Technical description and performance evaluation of different packaging plastic waste management's systems in a circular economy perspective
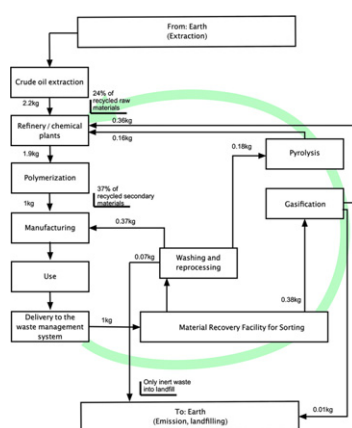
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HIGHLIGHTS

- Mechanical sorting and reprocessing processes alone are not sustainable and viable.
- Integration of thermochemical processes allows to fulfil the material recovery targets.
- Material recovery includes materials and fuels.
- Economic assessment demonstrates the present plastic waste management is unfeasible.

GRAPHICAL ABSTRACT

ABSTRACT

The plastic waste disposal strongly raised in importance in the recent past and it is approaching a critical situation worldwide, so requiring putting in practice the criteria of circular economy by avoiding meaningless policy responses against the plastic materials. The world of plastic materials includes a wide range of goods in all the sectors of our life: packaging, construction, biomedicals, etc. The answer to the plastic waste disposal is build an industrial network characterized by reliability, flexibility, sustainability, utility in the industrial cycle and ability to provide useful products to the market. The traditional processes including recycling and energy recovery fulfil only a part of these conditions and need to be assessed in correlation to their real effect on the circular economy such as the uses of the obtained products in the anthroposphere's life cycle, the burdens generated by the processes itself, etc. Among the possible processes that can be more sustainable if compared with the traditional ones, in a real circular economy perspective, those based on thermochemical exploitation of products obtained by plastics are discussed and assessed. The results show that it is possible to transform an expensive and non-resolving plastic waste management system in an industrial network having an intrinsic economic sustainability and, overall, a well-defined role in the economic chain of plastics. The results show that an integration between the present system with the production of petrochemical products, including oil/gasoline/syngas/..., strongly improve the economic performance of the overall waste system, allowing a larger savings of not-renewable resources, a limited greenhouse burden, a release in the market of valuable products instead of poor quality materials and the minimization of waste destined to be landfilled.

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1. Introduction and scope

Last data show that the plastic demand in Europe reached 51.2 million in 2018; 29 millions of tons of the post-consumer plastics have been collected as waste and addressed to the plastic packaging management system. The extensive production of plastics created a question related to plastic waste disposal that address to landfilling and to energy recovery a large amount of collected plastic waste (Plastics Europe, 2019). The European regulations about the so-called End of Waste (EoW) criteria should increase the fraction of recycled plastic waste by decreasing first of all the landfilling and second the energy recovery. The application of the EoW defined by the European Union at national level is targeted by early July 2020 (Langendorff, 2018); the new approach will be focused on the ambitious targets such as harmonized reporting of input data to recycling; increased targets for plastics (from 22.5% of 2008 to 50% by 2025 and 55% by 2030); new targets for aluminum. The lack of these criteria generated an unsustainable plastic waste management system characterized by a large export flow towards Asian market and almost negligible investments in infrastructure improvements. The statistics related to plastic waste have been also negatively affected by the impossibility to follow the actual fate of exported flows by generating inaccurate or misleading data. The question of how to obtain reliable data to evaluate the real impact of regulations, policy, infrastructures, etc. on the recycling/recovery rates is, at present time, an issue. By looking at the data provided by official sources such as European Commission, some information about the production rates and composition can be obtained (Fig. 1).

By observing the data related to the production and recovery rates for packaging waste in Fig. 1 it is easy to recognize such trends: a) the continuous increasing with time of both indexes; b) the slight variation of the ratio between the recovery and production rates.

Moreover, the overall recovery rate was 80% in 2017 with a limited increase (8%) respect to that of 2009. It is also noteworthy that, on the basis of data provided by Eurostat, (2019a), the ratio between the recycling rate and the total recovery rate also remained constant along the period. Actually, plastic recycling continues to be an economically marginal activity and recycling rates are thought to be 14–18% at the global level. The remainder of plastic waste is either incinerated (24%) or disposed in landfill or in the natural environment (58–62%) so that plastics recycling rates are substantially lower than those for other packaging materials such as glass, metals or paper (OECD, 2018).

The packaging waste data reported by the above mentioned the above cited sources are related to the waste streams produced by sorting plants by assuming that all the waste sorted is actually sent to recycling and recovery to be reintroduced in the market as secondary materials or energy to the grid: this assumption is the reasonable explanation of the almost constant value reported in the Fig. 1: once the waste is sorted out it is considered recovered and its end use is no more traced. Actually, this assumption is not realistic since the export flow of packaging waste to Asian market, East Europe and Third world Countries cannot be traced. In particular, until China was willing to buy the exported waste, the huge amount of packaging waste was recycled/recovered in China. Due to the possibility to export a large amount of waste without no specification's limits, the western nations had no incentive to invest in their own treatment facilities and the industrial network remained unchanged, not improved, totally inefficient. When China banned imports of 24 varieties of solid waste on January 1, 2018, including a host of plastics, global waste flows were overturned. Another 16 categories were prohibited from the end of the year 2018 and another 16 will be from the end of 2019 (Nichols and Smith, 2019).

The above reported situation of the global plastic waste management system has made the structural crisis of the system clear and has consequently raised the costs of disposal of plastic packaging waste to insurmountable values (CEA, 2019). Market data (WRAP, 2019) clearly shows that it is crucial for Europe as well as USA invest in a strong modification of the industrial network by increasing flexibility and reducing the cost of chain; the 5-year range trend of export to Asian markets reduced by more than two-third, that is more than enough to provoke a worldwide crisis. To this aim, the industrial system must be integrated with other processes able to make wider the resins types (plastics or elastomers) that can be recycled and converted into useful products; moreover, the products and intermediates obtained by these processes must be substitutive of raw materials in a life cycle prospective by leading to a real advantage to the circular economy. On these basis, the sorting and mechanical reprocessing of polymers suitable to be reintroduced in the market as secondary materials must be associated with other processes able to promote the recovery of a wider range of polymers such as chemolysis, pyrolysis, fluid catalytic cracking and gasification (Ragaert et al., 2017). The material recovery must be preferred
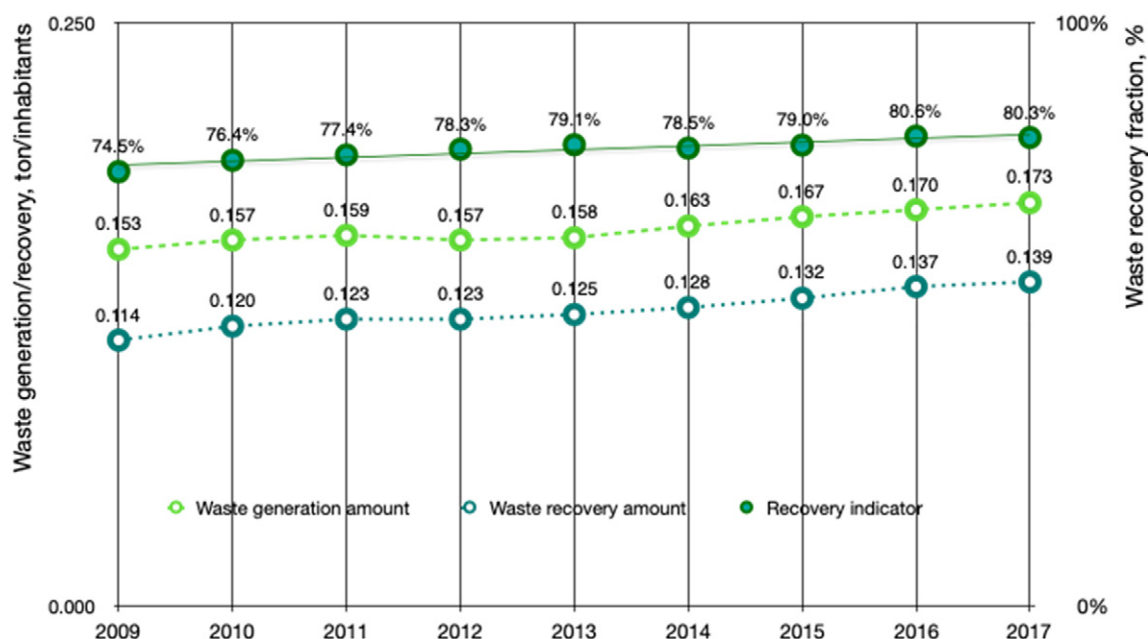

Fig 1. Production and recovery rates for packaging waste. Redrawn from Eurostat (2019a).

over the energy recovery even though the exploitation of waste by recurring to gasification or combustion allows to increase the sustainability of the waste management systems (Fernández-González et al., 2017). Anyway, the energy recovery needs to be limited to waste that cannot be recycled or recovered in any other ways; to this end other chemical and thermal processes needs to be assessed in order to verify if their application increases the economic and environmental sustainability of the overall management system (Al-Salem et al., 2017).

