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A feasibility assessment of an integrated plastic waste system adopting mechanical and thermochemical conversion processes

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ABSTRACT

The large variety and amounts of plastic waste produced worldwide requires to better organize the industrial network devoted to the exploitation of this material by including different processes that allow to recover the "material" as main target. This paper presents the results of the feasibility study developed for an integrated system for plastic waste management designed in such a way to deal with the real market and provide for reliable targets in term of material recovery yields, energy efficiency and waste minimization. The system under study is a combination of mechanical sorting, thermochemical processes and conversion into materials and energy. The quantified block diagrams are used to represent the mass and feedstock energy balances by allowing the calculation of yields of given products. The equipment list for each sub-system is provided together with the installed power for the main component and/or auxiliary; these data allowed to perform the energy balance and to obtain the net energy production by the integrated system. The energy balance demonstrated that the integrated system is feasible while, on the contrary, the single processes are not energetic self-sustainable.

1. Introduction and scope

1.1. Plastic waste management background

The amount of plastic materials produced worldwide reached 348 million of tonnes in 2017; about 18% of this amount was produced in Europe and more than 50% in Asia. A fraction larger than 50% of plastic converter demand is constituted by polypropylene (PP), low-density polyethylene (LDPE) and high-density polyethylene (HDPE) (Plastics Europe Annual Review, 2018). The extensive production of plastics and the indiscriminate disposal in the environment created also a question related to plastic waste disposal (Eriksen et al., 2014; López-López et al., 2018; Ritchie and Roser, 2019) and several alternatives for recovery and recycling have been proposed and assessed. The large variety of plastics and their various utilisation involves the necessity to find different processes able to obtain an environmental correct disposal and an optimised material and energy recovery. Depending on their physical and chemical characteristics, the collected plastic waste can be sent to mechanical reprocessing, to feedstock / chemical recycling process or to energy recovery and landfill (Al-Salem et al., 2009). A unique preferred process cannot be chosen for all the commercial plastics introduced in the market nowadays. The main non-

economic factors affecting the decision of which route is the most suitable for a certain waste are the composition in term of polymer type (HDPE, LDPE, PP, PS, EPS, PET, PVC, ...) and the fraction of non-polymeric materials (including multi-layered plastics and composites). These characteristics affect the design of the overall system, starting with the sorting facility where the commingled plastic waste coming from the separate collection of municipal and commercial waste is realised, until the recycling or recovery process, chosen as suitable tool to convert the waste into valuable materials (Zaccariello et al., 2015).

The most applied combination of processes in Europe consists of the centralised sorting at material recovery facility (MRF) followed by the mechanical recycling for given streams of PET and polyolefins (mainly HDPE) and by energy recovery for the remaining plastics mixed with the foreign matters. This latter fraction is not a small amount: the residual waste from MRF amounts to 40–60% of the input since it contains the foreign matter present in the collected waste and the plastics not sorted by the sorting line itself.

This residual waste is preferably sent to energy recovery or to production of secondary fuels for cement kilns and steel production industries as substitute of coals; unfortunately, despite of "landfill ban" existing in such Countries (Germany), a large of amount of this waste is sent to landfills. The cost for this final recovery/disposal in Italy, is, by

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referring to the gate fee only (i.e. transportation not included), reached very high values such as 145€/t (C.E.A. SpA, private communication, 2019).

The huge increase of cost to dispose this kind of waste in Europe was due to the so called “plastic ban” of China, following the so-called Green Fence, that introduced, for the first time after decades, quality specifications for secondary materials imported from Europe so dramatically limiting the plastic waste amount export from European Countries (Brooks et al., 2018). The restriction of use of landfilling for plastic waste imposed by the European regulation was another key point in the raising of disposal economic cost.

Nowadays, it is urgent to strengthen the industrial network devoted to the plastics’ recovery and recycling by including processes that asks for a lower degree of purity. The feedstock and the chemical recycling processes, once integrated in the recycling system, allow to use the same “equivalent petroleum amount” several times: as material, feedstock and fuel.

The mechanical recycling of plastics should be preferred when a mono-material collection of plastics must be treated, since the cost of the separation processes, as carried out in the material recovery facilities, is very high. Mechanical recycling consists of a series of physical operations where the recovered material is shredded, washed, melted and re-pelletized. In the case the mechanical recycling is not possible or convenient it is possible to refer to the feedstock and chemical recycling and, as last option, to the energy recovery processes. This last option is largely applied today for all the plastics that are not separately collected and for plastics that cannot be mechanically recycled. In fact, the heterogeneous mixture of plastic contaminated with other components (such as paper, biowaste, textiles, etc.) is sent to combustion process due to their large high-heating value¹ (about 31.8 MJ/kg for a household plastic mixture) (Themelis et al., 2011). Once energy recovery is applied, no other recovery is possible; in order to increase the number of life of fossil carbon, the preferred option is the material recovery that can be obtained by applying mechanical reprocessing and feedstock/chemical processes.

In all cases the mechanical recycling cannot be applied it is possible and convenient use the above cited alternative routes (Czajczyńska et al., 2017; Demirbas, 2004; Panda et al., 2010; Perugini et al., 2005). In particular, thermolysis processes of selected polymers and plastic waste mixture can lead to very good performances in term of energy recovery with a limited environmental impact. Most important, the pyrolysis and gasification processes can be applied even at smaller scale by making possible the integration with other facilities; for instance, gasifiers and pyrolyzers can be installed with thermal input capacities from 250 kW to several megawatts, by requiring small footprints and by favouring the real circular economy at local and regional scale. Several studies have been published on these processes applied to plastic waste and waste in general. Gasification processes differ for the applied technology of main reactor (gasifier), the method to minimize the tar formation, the cleaning/conditioning of syngas and its use. Gasification can be applied to heterogeneous plastic waste with good performances in term of syngas yield and cold gas efficiency (Gershman and B.I., 2013; Lopez et al., 2018). Pyrolysis of plastics aims to obtain preferably materials instead of energy or fuels. In this case the feeding composition is limited by strict specifications. The most applied and studied process for material recovery from plastic waste is pyrolysis of polyolefins

¹ High-heating value (HHV) indicates the energy content of one unit mass of matter that can be released during oxidation. HHV is an intrinsic property of matter since it is correlated to chemical composition. The low-heating value (LHV) is obtained starting from the HHV value by taking into account that the hydrogen contained in the matter is transformed into water at standard conditions (25°C) but, since the real temperature reached by oxidation is much larger than 100°C, the water is actually present under form of gas. The phase transition requires an amount of heat (2257J/g) that is subtracted to HHV by leading to the LHV. LHV is also an intrinsic property of molecules/compounds.

