 0.027 0.027 0.027

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criteria (EC, 2008): (i) to minimize use of landfills and ensure that no landfilled waste is biologically active or contains mobile hazardous substances. These aspects could become crucial due to the continuously reducing space for locating sanitary landfills, which is becoming a major concern worldwide (UNEP, 2012); (ii) to minimize operations that entail excessive consumption of raw materials and energy without yielding an overall environmental advantage (McDougall et al., 2001); (iii) to maximize recovery of materials, albeit in respect of the previous point; and (iv) to maximize energy recovery for materials that cannot be efficiently recycled, in order to save both landfill volumes and fossil-fuel resources (Azapagic et al., 2004; Rechberger and Schöller, 2006). The waste management systems that are operating successfully worldwide were all developed taking into account the above criteria (EAI, 2005; Dornburg et al., 2006). In particular, they demonstrate that no one process is suitable for all waste streams; and no single waste management practice (i.e. landfill, recycling, biochemical or thermochemical conversion) can handle the full array of waste types and, at the same time, satisfy the waste management planning criteria reported above (Mastellone et al., 2009: Izquierdo López, 2010).

In the light of such considerations and on the basis of a series of LCA investigations (Clift et al., 2000; McDougall et al., 2001; Arena et al., 2003: Azapagic et al., 2004: Biorklund and Finnyeden, 2005; Izquierdo López, 2010; Rigamonti et al., 2010; Giugliano 2011), an overall WM scheme was built and adequate techsolutions to be included in this scheme were identified. The led LCA studies assessed impacts for different sub-units comng a MSW management system generally concluding that the treatment for packaging materials is recycling, the best one ganic waste is anaerobic digestion, while the unsorted residvaste should be treated by thermo-chemical conversion with gy recovery (Rigamonti et al., 2010). The WM reference scesketched in Fig. 1 is thereby based on the fundamental and al role of source separation and collection, which works as irst stage of the recycling chain and then of material recovery, lso as unavoidable preliminary sorting to best prepare the dry numid waste fractions for downstream processes. These are: (i) cling of dry fraction mainly made of packaging waste, and conng glass, paper and cardboard, wood, plastics and metals; (ii) gical treatment of the organic wet fraction (OF_{MSW}), obtained eparate collection; (iii) thermal treatment of the remaining rted residual waste (URW), i.e. the dry fraction that cannot eniently be recycled, from both environmental and economic ts of view; and (iv) landfill disposal of all the residues from ecycling, biological and thermal processes. This WM scheme es each of these options to the extent that is compatible with technical, economic and environmental performance and that ted to the characteristics of the specific catchment area (waste position, population density, etc.). Importantly, source separaand collection is assumed to be applied at high quantitative s. The qualitative level is also assumed to be high, even at the est source separation levels (SSLs), such that the amount of ues to be taken into account is limited as much as possible. The s always sufficiently high to make the mechanical biological ment (MBT) neither environmentally nor economically feasi-Read and Godley, 2011). Moreover, when the SSL is larger than the residual waste often contains insufficient organic material tivate the biostabilization process (Ragazzi and Rada, 2009). poor environmental and economic sustainability of MBT plants ther supported by SFA studies showing the limitations of their bage in-gold out" approach: "waste inputs usually contain subes that are either detrimental due to their subsequent use (e.g., er in iron scraps) or hazardous for the environment (e.g., cadn in sorted fractions for compost production)" (Brunner and 2008).

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Fig. 1. Flow diagram of the MSW scenario assumed as reference.



Fig. 2. Compositions of separately collected and unsorted residual wastes in the scenarios with 35%, 50% and 65% of source separation level.

Table 2

Interception efficiencies assumed for the scenarios 35%, 50% and 65% of source separation levels (MSW production = 1000 t/d).

