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# Environmental assessment of the recycling process for polyamides - polyethylene multilayer packaging films

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**Abstract**

The plastic packaging, and in particular the multilayer flexible packaging, has several characteristics that make it essential in everyday life. However, the sustainability of used materials is undermined by the difficulties encountered for their recycling. The purpose of this study is to assess both the technical feasibility, and above all the environmental sustainability of an effective process enabling the recycling of polyamides-polyethylene multilayers packaging films. The technique used for the separation of the polymers is based on a selective dissolution, carried out using monoethylene glycol. The experimental tests made possible to identify the best conditions for treating the films and for maximizing yields and final products quality. The Life Cycle Assessment of the recycling process modelled at an industrial level, firstly allowed to determine the main process hotspots (i.e. the energy consumption). The LCA analysis was then extended, examining the life cycle of polyamides-polyethylene films with different end-of-life treatments, i.e. incineration, energy recovery and recycling. The results showed that the recycling process, carried out through the selective dissolution of the films, allows to reduce the overall environmental impacts of these materials along their life cycle. Therefore the recycling process analyzed here can be considered an effective approach to increase environmental sustainability and recovery of raw materials.

**Keywords:** Plastic recycling; Packaging; Life Cycle Assessment; Selective dissolution

## 1. Introduction

Since the beginning of the 20th century, with their introduction into the global market, plastics have gained a predominant position over many other materials. To the detriment of their excellent mechanical, thermal and physical properties, these materials have characteristics that, however, can undermine their use in a context of environmental sustainability. First of all, plastics are mainly made from non-renewable sources, mainly based on fossil fuels [1] and have a strong negative impact upon their final disposal. In particular, plastic packaging is perceived as an environmental problem, a source of waste and it has a strong impact on climate change [2]. One of the main solutions adopted to overcome these problems is represented by the improvement of the recycling efficiency of plastics; in fact, the recovery and the recycling processes can push towards a reduction of waste accumulations and, at the same time, allowing the re-introduction of materials in production chains [3,4]. According to some research work [5] closing the materials loop, already during the production process itself, has proved to be positive from an environmental point of view, as it allows for the reduction of burdens on various impact categories. As the plastic production has increased around the world since its introduction into everyday life [6], there is a strong concern about the environmental performance of these materials throughout their life cycle. Particular attention is paid to plastic materials exploited in the packaging sector. According to Plastic Europe, the vast majority of plastic producers would like mandatory legislation by the European Community setting a target of 30% recycled content for the production of plastic packaging [7].

Plastic packaging materials are based on various chemical compositions, have a short lifespan and a large production volume, all aspects that generate challenges for the end-of-life management [8]. Nevertheless, packaging allows to provide suitable mechanical stability and an adequate barrier against water vapour and oxygen, offering also the possibility to communicate with customers [9,10]. These features have been increasingly improved and modulated over time, thanks to the combination of multilayers made up of different plastic materials [11,12]. Given the chemical-physical properties, which make these materials resistant, moldable and with excellent barrier properties, the multilayer plastic packaging has become important for several applications and, already in 2017, it constituted around 26% by weight of the flexible packaging market [13,14]. However, products based on multilayer plastic packaging are still difficult to be recycled and this aspect, in addition to undermining the sustainability of these materials, slows the transition towards a circular economy. Main problems, which hinder a correct recycling path, are given by the fact that current standard technologies are unable to identify, select and easily separate the different layers that constitute these packaging materials [15]. Therefore, in the European countries, the main solution for the disposal of multilayer flexible packaging is represented by their incineration with energy recovery. Otherwise, in low-income countries, these wastes are sent to landfills [16]. According to Soares et al. [17] in the next ten years there will be a development of advanced technologies for the recycling of multilayer plastic packaging wastes; in detail, there are great expectations for the development of high-performance materials recycling technologies combined with advanced sorting technologies, together with an increase in the use of chemical recycling solutions, especially in high-income countries.

Nowadays, the recycling solutions for multilayer plastic packaging can be divided according to two main approaches: (I) detachment of the components making up the multilayer (through selective delamination or dissolution treatments); (II) processing of all layers together in one compatibilization step [17,18,19]. The first approach allows to separate the different materials, making them available for recycling in separate streams. On the contrary, compatibilization allows to treat the materials in one single flow without

separation, thanks to the addition of chemicals to increase the mechanical stability of the multilayers. An emerging recycling route, which does not require the separation of layers, consists in the pyrolysis treatment, which allows the production of petrochemical feedstocks such as naphtha or diesel [20,21].