(Adrados et al., 2012; Haufe et al., 2004; Sogancioglu et al., 2017).

1.2. Feedstock recycling of polymers

Polymers are the main component of the “plastics family”; they are constituted by a repeating structure of monomers basically composed by carbon and hydrogen and, in some cases, by heteroatoms like oxygen, nitrogen, chlorine, ...; they are generally classified, according to their structure and properties, on the basis of thermal-mechanical behaviour and on the basis of their processing characteristics in thermoplastics, elastomers and thermosets. They can be also be classified according to their mechanism of polymerization as either addition or condensation, where:

- a polyaddition consists in the repeating of the same monomer along the chain;
- b condensation requires instead the bond between two different molecules.

Thermoplastic polymers such as PE, PP, PVC, PS are examples of polyaddition polymers; PET is an example of polycondensed polymer.

The polyaddition polymers, with the exception of PVC that has a peculiar behaviour (Sheirs and Kaminsky, 2006) can undergo thermolysis in a controlled environment by producing a large spectrum of hydrocarbons having a number of carbon ranging from 1 (methane) to around 20. The thermolysis of plastic waste is in fact oriented to recover raw materials for petrochemical industries by means of processes such as liquid and gas phase hydrogenation, steam-cracking, catalytic cracking, pyrolysis, coking and gasification.

A classification of thermolysis process into “feedstock recycling” and “chemical recycling” (sub-category of feedstock recycling) also exists with reference to the different process outputs that are obtained. Chemical recycling consists in the depolymerization of certain condensation or addition polymers back to monomers. The chemical recycling allows the re-creating of the chemicals from which the polymers were initially made. If the treatment breaks the polymers into an assortment of chemical species, it can be decided whether to recover specific chemicals for feedstock use or to use the assortment of chemical species for fuel or to use some combination of both end products; in this case the process lays in the more general definition of feedstock recycling. A special class of feedstock recycling processes yields an important raw material called syngas (=synthesis gas, a mixture of hydrogen and carbon monoxide): in this case the common name to indicate the thermal conversion process is “gasification”. This latter process is carried out in an oxidative environment where the oxygen content is much less than the stoichiometric demand for complete combustion (Gartzen et al., 2018; Mastellone, 2015).

Hydrocarbons and syngas can be used as chemical feedstocks for further upgrading to commercial products at oil refineries and chemical plants.

The plastic conversion into a sort of synthetic crude oil (syncrude) can be obtained by using commercially available technologies that are reported and compared in term of reactor technology, process type (thermal/catalytic), yields of products, capacities. A list of suppliers that developed the above cited catalytic and non-catalytic thermolysis is reported in the Table 1. The common point of the largest part of the listed technologies is the plastic feedstock specifications: all the polyolefins can be accepted, polycondensed polymers must be avoided, cellulosic materials and moisture must be limited as much is possible. The differences between the technologies are related to the reactor used for thermolysis, the presence or not of a catalyst and the maximum capacity of a single reactor that normally does not exceed 25.000 ton/year.

Table 1
List of technological suppliers for plastic-to-oil plants.

#	Company Name	Country
1	Agilyx Inc.	USA
2	Alphakat GmbH	Germany
3	Anhui Oursun Environment & Technology Corporation	China
4	Agile Process Chemicals LLP	India
5	BlueAlp bv / (Petrogas, Gas-systems bv)	The Netherlands
6	Climax Global Energy	USA
7	Envion	USA
8	GEEP	Canada
9	JB I Global / Plastic2Oil	USA
10	Klean Industries, Inc.	Canada
11	MK Aromatics Limited	India
12	Nexus Fuels	USA
13	Plastic Advanced Recycling Corp. (PARC)	USA
14	Plastic Energy Ltd	UK + Spain
15	Polymer Energy LLC	USA
16	PRYME b.v. / (BTC b.v.)	The Netherlands
17	Pruvia Fuels GmbH	Germany
18	Pyrocrat Systems LLP	India
19	Renewlogy (Formerly: PK Clean Technologies Inc. (Salt Lake City))	USA
20	Res Polyflow	USA
21	Vadxx	USA

1.3. Feasibility assessment method

A feasibility study includes a series of stages related to different aspects of a project with the aim to give an orientation to the management devoted to take the “go/no go” decision for the following steps of the project development (generally referred as “gates”). A part of a feasibility study is dedicated to the “technical assessment” whose target is finding answers to questions such as: is the proposed technology or solution practical? Is the technology mature and reliable? If not, can it be obtained? At which risk? And so on. The main question is related to the practicality of the proposed solution, its effects on the market and/or on the system where it is applied, on the environment, on the economy, etc. Whatever are the methods and procedures used to perform the technical feasibility assessment, the material flow assessment (MFA) carried out at level of goods resulted instrumental for understanding how the proposed process or system (e.g. a waste management system) functions, facilitating connections and communications between the stakeholders, authorities, and involved companies. Allesh and Brunner (2015) demonstrated that “MFA has become a common, widely used tool to analyse waste management systems on different levels (goods and substances) and with various goals. MFA on the level of goods is highly useful for understanding WM systems. It represents a

tool for analysing, controlling, and managing material flows within a system”.

1.4. Scope

This paper aims to demonstrate that the integration of thermolysis processes such as pyrolysis of polyolefins mixture and gasification of plastic waste coupled with a mechanical sorting is technically and energetically feasible and reaches targets of material recovery yields, energy efficiency and waste minimization.

2. System description

The system under study aims to recover the largest amount of secondary materials and energy from plastic waste. As recalled before, the plastic waste taken as reference is the leftover of material recovery facilities processing the household/commercial mixed recyclable waste obtained by means of separate collection; this leftover is highly contaminated by non-recyclable plastics and foreign materials (paper, metals, textiles). The facility that can treat this kind of waste in a sustainable way must be integrated with processes that minimize the energy consumption, the waste production and that maximise the value and the yield of products. The proposed system is an example of an integration between mechanical and thermochemical processes aiming to obtain the maximum economic revenue with the minimum environmental burden.

The system is explained with reference to the block diagram of Fig. 1.