-				-		-			
Waste fractions	Organic fraction	Paper	Glass	Plastics	Metals	Aluminum	Wood + textiles	Bulky waste and WEEE	Total
In MSW, % SCENARIO SSL 35%	35.0	25.0	6.0	15.0	3.0	1.0	4.0	11.0	100
Interception efficiency, %	40.0	44.0	55.0	25.0	30.0	30.0	15.0	10.0	35%
Separate collection waste, t/d	140.0	110.0	33.0	37.5	9.0	3.0	6.0	11.0	349.5
Unsorted residual waste, t/d SCENARIO SSL 50%	210.0	140.0	27.0	112.5	21.0	7.0	34.0	99.0	650.5
Interception efficiency, %	65.0	50.0	65.0	45.0	35.0	35.0	20.0	17.5	50%
Separate collection waste, t/d	227.5	125.0	39.0	67.5	10.5	3.5	8.0	19.3	500.3
Unsorted residual waste, t/d SCENARIO SSL 65%	122.5	125.0	21.0	82.5	19.5	6.5	32.0	90.8	499.7
Interception efficiency, %	80.0	65.0	90.0	60.0	55.0	55.0	25.0	28.2	65%
Separate collection waste, t/d	280.0	162.5	54.0	90.0	16.5	5.5	10.0	31.0	649.5
Unsorted residual waste, t/d	70.0	87.5	6.0	60.0	13.5	4.5	30.0	79.0	350.5

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Table 3

Solid residues for each fraction of MSW, expressed as percentage of treated materials in the sorting and recycling units, for the scenarios SSL 35%, 50% and 65% of source separation levels.

MSW fractions	SSL 35%	SSL 35%			SSL 50%			SSL 65%		
	SR, %	RR, %	SR + RR, %	SR, %	RR, %	SR +RR, %	SR, %	RR, %	SR +RR, %	
Organic fraction	20.0	9.7ª	25.7	22.0	9.7 ^a	27.6	24.0	9.7 ^a	29.5	
Paper	5.0	11.0	15.5	6.5	11.0	16.9	8.0	11.0	18.1	
Glass	6.0	0.0	6.0	14.0	0.0	14.0	16.0	0.0	16.0	
Plastics ^b	35.0	25.5	51.6	40.0	25.5	55.3	44.0	25.5	58.3	
Metals ^b	6.0	9.5	14.9	6.0	9.5	14.9	6.0	9.5	14.9	
Aluminum ^b	15.0	16.5	29.0	15.0	16.5	29.0	15.0	16.5	29.0	
Wood and textiles	13.5	5.0	17.8	13.5	5.0	17.8	13.5	5.0	17.8	
Bulky waste and WEEE ^c	10.0	10.0	19.0	12.0	12.0	22.6	14.0	14.0	26.0	
Total	14.7	9.5	22.8	19.0	9.8	27.0	20.9	9.9	28.8	

^a This is the percentage of residues produced during the aerobic step of the digestate, as obtained by Grosso et al. (2009).

^b It has been assumed that plastics, metals and aluminum are collected together by means of a multi-materials collection system.

^c The data related to this fraction derive from Bianchi (2008) by assuming conservative average values, i.e. close to the lowest ones.

The WM scheme adopted was further divided into a number of scenarios, which differ only in the source separation level, assumed to be equal to 35%, 50% and 65%. These values were selected because they are able to describe a possible high-quality evolution of source separation and collection practices and because they have already been used by some LCA studies aiming to determine the best MSW management system (De Feo and Malvano, 2009) or to quantify the best level of source separation (Giugliano et al., 2011; Massarutto et al., 2011). Each of these SSLs has to be considered as an average of those that can actually be obtained for each waste fraction (Table 2). Provided that suitable collection and storage logistics are set up, in order to obtain a high or very high SSL, it is useful to considerably increase the interception efficiency of some waste fractions, such as wet organic waste, paper, and glass. The first, in particular, is predominant in MSW composition (about 1/3 of the total) and can be intercepted at very high levels since it is easily recognized by the public. Moreover, wet organic waste requires special downstream treatment and its interception greatly improves the quality of the remaining waste. On the contrary, fractions with low densities (such as plastics) or little abundance (such as ferrous and non-ferrous metals) are more difficult to collect at high rates, i.e. rates higher than those obtainable just with the interception of packaging waste. These considerations have to be taken into account particularly for scenarios assuming the highest average SSL: 65% of source separation and collection calls for very high interception levels also for plastics, metals, and "others" including bulky

wastes and WEEE, as indicated in Table 2. Data in Tables 1 and 2 were then processed to obtain the composition of separately collected waste (SCW) to be sent to the recycling chain and biological treatment, and that of unsorted residual waste (URW) to be sent to thermal treatment together with the residues of plastics, paper, and wood recycling chains (Fig. 2).

