Environmental Engineering and Management Journal October 2018, Vol.17, No. 10, 2437-2445
http://www.eemj.icpm.tuiasi.ro/; http://www.eemj.eu

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APPLICATION OF LCA METHODOLOGY IN THE ASSESSMENT OF A PYROLYSIS PROCESS FOR TYRES RECYCLING

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Abstract

According to recent estimations, the total yearly amount of end-of-life tyres (ELTs) arising in Europe exceeds 3 million tons; more than 95% are managed through mechanical and thermal treatments because ELTs represent a useful resource both of materials and energy. For this purpose, the goal of the present work is the assessment of the environmental impacts of a novel pyrolysis process and the comparison with alternative ELTs valorisation or disposal scenarios. Life Cycle Analysis (LCA) methodology has been applied to determine the most critical stages of the process under study, assessing the environmental benefits arising from the recovery of material and energy and the impacts, compared to the technologies already on the market, taking into account treatment processes, materials recovery and disposal of wastewater/residues. The chosen functional unit (FU) is 1 ton of ELTs treated by the plant. The different scenarios investigated have been analyzed through ReCiPe impact assessment method. Considering the pre-treatment, the pyrolysis process results in a lower environmental impact compared to the others, with the 1/3, 1/10 and 1/20 of energy consumption compared to the alternatives considered. The analysis of pyrolysis process showed that the avoided impact due to the recovery of carbon black, steel, oil and syngas exceeds the impact generated by the process, related to the energy consumption and to the emissions into the atmosphere. Compared to other energy-recovery scenarios, a greater advantage results from the pyrolysis process, mostly due to the recovery of valuable materials. Then, comparing it to other material recovery scenarios, a variable influence is given by the different options of recovery, considering which materials could actually be replaced and the commercial value of the materials that is replaced.

Key words: energy, life cycle assessment, material, recovery, tyres

Received: March, 2018; Revised final: July, 2018; Accepted: September, 2018; Published in final edited form: October 2018

1. Introduction

The flows of end of life tyres (ELTs) managed all around Europe has been recently quantified at more than 3 Mt in 2016 (about 3.51 Mt in EU-28, and 3.93 considering also Norway, Switzerland and Turkey) (ETRMA, 2018). Of this amount, more than 95% has been treated for the recovery of energy or materials. As for Italy, Ecopneus (National consortium company for ELTs collection and treatment) (Ecopneus, 2017) reports that approximately 265 kt of new tyres were introduced in the national market in 2015. The total collection of ELTs in 2015 amounted to about 252 Kt and, of these, about 245 kt were properly managed, according to the national legislation.

After collection, ELTs are subdivided between recycling and recovery plants; ELTs constitute an important source of valuable materials (about the 40%

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in weight of the tyre is composed of rubber, about the 30% is carbon black, while the third major component by weight is steel) and energy, with a lower calorific value similar to that of coal. The main treatments for ELTs are based on mechanical and thermal processes.

Mechanical processes generally start with the crushing of the tyres and the physical separation of its components, without changes in the chemical composition. The degree of grinding is a function of the desired final application. For low added value manufacturing, simple shredding technologies are adopted, which return chips varying in size from 20-50 mm up to 300 mm. They are suitable as coarse materials used in civil engineering works like modified asphalts or bitumen production (ETRMA, 2018; Ecopneus, 2017). Otherwise, a further grinding process, for example using mill technologies followed by sieving, allows to obtain homogeneous streams of granules and powders of different size (from 0.8 to 20 mm for granules and <0.8 mm for powders). These secondary raw materials can be used for a wide number of applications including elastic modifiers for sports and playgrounds surfaces or compound additives for new tyres manufacturing, insulation materials and urban furniture (ETRMA, 2018; Ecopneus, 2017). The use of specific process devices for cleaning the ELTs fed into the recycling plants, as well as other downstream technologies (granulators for the separation of steel from rubber, magnetically or gravimetrically, or of textile fibres through controlled aspiration), allows high quality in terms of purity of the recovered materials.

Thermal processes involve the use of the entire or crushed ELTs as alternative fuels for energy production, exploiting their high calorific value. Therefore, ELTs can be co–incinerated in cement kilns, paper mills, lime manufacturing plants and thermal power plants, i.e. in all the highly energy-intensive plants (ETRMA, 2018; Ecopneus, 2017).

An interesting alternative for treating ELTs is pyrolysis (a chemical process which is carried out at moderately high temperature and in the absence of oxygen) which allows the decomposition of the organic material. The resulting products are divided into three streams (Quek and Balasubramanian, 2013):

- solid fraction: this is a carbonaceous residue (char) in which it is also possible to find metals or inorganic fillers, if these were contained in the input material;
- liquid fraction: this is an oil rich in organic molecules (aliphatic and aromatic) having different molecular weight, such as alcohols, aldehydes, acids and ketones and also aromatic compounds;
- gaseous fraction: this is syngas composed of light hydrocarbons (C2-C6), in addition to CO, CO_2, H_2O and H_2.