The scope of this paper is modelling and comparing different scenarios designed to overcome the packaging waste management crisis in a structural way. The leading principle is improving the circularity of material and energy flows as much as is technically possible by considering the sustainability as the key parameter for comparison of alternatives. Starting from the present packaging waste system, alternative scenarios have been built taking into account the actual composition of waste and by focusing to unit processes able to exploit each stream at best. For each scenario the mass and energy balance have been fully developed and an economic assessment based on the secondary materials monetary values and on the estimation of operating expenditures, is associated to it. Indexing of the main parameters able to measure the

sustainability of the scenarios is used for an easy and effective comparison and for a decision makers' guidance.

2. Scenario proposal and description

2.1. General packaging waste management system and related scenarios

The processes to be included in the general packaging waste management system are summarized in the Fig. 2 representing which processes can be included and which products and waste can be correspond obtained. The reported streams of products have a different market value other than a different associated production cost; the economic value of the products obtained by packaging waste included in the general framework of Fig. 2 is reported in Table 1 (Economics Trading, 2019; EUROSTAT, 2019b; WRAP, 2016). The overall production cost depends on operating expenditures (opex) necessary to operate the process such as national prices of electricity, fossil fuels and man work costs but also items depending on specific sector's policy such as economic subsidiaries. An evaluation of an overall opex for selected processes is given in the Supplementary Materials section.
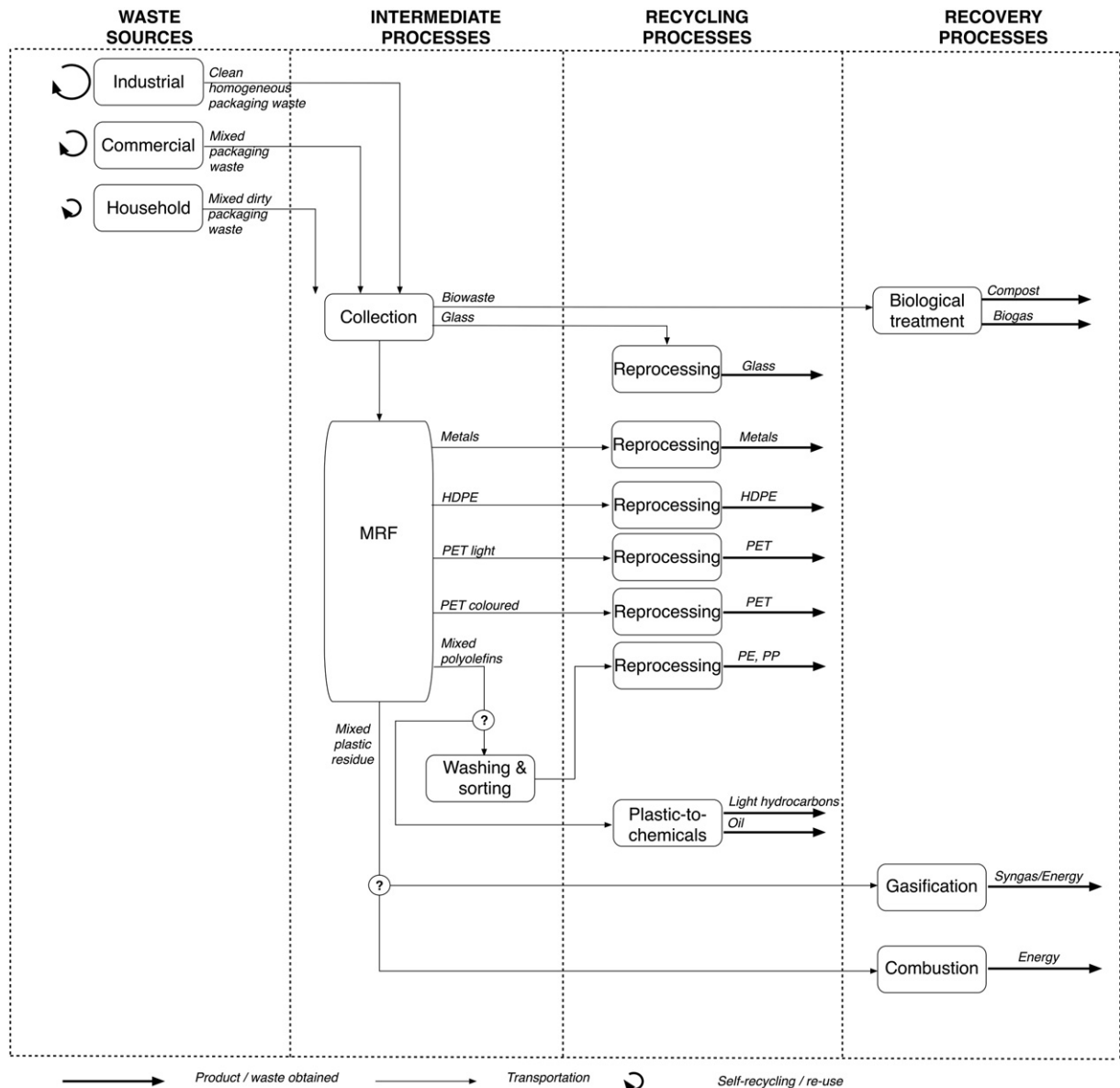

Fig. 2. Waste flow patterns between the main unit processes involved in the management system.

Table 1
Market prices for recycling and virgin materials.

Secondary material obtained from recycling	Market value of recycled material, €/t*	Market value of virgin material, €/t
PET light	220.6	1100
PET colored	36.0	–
HDPE	400.6	1250
LDPE film high-grade	210.2	1200
Steel	160.2	437
Aluminum	975.4	1600
Paper	140	Wide range
Glass	50	100–150 (cullet)
Mixed polyolefins	91.74	n.a.
Crude oil	–	446
Hydrocarbons diesel	–	613
Hydrocarbons gasoline	–	509

The collection of dry recyclable packaging waste as a separate stream from biowaste and residual waste is considered a mandatory and preliminary process; it is a common "unit process" that can be realized in different way and with different collection efficacy and collected stream composition so affecting the efficiency parameters of downstream processes. The stream collected by means of separate conferral can have a certain amount of foreign matter that affects the waste amount and composition fromobtaining the downstream processes; it is then important to establish which are the separated streams composition in order to evaluate the performances parameters of the following processes.

The core of the industrial system is the sorting plant, known as Material Recovery Facility (MRF), that receives the separated stream containing plastics packaging from the collection system, and that produces streams of secondary materials other than the mixed "plastic" residue (MPR). This latter stream contains the foreign matter incorrectly conferred to the collection system by waste producers in addition to the materials without a market value, not separate by choice, and the material which is not separable due to process inefficiencies. The layout of MRFs can be different case by case because of dependency of specific technical design and of packaging waste composition. A brief description of the MRF layout included in the scenarios is reported in Section 2.2.1 while a detailed assessment can be found elsewhere (Mastellone et al., 2017).

The waste resulting from sorting is a heterogeneous mixture mainly composed by plastics and it is then featured by a high-energy content; it is labelled in Europe with the code 19.12.10 and it is addressed to incineration or to landfill, depending on availability of plants. This waste represents the weakest point of the system reported in the Fig. 2 because it represents the overall "inefficiency" of the whole management system, including collection. A data confirming this weakness is that this waste code is the first one in the top 20 European codes for non-hazardous notified waste shipments exported from EU Member States by quantity (Eurostat, 2019a).

The erroneous assumptions made during the planning of waste management systems, justified by the need to achieve levels of collection, recycling and recovery defined at the political level, have led to under sizing of recovery and disposal facilities favoring recycling even when the latter was in fact neither possible nor sustainable. Until the Asian market accepted the European and U.S. waste as a commodity, the system has had a route to dispose it at an acceptable gate fee. Today, find a destination different from landfilling or incineration is quite difficult and the tipping fee reached unsustainable values such as 120€/t or higher; moreover, the need of long-distance transport requires fees of about 5c€/t/km, by road or train, prevailingly. The transportation unitary cost is a mere indicative value since the bulk density as well as the transportation type (road/train) strongly affect its value.

Based on these considerations, the landfilling of MPW, which remains the cheaper disposal tool even though not allowed by several national regulations, is considered as the destination process for this waste stream in the base case scenario A. This scenario represents the most applied one, even though the disposal of high-energy content waste should be avoided.

The alternative scenario B includes a recovery process instead of landfilling. This option allows to exploit the calorific value of plastics so producing electricity and heat. Anyway, it cannot be applied everywhere due to economic issues; transportation costs can be unsustainable for Countries that export the waste outside the national boundary towards those (few) Countries having an energy recovery demand not completely fulfilled. The scenarios A and B are not then truly alternatives but applied in the actual case by different Countries on their existing and in operation industrial infrastructures.

The alternative scenarios are indicated with the letters C and D; these scenarios are related to systems included thermochemical processes that integrate or substitute the traditional energy recovery by means of incineration. The scenario C differs by the scenario B because of inclusion of a feedstock recycling process applied to the polyolefin streams obtained by the MRF so increasing the recovery of materials instead on energy. The feedstock recycling processes are realized in small-medium scale plants and can be installed downstream the MRFs so encountering the principle of proximity. The feedstock recycling process selected to be included in the scenarios C and D is the pyrolysis for conversion of plastics into syncrude (i.e. a mixture of hydrocarbons) and selected chemicals (after proper refining processes); as indicated in the Fig. 2 this process is considered as a material recovery process.[1] The scenario D is a long-term scenario that consider the incineration no longer applied and the installation of smaller and fully integrated gasifiers as the final destination of not recyclable plastics flows. Gasification can be included in the feedstock recycling processes since, under certain operating conditions and with given waste composition, is able to produce synthesis gas. In the worst case it is anyway able to convert the organic fraction of waste into a producer gas, so replacing fossil fuel gas in power & heating production.