It is basically composed by the following sections:

- the sorting section (P1), where the plastic waste coming from the separate collection is sorted and separated into several streams including: a) the mono-polymeric streams destined to mechanical recycling (such as PET, HDPE, PP); b) the mixture of polyolefins (plastic feedstock) that is too difficult and expensive to separate into mono-material streams for mechanical recycling and that can be destined to feedstock recycling to oil and/or feedstock; c) the mixture of residue (plastic waste) containing the polymers not recognized during the sorting, the polymers that are not suitable to be recycled, the foreign matter (biowaste, cellulose, composites, multi-layered).
- the pre-treatment section (P2), where the plastic feedstock is shredded, dried and melted to be fed into the thermolysis section. In this section the densification and drying of the plastic feedstock is followed by de-halogenation by means of degradative extrusion. This process helps to reduce the halogen content, and chlorine in particular, lesser than 1%, as required by the process P3.

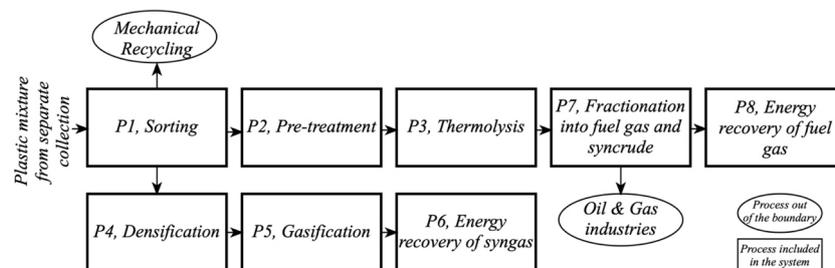


Fig. 1. Simplified sketch of the system under study.

- the thermolysis section (P3), where the molten polymer flow is converted into pyrolysis products (P3) and then, by using a series of condensers, is fractionated at least into syncrude (F18) and fuel gas (F15), or in more streams having different boiling points. The term syncrude is used to highlight that the destiny of this product is its use as blending component of crude oil in the refinery industry or as a raw material to be processed in the chemical industry.
- the gasification section (P5), preceded by a densification (P4) of the plastics and other burnable leftovers discharged from sorting section, where the waste, otherwise destined to landfilling of incineration, is converted into a synthetic gas i.e. syngas (F6);
- the energy recovery section, that includes an engine (P8) fed by the pyrolysis gas (F15) and an engine (P6) fed by syngas. The properties of the two fuel gas streams are quite different: the fuel gas from the process P3 is similar to LPG; the syngas is a mixture of carbon monoxide and hydrogen. Studies about their use as blended fuel gas in a unique engine are not available.

The mass and feedstock energy balance and the total energy assessment are presented in the following paragraphs. To obtain these data, a series of input data have been set up, the input data of the whole modelling are:

- plastic mixed waste (PMW) composition entering the integrated system;
- yields obtained by mechanical equipment and by chemical reactors (pyrolizer and gasifier);
- composition of pyrolysis products and syngas;

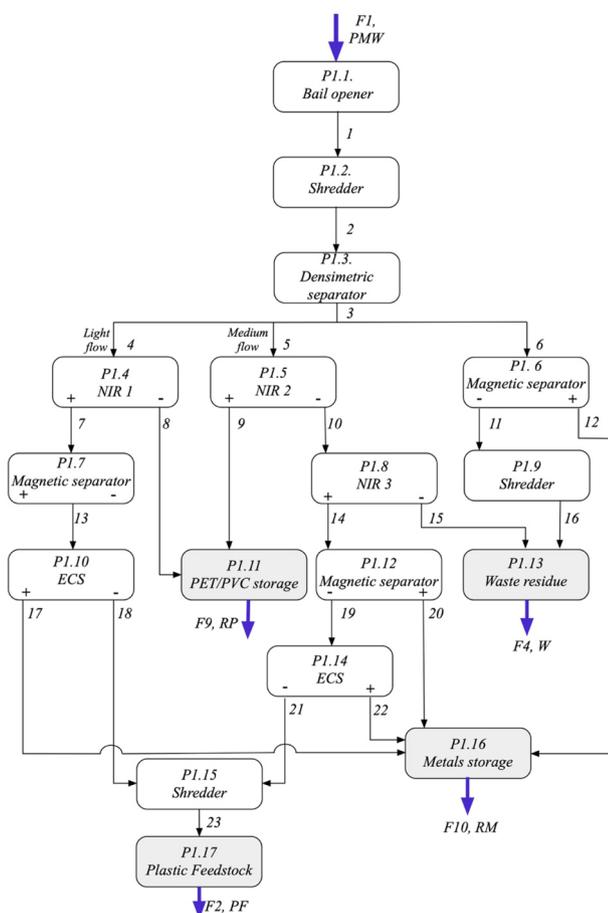


Fig. 2. Block diagram of sorting section P1. Details of main equipment composing this section of the overall system under study. The flows indicated by larger arrows are: input data = F1 (PMW); output data = F4 (W₁), F9 (RP), F2 (PF).

- electrical conversion efficiency for engine.

2.1. Mechanical sorting

The mechanical sorting process (P1) is realized by using both physical and chemical properties of the materials. First of all, the plastic waste bales are opened and roughly shredded. After this preliminary treatment a separation of flows by density is made by using an air drum separator. The flows having different density are addressed to optical sorting that is carried out by using Near-InfraRed (NIR) equipment that detects the chemical composition of materials on the belts and fires by high-pressure air jets those detected as “positive” flows. The rest of materials constitutes the “negative” flow. The sign “+” indicates that the equipment is able to detect as positive a given flow (Fig. 2); generally, the detection of materials as positive flow ensures a high level of purity of the flow if the distribution on the accelerating belt is guaranteed. The densimetric separation has been chosen because of the huge presence of films (LDPE, PP) that can envelope the heavy but smaller materials by decreasing the purity of the flows. The light density flow is sent to a unique NIR because the presence of PET/PVC/glass, etc. is expected to be in the higher density fractions that is sorted with more accuracy.

The composition of plastic feedstock needs to meet specifications so that the design of the sorting plant must be adapted until the obtained plastic feedstock composition complies with the specifications. For instance, PVC, PET, multi-layered compounds, etc. must be removed with large efficacy so that the optical sorters have to be placed with such a redundancy.

In the block diagram of sorting plant under study, the PET and PVC polymers are detected as positive flow in an optical sorter dedicated to the medium density flow. A second optical sorter is added in series to the first one in order to remove all the other impurities by detecting polyolefins as positive flow.