The pyrolysis process generally requires an initial phase of shredding to reduce tyres at 2 to 10 cm in size. These pieces are then loaded into the reactor, in which the pyrolysis reactions take place at temperature above 450°C, leading to the formation of a solid residue (coal and metals), gases and vapours. Vapours are then condensed and separated from the gases to obtain a pyrolytic oil, which can be furtherly refined. As regards the non-condensable compounds, these are burned to obtain electric and thermal energy. The exhaust gases are carried in a burner, in which a reduction of NO_x by selective non-catalytic reduction (SNCR) with urea and a reduction of sulfur oxides and HCl with soda are made. Once purified, the fumes are released into the atmosphere. The solid residue needs further treatment to separate the metal fraction from coal (Andreola et al., 2016).

For this purpose, the Life Cycle Assessment methodology (LCA) has been applied to determine the most critical stages of the process under study, the environmental benefits arising from the recovery of materials and energy and the greater or lower impact compared to the mechanical or thermal technologies on the market. Different previous works studied the environmental impact of recovery of tyres, through LCA methodology (ISO 14040, 2006; ISO 14044, 2006), comparing alternative scenarios of ELTs management (Corti and Lombardi, 2004; Li et al., 2010; Rafique, 2012). The assessment was performed considering the indirect impacts caused by energy production stage, the direct impacts caused by ELTs treatment process and the avoided impacts caused by valuable products (recycled materials and energy). Bartolozzi et al. studied the case of rubberized asphalt pavement in comparison to a conventional asphalt (Bartolozzi et al., 2015). In another paper (Clauzade et al., 2010) a comparative environmental evaluation of the various recovery alternatives was carried out, aimed at identifying the strengths and weaknesses of each recovery method. Even in Feraldi et al. (2013) and Ortiz-Rodrìguez et al. (2017) different treatment options have been compared as material recycling and tire-derived fuel combustion through co-incineration, finding that the material recycling scenario provides a greater impact reduction than the energy recovery scenario, but they did not analyze the case of pyrolysis process (Feraldi et al., 2013; Ortiz-Rodrìguez et al., 2017).

As for Italy (according to Ecopneus, 2017), about 150 kt of CO_2 eq. were emitted by the combustion of ELTs in cement plants, 57% of which in Italy (where there are 5 plants, mostly in the North) and 43% in foreign cement plants (located in Romania, Morocco, Turkey, Austria, Germany and Hungary (Torretta et al., 2015). This treatment allows the saving of other fossil fuels, such as coal and petroleum coke (56.8% of avoided emissions). On the other hand, emissions from ELTs recovery in power stations (11.6%) are not offset by the benefits resulting from the avoided production of an equal amount of electricity generated in Italy (Ciacci et al., 2014), considering the national energy mix.

As regards the resources balance (material footprint), 54% of their consumption is associated with collection and transportation of ELTs to the treatment systems, which involves the use of fossil fuels to feed hundreds of vehicles and dozens of ships, which annually cover millions of kilometres to carry

ELTs. In a scenario in which the treatment of ELTs is exclusively performed within the national territory, it would lead to a lower resources consumption and therefore greater environmental benefits.

Finally, ELTs recycling results in significantly greater water savings, compared to the energy recovery scenario, while carbon and material footprint show values of the same order of magnitude.

The goal of the study is the assessment of the environmental impacts of the pyrolysis process of ELTs performed with a novel plant (owned by the company Curti S.p.A., located in Northern Italy) (Bortolani et al., 2014) and to compare it with alternative valorisation and/or disposal scenarios.

2. Material and methods

2.1. Goal and scope definition

This novel pyrolysis plant is an upgraded version of a pilot plant designed and built by Curti S.p.A. The pilot plant was tested with ELTs (Giorgini et al., 2015a), glass fibres reinforced polymers (GFRPs) (Giorgini et al., 2016) and carbon fibres reinforced polymers (CFRPs) (Giorgini et al., 2015b). These experimentations have been crucial to obtain useful data required for the design of the new plant object of the study here proposed.

The environmental impacts associated to the life cycle of ELTs were evaluated giving credit to recycling for the avoided production from primary sources (Eckelman et al., 2014; ISO 14040, 2006; ISO 14044, 2006). For instance, the use of ELTs as a fuel in cement factory would avoid the use of coal and petroleum coke. Yet, steel and the combustion ash are incorporated into the cement, thus permitting a further saving of filler material. Similarly, sending ELTs to power plants for the production of electrical energy would avoid to produce the same amount of electricity from the national grid. The steel that is obtained by crushing ELTs and by combustion in power stations supplements the avoided impact, being recyclable as scrap iron. The ashes resulting from the combustion in power stations can be recycled as a binder for cement or as material for road infrastructure. Lastly, the rubber recycled as granules and powder, coming from a mechanical treatment of ELTs, avoids the consumption of virgin rubber for the creation of new compounds.