In summary, the scenarios are:

A. Present case: sorting of separately collected packaging waste, mechanical reprocessing of homogeneous streams, landfilling of residual plastic and residue.

B. Present advanced case: sorting of separately collected packaging waste, mechanical reprocessing of homogeneous streams, energy recovery of residual plastic and residue, landfilling of ashes and inerts.

C. Alternative case: sorting of separately collected packaging waste, mechanical reprocessing of homogeneous streams, feedstock recycling of selected plastic streams, energy recovery of residual plastic and residue, landfilling of ashes and inerts.

D. Alternative case: sorting of separately collected packaging waste, mechanical reprocessing of homogeneous streams, feedstock recycling of selected plastic streams, syngas recovery by gasification from residual plastic and residue, landfilling of ashes and inerts.

The main non-technical differences between the alternative scenarios C/D and the present scenarios A/B are the site location approach, the minimum required capacity, the footprint, the capital expenditures and the expected site-to-site distance. While the incinerators, cement kilns as well as steel production furnaces (scenario B) needs to be large-capacity plans to be economically viable, thermochemical processes included in the scenario C and D can be realized in small/medium-scale. Moreover, the footprint is quite smaller due to the reduced capacity but principally due to the smaller volumes of reactors and equipment necessary to operate pyrolysis and gasification processes. These features

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[1] *The EU directive 94/62/EC about packaging waste define 'recycling' as: the reprocessing in a production process of the waste materials for the original purpose or for other purposes including organic recycling but excluding energy recovery.*

Table 2
Composition of the input packaging waste from collection to MRF.

Commodity composition	Mass fraction, %	Mass rate in, t/h
PET	23.97%	3.36
PP (film)	0.44%	0.06
PP (containers)	0.58%	0.08
HDPE	7.21%	1.01
LDPE > A3	3.09%	0.43
LDPE < A3	14.75%	2.07
Paper	8.32%	1.16
Aluminum	1.02%	0.14
Ferrous metals	4.09%	0.57
Tetrapack	0.26%	0.04
Textiles	1.39%	0.19
Fines	6.96%	0.97
Other polymers including PS, PVC, PU, …	27.92%	3.91

Table 3
Composition of the output streams from MRF (Mastellone et al., 2017) and associate economic values (WRAP, 2016).

Stream type	Stream partition, % out/in	Mass rate in, t/h	Associated economic value, €/h
Light PET	6.8%	0.957	211.1
Light blue PET	8.7%	1.214	267.9
Colored PET	6.5%	0.914	32.9
Opaque PET	0.0%	0.000	0.0
HDPE	7.0%	0.980	392.0
LDPE > A3	1.7%	0.243	51.0
LDPE < A3	4.1%	0.571	0.0
PP	0.4%	0.056	0.0
Fe	4.3%	0.607	97.2
Al	1.2%	0.171	166.9
MPR	59.2%	8.286	–

allow to integrate these processes into MRF boundaries and/or not far from the waste sources by leading to the following advantages:

1. rational and balanced distribution of treatment facilities close to the production site (proximity)
2. strongly decreasing of transportation costs;
3. strongly decreasing of transportation impacts (distributed pollution, fossil fuel combustion, …);
4. increasing of efficacy in controlling illegal dumping, irresponsible long-term storage and criminal disposal of waste

In the following paragraph, the detailed description of the unit processes composing the alternative scenarios C and D is reported.

2.2. Description of unit processes

2.2.1. Collection & sorting

The plant receiving the collected waste at household and commercial sources is a material recovery facility (MRF) that sort out the material on the basis of such properties as density, size, shape, chemical composition. The detailed description of this facility is available elsewhere (Mastellone et al., 2017) and not reported here. The composition of the input stream has been obtained by analyzing several samples of the waste treated in a full-scale MRF having a total capacity of 14 t/h as input material (Mastellone et al., 2017); the resulting composition is reported in Table 2. The composition of output streams from the MRF, together with the potential economic value associated to these flows is reported in the Table 3.

The sorting residue (mixed plastic residue - MPR) is mainly composed by non-recyclable polymers and foreign matter but it also contains un-sorted valuable polymers such as PET other than polyolefins because of the non-unitary sorting efficacy of the MRF. Its composition can be obtained by carrying out a mass balance (Table 1.SM[2]) other than those obtained by the experimental analyses (Table 4). The composition of MPR is characterized by large uncertainties due to the difficulty to recognize the commingled polymer fraction composition (that represents 59% of the total sample); this is composed by multilayered polymers, polystyrene, PVC, etc. The polymer fraction that can be sorted and potentially recovered is in the range 5–30%.

2.2.2. Thermochemical processes: combustion, gasification and pyrolysis

The common point between the thermochemical processes applied to waste recovery/recycling is that they convert the original solid macromolecules into short and stable gaseous or liquid molecules: pyrolysis cracks the chain into a wide range of molecular weights (C1–C20); gasification partially oxidizes the chain by producing carbon monoxide, carbon dioxide and hydrogen, other than a small fraction of light hydrocarbons (C1–C7); the combustion totally oxidizes carbon and hydrogen

into carbon dioxide and water. The main difference, other than the operating conditions applied, is related to the different selectivity: combustion can easily convert almost all organic matter under suitable operating conditions; on the contrary, pyrolysis requires that the waste input composition strictly respects specifications, if useful material products are expected. Gasification is less selective compared to pyrolysis but the formation of poly-aromatics and unsaturated compounds led to tar production, in particular when fed by plastics, so limiting the application of this process (Arena et al., 2009, 2008; Gershman, 2013; Lopez et al., 2018; Mastellone, 2015; Wu and Williams, 2010). The coupling of gasification and thermal/catalytic cracking of tar is necessary with this material to realize a feasible process.

In the next paragraphs these processes are described by giving the details necessary for the assessment of the related scenario.

2.3. Combustion

The energy recovery is traditionally carried out by recurring to combustion. The combustion of packaging that cannot be recycled produce a certain amount of heat, converted into electricity by using HOB (Heat Only Boiler) or CHP (Combined Heat and Power) systems (Ola Eriksson, 2017). The low heating value of the MPR is 26.7 MJ/kg (see Table 2.SM), well above the energy content of municipal solid waste. By assuming a power-to-fuel ratio of 0.184 (Ola Eriksson, 2017), the contribution of this stream to the power generation can be estimated. The bottom ashes have been considered to be 15% of the input mass flow in case the MPR is not sent to a further sorting (scenario B) while it is considered 4% if the additional sorting is realized (scenario C).

2.4. Gasification

The gasification system technology is based on that applied to the 100 kg/h pilot plant MNIS (www.mnis.it) owned by Bell Production SpA and operated in the framework of a research program under the supervision of University Luigi Vanvitelli. The core of the MNIS technology, whose block diagram is reported in Fig. 3, is a dual-stage system composed by two plasma-assisted reactors, one where the solid waste is converted into a raw syngas and a second one where the syngas undergoes a thermal cracking in a ionized environment. The main gasifier unit (block C in the block diagram) is basically an updraft gasifier where the waste is fed together with a stream of air injected by two series of

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[2] In the text the SM suffix indicates the Supplementary Materials section.

Table 4
Commodity composition of mixed plastic residue.

Polyolefins	47%
Metals	2%
PET + PVC + other polymers	42%
Foreign materials (biowaste, glass, …)	9%
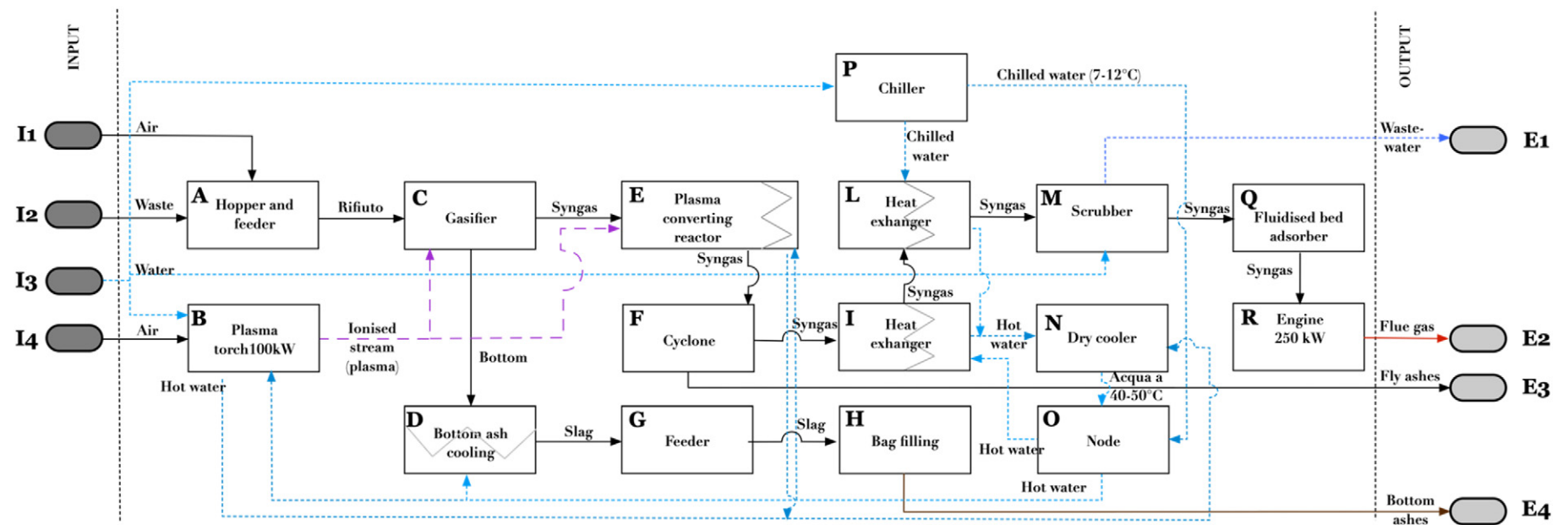
Fig. 3. Block diagram of the MNIS plasma gasifier. I: input; E: emissions & outputs. Dot lines: water.