The composition of material flows obtained by the sorting has been obtained by applying a mathematical modelling reported in the paragraph “Materials” with reference to the detailed block diagram of Fig. 2.

The magnetic separator removes ferrous metals while the ECS is able to remove the non-ferrous materials by allowing to obtain a clean flow of polyolefins. After an intense shredding and homogenization, the flow of polyolefins becomes a plastic feedstock suitable to be addressed to a plastic-to-oil (PtO) section.

2.2. Plastic-to-oil section

The thermolysis section is organized to produce a mixture of hydrocarbons ranging between C1 and C30 that undergoes a fractionation based on the boiling temperature (T_b) of components; three fractions are obtained: a fraction from C1 to C4 (non-condensable gas or NCG), a fraction from C5 to C21 (similar to the crude oil and then utilized for blending with it at the refinery inlet named syncrude) and a heavier fraction that constitutes a bottom residue ($T_b > 350$ °C) recirculated to the cracking reactor. The reactor is fed by polyolefins with a limited amount of foreign matter (cellulose, glass, paper, etc.) and traces of undesired polymers such as PET or PVC. The thermal cracking in a reactor is operated at a temperature between 450 and 480 °C. More specifically a temperature of 480 °C is used in the primary cracking zone (heterogeneous zone) and a slight lower temperature (450–460 °C) is established in the homogenous zone. The kinetic model used to predict the products distribution and the reaction time is based on model available in the literature (Al-Salem and Lettieri, 2010) as modified on the basis of a global reaction model not presented here. The resulting yields of products are: 61.3% of waxes, 18.5% of liquids, 7.6% of aromatics, 10% of NCG. On the basis of industrial experiences, a certain amount has been considered as a process loss due to tar formation during cooling and partial condensation; this led to consider, for

the following calculations, a fraction of liquid product (syncrude) equal to 80% and NCG equal to 10%. The rest of the matter is converted into char and tar (Mastellone et al., 2002; Scheirs and Kaminsky, 2006).

The PtO plant (based on the Pruvia Fuels GmbH technology) is preceded by a pre-treatment that removes moistures and halogens from the plastic feedstock. This pre-treatment is carried out by using a combination between densifiers and degassing extruders (AMUT Group, 2019). The densifiers allow to manage polyolefins having very light bulk density (LLDPE) and heterogeneous mechanical properties. The extrusion is a pre-treatment that allows to remove moisture with 99% efficacy if the initial content is less than 10%, to remove organic chlorine by breaking the C-Cl bonds in the initial part of machine by allowing the degassing together with moisture, to bring the temperature up to 350 °C so favouring the input to the thermolysis reactor (pyrolizer). Downstream the extruder, the following equipment is installed:

- 1) Heated/insulated feeder to bring the plastic feedstock in the reactor over the melting temperature
- 2) Thermolysis reactor
- 3) Carbon/ash discharge system
- 4) Cooling of pyrolysis products
- 5) Fractionation of pyrolysis products
- 6) Light fuel gas energy recovery (heat, electricity or both)
- 7) Auxiliaries (scrubber, cooling water production, ...)

The reaction enthalpy necessary to break the chemical bonds of polymers can vary (Brems et al., 2011); a mean value of 250 MJ/t has been used by considering a prevailing amount of polyethylene. Starting from ambient temperature, a sensible heat of 1200 MJ/t needs to be added to reach the reaction temperature (total energy: 1450 MJ/t). It is noteworthy that the sensible heat acquired by the molten polymer in the extruder is in the range 60–70% of the total need. This means that the electricity used to operate the extruder is used to increase the enthalpy of the plastic flow and only a limited amount is transferred to the pyrolizer by using waste heat from engines (e.g. by using heat exchangers crossed by hot diathermic oil as heat vector).

2.3. Gasification process description

Gasification is a thermochemical process that transforms a carbon-based material into a gaseous mixture of low molecular weight species. What's left is a clean "synthesis gas" that can be converted into valuable products and electricity. In particular, gasification of waste is an economical and environmental viable solution to produce cleaner energy together with a remarkable waste weight reduction. The most simplistic way to consider gasification is as an alternative to the combustion to obtain heat and power with a lesser environmental impact. In this case the gasification process is used to transform a heterogeneous fuel (solid waste, sludge, biomasses, low-rank coal, ...) into a homogeneous synthetic gaseous fuel to be utilized in an industrial burner, an engine or a gas turbine to produce electricity and heat. The loss of chemical energy necessary to promote the endothermic reactions of gasification is balanced by the higher performance of the homogeneous combustion. The general concept and the technologies utilized to gasify a material is similar to that reported for incineration facilities, but the operating parameters of the plant are completely different.

First of all, the gasification process cannot be represented by a single main reaction, as for the combustion process, but by reactions involving different reactants (oxygen, carbon dioxide, water) and characterized by different reaction's enthalpies (Mastellone, 2015). Moreover, the interaction between intermediates and minerals must be considered,

particularly for plastics (Mastellone and Arena, 2008; Sjöström et al., 1999).

The heat released by exothermal reactions increases the environment temperature until values depending by the equivalence ratio, proximate and ultimate analysis of waste and its net chemical energy. The operating temperature of the waste gasifiers is generally in the range 850–1200 °C depending by the above recalled parameters and by heat losses. The oxygen supplied to the gasifier is generally 25–40% of the stoichiometric demand. The reducing environment promotes the partial oxidation of carbon and hydrogen element in the fuel by providing the heat necessary to the cracking of waste/fuel component. The fraction between the oxygen provided for gasification reactions and the stoichiometric demand is known as equivalence ratio and its value depends on ultimate analysis of the waste. The main product of gasification process is the synthesis gas also identified as "synthetic gas or syngas". This term is used when it is used as a feedstock for production of synthetic natural gas (SNG), Fischer-Tropsch liquids (FTL), hydrogen or any other material or fuel; the term "producer gas" is used when the gas is used to energy production. Anyway, whatever is the final utilization, the term syngas can be used to indicate the gaseous product of gasification. The syngas composition can vary depending on waste and operating conditions, but the main constituents are carbon monoxide, hydrogen, methane, C₂-C₆ hydrocarbons, carbon dioxide and water.

The general layout of a gasification plant includes:

- 1) Mechanical feeder at ambient temperature
- 2) Gasification reactor
- 3) Bottom ash discharge system
- 4) Cooling of syngas
- 5) Cleaning / upgrading of syngas (thermal/catalytic cracking, tar absorption, ...)
- 6) Syngas energy recovery (heat, electricity or both)
- 7) Auxiliaries (scrubber, cooling water production, ...)