The boundaries of the system are "from gate to gate", considering the life cycle phases regarding the following operations:

- treatment process (including all input and output streams for the supply and distribution of materials and energy);
- materials recovery (sent to recycling plant);
- disposal of wastewater/residues.

The functional unit is the physical quantity to which all flows and impacts (input and output) are reported: 1 ton of ELTs treated by the plant of pyrolysis has been chosen as the functional unit, dimensioned on a 4 tons/h capacity (according to the primary data provided by the plant). As regards the alternative scenarios, they also have been evaluated with the same reference unit in such a way that they can be compared in a univocal way.

2.2. Life Cycle Inventory (LCI)

In the Life Cycle Inventory step all mass and energy flows of the processes investigated were considered. First of all, steps and material flow of the pyrolysis plant managed by the company Curti s.p.a. has been analyzed, considering the flow diagram shown in Fig. 1.

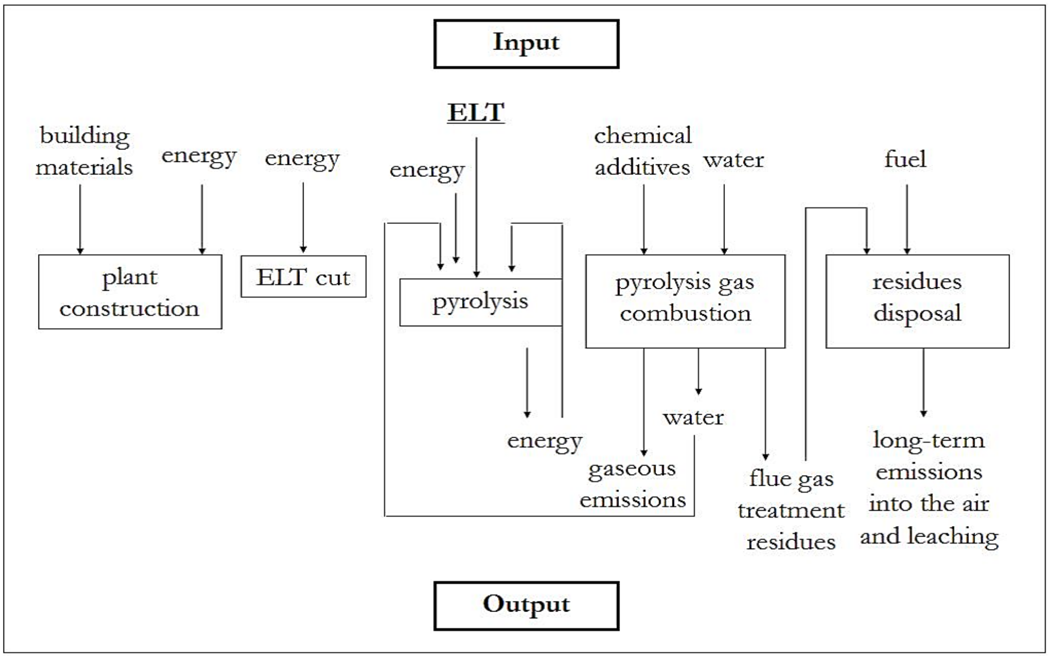

Fig. 1. Flow diagram of the pyrolysis plant studied

This is a pyrolysis process for the recovery of ELTs able to recover three main products having a commercial value: carbon black, oil and steel. The peculiarity of this process is that, in contrast to the common pyrolysis processes, in the pre–treatment step it is not necessary to crush the whole tyre, but only a circumferential cutting of ELTs (a "single cut") is needed (the diameter must be less than 1400 mm), (Bortolani et al., 2014; Giorgini et al., 2015a).

Input and output flows included in the model are shown in Table 1.

Table 1. Input and output of the PFU pyrolysis process by Curti S.p.A.

Flows	***Quantity***	***Unit***
Input		
PFU	1.00E+00	t
Electricity for pre treatment	1.41E+01	kWh
Electricity for pyrolysis	5.69E+01	kWh
Air	1.20E+04	kg
$CO(NH_2)_2$	1.33E+00	kg
$NaHCO_3$	2.67E+00	kg
Output		
Carbon black	3.15E+02	kg
Oil	2.57E+02	kg
Steel	1.30E+02	kg
CO_2	8.08E+02	kg
NO_2	2.03E-01	kg
SO_2	1.40E+00	kg
H_2O	1.16E+01	kg

By this system it is possible to obtain the recovery of the following materials:

- carbon black. In the modelled scenario an equal amount of *Carbon black, at plant/GLO U* has been inserted as an "avoided product" (process already present in the reference database, Wernet et al., 2016);
- oil (a low sulphur diesel). In the modelled scenario an equal amount of *Diesel, low-sulphur, at refinery/CH U* has been considered (Wernet et al., 2016);
- steel. In the modelled scenario an equal amount of *Steel, low-alloyed, at plant/RER U* has been accounted (Wernet et al., 2016).