The purpose of this study is to assess, from a technical and an environmental point of view, a simple but effective process enabling the recycling of a specific group of multilayer plastic packaging. Along the path towards greater environmental sustainability of human actions, it is of primary importance to evaluate and verify that the strategies adopted, such as greater use of recycling processes for the recovery of materials, actually allow to reduce the human footprint on the planet. For this reason, as has been done in this study, the analysis of new processes (e.g. recycling of polymeric materials), should be accompanied by precise assessments of the associated environmental impact. In detail, the analysed process allows the initial separation of multilayers consisting of a combination of polyamides (PA), linear low density polyethylene (LLDPE) and low density polyethylene (LDPE). The process allows to separate PA and PE with high efficiency and yield, allowing to obtain distinct polymers. The obtained polymers could then be used as they are as virgin materials (polyethylene and partially polyamides) or recycled through standard treatment methods. The separation treatment is carried out by exploiting a selective dissolution approach. As reported by Mieth et al. [12], this approach exploits the different solubility of polymers: a specific solvent allows to dissolve the targeted component and therefore gradually removes it from the system. The recycling process that is proposed in the present work is related to the separation of PA and PE layers from a multilayer plastic packaging. The industrial process enables the dissolution of PA into the solvent on which PE is remaining as a solid, that can be easily filtered, while PA can precipitate at lower temperatures. In details, the process can be divided into different steps: i) selective dissolution of PA; ii) filtration of the remaining PE and washing; iii) cooling of the solution containing PA; iv) PA filtration and washing. The filtration and washing steps allow the recovery of polymers (both PA and PE) together with the recovery of the solvent. Recovered solvent is then re-used in the process, improving the sustainability of the whole industrial process.
This work reports both the results of experiment carried out at laboratory scale and the analysis of the environmental impacts associated with the industrial process. The goal of the laboratory tests is to identify the best selective dissolution conditions (i.e. temperature and treatment times), in order to optimize both the separation yield and the quality of the obtained products (i.e. PA and PE). The best experimental conditions, determined at the laboratory level, were then used to model the separation treatment of PA-PE films at an industrial scale. These data were then used as a basis for the assessment of the environmental impacts, conducted with the Life Cycle Assessment (LCA) methodology. Results allowed not only to determine impacts associated with the specific process examined, but also to evaluate the effective sustainability of the recycling of these materials compared to other solutions for their disposal.

## 2. Material and methods

### 2.1 Experimental part

At the laboratory scale, selective dissolution tests were carried out on a multilayer plastic packaging based on PA-PE materials. The used materials are process industrial waste from B-Pack S.p.A., a well-known company leader in the production of multilayer flexible coextruded barrier films for the pharmaceutical/medical and food packaging sectors. For the experiments, the multilayer was first reduced into small flakes by using a two-shaft electrically operated shredder (7,5 kW with blades thickness of 10 - 30 mm), in order to improve the efficiency and processability of dissolution reactions.

Monoethylene glycol (MEG) (Sigma Aldrich, purity ≥ 99.5%) was used as a solvent for the selective dissolution treatment, allowing the separation of the two polymers, i.e. PA is dissolved in the reaction conditions into MEG, while PE is remaining as a solid film floating into the solution.
Selective dissolution experiments were carried out in a beaker heated by a hot plate under stirring. After a specified time interval, the PE polymer was separated from the solution by filtration, washed first in hot MEG at 95 °C and then in hot water at 90 °C. The PA was recovered from the MEG solution at the temperature of 35 °C by filtration on buchner. The PA was then dispersed in water at 90 °C in order to remove MEG residue and successively filtered. Both the PA and PE polymers were dried at the end of the process in order to remove the water remained after washing and weighed.
Preliminary tests were performed to check the dissolution of flaks of virgin PA in MEG to determine the minimum operating temperature value of 140 °C that enable the selective dissolution of PA while leaving PE as a solid. The identified temperature is confirmed in the literature [22]. In order to find the best reaction conditions in terms of polymer separation and process energy efficiency, studies were performed varying three parameters: the operating temperature (from 140 to 190 °C), the time of treatment (from a minimum of 30 minutes to a maximum of 120 minutes) and the ratio (R), in weight, of the amount of MEG used divided by the weight of PA-PE film immersed in the solvent.
The experimental conditions of the different dissolution tests are shown in Table 1. In these experiments, the lowest temperature of 140°C was used in order to limit the thermal impact of the process, but in this case, in order to obtain the optimal polymer separation, it was necessary to increase both MEG amount and reaction time. Therefore, different combinations of temperature and reaction times were tested in order to individuate the best processing conditions as described in detail in the experimental part.

**Table 1: Experimental conditions of PA-PE film selective dissolution carried out in monoethylene glycol**

| Experiment | T (°C) | Time (min) | R (MEG/film) |
|---|---|---|---|
| 1 | 140 | 40 | 15 |
| 2 | 190 | 30 | 10 |
| 3 | 160 | 40 | 10 |
| 4 | 160 | 60 | 10 |
| 5 | 160 | 90 | 10 |
| 6 | 160 | 120 | 10 |

The characterization of the separated polymers was carried out through infrared spectroscopy measurements with an Agilent 630 FT-IR spectroscope in attenuated total reflection (ATR).

### 2.2 Life Cycle Assessment

LCA is a well-established methodology that allows to evaluate the environmental impacts associated with a product, a process or a service along its entire life cycle. According to ISO standards [23,24], LCA consists of four phases, including goal and scope definition, life cycle inventory (LCI), life cycle impact assessment (LCIA), and interpretation of the results. The whole analysis was carried out using the SimaPro 9.2 software and the Ecoinvent v.3.7.1 database. The treatment of complete separation of 1 kg of PA-PE packaging material was set as functional unit (FU) for the assessment of the environmental burdens of the process.

In the goal and scope definition phase, the object of the study, the system boundaries and the functional unit must be adequately defined [25]. As mentioned above, the aim of the study is to evaluate the environmental burdens generated during the separation process of PA-PE multilayers packaging materials. In addition, to make the analysis more informative, a comparison was carried out on different solutions for

the end-of-life of these materials. The treatment of complete separation of 1 kg of PA-PE packaging material was set as functional unit (FU) for the assessment of the environmental burdens of the process.