The data used to model the performance of the gasifier section in the following paragraphs come from an extensive series of experiments carried on the plastic waste by using a pilot-scale fluidized bed gasifier (Arena et al., 2010, 2008; Mastellone and Zaccariello, 2013). The main performance parameters for the gasification of plastic waste mixture are reported in the following (§4.3) and are related to a specific pilot plant operated with an oxidizing stream constituted by air having an equivalent ratio of about 0.3. The tar content in the syngas has been considered negligible thanks to use of catalytic reaction in the gasifier (Arena et al., 2009). In the case of gasification, the absorbed process energy can be evaluated by considering that the exothermic reactions uses a fraction of the feedstock energy of the input waste to guarantee a given temperature and to promote the endothermic reactions; this fraction of energy is the complement of the Cold Gas Efficiency parameter (CGE).

3. Materials

The input material to which this paper refers is the plastic waste residual from the centralized sorting at material recovery facility (MRF). This plastic waste has a low content of PET and a high content of polyolefins, other polymers and foreign matter. The sorting of this waste with the only aim to recover the PET and the HDPE, that are the only polymers suitable for the mechanical recycling, would be not economically convenient because of the largest part of the flow (about 90%) that should be addressed to disposal (landfilling or incineration). By referring to the integrated system under study, where the sorting is finalized to prepare the feedstocks for PtO and gasification processes,

Table 2
Material flows and brief description.

Name	Short name / flow code in the block diagrams	Description
Plastic mixed waste	PMW / F1	Mixture of materials with a high predominance of plastics but with presence of metals, paper, textiles and composites
Plastic feedstock	PF / F2	Mixture of polymers fed to the thermolysis reactor in order to be converted into syncrude and by-products
Waste residue from sorting	W_1 / F4	Mixture of materials having a high feedstock energy that cannot be mechanically recycled or converted into syncrude
Synthetic gas	Syngas / F6	Mixture of carbon monoxide, hydrogen, hydrocarbons and a variable content of nitrogen
Synthetic crude oil	Syncrude / F18	Mixture of heavy hydrocarbons having a molecular weights range between 72 and 400 g/mol that can be fractionated in a distillation column to obtain kerosene, gasoline, jet fuel, etc.

Table 3
Properties of polymers of interest.

Polymer name	Acronym	Monomer molecular weight g/mol	Density range kg/m ³	Melting point °C	Specific heat capacity J/g/K	Heat of fusion kJ/mol	Lower Heating Value*** MJ/kg	Tensile strength**MPa
Polyethylene (mix)	PE (LDPE – HDPE)	28.05	825 - 1000	110	1.55 – 1.76	7.87	24 - 38	10 – 32
Polypropylene	PP	42.08	850 - 936	176	1.63 – 1.76	9.92	44	26
Polyethylene terephthalate	PET	192.16	1355 - 1455	265	1.13	24.12	24	55
Polystyrene*	PS	104.15	1050 - 1060	264 - 272	1.20 – 1.28	5.54	41	34

References: (Mastellone, 1991); * (Pasztor et al., 1991); ** (AA.VV., 2019; Landel and Nielsen, 1993); ***(Themelis et al., 2011).

the mass rate and composition of given flows has been obtained.

The main streams connecting the processes included in the system boundary are reported in the Table 2, identified by a short name and briefly described.

The main properties of the main polymeric materials to which the paper refers in the P-to-O process are reported in the Table 3.

About viscosity, its value depends on the average molecular weight of the polymer, the temperature, the shear rate and the hydrostatic pressure. Semiempirical relationships for these dependencies permit estimations of melt viscosity that cannot be reported in a general table.

4. Results and discussion

4.1. Mass and energy data evaluation of main streams

The mass flow rate and the composition of the plastic feedstock (PF) and of the waste constituted by the sorting residues (W_1) have been obtained by performing a mass balances on each material over the sorting section on the basis of the designed layout (Fig. 2) and of the performances' parameters (yield and purity) for each equipment. The performances' parameters set up for the equipment are reported in Table 4; they have been set on the basis of technology provider's information (TOMRA, 2019) and on the basis of on-site data taken at operating MRF (Zaccariello et al., 2015).

The partition parameters reported in the Table 4 have been used in the material flow analysis of a sorting plant to predict the mass rate and

Table 4

Performance's parameters of equipment installed in the sorting section. L: lightweight fraction; M: medium density fraction; H: heavy fraction. PS: positive flow; NF: negative flow. POL: polyolefins; Fe: ferrous metals; AL: aluminium metals.

	Dry Separator	NIR Polyolefins (Light)	NIR PET/PVC	NIR Polyolefins (Medium)	Ferrous metal separator	aluminium separator
Efficacy's parameters	L → L 80%	Pol → PF 90%	PET → PF 90%	Pol → PF 90%	Fe → PF 90%	Al → PF 90%
	L → M 15%	Pol → NF 10%	PET → NF 10%	Pol → NF 10%	Fe → NF 5%	Al → NF 5%
	L → H 5%	Other → PF 10%	Other → PF 10%	Other → PF 10%	Other → PF 5%	Other → PF 5%
	M → L 5%	Other → NF 90%	Other → NF 90%	Other → NF 90%	Other → NF 90%	Other → NF 90%
	M → M 85%					
	M → H 10%					
	H → L 0%					
	H → M 10%					
	H → H 90%					

Table 5

Composition of plastic-based flows: plastic mixed waste (PMW), plastic feedstock (PF) and waste (W₁).

Commodity item	PMW Fraction, %	PF Fraction, %	W ₁ Fraction, %
PET	8.5%	0.3%	4.0%
PP (film)	6.9%	13.7%	2.1%
PP (containers)	1.1%	1.8%	0.4%
HDPE	1.8%	3.1%	0.7%
LDPE > A3	1.7%	3.4%	0.5%
LDPE < A3	35.4%	69.7%	10.9%
Paper	4.8%	1.0%	9.3%
Aluminium	0.8%	0.0%	1.4%
Ferrous metals	0.6%	0.1%	0.9%
Tetra pack	0.4%	0.1%	0.6%
Textiles	1.7%	0.3%	3.1%
Fines	2.3%	0.0%	4.6%
All other polymers including PS, PVC, PU, ...	34.1%	6.5%	61.5%

Table 6

Ultimate composition and calorific values of waste (W₁).