In the model, the consumption of air, sodium carbonate and urea (necessary for the SNCR treatment) has been inserted, as well as two different input flows of electricity consumption, one necessary for the circumferential cut of ELTs and the other for the pyrolysis process considering the national energy mix (Electricity, medium voltage, production IT, at grid/IT U).

Moreover, the emissions of CO_2, NO_2 and SO_2 into the air due to the combustion step, and the wastewater output were considered.

After the analysis of this process, a comparison between different pre-treatment scenarios has been performed, considering different processing steps (Corti and Lombardi, 2004; Rafique, 2012):

- a single circumferential cut realized by the new pyrolysis plant;
- grinding, with the production of ground particles of about 7-10 cm, that could be used for energy recovery (eg. electricity production, cement plant; Ecopneus Report, 2017);
- crushing, that is a further grinding to a size of about 2 cm, that could be used for energy and material recovery purposes too;
- pulverization, to a size lower than 1 mm, that could be aimed at material recovery (eg. sports floors, insulating, rubber goods).

In every scenario, the electricity consumption and the steel consumption related to the wear of the cutting blades have been considered, referred to 1 ton of ELTs treated.

After this step, the pyrolysis process under investigation was compared to other scenarios of recovery of energy or material.

Considering the energy recovery processes, the management of 1 ton (the functional unit) of ELTs has been compared amongst:

- pyrolysis plant;
- cement plant;
- waste-to-energy plant.

In the cement plant scenario, the following input and output have been considered (Corti and Lombardi, 2004):

- the avoided use of coal (as *Hard coal supply mix/IT U*) and iron (as *Iron scrap, at plant/RER U*) due to the use of ELTs (already containing the steel necessary as reinforcement for the cement);
- the input of energy necessary for the co-combustion (diesel and electricity) and the electricity for the grinding step;
- atmospheric emissions of CO, chromium, lead, NOx and non-methane volatile organic compound (NMVOC).

Concerning the waste-to-energy process scenario, a previous life cycle assessment model containing primary data regarding a municipal solid waste (MSW) incineration process (Passarini et al., 2014) has been used.

In the second step, the comparison of different scenarios for material recovery through the pyrolysis process was carried out. Since the recovery of secondary rubber from ELTs could result in different uses (e.g., modified asphalts, sports surfaces, anti-trauma surfaces for playgrounds, etc.), its recycling could substitute different kinds of material, and cannot be simply considered as a replacement of an equivalent amount of primary (synthetic or natural) rubber (which has a very versatile use, in many different applications). However, the recovery of secondary rubber from ELTs has been considered as an extreme hypothesis of recycling, which can settle the highest benefit deriving from mechanical recovery. In order to take into account, the avoided impact by the recycling of ELTs two different scenarios were considered and compared to the pyrolysis process under investigation:

- Scenario 1. In the first scenario the avoided impacts associated to the following recovery processes were considered:
  - o recovery of iron (*Iron scrap, at plant/RER U*);
  - o recovery of polymeric material as an avoided production of synthetic rubber (*Synthetic rubber, at plant/RER U*);
  - o recovery of the fibers as an avoided production of material for road paving such as bitumen (*Bitumen, at refinery/RER U*).
- Scenario 2. In the second scenario the avoided impacts associated to the following recovery processes were instead considered:
  - o recovery of iron (the same as above);
  - o recovery of the polymeric material and of the fibre as an avoided production of an equivalent amount of synthetic rubber, bitumen and sand (with the proportion of one third, each).

All the two scenarios have been modelled considering the same functional unit (1 ton of recovered ELTs); in all models, the input of electricity necessary for the pulverization step and the emission of particulate matter into the air were included.

3. Results and discussion

The Life Cycle Impact Assessment (LCIA) stage was carried out using the ReCiPe analysis method (Goedkoop et al., 1999; Goedkoop et al., 2008; Goedkoop et al., 2012), considering five impact categories at the midpoint level (Climate Change, Human Toxicity, Particulate Matter Formation, Fossil Fuels Depletion and Metal Depletion) and three damage categories at the endpoint level (Human Health, Ecosystem Quality and Resources Depletion). According to the methodology, to each input and output of the models created, one or more "midpoint" impacts have been associated, using the method of analysis ReCiPe and the software SimaPro. Following ReCiPe procedures of normalisation and weighting (hierarchical perspective) a single score (Pt) was calculated, that is an index summarising the global impact of each scenario (Goedkoop et al., 2012). In the following paragraphs, the results related to different scenarios are reported and discussed (Fig. 2 and Fig. 3).