3.2. Material and substance flow analysis of the WM options

All the waste treatment options that compose the WM scenario assumed as reference were defined on the basis of the above-mentioned LCA studies and Best Reference Documents of the European Community (EC-IPPC, 2006a, 2006b). A specific MFA/SFA was developed for each of these recycling, biological, and thermo-chemical technologies. The results are schematically reported below.

Recycling chain. Recyclable dry fractions of separate collections, mainly made of packaging waste, are sent to a sorting stage and then to the specific recycling process. The whole chain of the separate waste recovery/recycling process of paper, plastic, metals, wood, bulky waste and WEEE as well as that of the wet organic fraction have different process efficiencies. In other words, there is a non-negligible amount of material discarded in the sorting process (SR, selection residues) and of residues from the final recycling process (RR, recycling residues). The SSL is thus always higher than the fraction of materials actually recycled. The amounts of sorting



Fig. 3. Material flow analysis of the recycling chain of the multi-material dry recyclable fraction obtained from source separation and collection, in the scenario SSL 50%. Data are in t/d.

Main sources of input data are: Perugini et al. (2005); Giugliano et al. (2011).



Fig. 4. Material flow analysis of the recycling chain of the paper and cardboard fraction obtained from source separation and collection, in the scenario SSL 50%. Data are in t/d.

Main sources of input data are: Arena et al. (2004), Giugliano et al. (2011).

residues were determined on the basis of investigations carried out in Italian areas characterized by "best practices" of separate collection (Giugliano et al., 2011), while those of recycling residues derive from a number of LCA studies focused on the specific recycling chains (McDougall et al., 2001; Arena et al., 2004; Perugini et al., 2005; Giugliano et al., 2011). In particular, Table 3 indicates that the percentages of RR are constant for all the SSLs taken into account, while those of SR increase when the level of source separation and collection increases, as reported by different sources (Bianchi, 2008; Giugliano et al., 2011). The results of the material flow analyses are reported here for the two main waste fractions. They are shown in Fig. 3 for the recycling chain of the multi-material waste stream, which includes recyclable packaging made of plastic, ferrous metals and aluminum, and in Fig. 4 for the recycling chain of the paper waste stream. Both flow diagrams refer to an SSL of 50%. The input data for all these MFAs were mainly derived from the inventory tables of some of the LCA studies cited above (Arena et al., 2004; Perugini et al., 2005; Giugliano et al., 2011).

Biological treatment. The organic fraction from household source separation and successive separate collection (O_{MSW}) is sent to a preliminary sorting stage and to an integrated anaerobic digestion (AD) plant, including a final aerobic treatment. At the AD plant, the obtained biogas is used for energy generation and the resulting digestate is post-composted in order to obtain a material for agriculture, landfill capping or restoration of contaminated sites, thereby further reducing the mass of solid residues to be landfilled. AD is considered the best available process for biochemical conversion for several reasons: minimization of greenhouse gas emissions, stabilization of the organic fraction, energy recovery from the produced biogas, absence of emissions of bio-aerosols and bad odors, limited land surface use, and economic sustainability (Mata-Alvarez et al., 2000; Defra, 2007; Zhang et al., 2007; Khalid