	Paper	Plastics	Biowaste	Waste (W ₁)
C	32.52%	63.57%	12.36%	60.34%
H	4.46%	12.00%	1.65%	11.21%
O	30.38%	9.02%	9.68%	11.24%
N	0.19%	0.90%	0.67%	0.83%
Cl	0.32%	3.38%	0.21%	3.06%
S	0.19%	0.34%	0.10%	0.32%
Moisture	25.00%	4.20%	74.25%	6.37%
Ash	6.94%	6.59%	1.08%	6.63%
Lower Calorific Value	18,911.54	MJ/t		
Higher Calorific Value	21,184.48	MJ/t		

Table 7

Mass balance over the densimetric equipment (DRY BASIS).

Plastic Mixture Waste = 15.5 t/h (db)				
	Mass flow, t/h	Light flow t/h	Medium flow t/h	Heavy flow t/h
PET	1.32	0.07	1.12	0.13
PP (film)	1.08	0.86	0.16	0.05
PP (containers)	0.17	0.01	0.14	0.02
HDPE	0.28	0.01	0.24	0.03
LDPE > A3	0.27	0.21	0.04	0.01
LDPE < A3	5.51	4.41	0.83	0.28
Paper	0.75	0.60	0.11	0.04
Aluminium	0.12	0.01	0.10	0.01
Ferrous metals	0.09	0.00	0.07	0.01
Tetra pack	0.06	0.00	0.05	0.01
Textiles	0.27	0.01	0.23	0.03
Fines	0.35	0.00	0.04	0.32
Other polymers including PS, PVC, PU, ...	5.30	0.26	4.50	0.53
TOTAL t/h	15.54	6.46	7.63	1.46

Table 8

Output flows from optical sorter fed by light flow produced by the densimetric equipment (DRY BASIS).

Light flow in NIR 1		
	Output positive mass flow (F _{pot1})	Output negative mass flow
PET	0.01	0.06
PP (film)	0.78	0.09
PP (containers)	0.01	0.00
HDPE	0.01	0.00
LDPE > A3	0.19	0.02
LDPE < A3	3.96	0.44
Paper	0.06	0.54
Aluminium	0.00	0.01
Ferrous metals	0.00	0.00
Tetrapack	0.00	0.00
Textiles	0.00	0.01
Fines	0.00	0.00
Other polymers including PS, PVC, PU, ...	0.03	0.24
TOTAL, t/h	5.05	1.41

the composition of each flow starting from the characteristic of plastic waste entering the plant. Each coefficient represents the ratio between the amount of flow having a given property (high density, magnetism, molecular structure, etc.) that can be sorted out from the sorter and that contained in the input flow.

By assuming the maximum capacity of the sorting facility previously described at about 15 t/h (dry basis) the mass flow rate of the main output flows are:

	PMW	PF	W ₁
Mass feed rate (moisture included)	17.9 t/h	6.9 t/h	8.2 t/h

The plastic mixed waste (PMW), the plastic feedstock (PF) and the waste (W₁) from sorting have the compositions reported in the Table 5. The effectiveness of sorting process to prepare a good plastic feedstock leads to have a considerable amount of residue that is mainly constituted by plastics and organic materials (the W₁ flow).

This specific stream has been characterized to evaluate the calorific value on the basis of mean ultimate composition of each commodity category (plastic, paper, textiles, ...). The calculation of expected commodity composition of each flow is useful to estimate the ultimate composition and, from this, the feedstock energy value (Table 6).

The plant used to carry out the sorting and the pre-treatment is specifically designed to produce a plastic feedstock that complies with the quality specifications required at the inlet of the plastic-to-oil line that are: 75% polyolefins, 20% polystyrene, 5% max of other polymers such as polyurethane (PUR), poly (methyl-methacrylate) (PMMA) etc., 5% max of moisture, < 0,5% of chlorine-containing plastics, < 0,5% organics, < 1% inerts.

In the following tables, the mass flow rate and flow composition are reported for the main equipment. The setting-up of the sorting plan has to be able to produce a plastic feedstock that complies with the just

Table 9
Mass balance over the NIR 2 fed by the medium flow coming from the densimetric equipment and NIR 3 fed by the negative flow of NIR 1 (**DRY BASIS**).

	From: medium flow TO: NIR 2		From: NIR 2 (negative flow) TO: NIR 3	
	Output positive flow, t/h	Output negative flow, t/h	Output positive flow (F_{pol2}), t/h	Output negative flow, t/h
PET	1.01	0.11	0.01	0.10
PP (film)	0.02	0.15	0.13	0.01
PP (containers)	0.01	0.13	0.11	0.01
HDPE	0.02	0.21	0.19	0.02
LDPE > A3	0.00	0.04	0.03	0.00
LDPE < A3	0.08	0.74	0.67	0.07
Paper	0.01	0.10	0.01	0.09
Aluminum	0.01	0.09	0.01	0.08
Ferrous metals	0.01	0.07	0.01	0.06
Tetrapack	0.00	0.04	0.00	0.04
Textiles	0.02	0.20	0.02	0.18
Fines	0.00	0.03	0.00	0.03
Other polymers including PS. PVC, PU, ...	0.45	4.05	0.41	3.65
	1.66	5.97	1.61	4.36

given specifications.

Table 7 reports the data obtained by carrying out the calculation for the densimetric equipment. The calculations are based on the characterization of plastic mixed waste by the point of view of density distribution that has been made experimentally.

The equipment following the densimetric separator is the NIR for both light flow and medium flow (two NIR sorters in series configuration) (Fig. 2). The Tables 8 and 9 report the calculations related to this stage of the sorting process.

The medium flow sent to NIR 2 detects the PET polymer as a positive flow while the second one (NIR 3) selects the polyolefins as positive flow so reaching a very high interception and separation efficiency.

The flows F_{pol1} and F_{pol2} are then further sorted to remove metals by producing the final flow “plastic feedstock” as reported in the Table 4. The flow remaining after the sorting of polyolefins, ferrous metals, aluminium and PET, is that indicated as “sorting leftover – W_1” in Table 5.

The calculations made for magnetic and para-magnetic equipment are not reported in detail but the final results are represented by the

compositions of Table 5 and by the mass flow rate reported in the following paragraph under form of overall mass balance.

4.2. Material and feedstock energy assessment

The overall material flow assessment is represented as a multi-layered block diagram created with the software STAN developed with reference to the “good” layer (total mass balance) and the “feedstock energy” layer (Brunner and Rechberger, 2016; Cencic and Rechberger, 2008).