Results show that electricity consumption for tyres cutting and co-combustion accounts for about 10% of the total impacts of the process, while atmospheric emissions are responsible for the most of the total impacts generated. However, the avoided impact due to the recovery of carbon black, steel and oil fuel exceeds widely the impacts estimated (more than an order of magnitude), with a gain associated to the damage category "Resources Depletion" (Fig. 3). Therefore, environmental benefits are greater than impacts, especially considering the impact categories of Climate Change, Fossil Fuel Depletion and Metal Depletion (directly related to the recovery of steel). Fig. 4 shows the results from the scenario analysis

Considering only the pre-treatment, the process consisting of a single cut got an environmental impact equal to 1/3, 1/10 and 1/20 respectively of the alternatives considered (i.e., grinding, crushing and pulverization). In particular, lower impact resulted for the categories related to Climate Change and Fossil Depletion. This is likely due to the very low consumption of electricity required by the single cut step (130 MJ/t.

For the other recovery and recycling processes, a finer grinding is required, with a higher consumption of electricity, from a size of few centimeters to less than 2 millimeters (the finest fraction is generally employed for material recycling for which 2.81E+03 MJ/t are required). Considering energy recovery processes, the management of 1 ton of ELTs in the pyrolysis plant investigated was compared with a cement factory and a waste-to-energy plant (Figs. 5 - 6).

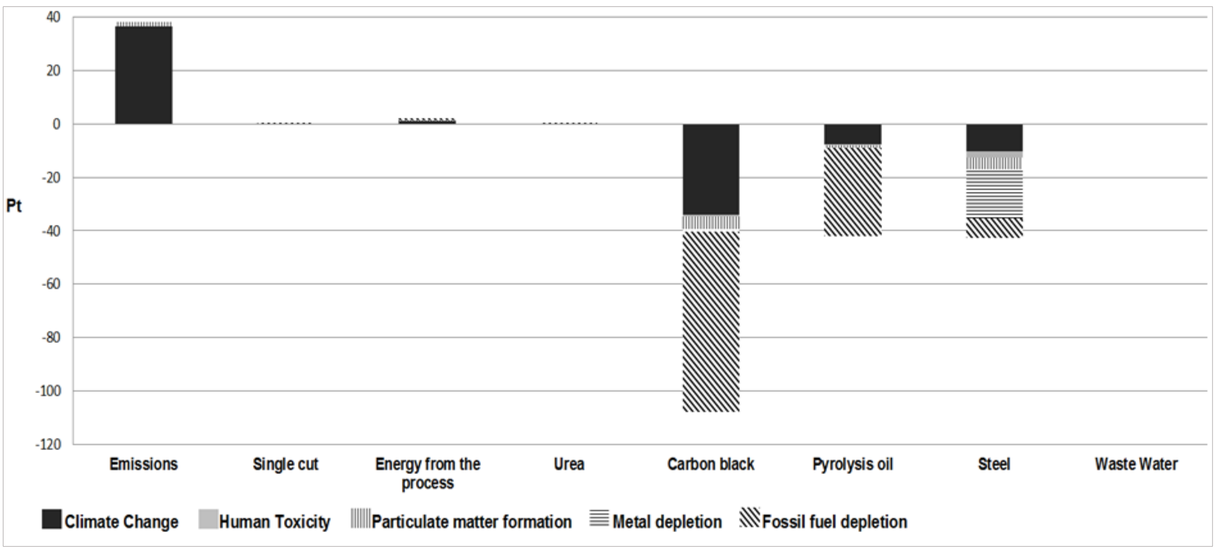

Fig. 2. Impacts of the pyrolysis process (Single Point, midpoint impact categories)