et al., 2009). From an LCA perspective, all these reasons contribute to largely higher environmental sustainability with respect to other biological treatments, like aerobic composting (Arena et al., 2005; Hermann et al., 2011; Yoshida et al., 2012). MFA applied to the composition reported in Table 1 takes into account the amounts of SR and RR indicated in Table 3 and the transfer coefficients inferred from the scientific literature for a dry process (Mata-Alvarez et al., 2000; Grosso et al., 2009; Zhang et al., 2011). The biogas produced (about 60% methane and 39% carbon dioxide) is then burnt for electrical and thermal energy production. The estimated biogas production depends on a series of factors, such as the quality of the substrate, the hydraulic retention time, and the specific technology of the digester. Evaluation of electricity exported has to take into account that roughly a quarter of the total produced electricity is used internally for running the digester and the cleaning section (scrubbers and membrane filters) upstream of the internal combustion engine (Izquierdo López, 2010; Hermann et al., 2011; Banks et al., 2011). A conservative value of 0.12 MWhe per ton of OF_{MSW} was used for the net production of electricity while that of total (electrical and thermal) energy produced was assumed to be 0.5 MWh per ton of OF_{MSW}. Digestion also leads to the production of a digestate, which is sent to a post-composting section, considered as a typical industrial composting unit. The results are shown in Fig. 5. It should be noted that the post-composting process typically includes an addition of ligneous cellulosic organic materials (such as cardboard and wood), which was not considered in the analysis and therefore does not appear in the quantified flow diagram.

Thermal treatment. The unsorted residual waste is sent for thermal treatment as it is, i.e. without preliminary MBT, as a positive consequence of the source separation levels that characterize the proposed scenarios and work as a preliminary and efficient sorting stage. It was also assumed that all the combustible residues





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Fig. 6. Material flow analysis for thermal treatment by means of a combustion-based process of URW. Data are in t/d. Source of input data: Arena and Di Gregorio (2013b).

from recycling chains of paper, plastics and wood have to be sent to thermal treatment, for the following reasons: (i) the opportunity for a further reduction in landfill use. (ii) the possibility to reduce the wastage of feedstock energy (McDougall et al., 2001), and (iii) compliance with the European Directive 1999/31/EC prohibiting landfill disposal of any material having a heating value greater than 13 MJ/kg (EC, 1999). Thermal treatment is recognized as an essential component of any sustainable integrated MSW management system (Porteous, 2005; Psomopoulos et al., 2009). It can play a number of important roles: it reduces the mass and volume of waste, thereby preserving landfill space; it recovers energy from the solid waste stream; it allows the recovery of materials from solid residues; it destroys a number of organic contaminants that may be present in the waste stream; and it reduces greenhouse gas emissions with respect to anaerobic decomposition in landfills (Arena et al., 2012). It affords a further fundamental benefit, namely that of being able to separate inorganic components (metals such

Table 4

Summary of material, volume and energy flow analysis for the scenarios 35%, 50% and 65% of source separation levels (MSW production = 1000 t/d).

Scenario	SSL 35%	SSL 50%	SSL 65%
Mass of waste to landfill, % _{entering MSW} From sorting and recycling chain	0.7	1.2	2.0
From biological treatment	3.6	6.3	8.2
From thermal treatment	17.0	13.8	10.7
Total	21.3	21.3	20.9
Volume of Waste to Landfill, m ³ /d (% entering M	_{SW} ^a)		
From sorting and recycling chain	10.8	20.3	32.8
From biological treatment	60.0	105	137
From thermal treatment	101	82.4	64.0
Total	172(8.3)	207(10.0)	234(11.2)
Energy net production, GWh/y			
Electric energy	126	108	92
Thermal energy (in cogeneration)	313	265	211
Total	439	373	303
Lost and available feedstock energy, GWh/y (% entering wast	e energy)	
Converted in electric and thermal energy	770(78.1)	691(70.1)	612(62.1)
Lost in landfill	45(4.5)	68(6.9)	85(8.6)
Recovered materials, t/d (% entering MSW)			
Glass	31.0	33.5	45.4
Plastics	19.5	32.5	41.2
Metals	12.8	17.6	27.8
Aluminum	2.7	3.4	5.3
Paper	93.0	104	133
Textiles	2.5	3.3	4.1
Wood	4.2	6.2	8.7
Compost	24.3	38.5	46.1
Total	190(19.0)	239(23.9)	312(31.2)

 a Assuming a bulk density of collected MSW of 0.48 t/m³.

as iron, cadmium, lead, and non-metals such as chlorine, bromine, etc.) from the organic fraction (consisting of carbon, hydrogen and oxygen). This allows reuse or inertization, thereby preventing dispersion and accumulation of hazardous constituents not only in the environment but also in recycled products, where they can reach

Table 5

Summary of substance flow analysis for the scenarios 35%, 50% and 65% of source separation levels (MSW production = 1000 t/d).