The total mass balance (Fig. 3) indicates that the amount of secondary materials is 15% of the total input. The polymer flow addressed to recycling (RP) is mainly constituted by PET while the other flow RM contains aluminium and ferrous metals. The flow PF_0 contains polyolefins and polystyrene as reported in the Table 4 and is addressed to the plastic-to-oil plant; the reject flow W_1 should be destined to landfilling or incineration. In the system under study the flow W_1 is sent to a densificator with the aim to homogenize the composition, increase the bulk density and makes the material suitable to be mechanical fed into the gasifier.

With reference to the Fig. 4, that reports the feedstock energy balance, it is highlighted that the thermal input capacity of this latter line is about 48 MW (W_2); by assuming that the gasification process has a Cold Gas efficiency of about 0.70, the syngas chemical energy is expected to be equal to 33.4 MW. The syngas production is 21.5 t/h corresponding to 16,926Nm³/h and to a yield of 2,287Nm³ for each ton of input to the gasifier. The calorific value of syngas, as calculated by the energy balance, is 5588 MJ/t (about 7 MJ/Nm³). The use of an engine, specifically designed to burn syngas, allows to convert the chemical energy into electricity and heat by co-generation. By considering 35% as efficiency for conversion, an electrical generated power of 11.7 MW is obtained.

The plastic feedstock corresponds to about 6 t/h; the pyrolysis products are constituted by char, having a yield in the range 5–10% of the input, and vapours with a molecular weight up to 400 g/mol. This latter flow is fractionated into two streams: one at 25 °C, containing the non-condensable gases (up to C₄), and another that can be considered as syncrude. This latter is sent to a refinery process (distillation) in order to obtain fractions having the boiling temperature larger than 350 °C (off-specification), between 260 °C and 350 °C (kerosene), etc.

The gas is mainly constituted by propylene, with presence of methane, hydrogen, ethylene. Its calorific value is higher: 43 MJ/kg. The use of an engine, specifically designed to burn heavy fuel gas (LPG), allows to convert 9.3 MW of chemical energy into electricity and heat by co-generation. By considering 35% as efficiency for conversion, an electrical generated power of 3.3 MW is obtained.

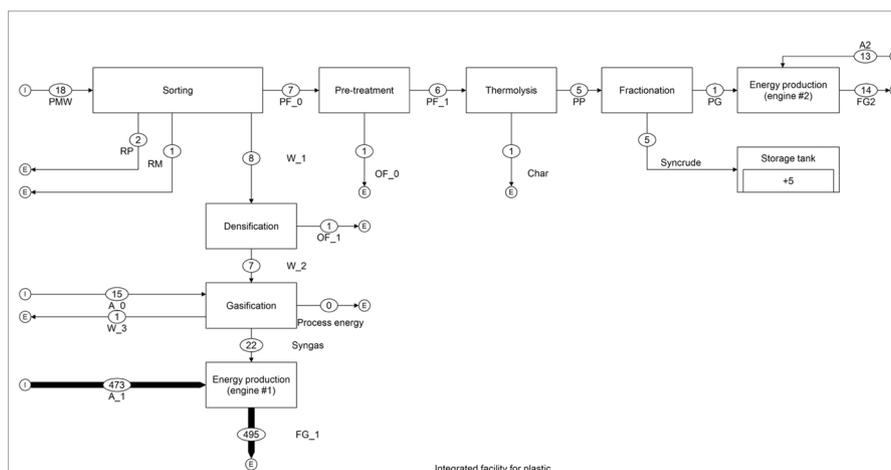


Fig. 3. Material mass balance (layer: good).

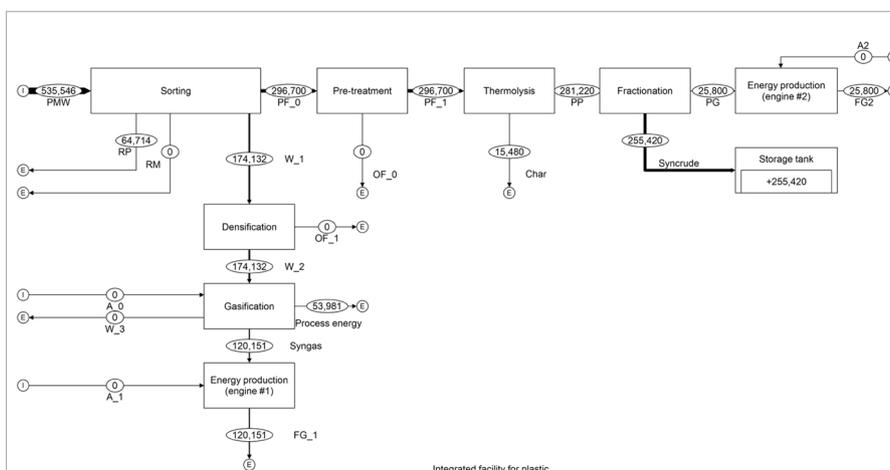


Fig. 4. Feedstock energy balance [MJ/h].

Table 10
Energy balance developed by assuming 7.920 h/year of operation.

power			energy		
Sorting plant	1,177	kW	Sorting plant	9,322	MWh/year
Pre-treatment	1,900	kW	Pre-treatment	15,048	MWh/year
Thermolysis + fractionation	682	kW	Thermolysis + fractionation	5,401	MWh/year
Densification	1,510	kW	Densification	11,959	MWh/year
Gasifier	650	kW	Gasifier	5,148	MWh/year
TOTAL installed power	5,919	kW	TOTAL energy consumption	45,477	MWh/year
TOTAL produced power	14,190	kW	TOTAL energy production	112,383	MWh/year
NET produced power	8,340	kW	NET produced power	66,906	MWh/year

The feedstock energy balance (Fig. 4) shows how the integrated system promotes the maximum valorisation of the feedstock energy contained in the plastic waste entering the system: 27% of the plastic waste chemical energy is stored into fuel gases that can be used to produce heat and/or electricity with high efficiency and low environmental impact; 49.6% is converted into syncrude that can be used to produce new polymers, fuels, chemicals, etc. without any limitation for the new materials; 10% is used to sustain the endothermic reactions of gasification while more than 10% is stored into secondary materials addressed to mechanical recycling.