In detail, the separation process is described considering monoethylene glycol as solvent. At a certain temperature (around 160 °C), the solvent begins to solubilize the polyamides. By removing the solvent in the reaction solution, it is therefore possible to remove the polyamide and obtain polyethylene alone as a product. Subsequent washes with MEG and water allow to recover all the polyethylene present in the initial film. The polyamide is instead recovered by reducing its solubility in the MEG by simply lowering the temperature of the solution. The process generates as waste: monoethylene glycol (lost along the steps of the process), wastewater and emissions of volatile organic compounds (VOC). MEG and water can be partially recovered through distinct distillation processes.

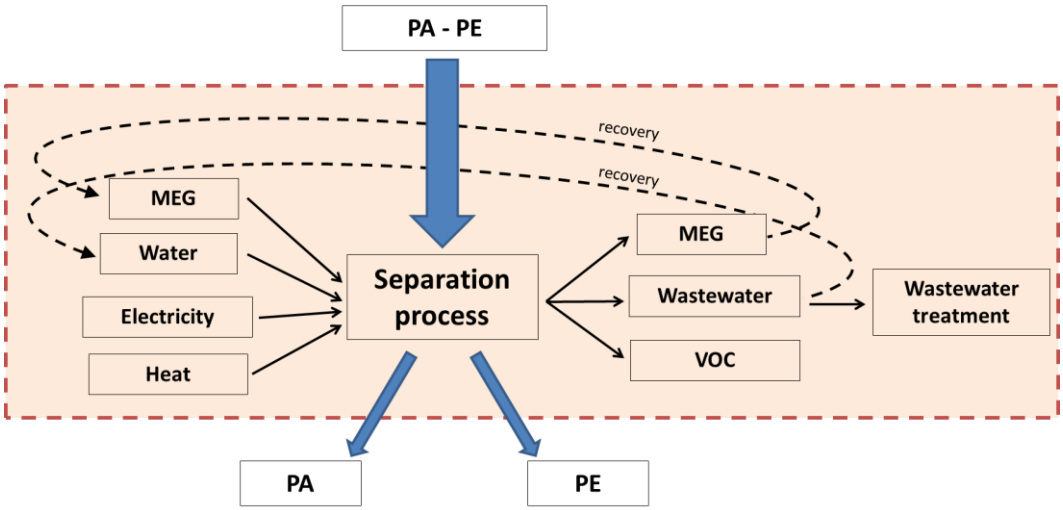


**Figure 1 Scheme of the materials and processes that allow the separation process of PA-PE packaging; the box indicates the system boundary for the LCA.**

Figure 1 shows a scheme of the separation process, with an indication of the system boundary considered during the environmental analysis; only the burdens of the materials and processes enclosed in the orang box have been considered during the Life Cycle Assessment. As shown in Figure 1, the process uses MEG as a solvent for selective dissolution; the water is used in the washing and purification of the two separate polymers. At the end of the selective dissolution process, both the MEG and the water are recovered and reused, thanks to distillation processes after which they are inserted again as solvents for the separation.

An inventory of inputs and outputs (i.e. resources, materials, emissions, waste, etc.) was compiled to create a representative model of the system investigated. The data used to model the system (see Table 2) were estimated for an industrial application (i.e. treatment of 1 ton of multilayer film per hour); this estimate was based on initial tests carried out both at the laboratory and at the pilot plant level.

**Table 2: Inventory of the materials, energies and emissions used to model the separation process (treatment of 1 ton of PA-PE film per hour).**

| INPUT | Amount | Unit of measurements |
|---|---|---|
| MEG | 23.29 | kg |
| Water | 1.351 | kg |
| PA - PE film (ratio PA:PE =30:70) | 1.000 | kg |
| Electricity | 180.00 | kWh |
| Heat | 0.976 | kWh |
| **OUTPUT** | **Amount** | **Unit of measurements** |

| | | |
|---|---|---|
| MEG | 23.0 | kg |
| Wastewater (to distillation process) | 1.350 | kg |
| VOC | 0.50 | g |
| PA | 300.0 | kg |
| PE | 700.0 | kg |

A plant lifespan of 20 years has been estimated for a total of 40000 working hours. The quantities reported as inputs take into account both the contribution of solvent, always present in the plant (thanks to the distillation recirculation), and the amount of solvent added every hour for the treatment of 1 ton of PA-PE film. In fact, 11400 kg of MEG are constantly present in the plant, which are recovered by distillation and recirculated. Of these 10000 kg of MEG take part in the solvolysis reaction, the remaining 1400 kg are used for washing the polyolefin. For each hour of the process it is necessary to add 23 kg of MEG to replace those lost during solvolysis. The amount of MEG, indicated in Table 2 as input, it is thus composed of the quantity of MEG that is always present in the plant, normalized for the total operating hours of the plant along its life cycle and of the contribution of MEG that must be added every hour of processing (i.e. 23 kg). According to this approach, the quantity of MEG constantly recirculated in the plant and allocated for a single hour of use is 0.29 kg/hours. A similar approach applies to the quantity of water, in fact there are approximately 2599 kg of water in the plant, 1249 of which are always recycled (i.e. 0.031 kg of water allocated considering the total life of the plant) instead 1350 must be added every hour of the process. The electrical energy input, for the operation of the plant (i.e. pumps, stirrers and cooling systems), was modelled using electricity data obtained from the Italian national grid (Ecoinvent dataset: Electricity, medium voltage {IT} | market for | Cut-off, U). The input of heat includes both the contribution of energy used to heat the reactor during the dissolution process and the energy required for the distillation processes of MEG and water. The wastewater generated at the end of the process is sent to a small treatment plant, modelled using the Ecoinvent dataset: Wastewater, average {Europe without Switzerland}| treatment of wastewater, average, capacity 1E9l/year | Cut-off, U.