Scenario	SSL 35%	SSL 50%	SSL 65%
Carbon to landfill, t/d (% input C)			
From sorting and recycling chain	0.8 (0.3)	1.4 (0.5)	2.2 (0.8)
From biological treatment	11.0 (4.2)	18.3 (7.0)	23.0 (8.7)
From bottom ash	2.0 (0.8)	1.7 (0.6)	1.5 (0.6)
From APC residues	0.9 (0.3)	0.8 (0.3)	0.7 (0.3)
Total	14.7 (5.6)	22.2 (8.4)	27.4 (10.4)
Cadmium to landfill, g/d (% input Co	a)		
From sorting and recycling chain	898(8.9)	1,577(15.6)	2,536(25.1)
From biological treatment	249(2.5)	405(4.0)	499(4.9)
Partial total	1150(11.4)	1980(19.6)	3030(30.0)
From bottom ash	831(8.2)	721(7.1)	589(5.8)
From APC residues	7477(74.0)	6488(64.2)	5293 (52.4)
Total	9450(93.6)	9190(91.0)	8920(88.3)
Cadmium in recycled product, g/o	d (% input Cd)		
Glass	81(0.8)	87(0.9)	118(1.2)
Plastics	312(3.1)	521(5.2)	660(6.5)
Metals	56(0.6)	78(0.8)	122(1.2)
Aluminum	3(0.03)	3(0.03)	5(0.05)
Paper	177(1.8)	198(2.0)	253(2.5)
Textiles	5(0.05)	7(0.07)	9(0.09)
Wood	4(0.04)	6(0.06)	8(0.08)
Compost	2(0.02)	4(0.04)	5(0.05)
Total	640(6.3)	903(8.9)	1180(11.7)
Lead to landfill, kg/d (% input Pb)			
From sorting and recycling chain	10.2 (5.5)	16.3 (8.9)	26.7 (14.5)
From biological treatment	1.2 (0.7)	2.0(1.1)	2.5 (1.3)
Partial total	11.4 (6.2)	18.3 (10.0)	29.2 (15.8)
From bottom ash	64.9 (35.3)	51.8 (28.1)	26.4 (14.4)
From APC residues	53.1 (28.8)	42.3 (23.0)	21.6 (11.7)
Total	129(70.3)	112(61.1)	77.2 (41.9)
Lead in recycled product, kg/d (%	innut Ph)		
Glass	13.3 (7.2)	14.4 (7.8)	19.5 (10.6)
Plastics	3.3 (1.8)	5.5 (3.0)	7.0 (3.8)
Metals	29.3 (15.9)	40.5 (22.0)	63.9 (34.7)
Aluminum	6.1 (3.3)	7.9 (4.3)	12.2 (6.6)
Paper	1.0 (0.6)	1.1 (0.6)	1.5 (0.8)
Textiles	0.2 (0.1)	0.3 (0.2)	0.4 (0.2)
Wood	0.8 (0.5)	1.2 (0.7)	1.7 (0.9)
Compost	0.3 (0.2)	0.5 (0.2)	0.5 (0.3)
Total	54.5 (29.6)	71.5 (38.9)	107 (58.0)



Fig. 7. Substance flow analysis of lead element for thermal treatment by means of a combustion-based process of URW. Data are in g/d. Source of input data: Arena and Di Gregorio (2013b).

hazardous concentrations (Brunner et al., 2004; Porteous, 2005; Kellner et al., 2011; Arena and Di Gregorio, 2013b).

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The thermo-chemical conversion technologies can be grouped into two main categories: combustion-based and gasificationbased thermal treatments. The specific thermo-chemical

technology considered in this study was the mass-burn movinggrate incinerator, which is that most utilized worldwide (CEWEP, 2009). The developed MFA/SFA quantifies the partitioning of total mass as well as that of some crucial elements throughout the different output streams, by taking into account the ultimate



Fig. 8. Material flow analysis of the waste management system for the scenario 50% of source separation level, for a 1000 t/d MSW production (I: import flow; E: export flow; dStock: amount of accumulated material).

