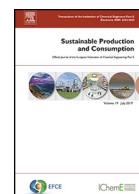




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Can plastics from end-of-life vehicles be managed in a sustainable way?

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ABSTRACT

Plastic from end-of-life vehicles (ELVP) are currently managed in European Union without any attention to polymer recovery. The study analyses novel treatments of sorting, dissolution/precipitation, extrusion, catalytic pyrolysis, and plastic upgrading, which could contribute to define a sustainable ELVP management scheme. The environmental performances of each of these treatments have been quantified by an attributional Life Cycle Assessment, allowing to compare a possible innovative recycling scheme with that of the European currently adopted options. The new scheme greatly enhances ELVP management performances, by hugely increasing annual amounts of polymers sent to recycling (from 26 kt/y up to 509 kt/y), drastically decreasing residues to be sent to combustion or landfill (from 984 kt/y down to 232 kt/y), and improving the impact of main environmental categories. Carcinogens, Non-Carcinogens, Global Warming and Non-Renewable Energy reduce of 138%, 100%, 42% and 114%, with reference to the current scenario. These promising results are mainly related to the utilisation of a dissolution/precipitation process (Crea-Solv®), whose introduction could allow recovering large part of target polymers (PE and PP). The recovery of PE in fuel tanks by a supercritical extrusion process (Extruclean) and the treatment of residues and non-target polymers by a catalytic pyrolysis process also contribute to improve the environmental performances. A sensitivity analysis quantifies the role of some key parameters, indicating that the results could be affected by energy consumption of dissolution/precipitation process, oil yield of catalytic pyrolysis treatment, but also by the substitutability factor utilised to quantify the avoided burdens associated to the recycled polymers.

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1. Introduction

Every year in Europe about 6 million of vehicles reach their end-of-life and are deregistered through official schemes (Eurostat Database, 2020), becoming end-of-life vehicles (ELVs). The treatments of this waste stream are regulated by the Directive 2000/53/EC, which establishes a minimum reuse and recycling rate of 85% of vehicle total weight and a minimum reuse and recovery rate of 95% of the same total weight (EC, 2000). To satisfy these ambitious targets, current management schemes focus mainly on recovery of metals (Stena Metall AB, 2020), which are about 70% of ELVs weight and easily resold to companies of the metallurgical sector (EC-JRC, 2018). However, the use of plastic materials is always more widespread in automotive sector, for

important environmental and economic aspects. Their low density reduces car weight, helping to minimise fuel consumption and gas emissions, while their well-known flexibility and durability allow many design solutions, saving production and maintenance costs (EC, 2020). A vehicle is made up for about 15%_w of its total mass of plastics (Schönmayr, 2017), hence their recovery and recycling could remarkably enhance the current EU Plastics Strategy (EC – European Commission 2018a). On the other hand, the management schemes for plastic fraction of ELVs (here indicated as ELVP) are complex and non-harmonised among different countries (Ramboll Deutschland GmbH 2020). They commonly include dismantling (depollution), shredding, and, only sometimes, post-shredding treatment (PST). The latter is commercially available but still not common in the ELVP management practices, which usually send non-metal fractions directly to energy recovery (EC, 2020) for two main reasons. First, ELVP are strongly commingled with other fractions, thus their recovery is technically possible only after a series of complex sorting steps. Moreover, the recovery of the different polymers used in ELVP cannot be achieved by conventional

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List of Acronyms

ABS	Acrylonitrile–Butadiene–Styrene
APC	Air Pollution Control
ARN	Autorecycling Nederland
ASR	Automotive Shredder Residue
BAT	Best Available Technologies
BAT-AEPLs	BAT Associated Environmental Performance Levels
BFR	Brominated Flame Retardant
BREF	BAT Reference document
CHP	Combined Heat and Power
daf	Dry and Ash Free
DecaBDE	Decabromodiphenyl ether
EC	European Commission
ELV	End-of-Life Vehicle
ELVP	Plastics from End-of-Life Vehicles
EU	European Union
LCA	Life Cycle Assessment
LCI	Life Cycle Inventory
LCIA	Life Cycle Impact Assessment
MFA	Material Flow Analysis
PA	Polyamide
PBDE	Polybrominated Diphenyl Ether
PBT	Persistent, Bioaccumulative, and Toxic
PC	Polycarbonate
PE	Polyethylene
PET	Polyethylene terephthalate
PMMA	Poly Methyl Methacrylate
PP	Polypropylene
PS	Polystyrene
PST	Post-Shredding Treatment
PUR	Polyurethane
PVC	Polyvinyl Chloride
REACH	Registration, Evaluation, Authorisation and Restriction of Chemicals
sc-CO ₂	Supercritical carbon dioxide
SFA	Substance Flow Analysis
SHF	Shredder Heavy Fraction
SLF	Shredder Light Fraction
VF	Variation Factor
VOC	Volatile Organic Compound
vPvB	very Persistent, very Bioaccumulative
WEEE	Waste of Electrical and Electronic Equipment
WtE	Waste-to-Energy