4.3. Energy balance

The energy balance has been developed by considering the installed power for the main equipment of: sorting; pre-treatment; plastic-to-oil; densification; gasification. In the following tables, the installed power for the given plant, are reported.

The sorting section includes:

N°	Name	Installed power, kW
1	Double-shaft shredder	264
1	Drum air separator (densimetric sorter)	44
5	Near Infrared Analysers	35
3	Magnetic separator	9
3	ECS aluminium separator	20
1	Shredder	264
1	Air compressor line	130
-	Belts	213

The pre-treatment section includes:

N°	Name	Installed power, kW
1	Dryer	300
4	Degassing extruder	400

The plastic to oil section includes:

N°	Name	Installed power, kW
1	Carbon discharge system	100
1	Distillation system	32
1	Gas washer	5
1	Air treatment	150
2	Storage system	50
1	Gas buffer	50
1	Water cleaning system	10
1	Cooling water system	85
1	Chilled water system	20
1	Tracing	41
	Other	89

The densification section includes:

N°	Name	Installed power, kW
1	Buffer	5
5	Agglomerators	300
1	Gas washer	5

The gasification section includes:

N°	Name	Installed power, kW
1	Feeder	75
1	Ash discharge	100
2	Thermal cracker (plasma)	100
3	Pumps	25
1	Air compressor	85
1	Analysers	5
	Cooling water system	110

The engines can reach 35% of conversion efficiency in electricity by self-producing all the electric power and by feeding the excess into the grid.

The energy balance (Table 10) helps to assess the economic feasibility of the integrated system with regard to the operating cost and, specifically, to the electricity cost that constitute the most relevant contribution to the operating cost (opex). The operating cost can be considered as obtained by adding the following items:

- human resources
- electricity
- waste disposal
- raw materials\additives

All the financial items are not considered.

The economical assessment has been made by comparing the opex calculated for the integrated facility and for the three separated subprocesses: mechanical sorting, plastic-to-oil, gasification. Each facility opex is obtained by assuming that the operating time is 7920 h/year (i.e. three shifts for day, 330 days/year), the annual gross mean salary is 48.000 €/year, the electricity cost is 120€/MWh. The value of opex has been calculated for the facilities considered as standalone installations (columns 2, 3 and 4 of Table 11) and for integrated facility. The human resources for this latter have been considered as the sum of those necessary for the three process's sections but this assumption can be considered conservative because the shift supervisors, extraordinary maintenance workers, administrative people can be shared.

The data of Table 11 demonstrate that the integration between the mechanical sorting of low-quality waste with the gasification and plastic-to-oil units allow to decrease the cost for waste disposal and for electrical energy. All the waste that is not suitable to be converted into syncrude (from which kerosene and other petrochemical feedstocks are produced) can be converted into syngas (from which energy and/or other petrochemical feedstocks can be produced).

5. Summary and conclusions

The assessment of an integrated system allowing the exploitation

Table 11
Operating cost evaluation for single installations and for the integrated facility.

ITEM		SORTING	PRETREATMENT-P2O-ENERGY RECOVERY	DENSIFICATION-GASIFICATION-ENERGY RECOVERY	INTEGRATED FACILITY
Capacity	t/h	17.87	6.90	8.22	17.87
Human resources (technical staff)	#	15	9	9	33
	€/t	5.09	7.91	6.64	11.19
Electricity	kWh/t	65.9	374.2	262.9	-462.8
	€/t	7.90	44.91	31.55	-55.54
Waste disposal	t/h	15.12	0.55	0.58	1.13
	€/t	122.6	32.0	28.0	25.2
Raw materials\additives	€/t		5	5	4.2
Total	€/t	135.64	89.81	71.19	-14.89

of plastic waste by using thermolysis processes, applied to the flows that are not mechanically recyclable to new valuable goods, has been carried out. The data sources are mainly obtained by literature, industrial applications and Companies active in the related fields with demo plants installed; the model to simulate the mass flows is the Material Flow Analysis. A feedstock energy flow assessment is also applied in order to evaluate which part of material energy content is exploited and transferred to fuels and secondary materials. Data about the installed power necessary to operate a modelled integrated plant is also provided to establish which is the external energy request and, then, the opex.

The most important result of the feasibility study is that the plastic waste used as reference input to the system is the residue coming from the existing material recycling factories that sorts the plastics collected at households and commercial sources; this residue is addressed to disposal such as landfilling or incineration with municipal solid waste with a corresponding high costs and impacts. The conversion into secondary fuels is also applied to substitute the fossil fuels into cement kiln or (rarely) in the steel production factories; in this case the sorted plastics substitutes the coal as fuel for production of heat after a sorting and densification stage (so called CSS production). Today the cost of these stages is about 80€/t that have been added to the transportation costs between the MRF and the CSS facility and from CSS to cement facility licensed to use the CSS fuel. The large cost of treatment of plastics and the limited number of authorized facilities, led to a massive landfilling

The material and energy balances allowed to verify that a combination between energy-intensive processes, like mechanical sorting, can be energetically and economically sustained by integrating them in a network where the non-recyclable materials can be exploited in form of materials and energy. The complete exploitation of mixed plastic waste, containing polymers that cannot be mechanically recycled with a sustainable industrial cost or having no real market to be sold, cannot be obtained by recurring to a unique process that inevitably results in a not sustainable gate fee. The building of an integrated and sustainable network allows to apply for real the circular economy principles and the results obtained in this paper give a demonstration of this statement.

In the examined case, two thermochemical processes have been used for the flows (polyolefins-based flow and commingled plastic & waste flow) resulting from the sorting of a typical plastic waste coming from household and commercial separate collection: a) a plastic-to-oil process based on pyrolysis of polyolefins and fractionation of obtained vapor products; b) a gasification process of the flow containing all the other components of the sorting line, including composites, elastomers, foreign matter, etc., with the aim to produce a high-calorific syngas.

The use of the pyrolysis gas from the plastic-to-oil and the syngas from gasification to produce electricity, allows to cover integrally the

energy cost and, then, minimizing the opex. The surplus of electric energy can be delivered to the grid or used in the industrial district. The syngas can be used for other processes, not covered in this paper, such as: a) production of combined heat and power, by using the produced heat for heating/cooling; b) addition of a methanation process with a corresponding production and delivery of methane in the public grid (Rönsch et al., 2016). These options are reliable and can be assessed at front-end engineering level. of a potentially valuable material with a huge environmental, social and economic cost.

Declaration of Competing Interest

The authors declare that there are no conflicts of interest.

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