The third phase of LCA aims at quantifying the environmental impacts associated with the inputs and outputs data used to model the inventory. The environmental indicators, evaluated at midpoint level using the CML-IA baseline method [26,27] are: Global warming (GWP100a), Ozone layer depletion (ODP), Photochemical oxidation, Acidification, Eutrophication, Abiotic depletion, Abiotic depletion (fossil fuels).
Firstly, the impact analysis has been carried out on the separation process according to the system boundary identified in Figure 1. This analysis allowed to highlight the main contributions to the impact for each category examined. Subsequently, for a better understanding of the burdens of suggested recycling process along the life cycle of PA-PE multilayer films, the analysis was enriched by comparing it with different end-of-life (EoL) solutions (i.e. incineration and incineration with energy recovery). As shown in Figure 2, to assure comparability among considered recycling technologies, the system boundaries for this second part of the analysis include: polymers production, film extrusion, EoL treatment and environmental credits. These credits correspond to recovered materials obtained at the end of the recycling process. By means of the substitution approach, these materials are credited as “avoided products” [28]. The use phase and the steps of waste collection and pre-treatment (i.e. sorting, cleaning) are not included in the analysis.
The datasets present in the Ecoinvent database were used to model the production of PE, PA and the plastic film extrusion. To model the PE, an equal mixture in weight of LLDPE and LDPE has been considered. The PE incineration process was employed as a proxy to represent the entire multi-plastic film. In the case of incineration with energy recovery, the quantities of electricity and heat released during the incineration of PE were added. The recycling process, carried out by selective dissolution process, was modeled using

the previously described data. The contribution of the avoided products was modeled assuming that, respectively, 70% and 30% of the PE and PA, recovered from recycling, could be used as virgin materials.

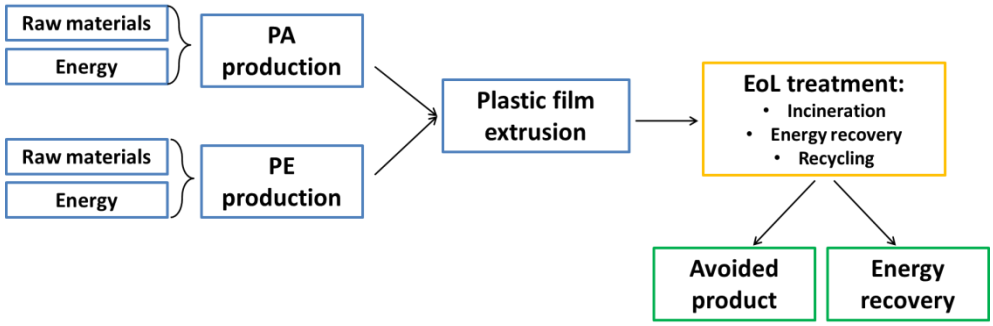


**Figure 2 System boundary for the comparative analysis of different end-of-life solutions. The blue boxes indicate the production phases; the yellow box indicate the EoL treatment (three different solutions are evaluated: incineration, incineration with energy recovery and recycling); the green boxes represent the environmental credits generated through the energy recovery and recycling solutions.**

## 3. Results and discussion

### 3.1 Experimental part

Table 3 shows the summary of the experiments performed to determine the best selective dissolution conditions for separating PA-PE films. The table shows the amount of separate PA and PE polymers (amount reported as % by weight) for each of the experiments performed.

**Table 3: PA and PE conversion (%) obtained after selective dissolution in MEG under different experimental conditions.**

| Experiment | PA % (gravimetric determination) | PE % (gravimetric determination) | Tot % (gravimetric determination) |
|---|---|---|---|
| 1 | 28.4 | 71.5 | 99.9 |
| 2 | 29.6 | 70.4 | 100.0 |
| 3 | 28.9 | 69.4 | 98.3 |
| 4 | 29.7 | 70.2 | 99.9 |
| 5 | 29.5 | 70.5 | 100.0 |
| 6 | 30.1 | 69.6 | 99.7 |

Figure 3 reports the IR spectra of the PE separated fractions after each experiments to test the different conditions for the dissolution process.