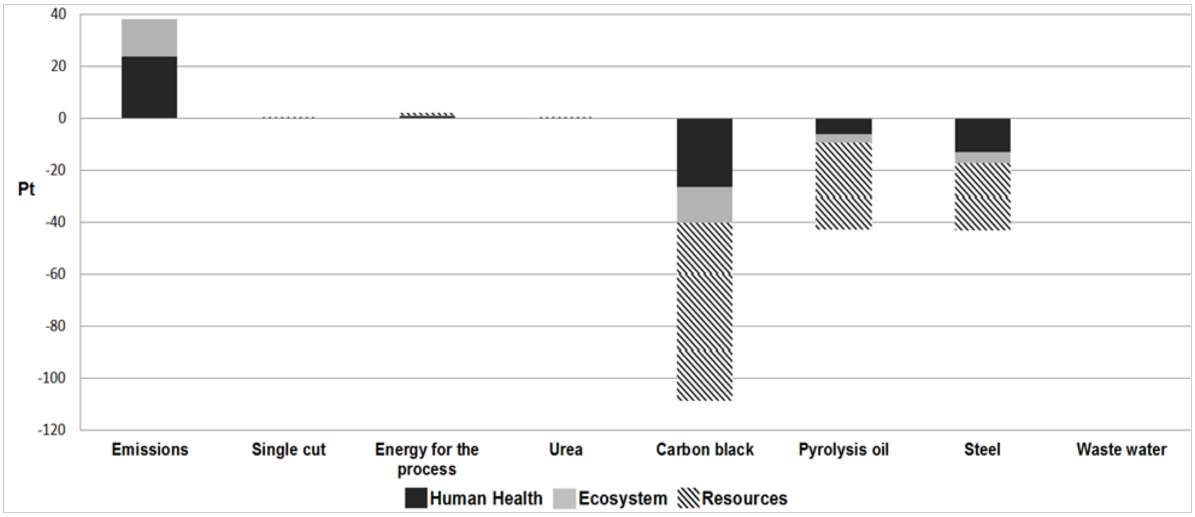

Fig. 3. Impacts of the pyrolysis process (Single Point, endpoint impact categories)

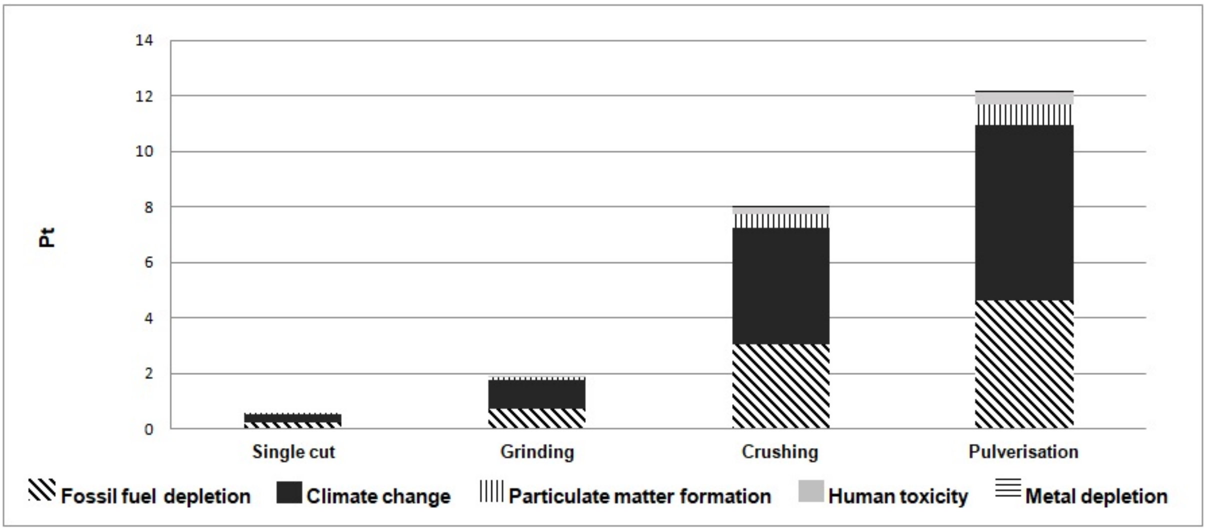

Fig. 4. Impacts of different pre-treatment process (Single Point, midpoint impact categories)

Compared to other energy-recovery scenarios (recovery in cement plant and in a solid waste incineration plant) the environmental preference is given to the pyrolysis process, especially for the Fossil Fuel Depletion midpoint category (Fig. 5) and the Resources Depletion endpoint category (Fig. 6) thanks to the recovery of diesel performed. It is worth noting that for energy recovery in a cement factory, coal was considered as energy carrier providing the same amount of energy; this has been considered as an avoided impact. If another fuel less impacting than coal was considered, the avoided impact would be likely lower for the categories Fossil Fuel Depletion and Human Toxicity. The avoided impact for the Fossil Fuel Depletion category is due to the partial replacement of the diesel with ELTs, while that of Human Toxicity is the result of the avoided emission of NO_x and NMVOC during the production of the clinker in the cement plant.

It can be observed, on the other hand, that the waste to energy process results in a net-positive impact since the energy recovery with an avoided impact for the category Fossil Fuel Depletion does not offset the damages coming from the different emissions and consumptions. A higher impact for the category Climate Change (722 $kgCO_{2eq}/t$) was computed. Considering the endpoint categories (Fig. 6), the waste-to-energy of ELTs has a higher damage related to Human Health and the pyrolysis resulted in the lower environmental burden thanks to the recovery of material and the avoided damage for Resources Availability.

Then, a comparison of the pyrolysis process with different recovery of material scenarios was performed (Figs. 7 - 8). Compared to other material recovery scenarios, a better performance is due to the different recovery options of the granulate/powder, depending on which materials they could actually replace. The efficient recovery of rubber (to replace the same amount of synthetic rubber), and metals would bring to a greater environmental benefit especially related to the impact categories of Climate Change and Fossil Depletion (Fig. 7). However, as previously said, this could be considered an extreme hypothesis: even though a quantitative recovery of secondary rubber could be reached, a complete substitution of primary synthetic rubber would also depend on comparable quality of the recycled material for the same range of applications. A more realistic option, which considers that only a part of material could effectively replace primary synthetic rubber, would generally bring to a lower gain from the environmental point of view, compared to the pyrolysis technology.