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analyses of URW and combustible residues from recycling chains of plastics, paper and wood. Particular attention was paid to the partitioning of volatile heavy metals and to their concentration in output solid streams with reference to reuse or disposal scenarios (Jung et al., 2004, 2006; Rocca et al., 2012; Arena and Di Gregorio, 2013b). This aspect is crucial to define the potential for reducing the final amounts to be sent to landfill disposal, which used to be particularly relevant to densely populated areas but is now becoming increasingly important worldwide (UNEP, 2012). For the sake of simplicity, only two of the quantified flow sheets obtained by the MFA/SFA (and generally named "layers") are reported: that related to mass flow rates of the URW (Fig. 6) and the one pertaining to lead (Fig. 7). The net production of electricity has been assumed to be equal to 0.55 MWhe per ton of burned waste, which the Best REFerence Document of European Community on Best Available Technologies for Waste Incineration (EC, 2006a) reports as an average value. It is below the range 0.65-0.75 MWhe that can be obtained by an optimized installation for heat and power, which could be taken into account for WM planning for large catchment areas (Consonni and Viganò, 2011; Simoes and van Berlo, 2013).

3.3. Material and substance flow analysis of the alternative WM scenarios

A material and substance flow analysis was developed for each of the alternative scenarios of waste management. Tables 4 and 5 summarize most of the obtained results in terms of total mass, total volume, feedstock energy, carbon, cadmium, and lead, which are assumed as main environmental indicators on the basis of previous studies on goal-oriented WM planning (Brunner and Ma, 2008; Mastellone et al., 2009). The overall set of data allows to compare the scenarios and quantify the recycling, biological, thermal, and landfill facilities required for the good operation of a WM system. For space limitations, only 3 of the 18 quantified flow diagrams which result from MFA/SFA analysis are reported here, in Figs. 8-10: they refer to total mass, carbon and feedstock energy flow rates, as obtained for the scenario SSL 50% and a hypothetical MSW production of 1000 t/d. As shown in Fig. 8, wet organic waste is the largest fraction of materials from source separation and collection, due to the high interception level (about 45%, as reported in Fig. 2), and is sent for co-digestion treatment. The dry recyclable fractions (plastics, glass, paper, metals, etc.) are sent to



Fig. 9. Substance flow analysis of the waste management system for the carbon element for the scenario 50% of source separation level, for a 1000 t/d MSW production (I: import flow; E: export flow; dStock: amount of accumulated material).

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the appropriate sorting and reprocessing chain. Residues of the sorting and recycling processes of plastics, paper, and wood, which together account for 6% of total MSW, are sent to the WtE process along with the URW, thereby obtaining energy recovery and a mass reduction of 77% (138 t/d instead of 560 t/d). Only the residues that are not energetically valuable due to their low heating value or very limited amount (glass, ferrous metals, aluminum, organic and textiles) are sent for landfill disposal: they represent the 7.5% of total MSW, and then 15% of the stream obtained by household separation. It is noteworthy that the residues from the pre-treatment of the organic waste for anaerobic digestion could be energetically recovered, in order to further reduce the amount of waste in landfill. This option appears interesting but it has not been taken into account in the examined scenarios since it is strongly affected by the quality of household source separation, and the available information is not sufficient to develop a material and substance flow analysis. With reference to the whole SSL 50% scenario, 21.3% of total MSW is sent to landfills while the materials actually recovered, i.e. the fraction that comes back to the process industries,

amounts to 23.9%, hence less than half of the source separation level.