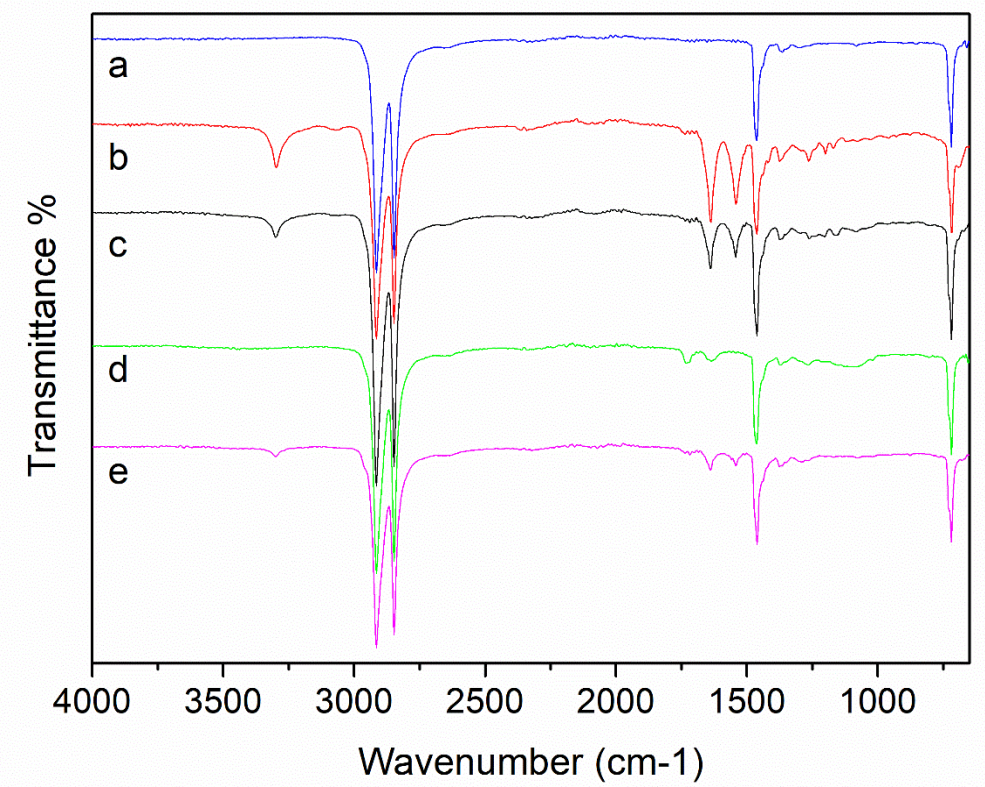


**Figure 3 IR spectra related to separated PE after the following processes: a) virgin PE, b) experiment n°1, c) experiment n°3, d) experiment n°4, e) experiment n°2.**

Experiment n°1, performed at 140 °C, 40 min and R MEG/film = 15, showed that PE % yield obtained is greater than 70 % because not all the PA is able to go into solution. The IR spectrum (Fig. 3 b) shows the presence of PA identified by the characteristic peaks at 3300 $cm^{-1}$, associated with amide N-H stretching, and C=O amide stretching to the value of 1630 $cm^{-1}$.

Increasing the temperature up to 190 °C and reducing the reaction time to 30 min, the PA yield became 30.2 % (experiment n°2, Table 3). The PA dissolved into the solution increased and, at the same time, the quantity remained adherent to PE decreased compared to experiment n°1. This can be seen by comparing the peaks at 3300 and 1630 $cm^{-1}$ of Fig. 3 e and Fig. 3 b. The % PE is greater than 70 % because, at high temperatures, the PE tends to agglomerate, incorporating a high quantity of MEG, that is hardly washed away in the following processing phases.

Excellent reaction yields have been obtained in experiment n°3 (Table 3 and Fig 3 c) even if the temperature was decreased to 160 °C and the reaction time was increased from 30 to 40 minutes. Moreover, going to 60 minutes in experiment n°4 (Table 3 and Fig. 3 d), by maintaining T=160 °C, a further increase in yields was observed. Experiments n°5 and n°6 showed how the yield of PA% increases with increasing reaction times, at the same temperature value of 160 °C. However, the increased reaction time does not significantly improve the efficiency of the process. Therefore, the best reaction conditions have been defined as those determined in experiment n°4, as confirmed by the absence of extra peaks in the corresponding IR spectrum (Fig. 3 d).

### 3.2 Life Cycle Assessment

The selective dissolution process for the separation and recycling of PA – PE packaging films was analysed, with the aim to highlight the main environmental hotspots. Figure 4 shows the percentage impact for each category broken down by input/output contributions to the overall process. The value for each process contribution is reported as a percentage of the total value obtained at the level of characterization for each category analyzed. In turn, the total numerical value, for each category, at the characterization level, is reported with the specific units of measurement in Table 5. For all the impact categories, with the exception of Abiotic depletion, the energy demand, both as electricity and heat, is the main responsible for the impacts. In particular, heat consumption is responsible for more than half of the burdens for various categories: Global warming, Photochemical oxidation, Ozone depletion and Abiotic depletion (fossil fuels). The impact associated with the electricity is particularly significant for the categories Acidification and Eutrophication, corresponding to, respectively, 42% and 38% of the overall impact. The third cause of impact is the use of monoethylene glycol, which determines the main impact for the category Abiotic depletion (53% of the overall impact). Wastewater treatment generates significant impacts (about 9%) only for the Eutrophication category.