4. Conclusions

The LCA analysis carried out on a new pyrolysis plant for ELTs treatment highlighted the effective influence of the added value of recycled products in determining the environmental sustainability of a process. Taking the case of the scenarios investigated in this study, carbon black, oil, steel and pulverized ELTs ready for material recovery have all a high added value, as they can be reused as secondary raw materials in many applications. In order to optimize a process, it is therefore essential to analyse the industrial demand to ensure the use of the recycled products and thus the accounting of avoided impacts.

The study identified the most environmentally sustainable solutions among the pre-treatments and the recovery processes investigated, and performed preliminary assessments to support the design phase of a new pyrolysis pilot plant. At the end of the analysis, it was possible to identify the critical aspects and the strengths of each scenario, computing the associated environmental impacts.

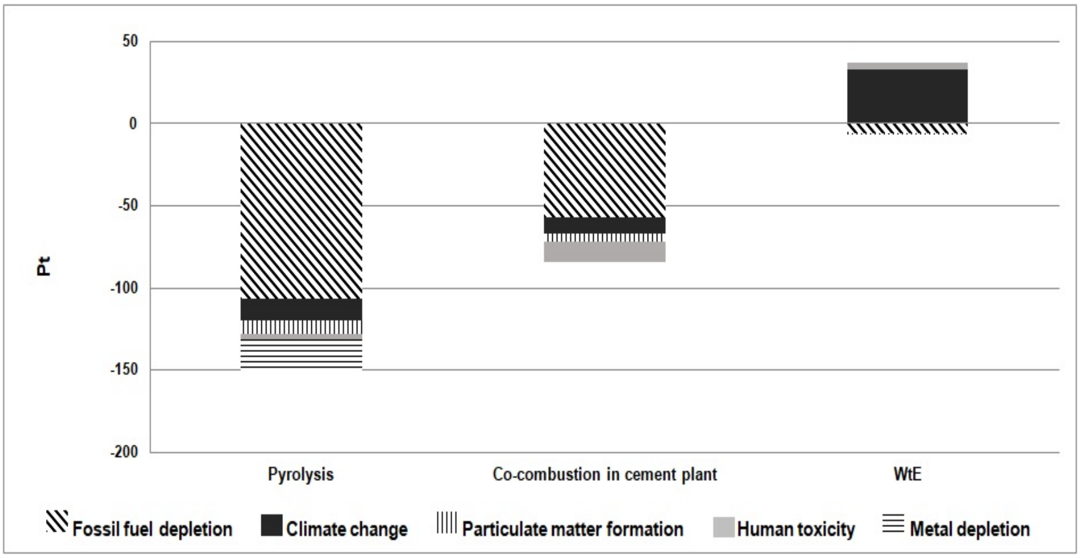

Fig. 5. Impact of the energy recovery processes (Single Point, midpoint impact categories)

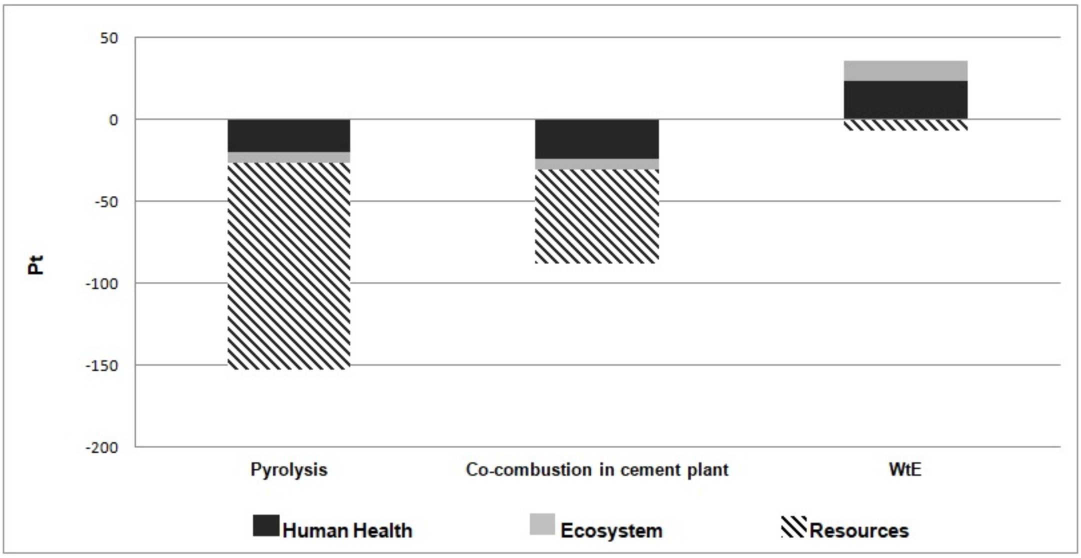

Fig. 6. Impacts of the energy recovery processes (Single Point, endpoint impact categories)