Fig. 9 quantitatively reports the mass flow rates for the carbon element for scenario SSL 50%, the carbon data for the other scenarios being summarized in Table 5. Carbon is an indicator of resource potential, such as energy and biomass, but also of environmental hazard, such as greenhouse gases and persistent and toxic organic substances (Mastellone et al., 2009). The main goals are to transform hazardous organic compounds into relatively harmless substances such as carbon dioxide and water, and to produce energy while mineralizing carbon to CO2. The flow diagram in Fig. 9 indicates that carbon in scenario SSL 50% is mainly transferred to the export streams from thermal and biological energy recovery processes (68.4%) and to a lesser extent (23.1%) to recycled materials. This means that only 8.4% of the carbon present in total MSW produced is disposed via landfill (see also Table 5), thus preventing its accumulation and reducing greenhouse gas production. With reference to all the proposed scenarios. substantially no organic carbon is landfilled and the amount of



Fig. 10. Energy feedstock flow analysis of the waste management system for the scenario 50% of Source Separation Level, for a 1000 t/d MSW production. All the data are reported in GJ/d (I: import flow; E: export flow dStock; amount of accumulated material).

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inorganic carbon in incineration residues to be landfilled is limited. This confers various benefits. The amount of greenhouse gas is reduced through high recycling rates (which in turn depend on the SSL and the best practices of separate collection and recycling processes): the carbon that is reutilized in the form of compost (from integrated biological treatment), polymers (from the plastic recycling chain) and cellulose (from the paper recycling chain) is not landfilled and will not contribute to greenhouse gases. The energy produced by thermal treatment and, to a lesser extent, by the AD process, replaces other energy sources, thereby reducing related environmental burdens (Clift et al., 2000; McDougall et al., 2001). Moreover, about half of the energy produced by WtE and AD plants derives from non-fossil materials and therefore does not contribute to climate change. With reference to hazardous organic carbon compounds, the WtE process is able to degrade them in a controlled way (i.e. in a plant equipped with an advanced APC system), while during landfilling and biological treatment most of these substances are released into the environment (Kellner et al., 2011).

The feedstock energy analysis layer reported in Fig. 10 shows that only 6.9% of chemical energy entering with MSW is wasted while 70.1% is available for power and heat generation and the remaining 23% is stored in the recycled materials. The data related to all the scenarios, reported in Table 4, confirm this positive aspect. It is also evident that the amount of feedstock energy converted to electric and thermal energy decreases (from 78.1% to 62.1%) when SSL increases from 35% to 65%, due to the reduced flow rates of URW; correspondingly, the feedstock energy lost in landfill increases, as a consequence of the larger amounts of sorting and recycling residues.

Data in Tables 4 and 5 allow the scenario evaluation to be extended also to volume, cadmium and lead. First of all, the amount of material sent back to the process industry, i.e. effectively recovered, is about half of that separated and collected at source (Table 4). The amounts recovered include the compost produced by the integrated digestion plants and are equal to 19.0%, 23.9% and 31.2% for SSL 35%, SSL 50%, and SSL 65%, respectively. Moreover, for higher SSLs, a larger volume of waste has to be sent to landfill (11.2% entering MSW instead of 10.0% entering MSW and 8.3% entering MSW), since the higher amounts of residues generated from the recycling chains and from biological treatments are not balanced by the lower amounts generated from thermal treatments (Table 4). This consideration applies (Table 5) also to cadmium and lead. Importantly, the Cd sent to landfill from the sorting and recycling chain increases from 8.9% inputCd to 25.1% inputCd when the SSL increases from 35% to 65% and, correspondingly, the Pb sent to landfill from the sorting and recycling chain increases from 5.5% inputPb to 14.5% inputPb. There is also a similar increase in the amount of these toxic substances in the totality of recycled products, as a direct consequence of the larger amounts of waste that are sent to the recycling chain. It is worth pointing out that, when higher SSLs and recycling rates have to obtained, long-lasting non-packaging plastic waste has to be included in the recyclable fractions. These materials are typically stabilized with metals, such as lead and cadmium but also zinc, antimony, PCBs, PBDEs, in order to fulfill their functions over a certain lifetime (Schlummer et al., 2007). Over time the recycling of waste containing these hazardous substances could lead to new material stocks that, in turn, could generate not only risks for its users but also problems for future waste management as a consequence of successive accumulations (Brunner, 2009). Thus, it appears crucial that waste management systems must take due measures to allow for the potentially high growth of such additives. The results and considerations reported above, together with the complex logistics and high cost necessary to collect materials with high interception levels (Giugliano et al., 2011; Massarutto et al., 2011), indicate



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Fig. 11. Recovered materials, recovered energy and required landfill volume for the scenarios 35%, 50% and 65% of source separation levels.

that levels of source separation and collection as high as 65% could have a limited sustainability. A great innovation and improvement in the recycling technology of each waste fraction (for instance, to obtain an efficient separation of toxic additives) appears to be the real challenge for the near future, with a view to maintaining or, better, to improving the environmental sustainability of recycling.