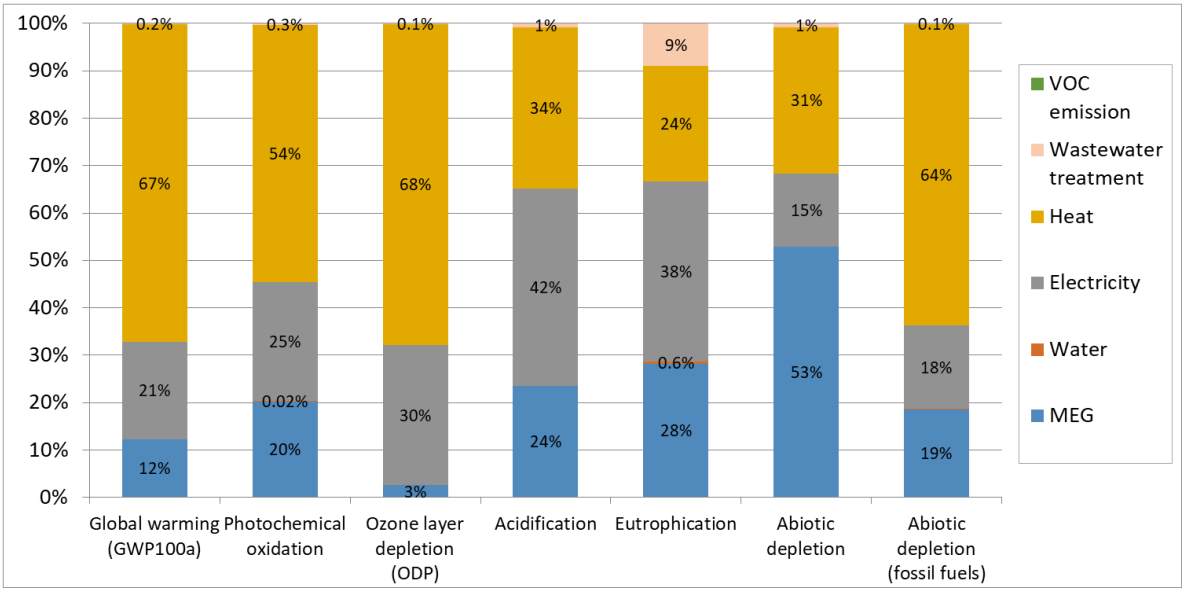


**Figure 4 The absolute environmental impacts generated by the selective dissolution process of PA-PE films, i.e. the separation and recycling process of PA-PE; the impact for each category is split into the contribution of the inputs and outputs to the process.**

Table 4 reports the numerical value of the impacts determined for the selective dissolution process of 1 kg of PA-PE films for packaging. The table shows both characterisation and normalized results. The normalization scores for each category are those used by the CML-IA method for the EU 25 reference situation. It can be observed that the impacts for the categories Photochemical oxidation and Ozone layer depletion (ODP) are one or two order of magnitude smaller than the others, therefore the general burdens on these categories are very limited. The most affected category among those analyzed is Abiotic depletion (fossil fuels); this result is due to the considerable amount of energy required by the dissolution process and also by the thermal energy spent to recover the solvents by distillation. A hypothetical decrease in environmental impacts could be obtained by replacing the use of electricity taken from the Italian national grid with electricity generated from renewable sources. Even a further optimization of heat exchanges could further reduce environmental impacts.

**Table 4: Numerical environmental impacts, at characterisation and normalization level, generated by the selective dissolution process of 1 kg of PA-PE packaging films.**

| Impact category | Unit | Characterisation | Normalization (EU 25) |
|---|---|---|---|
| **Global warming (GWP100a)** | kg $CO_2$ eq | 0.38 | 7.64E-14 |
| **Photochemical oxidation** | kg $C_2H_4$ eq | 5.72E-05 | 6.75E-15 |
| **Ozone layer depletion (ODP)** | kg CFC-11 eq | 3.24E-08 | 3.63E-16 |
| **Acidification** | kg $SO_2$ eq | 7.09E-04 | 2.52E-14 |
| **Eutrophication** | kg $PO_4^{---}$ eq | 2.02E-04 | 1.53E-14 |
| **Abiotic depletion** | kg Sb eq | 1.08E-06 | 1.27E-14 |
| **Abiotic depletion (fossil fuels)** | MJ | 5.86 | 1.86E-13 |

The results reported in Figure 5 show the comparison between the life cycle of 1 kg of PA-PE packaging films by testing different end-of-life solutions. The comparison among incineration, incineration with energy recovery and recycling is reported for seven impact categories. The environmental burdens are split between the contribution of the polymers production and film extrusion, EoL treatments and possible environmental credits. For a detail of the numerical values of the impacts, see Table A1 in the Appendix section.