Fig. 4. Block diagram of plastic-to-chemical plant MLM-RTM. I: Input; P: Product; E: Emission/waste; Dot-solid line: Heat flow.

nozzles at different heights, with a flow rate such as to obtain a given ratio "air-to-waste", so realizing the partial oxidation of the waste; the syngas converting unit (block E in the block diagram) works in homogenous phase as tar cracker.

Reaction temperature of the solid phase gasification is controlled by using the plasma torch located at bottom just below the waste feeder entrance. The plasma generation system can be fed by an air/steam mixture so allowing the provision of heat and ionized molecules of oxygen and hydrogen at same time. The provided heat allows to operate the gasification process at a temperature level independent by thermodynamics constraints and by input waste composition such as moisture and/or inorganics content. This way to operate the temperature process control does not modify the syngas composition and does not use additional fuels. The electricity absorbed by plasma torch is a variable value depending on the waste calorific value and composition; it can be obtained by the energy balance reported in the following paragraph and it is in the range 3–10% of the input feedstock energy flow. The possibility to vary the air/steam ratio of the ionizing gas fed to the plasma torch increase the flexibility of the process in relation to both waste and syngas composition. In fact, the H/C molar ratio of the waste fed into the gasifier affects the syngas composition and in particular the H_2/CO ratio as well as tar and carbonaceous residue yields. Several studies related to the co-gasification of plastics-based waste mixed with cellulose-based waste, demonstrated that the release of H and OH from the polymer cracking acts as hydrogen donor species so promoting the cracking of the intermediates and radicals by increasing yield and quality of the syngas (Burra and Gupta, 2018; Mastellone et al., 2010; Pinto et al., 2003, 1999). The technology chosen as reference for gasification process is also flexible enough to adjust the H/C molar ratio in the ionizing medium (acting also as reactants gas) and to maintain the temperature level in an optimal range in order to guarantee the intermediates cracking so limiting tar formation whatever is the waste composition (e.g. moisture and ash content).

The data related the syngas composition have been obtained by a simplified equilibrium model and are reported in the Fig. 1.SM.

The presence of tar in the syngas is due to the highly reactive hydrocarbons produced by cracking of polymeric chains; the cracking of tars in vapor phase is obtained by recurring to thermal cracking in the secondary cracker (item E in Fig. 3). The raw syngas produced in the gasifier is in fact sent to a cracking chamber where a plasma plume is generated and a temperature larger than 1250 °C is guaranteed.

The layout is completed by heat exchangers, a scrubbing unit and a fluidized bed adsorber (Fig. 3).

2.5. Pyrolysis

The plastic-to-chemical system is based on the MLM-R™ technology developed and owned by PRUVIA Fuels GmbH; the typical block diagram is composed by three main sections: 1) the feeding and pyrolizer reactor section; 2) fractionation of pyrolysis products into heavy, medium and light hydrocarbons section; 3) energy recovery and production section.

The process that converts the polyolefins into hydrocarbons with wide range of molecular weights is the thermal cracking i.e. pure pyrolysis. The cracking of polymeric materials is disadvantaged by the poor thermal conduction capacity of polymeric materials; in order to make the process faster and more efficient, the heating / melting stage is separated from the cracking stage. During the melting phase the dehalogenation is also realized in order to remove chlorine and avoid corrosion in the plants. Several studies report the different aspects of thermal cracking/pyrolysis, catalytic cracking and hydro-pyrolysis as well as describe the differences between the main proposed technologies such as fluidized bed reactors, screw reactors, conical spouted bed reactors (Butler et al., 2011; Dwivedi et al., 2019; Lopez et al., 2017; Ragaert et al., 2017). The selected process is described in the following by means of the block diagram reported in Fig. 4.

The section composed by blocks A, B, D and M realizes the melting and cracking of polymer mixture in a pyrolizer connected to a heated feeder (A) where the polymers mixture is dried and de-halogenated. The section composed by C, E, F and G promotes the fractionation into four streams: the heaviest is recycled back to the reactor to be cracked again while the lightest, composed by non-condensable gases, is sent to an engine for electricity production. The two streams P3 and P4 are high-market value products that are stored in tanks after cooling down. The energy recovery is completed by the process N, that realize the combustion of char and tar removed from the reactor in order to use the energy content of this fraction. In case this process is not used, the carbonaceous residue has to be disposed off.

The composition of products obtained by pyrolysis of mixed polyolefins depends by the operating conditions, in particular by temperature, and by the polymers' composition. At 500 °C the results based on the kinetic modelling (Al-Salem and Lettieri, 2010) allow to develop the mass balance for the plastic-to-oil plant. The composition of products as expected on the basis of the kinetic model is reported in Fig. 2.SM. The plant is completed by the partial condensation of the raw pyrolysis product stream with the aim to obtain: fuel gas (non-condensable gases), diesel range products, gasoline range products, heavy condensate.

3. Results

3.1. Material flow assessment of scenarios

Each scenario has been sketched by utilizing a block diagram representation and assessed by building a mathematical model based on mass and energy balances supported by the software STAN (Cencic and Rechberger, 2008).

The flows assessment is reported with reference to the total mass (layer "goods") and the feedstock energy (layer "energy") balances. The layer referred to the total mass is useful to evaluate the yields of both unit processes and whole system as well as it is the a valid support for the economic balance calculation. The layer based on the feedstock energy allows to understand how the internal energy of the matter is distributed along the system for each material stream. In this kind of layer, the external sources of energy (electric, mechanic, heat, …) are not reported and the reaction energy developed or absorbed by chemical reactions is indicated as a positive/negative accumulation of the unit process (block).

3.1.1. Base case scenario description

The first unit process of the base case scenarios is the MRF; this is fed by 14 t/h of separately collected packaging waste having the composition reported in Table 2. The output streams are grouped into two main categories: secondary materials addressed to mechanical reprocessing to be used in substitution of virgin products, that are mono-material streams of PET, HDPE, high quality LDPE, aluminum, iron/steel, mixed polyolefins, and the mixed plastic residue (MPR) containing polyolefins such as the unsorted LDPE, PP, PS other than non-thermoplastic polymers, layered and coated plastics, wires, textiles, etc. The MPR stream has a mass flow and composition that depends on the performance of sorting equipment and on the initial composition of separated packaging waste. In the case of the MRF taken as reference, the composition of the MPR is reported in Table 4 (and, in more detail, in the Table1.SM). This highly heterogeneous flow is sent to disposal into landfills (scenario A) or energy recovery by combustion (scenario B) in the base cases. The scenario A is largely applied, although by way of derogation from the regulations on waste with a high calorific value, whenever energy recovery equipment is not available.

The mass balance for all the cases where energy recovery is unavailable is reported in Fig. 5. The scenario shows how the sorting carried out by the MRF (P1) produces flows of secondary materials (SM) including PET, HDPE, aluminum and iron/steel addressed to mechanical
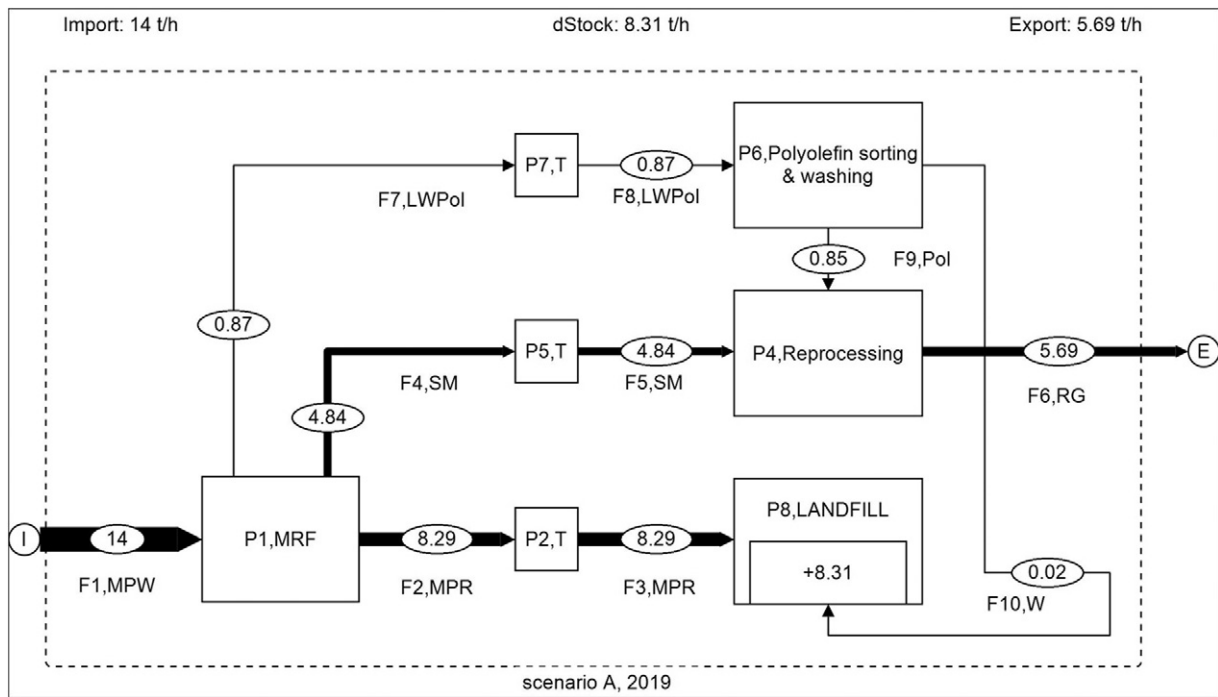
Fig. 5. Scenario A: base case with MPR landfilling – total mass layer.