In order to make it easier to compare the three scenarios proposed, and thus the quality of the technical input provided for the WM decision-making process, Fig. 11 shows the results in terms of recovered resources (materials and energy) and required volume of landfill. SSL 35% maximizes energy recovery (126 GWh/y for power generation, 439 GWh/y in cogeneration) but has lower amounts of recycled products (69 kt/y). By contrast, SSL 65% represents the maximum source separation and collection effort, permitting larger recovery (92 GWh/y for power generation, 303 GWh/y in cogeneration). The implementation of a high quality source separation and collection system, together with the availability of up-to-date plants for biological and thermal treatments, strongly reduces the need of landfill volumes. Anyway, there is a slightly higher requirement for landfill volume in the SSL 65% scenario (234 m³/d, i.e.

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36.1% more than that for SSL 35%), due to the necessity of disposing the increased residues from the recycling chain (186% more than in SSL 35%) and from biological treatment (128% more than in SSL 35%).

4. Conclusions

A waste management planning was developed on the basis of an extensive utilization of material and substance flow analysis, taking into account a series of life cycle assessment studies on waste management systems and specific technological options. The approach appears able to compare alternative waste management technologies and scenarios, even though the results obtainable are just part of the input data to the decision-making process, which should further take into account a variety of economic and social aspects. Future work will investigate the possibility to complete the set of input data for the scenarios selected and assessed by the proposed approach. A Life Cycle Assessment, able to include the main social implications, and a Life Cycle Costing, carried out in a welfare economy perspective, could be adequate tools for this aim

The study quantified the benefits afforded by the application of high-quality household source separation and collection, biological treatment of the organic fraction from this separate collection, and thermal treatment of unsorted residual waste. Landfill volumes and greenhouse gas emissions are minimized, toxic organic materials are mineralized, heavy metals are concentrated in a small fraction of the total former MSW volume, and accumulation of atmophilic metals in the APC residue allows new recycling schemes to be designed for metals. An efficient household source separation level (i.e. 50% or more) works as a pre-sorting stage for MSW, preparing it for the different material and energy recovery chains. As a consequence, the resulting unsorted residual waste contains too limited an amount of both dry recyclable waste (to be sent to material recovery) and wet organic waste (to be sent to biological stabilization) to make the choice of an MBT sustainable. Moreover, the contamination level of the wet fraction produced by the MBT plant is usually so high that the "stabilized" organic fraction cannot be used for any purpose other than landfill cover

A significant reduction in the requirement of landfill volume can only be achieved by sending the unsorted waste, which is residual from source separation and collection, to a waste-to-energy process. As regards greenhouse gas emissions, the scenarios with high energy recovery provide more benefits, since they allow full utilization of the energy produced by carbon oxidation. On the other hand, landfills of residues from thermal treatments do not emit any greenhouse gases. Moreover, the combination of high recycling rates and efficient thermal and biological treatment means that inorganic materials are mainly concentrated in the residues of WtE plants while the hazardous organic compounds are completely destroyed

The reported results and considerations also suggest that the maximum level of source separation and collection should be defined on the basis of the overall sustainability, and then also on basis of the quantities of sorting and recycling residues, complexity and cost of logistics to obtain very high interception levels, and the amounts of toxic substances that could be present in the recycled products. This means that very high levels of separate collection could ensure the desired sustainability only with considerable improvements in sorting and recycling technologies, which should become also able to separate toxic additives (such as cadmium, lead, zinc, antimony, but also PCBs, PBDEs, etc.) from materials of value, thereby preventing their accumulation in recycled products.

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