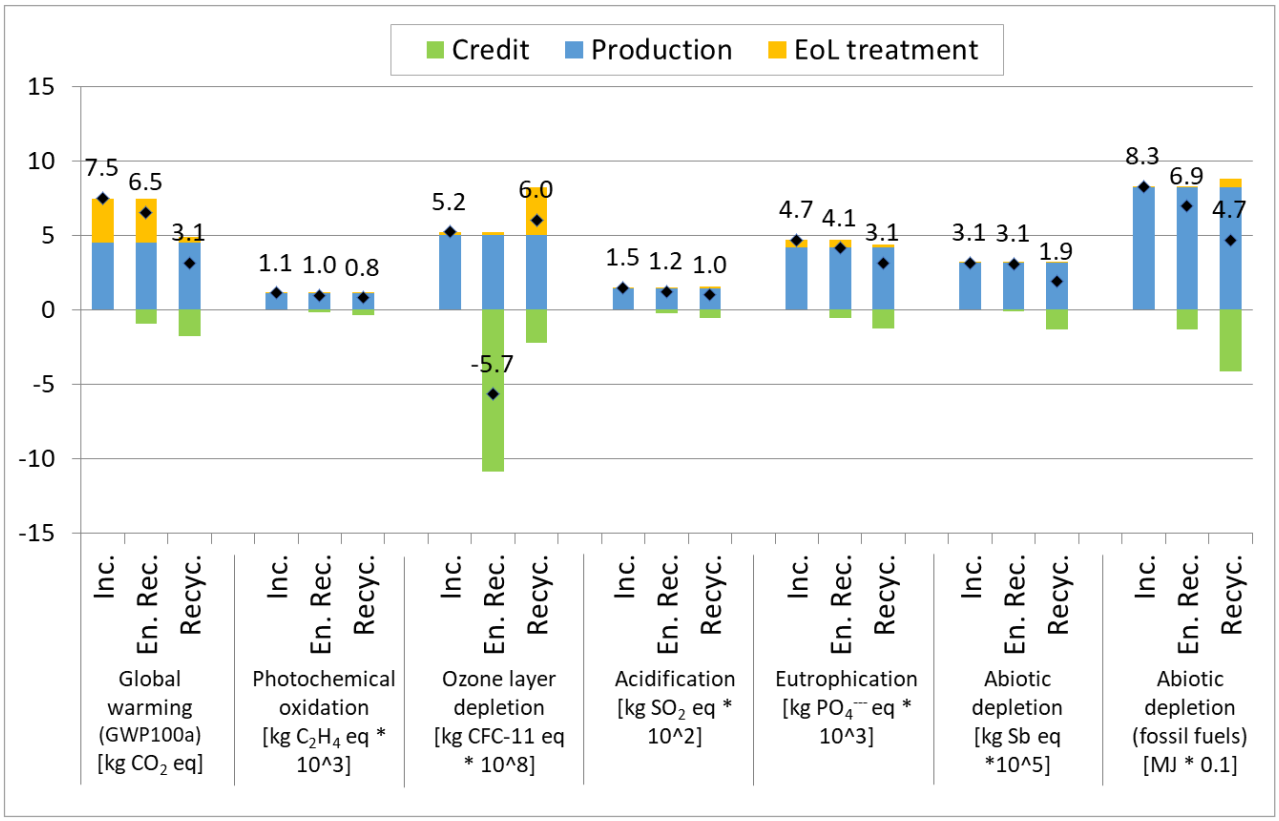


**Figure 5 Environmental impacts associated with the Production and EoL stages of 1 kg of PA-PE packaging films, comparing three different EoL solutions: incineration, incineration with energy recovery and recycling through a selective dissolution process. The analysis took into consideration polymers and film production, EoL treatment and environmental credits, which are visualized as negative values. Numerical values of the impacts associated with the life cycle of PA-PE packaging films were added: details are reported in Table A 1.**

The impacts related to the Global warming (GWP 100a) indicator reported in Figure 5 show that the recycling solution, represented by the selective dissolution process analysed in this study, significantly reduces the overall impact of these materials along their life cycle. In fact, the possibility of recovering and reusing secondary polymers, as virgin material, generates a consistent positive effect on the environment.

The incineration process generates a marked emission of $CO_2$ eq. into the atmosphere, with a value similar to that generated during the production phase of plastic films. The carbon dioxide emission is only partially offset by the energy produced by the combustion of the packaging films in the case of energy recovery.
In the case of the photochemical oxidation indicator (Fig. 5) the main burdens are generated during the production phase of the materials, explaining why the solution represented by recycling, with recovery of polymers, allows to reduce the overall impact. It should be noted that in this case the impact generated during the selective dissolution process is greater than that of incineration. As already shown in Figure 4 this environmental impact is strongly correlated with the amount of energy consumed by the recycling process.
The Ozone depletion potential (ODP) indicator (Fig. 5) shows a different trend among the compared cases. The solution represented by incineration with energy recovery is the best EoL treatment for the final disposal of PA-PE packaging films. This result is due to the replacement of the energy produced according to the Italian national mix, strongly based on natural gas, with electricity and heat obtained from the combustion of polymers. Looking at the impact generated by the selective dissolution process, it can be seen that even the normal incineration process, without any type of environmental credit, is slightly better than the recycling solution.
As for the impact on the category Acidification reported in Figure 5, the recycling process, despite the highest burdens of the EoL treatment, prove again to be the better solutions among the compared ones. The possibility to recover polymers and use as new materials allows to limit the overall impact along the life cycle of the PA-PE films.
The Eutrophication indicator (Fig. 5) shows how the incineration process generates a greater impact than selective dissolution. It can also be observed how the environmental credits, obtained thanks to the recovery of materials, allow to further reduce the burdens on this impact category.
The impact on the Abiotic depletion indicator (Fig. 5) is strongly dominated by the burdens generated during the production phase. The analysed EoL solutions does not show marked impact and again the recycling process represent the best treatment to reduce the environmental impact of PA-PE films along the life cycle.
The second indicator that evaluates the depletion of resources, i.e. the category Abiotic depletion (fossil fuels) (Fig. 5), shows how the recycling process allows to halve the environmental impact respect to the simple incineration process. Even if incineration causes very limited environmental burdens, simple energy recovery is not sufficient to reduce the impact generated during the production of PA-PE films.
In general terms, for basically all categories, it can be noted that the production phase is the main responsible for the impacts along the life cycle of PA-PE packaging films. Therefore, the possibility of recovering the polymers and reusing them as virgin materials through a new cycle, is the best solution to reduce environmental impacts. Others end-of-life solutions were not taken into consideration since nowadays this class of materials it is collected, separated from other plastic materials and mainly send to incineration or landfill. Landfilling was not analyzed in the comparison as an end-of-life solution for materials; as it represents the last desirable solution according to the hierarchy recognized by the European community [29]. Solutions considered innovative such as pyrolysis were not taken into consideration as it was not possible to collect data that would allow valid modeling.