reprocessing in substitution of virgin raw materials. A flow of mixed polyolefins, containing mainly LDPE, PP and PS is sent to further processing (P6) that is known as "washing" because of the use of water and water-added equipment to realize the additional sorting process;

the dry sorting processes such as aeraulic, ballistic, density separators and near-infrared sorters resulted in fact unable to remove the undesired foreign matter from these streams and sort it adequately, so excluding to assign a positive value in the economic chain for these
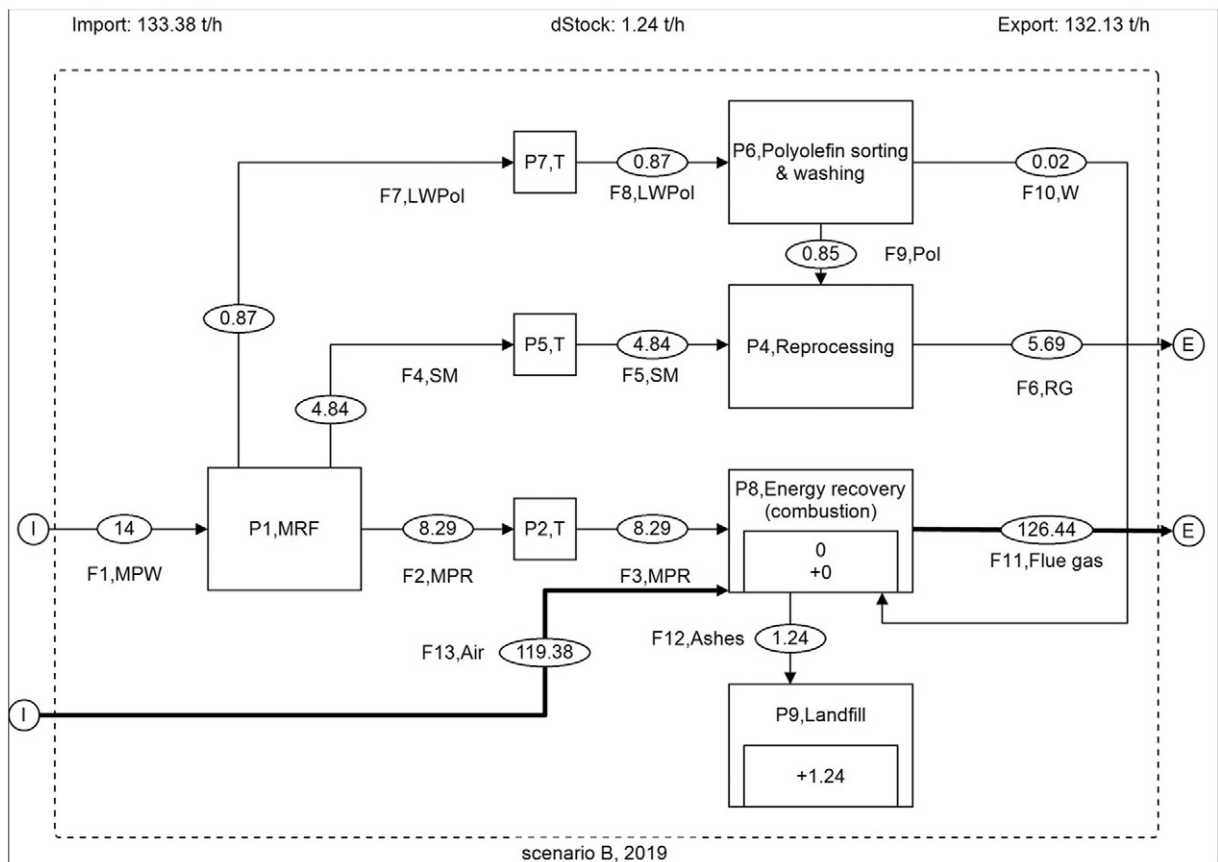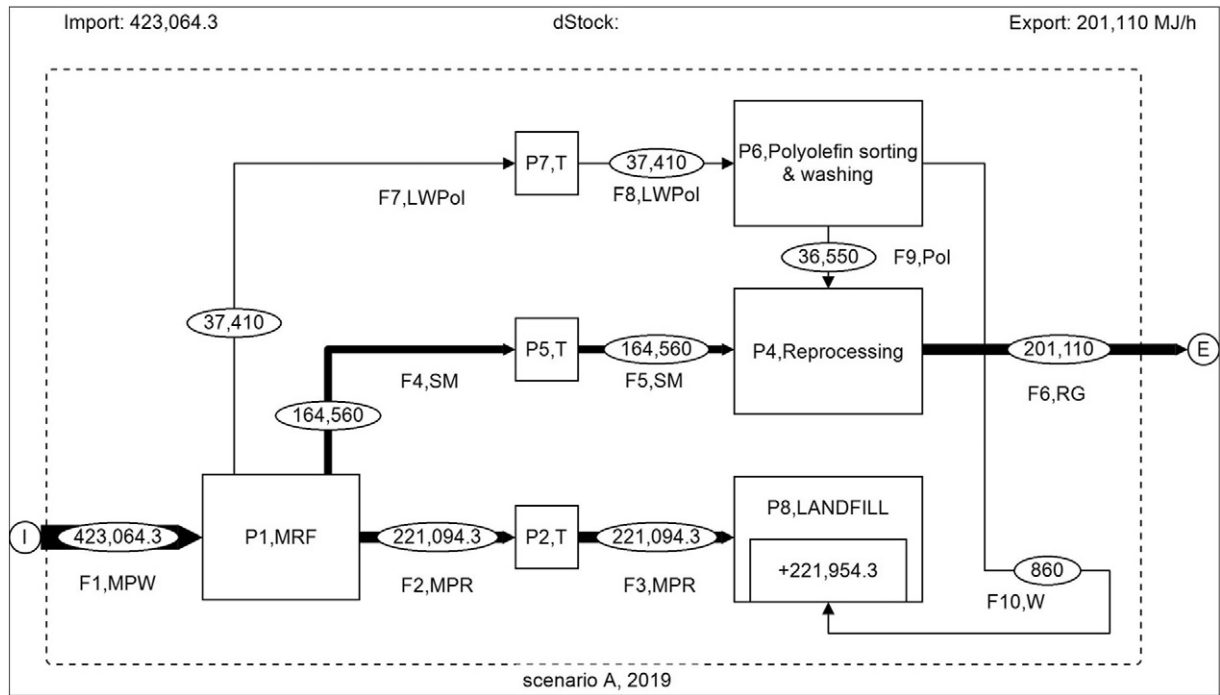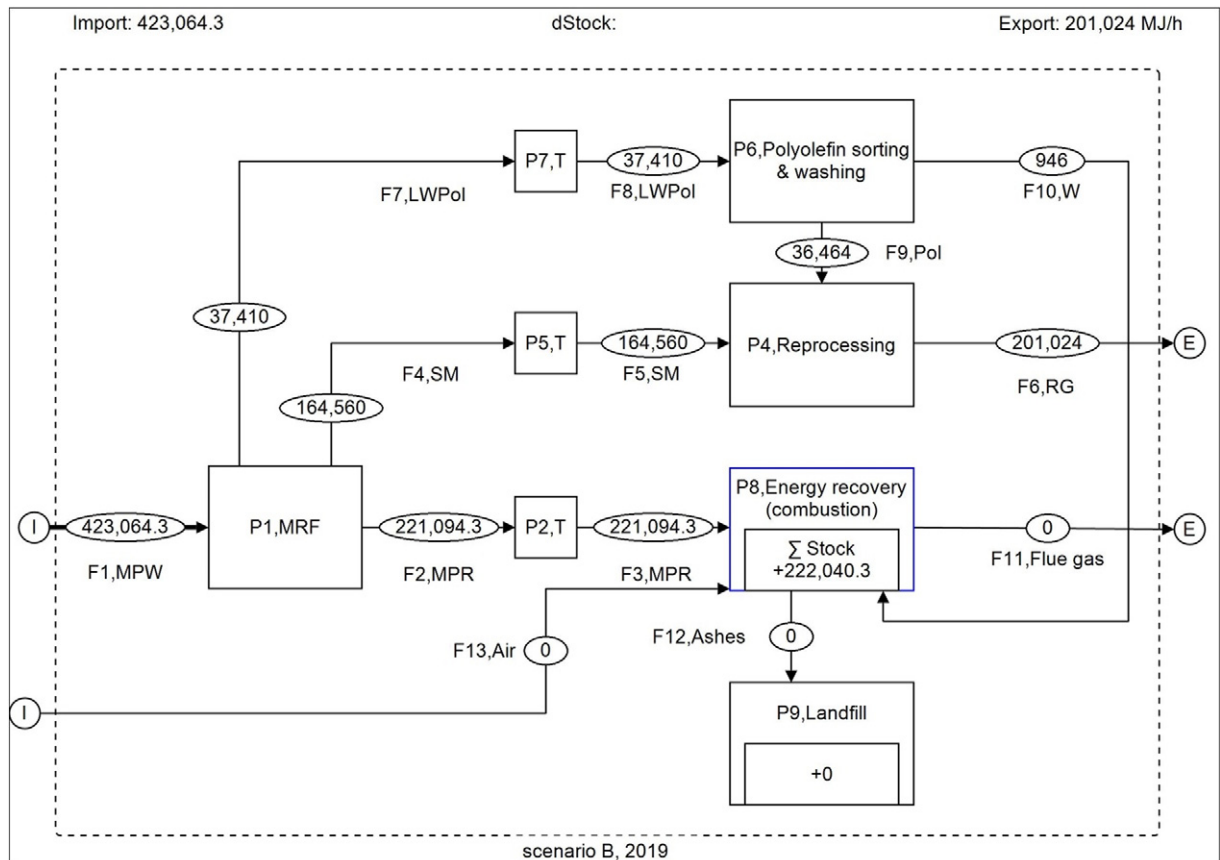