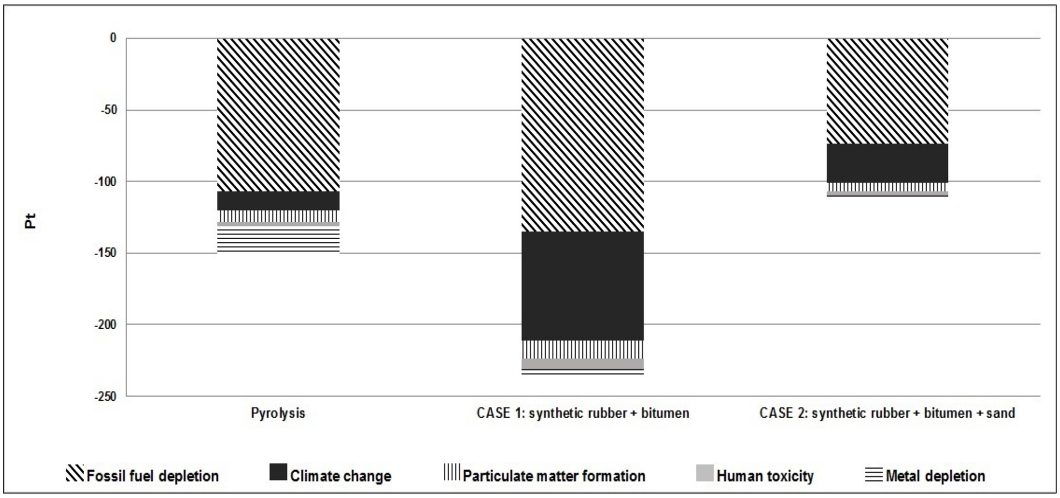

Fig. 7. Impacts of the material recovery processes (Single Point, midpoint impact categories)

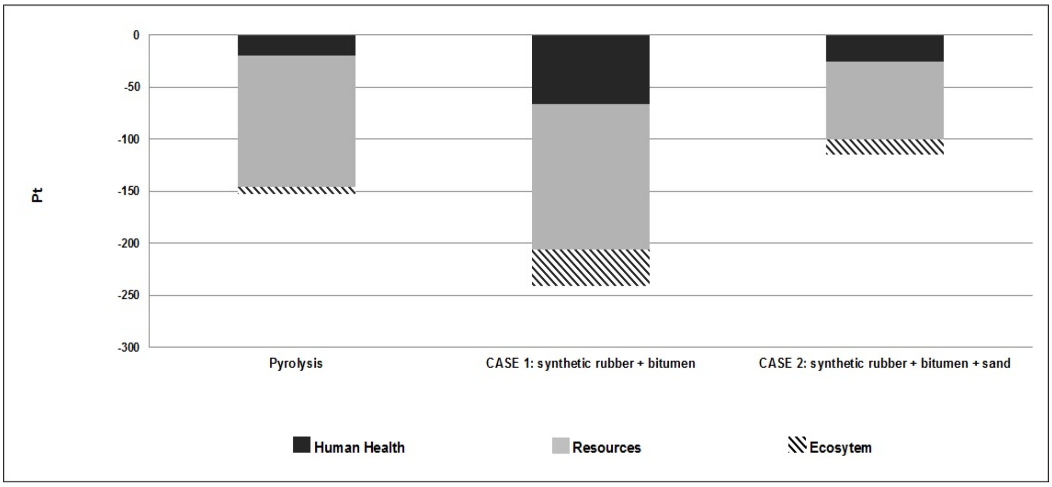

Fig. 8. Impacts of the material recovery processes (Single Point, endpoint impact categories)

The pyrolysis process investigated resulted in lower environmental impacts if compared to different energy recovery options thanks to lower energy requirements for the pre-treatment phase (only "single cut", with a cumulative energy demand of 1.30E+02 MJ_{eq}/ton, with respect to the classical crushing of the tyres, with a cumulative energy demand of 1.83E+03 MJ_{eq}/ton), and to a more efficient recovery of secondary materials (carbon black, oil and steel). The same process resulted also competitive, in terms of environmental performance, with other recyclingtechniques, even if it is important to understand which materials the recovered resource can replace (sand, bitumen or synthetic rubber) and in which fraction. However, quality issues of the recovered materials can affect the replacement of primary resources for given applications.

LCA confirmed to be a very useful methodology investigate the environmental performance of industrial processes and to compare alternative technologies. Identifying of the most advantageous options for material and energy recovery provides an evidence-based knowledge for safeguarding the environment, natural resources and the human health.

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