## 4. Conclusions

This study examined both the efficacy and environmental impacts, hence environmental sustainability, of a selective dissolution process applied to enable recycling of PA-PE packaging films. Main conclusions can be summarized as follow:

- Selective dissolution test allowed to determine the best condition for the process: T = 160°C, t = 60 min, R MEG/film = 10, with an excellent separation ratio of the two polymers.
- The detail LCA analysis of the selective dissolution process applied to PA-PE packaging films highlight that the main contribution to the process, for all impact categories, is due to energy consumption.
- The comparison of different EoL solutions was carried out by taking into account the life cycle of PA-PE packaging films, by considering the contribution of: production phase, EoL treatments and possible environmental credits. The results showed that for all the analysed categories, with the exception of Ozone layer depletion, the recycling solution allows to reduce the environmental impact of this specific type of multilayer packaging. The main contribution to the impact reduction derives from the possibility to recover polymers and used them as virgin materials. The approach of recycling plastic materials allows to obtain the greatest reduction in impacts (compared to the worst EoL solution) for the categories: Global warming, with a reduction of 58%, Abiotic depletion, with a reduction of 40% and Abiotic depletion (fossil fuels) with 43% of impact reduction.

As a general comment the selective dissolution process can be considered a viable and sustainable solution for the treatment of PA-PE packaging films at the moment of their disposal. Further studies should focus on expanding the type of multilayer plastic packaging investigated and at the same time conducting a better investigation of techniques/solutions to remove such materials from the stream leading to incineration or landfill.

**Acknowledgments**

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## Appendix A

**Table A 1 Numerical values of the impacts associated with the life cycle of PA-PE packaging films, comparing three different EoL solutions: incineration, incineration with energy recovery and recycling through a selective dissolution process. The analysis considers production, EoL treatment and environmental credits.**

| Impact category | EoL solution | Production | EoL treatment | Credit | TOTAL |
|---|---|---|---|---|---|
| **Global warming (GWP100a) kg $CO_2$ eq** | Incineration | 4.49 | 2.99 | / | 7.48 |
| | Energy Recovery | 4.49 | 2.99 | -0.96 | 6.53 |
| | Recycling | 4.49 | 0.38 | -1.74 | 3.13 |
| **Photochemical oxidation kg $C_2H_4$ eq** | Incineration | 1.12E-03 | 9.02E-06 | / | 1.12E-03 |
| | Energy Recovery | 1.12E-03 | 9.02E-06 | -1.51E-04 | 9.74E-04 |
| | Recycling | 1.12E-03 | 5.72E-05 | -3.64E-04 | 8.09E-04 |

| | | | | | |
|---|---|---|---|---|---|
| **Ozone layer depletion (ODP) kg CFC-11 eq** | Incineration | 5.01E-08 | 2.03E-09 | / | 5.22E-08 |
| | Energy Recovery | 5.01E-08 | 2.03E-09 | -1.09E-07 | -5.67E-08 |
| | Recycling | 5.01E-08 | 3.24E-08 | -2.22E-08 | 6.03E-08 |
| **Acidification kg $SO_2$ eq** | Incineration | 1.46E-02 | 2.26E-04 | / | 1.49E-02 |
| | Energy Recovery | 1.46E-02 | 2.26E-04 | -2.59E-03 | 1.23E-02 |
| | Recycling | 1.46E-02 | 7.09E-04 | -5.41E-03 | 9.94E-03 |
| **Eutrophication kg $PO_4^{---}$ eq** | Incineration | 4.17E-03 | 5.18E-04 | / | 4.69E-03 |
| | Energy Recovery | 4.17E-03 | 5.18E-04 | -5.58E-04 | 4.13E-03 |
| | Recycling | 4.17E-03 | 2.02E-04 | -1.22E-03 | 3.15E-03 |
| **Abiotic depletion kg Sb eq** | Incineration | 3.13E-05 | 5.92E-08 | / | 3.14E-05 |
| | Energy Recovery | 3.13E-05 | 5.92E-08 | -6.88E-07 | 3.07E-05 |
| | Recycling | 3.13E-05 | 1.08E-06 | -1.34E-05 | 1.90E-05 |
| **Abiotic depletion (fossil fuels) MJ** | Incineration | 82.43 | 0.12 | / | 82.55 |
| | Energy Recovery | 82.43 | 0.12 | -13.08 | 69.47 |
| | Recycling | 82.43 | 5.86 | -41.58 | 46.71 |

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