Fig. 6. Scenario B: alternative case with MPR energy recovery via incineration – total mass layer.

Fig. 7. Scenario A: base case with MPR landfilling – energy layer.

polymers. The reprocessing (P4) of selected mono-material streams is realized by recurring to processes such as melting and granulation (for polymers), melting and shaping (metals), water dissolution and paper forming (cellulose based materials) realized in different facilities, possibly integrated with those using the virgin materials; these processes have been grouped into one block (P4) and no detail is given here; a deep investigation of these processes can be found elsewhere (Perugini et al., 2005; Singh et al., 2017; Welle, 2011).

Fig. 8. Scenario B: base case with MPR energy recovery via incineration – energy layer.
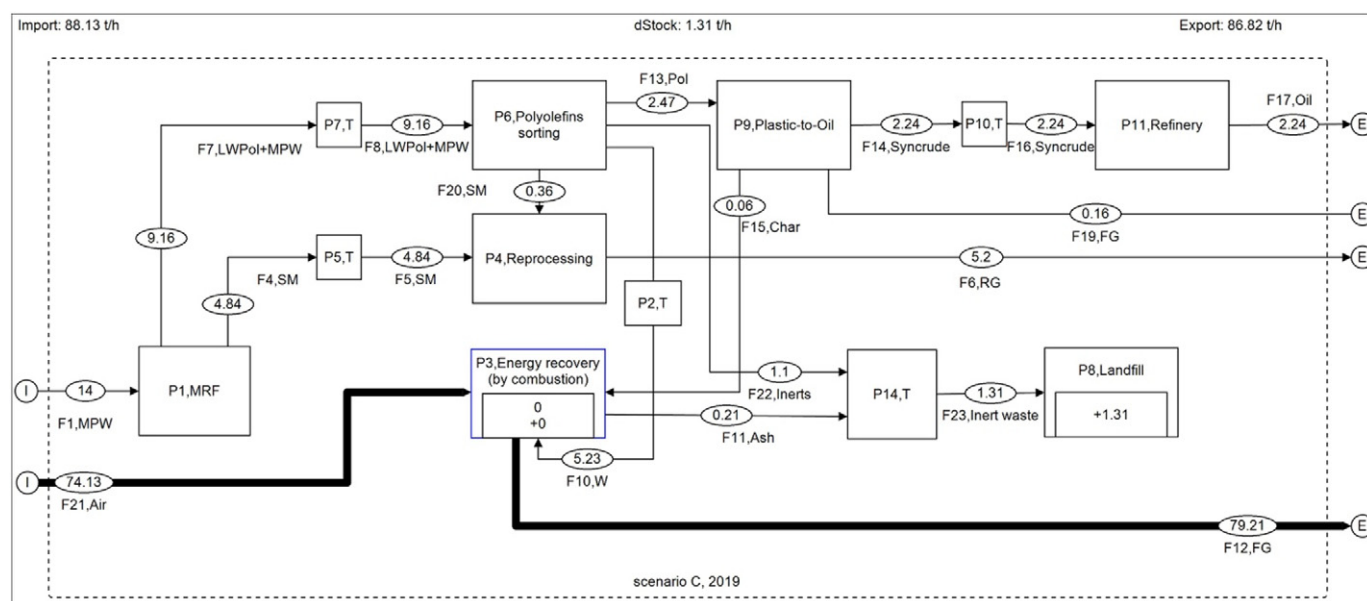
Fig. 9. Scenario C: alternative scenario with plastic-to-oil process – total mass layer.

The scenario including the energy recovery (Fig. 6) clearly allows to reduce the landfill disposal and fulfil with the landfill bans criteria for high-energy content waste. By looking at environmental issues, it is useful assess how the feedstock energy is distributed along the different streams in the system; the feedstock energy flows [MJ/h] are therefore reported for both scenarios A and B in Figs. 7 and 8. The scenario A promotes the loss of 52% of the energy content of the input flow due to landfilling; this is a negative issue for a lot of aspects also by considering the plastics undergo a slow, but inesorable, degradation once dumped in the landfill.

3.1.2. Alternative case scenarios description and assessment

The scenarios related to the base case can be improved by looking at the following targets:

a) Increasing of performances of existing processes
b) Addition of processes that allow to increase the amount and types of polymers and resins addressed to material recycling
c) Production of goods having a high-market value and included in a real economy loop, not sustained by incentives (virtual economy)

With reference to the polyolefin flow, the inclusion of the plastic-to-oil or to-chemical plant is considered. In fact, the polyaddition polymers can be converted with good yield into a petrochemical feedstock; this latter can be refined by means of fractionation to obtain commodities such as diesel, gasoline and fuel gas (similar to LPG). The inclusion of this process in the plastic waste management scenario allows to change the economic value of the whole chain and increase the sustainability of the entire system. The mass balance of this plant is related to an input corresponding to the fraction of polyolefins that have no market and are landfilled, used as fuel or sent to Asian markets, that corresponds to 2.47/h for a base of 14 t/h of packaging waste collected; this stream is obtained by sorting the mixed plastic residue and the polyolefin mixture of the main sorting plant with the double target to recover the HDPE and PET (that have very good prices in the market of secondary materials as shown in Table 1) and to meet specification for plastic-to-oil/chemicals plant. The restrictions are in fact mainly related to polymers containing heteroatoms like PET other than PVC. The PET is a valuable product so it can be sorted out by recurring to optical sorter and sell as secondary materials. The metals must be removed since their

presence catalyzes undesired reactions and produce more char and ashes. Biowaste, paper, textiles, rubber can be limited at maximum 5% total. All these requirements can be fulfilled by recurring to a specific sorting plant specifically designed to this end (AMUT Group, 2019).

The scenario C is then derived by scenario B by including the plastic-to-oil plant for a flow of mixed polyolefins (F13 in Fig. 9). The main differences between the scenarios B and C are: the different fate of mixed polyolefins (F13) addressed to material recovery by means of pyrolysis (P9) and the removal of inert fraction during the sorting (P6) from the waste destined to energy recovery by combustion. This allows to produce a lower amount of bottom ashes from the combustion process even though, by looking at mass balance, the final destination is the landfill in both cases.

The detailed mass balance for the plastic-to-oil process by pyrolysis is reported in the Table 5.

The heavy oil can be fractionated to obtain diesel while the light oil includes gasoline and jet fuel; in the total mass balances these streams are indicated with the name of "synthetic crude", before conditioning in the refinery, and "oil", after proper conditioning.

The feedstock energy balance is reported in Fig. 10; it is evident that 23% of the feedstock energy of the input F1 is stored in commodities that can substitute fossil fuels with a 1:1 substitution factor. This is quite important in the optical of circular economy because the recycled goods obtained by using reprocessed granules are downgraded due to the lacking properties. There are several examples of materials obtained by reprocessing of the secondary polymers that can or cannot be used for the production of goods in substitution of virgin polymers. For example:

Table 5
Technical data related the plastic-to-oil process in scenario C and D.

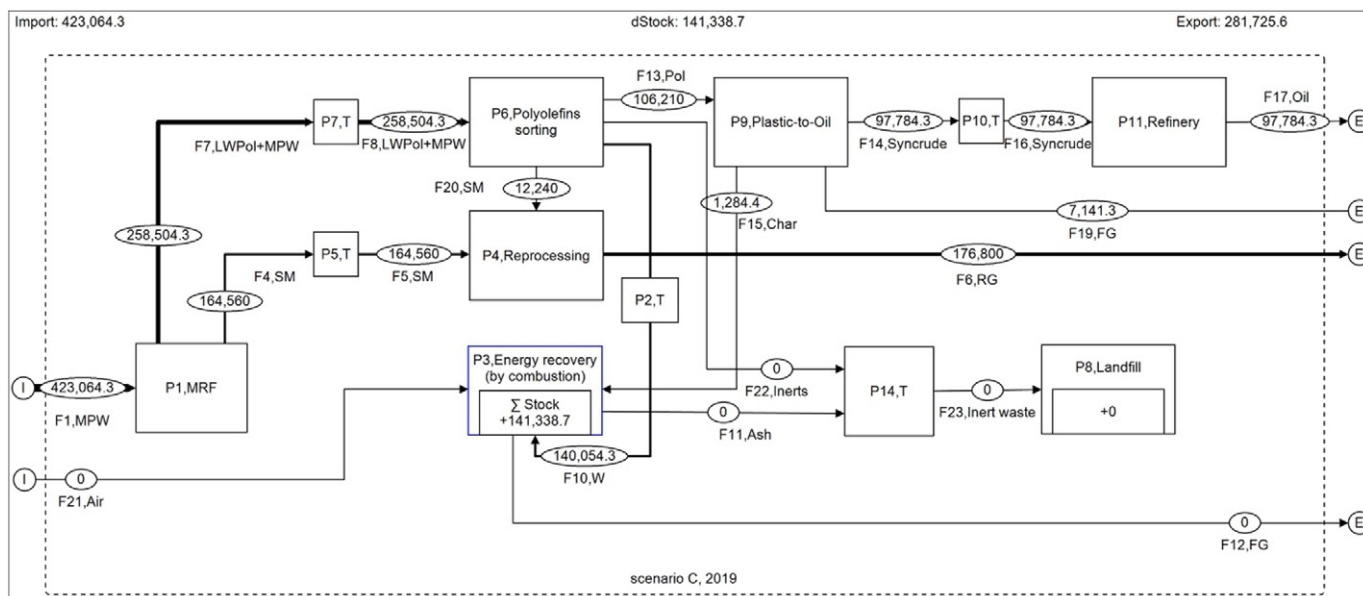|		
Plastic feedstock rate in	2.47	t/h
Fuel gas yield	6.6%	t/t
Fuel gas mass rate	0.16	t/h
Char yield	2.6%	t/t
Char mass rate	0.07	t/h
Light oil yield	26%	t/t
Light oil mass rate	0.65	t/h
Heavy oil yield	65%	t/t
Heavy oil mass rate	1.60	t/h

Fig. 10. Scenario C: alternative scenario with plastic-to-oil process – energy layer.

a) the recycled PET cannot be used for more than one time to produce bottle grade goods due to loosing of the necessary physical chemical properties during extrusion and reprocessing. The value of secondary PET light (i.e. not colored or opaque) is around 220€/t while the virgin PET price is 1100€/t; the large gap between the prices of the virgin and secondary PET drives the industrial users to find ways to use the secondary PET for application where the bottle-grade PET should be use. The applications such as of secondary PET used as internal liner between two sheets of virgin PET is a typical example of 1:1 substitution ratio. Also, the use of secondary PET for synthetic fabric can give a 1:1 substitution factor because of the price of polyester that is about the same of that indicated for PET bottle grade (https://emergingtextiles.com).

b) The LDPE used for agricultural purposes is degraded by light and heat, so that the reprocessed granule can be used for low grade goods such as bags. By considering that the plastic bags are prohibited in many Countries, the value of the largest part of

secondary LDPE (and LLDPE) is very low or negative, by considering the sorting and reprocessing costs. The exception consists in the large film packaging (LDPE > A3), for example used as tertiary packaging, whose value is saved due to short-life time.

c) All the polyolefin-based goods, even not packaging, that are already been recycled one or more time have no value as material due to the contamination of pigments and metals and very bad physical properties. For these polyolefins the way towards mechanical recycling has no sense.

In the scenario B more of 50% of the feedstock energy of plastic waste entering the system is converted into thermal energy by means of combustion while, in the scenario C, it is stored as energy content in commodities that can be used for several purposes, not limited to energetic uses.
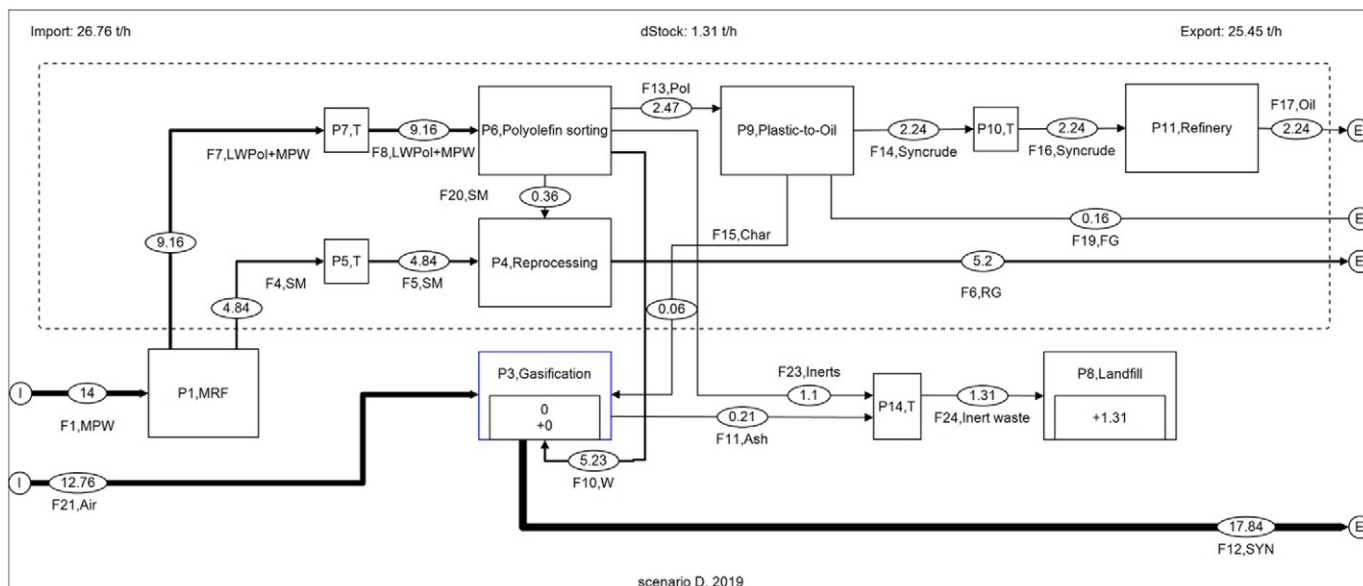

Fig. 11. Scenario D: alternative scenario with plastic-to-oil process and gasification of residue – good layer.

The sustainability of the whole system can be further improved by avoiding the use of large-scale plants for energy recovery; the traditional incinerators are in fact difficult to be localized and socially inacceptable. A different approach is the use of smaller facilities distributed over the territory by respecting the criterion of proximity for waste disposal; since the processes based on combustion requires a minimum scale (100.000 tons/year) to be viable (Fernández-González et al., 2017), it is necessary to select processes that can be carried at small-medium scales (10.000 tons/year). To this end, the gasification can be an option. The gasification process requires volumes that are much lower than those involved during combustion and, as a consequence, the capital expenditure is quite smaller at similar capacity. Moreover, once the critical aspect of syngas cleaning and conditioning is faced off and solved, it is the best option by an environmental point of view (Dong et al., 2018). The selected process (Section 2.4) can be applied to capacity starting from about 100 kg/h up to 1 t/h, even though can reach larger scale by using more modular plasma-gasifier in parallel configuration. By including these facilities in the system, the scenario can be modified as reported in the Fig. 11.

The gasification process requires less air compared to the energy recovery by combustion and produces a syngas stream that can be utilized not only for energy production. The mass balance in the figure is related to humid basis; in case of utilization of syngas in engines or turbines, a drying system needs to be installed.

The main process data related to the gasification process are reported in the Table 6 while the energy balance is reported in the Fig. 3.SM. The cold gas efficiency of the gasification process is about 0.72 if an air-to-waste ratio of 2.4 is chosen; this means that the chemical energy used by the exothermic reactions is about 5207 MJ for each input ton of waste i.e. the 26% of the input feedstock energy (Fig. 3.SM).

The electricity required by plasma torches is about 7% of the total produced; this latter value, referred to the syngas contribution, corresponds to 4300 kW of electric power generated by a dual-fuel engine fueled by a mixture of syngas and gasoline/diesel. The plasma self-consumption is reasonable by considering that the use of the plasma-assisted gasification allows to carry out the process with high flexibility and good control of operating conditions so allowing a stable operation.

3.2. Indexing and summary of results

The comparison between the scenarios A, B, C and D has been made by defining some performance's indexes. The first set of indexes are related to the mass flows of: material recycled as new goods ($Y_{M,MR}$), materials used as fuel in processes for energy production ($Y_{M,ER}$) and the materials landfilled ($Y_{M,L}$). The exact definitions of indexes are reported in the following:

$$Y_{M,MR} = \frac{\text{mass flow of recycled goods } (\sum_i F_i)}{\text{mass flow of F1}}$$

$$Y_{M,ER} = \frac{\text{mass flow of materials utilised as fuels } (\sum_j F_j)}{\text{mass flow of F1}}$$

$$Y_{M,L} = \frac{\text{mass flow of materials addressed to landfill } (\sum_k F_k)}{\text{mass flow of F1}}$$

These indexes are reported in Table 7 and demonstrate that the highest material recycling yield is obtained for scenario D as well as the minimum landfill demand.

The same indexes measuring the scenario performance reported with reference to the mass flows have been defined and evaluated regarding the energy flows. These "energy yields" (Table 8) are defined as:

$$Y_{E,MR} = \frac{\text{energy flow of recycled goods } (\sum_i F_i)}{\text{energy flow of F1}}$$

$$Y_{E,ER} = \frac{\text{energy flow of materials utilised as fuels } (\sum_j F_j)}{\text{energy flow of F1}}$$

$$Y_{E,L} = \frac{\text{energy flow of materials addressed to landfill } (\sum_k F_k)}{\text{energy flow of F1}}$$

The assessment of feedstock energy flows is quite interesting to evaluate the performance and the sustainability of the proposed scenarios: the best scenario results to be the scenario D because of the production of synthetic gas, that can be assumed as a feedstock and not necessarily as a fuel, and the maximization of production of valuable materials.

3.3. Economic comparison

The different scenarios can be compared also in relation to their impact on the economy of the waste management system. The target of realizing a real circular economy that is intrinsically sustainable is in fact reached when the system is part of an industrial system producing goods and commodities. Starting from the consideration that the MRFs represent a part of the overall system that do not produce final products but only intermediates and waste, it is clear that these processes needs of a tipping fee. The tipping fee depends on the potential values of secondary streams that the MRFs are able to obtain by managing a given input waste and by the downstream processes that convert those waste into marketable products. Two cases are discussed: one related to the scenarios A/B and another one related to the scenario C/D. The scenarios A and B differ each to other for the environmental sustainability, but they do not differ in a relevant way by an economic point of view since the disposal cost for landfilling and incinerators (that should be quite different) is levelled due to the large demand for disposal. The economic comparison has been made on the basis of the following assumptions:

a) the market values of commodities and secondary materials are those of Table 1;
b) the operating expenses of all single processes are those reported in the Table 2 of Supplementary Materials;
c) the electricity cost is 120€/MWh;
d) the value of syngas is proportional to that of LNG on the basis of its energy content (criterion of energetic equivalence);
e) the value of oil is that of Brent crude oil;

Table 6
Technical data related the gasification process in scenario D.

Input	5.29	t/h
Air-to-Waste ratio	2.41	t/t
Equivalence ratio	0.25	–
Cold Gas Efficiency (CGE)	0.72	MJ/MJ
Reaction temperature	950	°C
Syngas yield (dry)	2409	Nm³/t
Syngas density (dry)	1.25	kg/Nm³
Syngas low calorific value (dry)	4.91	MJ/kg
Syngas specific heat	1.48	MJ/t°C

Table 7
Performance mass-based indexes for scenarios A, B, C and D.

Scenario	Material recycling yield ($Y_{M,MR}$, t/t)	Energy recovery yield ($Y_{M,ER}$, t/t)	Landfill yield ($Y_{M,L}$, t/t)
A	0.41	0	0.59
B	0.41	0.50	0.09
C	0.54	0.36	0.097
D	2.08	–	0.019

Table 8
Performance energy-based indexes for scenarios A, B, C and D.

Scenario	Material recycling yield ($Y_{E,MR}$, t/t)	Energy recovery yield ($Y_{E,ER}$, t/t)	Landfill yield ($Y_{E,L}$, t/t)
A	0.47	0	0.61
B	0.47	0.53	0
C	0.67	0.33	0
D	0.86	0.14[a]	0

[a] It is referred to the fraction of organic matter used for exothermic reactions during gasification.

Table 10
Evaluated costs for the scenarios.

	Scenario A/B	Scenario C	Scenario D
System cost (no transportation), €/t	−119.64	−81.33	+8.01
Total distance travelled, km	500.0	1200.0	900.0
Total amount transported, t	14	22.78	17.55
Total transportation costs, €/t	−25.00	−97.63	−56.41
Total system cost, €/t	−144.64	−178.96	−48.40

First of all, the value of secondary materials addressed to reprocessing has been evaluated with respect to the incoming MPW. The results are reported in Table 9.

On the basis of the given data and on the mass balances of the reported scenarios, the tipping fee can be calculated by considering the operating costs for the unit processes included in the system boundaries of the different scenarios. The detailed calculations are reported in the Supplementary Materials section, the Table 10 summarizes the obtained results. These include the transportation costs by considering that:

- Scenarios A and B request short-medium transportation distances for processes P7 and P5 and long-distance transportation route for P2.
- Scenario C requests the same distances P2, P7 and P5 and, in addition, the transportation labelled with P10 and P14. The P14 is towards a landfill for inert waste while P10 represents the transportation of syncrude from PtO to the refinery site. This latter distance is quite long since refineries are present in few sites for each Country.
- Scenario D presents the same P7, P5, P10 an P14 transportation processes but, since it is integrated or very close to MRF, like in the case of PtO, the P2 is missing.

By considering assigned distance travelled for each transportation unit process, whose values have to be considered as a mere indication for comparison purposes, the values of system costs have been obtained (Table 10).

All values are referred to the input mass rate at 14 t/h. The value estimated for the system cost of scenario A/B is in agreement to that given by the waste traders (CEA, 2019). The considerable effect of transportation demonstrates that the installation of several medium-scale fully integrated facilities should be favored compared by few large-scale plants at long distance from each to other.

4. Conclusions

The so-called "plastic" waste is composed by polymers, including thermoplastics and other resins (elastomers, multilayered,

Table 9
Evaluation of the specific income value of the secondary materials produced by the MRF.

Material	Market price, €/t	Mass flow yield measured, t/t	Fract. value, €/t
Light PET	220.6	6.8%	15.08 €
Light blue PET	220.6	8.7%	19.13 €
Colored PET	36.0	6.5%	2.35 €
Opaque PET	36.0	0.0%	0.00 €
HDPE	400.0	7.0%	28.00 €
LDPE > A3	210.2	1.7%	3.65 €
LDPE < A3	0.0	4.1%	0.00 €
PP	0.0	0.4%	0.00 €
Fe	160.2	4.3%	6.94 €
Al	975.4	1.2%	11.92 €
Specific income value			87.08€

multipolymer, …). The mechanical recycling can be applied to a large fraction of packaging polymers but not to all resins and even not to all thermoplastic ones, due to technical, economic and commercial reasons.

The present system applied to "plastic" waste management is totally based on sorting into MRF followed by mechanical recycling and disposal of unsorted waste. The amount of this latter fraction is huge, and the landfilling is the worst (largely applied) option to refer to. Energy recovery by incineration allows to save space in landfills and to recover the high energy content of polymers, but it is not sustainable and opposed by population other than limited by new regulations and guidelines.

Generally speaking, pyrolysis and gasification are in development, stimulated by the request of more sustainable waste treatment options; in this paper, the application of these processes to polymers that cannot be conveniently recycled as secondary materials is highly specific, in particular:

- the clean polyolefins are converted into oil (and its commercial fractions such as diesel and gasoline). The selected process is based on pyrolysis (thermolysis under inert atmosphere) followed by fractionation into two commodities: an LPG stream usable for self-consumption and a synthetic oil to be added to fossil raw oils or fractionated downstream the plant itself;
- the commingled and/or poly-condensed polymers together with other resins are transformed into syngas. Due to the tar production during gasification of polymers, a plasma-assisted gasification process has been chosen in order to promote the cracking of intermediates and of heavy hydrocarbons chain.

The mass and feedstock energy balances show that the scenarios including the above cited processes allows to meet the present and future regulations about the plastic waste management and end-of-waste criteria. The scenario D is a long-term scenario that would allow to put apart the incineration of plastics in traditional large-scale facilities. It represents the best option because it guarantees the highest values for material recovery and the minimum values of landfilled matter with a reasonable cost.

An economic assessment demonstrates that the present scenario is sustained by tipping fee that are continuously arising due to the high costs of transportation towards the treatment processes, both those for recovery as well as those for disposal. The evaluation of total travelled distance is indicative since it is Country-specific but it is helpful to understand how the integration between plants and the progressive scaling own of facilities can help to decrease the economic costs of the packaging waste system. The introduction of the suggested processes, due to the production of commodities such as diesel, gasoline (or simply oil) and syngas allow to increase the net income at such values to put to zero all the fees. These scenarios (C and D) are then actually able to promote the realization of circular economy without incentives that sustain a virtual economic chain, destined to fail.

On the basis of the economic assessment, even if realized without realizing a complete business plan, the target of zero-approaching plastic waste and real circular economy can be reached in a medium term by enlarging the industrial system to plastic-to-oil and plastic-to-

chemicals processes, by substituting the large-scale combustion plants by smaller-scale integrated gasification plants and by using the electricity produced by these plants to lower the opex of treatment facilities operating in the industrial site. The evaluation of operating expenses for these systems has clearly shown that the electricity represents the main cost; its production should be incentivized for self-use instead of putting it into the grid.

A general conclusion should be that the waste system planning that actually address to circular economy needs to consider the smart integration of different technologies in industrial districts as a preferable target.

Acknowledgements

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.137233.

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