mechanical recycling (Ragaert et al., 2017), due to the presence of additives – such as brominated flame retardants (BFRs) (Strååt and Nilsson, 2018; Mehlhart et al., 2018), plasticisers, stabilisers (including heavy metals) (Wagner and Schlummer, 2020), glass fibres (Gallone and Zeni-Guido, 2019) – and contaminants, such as volatile organic compounds (VOCs), fuels, non-plastic materials and oil residues. However, resource recovery from ELVP could provide important economic and environmental advantages (Gallone and Zeni-Guido, 2019; Ciacci et al., 2010).

This study describes novel treatments of sorting, dissolution/precipitation, extrusion, catalytic pyrolysis, and plastic upgrading, which can be combined together in a new ELVP management scheme. The environmental performances of the current management scheme in Europe have been quantified and compared with those of the new management scheme, by means of the standardised and holistic tool of environmental life cycle assessment (LCA). The implemented LCA refers to the annual generation and average composition of ELV plastics in Europe, and is supported by specific

Material and Substance Flow Analyses (MFA/SFA) to exhaustively describe the system under analysis and to quantify the maximum amounts of materials obtainable from officially collected ELVP in the current and proposed management schemes. The reliability of the obtained results is increased by the high-quality of data used, which mainly come from 12 research centres and companies active in the ELV plastics recycling sector. All of them operate together in the framework of a Horizon 2020 project, called Nontox (Nontox Project, 2020), which aims at validating new and sustainable treatments for ELVP management and suggesting reliable alternative options for European recyclers.

2. Current European management scheme and some innovative treatments

Fig. 1 reports the management pathways of the current European management scheme of ELVP, and those of a proposed new scheme, indicated as Innovative. The first takes into account that a large amount of ELVP cannot be treated by mechanical recycling and is disposed of by means of thermal treatments or landfilling (Mehlhart et al., 2018; Eurostat Database, 2020; UNEP, 2017). The proposed new scheme involves novel treatments of sorting (based on density and optical differences), dissolution/precipitation (Crea-Solv®), extrusion – either with (Extruclean) or without (Modix) supercritical CO₂ (sc-CO₂) –, catalytic pyrolysis, and plastic upgrading. An essential description of the processes is given in the following sections while technical details are provided in the life cycle inventory (LCI) paragraph.

2.1. Current European management options for ELV plastics

2.1.1. ELV dismantling

The ELV dismantling provides a first separation of the plastic fraction. This stage is also known as depollution, as it comprises the separation of hazardous substances and liquids such as batteries, brake fluids and fuels (Cossu and Lai, 2015; EC – JRC, 2018). Other exit streams are represented by engines, tyres, metals, glass and everything else that can be easily manually removed (stream F3 of Fig. 1). Some functional components can be separated for reuse and sold as second-hand spares (stream F2 of Fig. 1), including large plastic parts, which can derive from bumpers, hubcaps, grilles, but also PUR parts, back lights (usually made of PMMA), seatbelts (generally made of PA) and others (Leslie et al., 2013). A not-negligible amount of plastic is separated during this stage and is directly sent to plastics re-manufacturing, thermal treatments or landfill (streams F4, F5, and F6 of Fig. 1), with a partitioning varying from country to country (Eurostat Database, 2020). In some European countries, as the Netherlands, the amount of plastic separated for reuse during dismantling is very high (up to 21% of total plastics) (Leslie et al., 2013), but in these cases there are always site-specific economic incentives (EC, 2020). In the rest of Europe, most of plastics fractions are shredded and then end up in the so-called automotive shredder residues (ASR) (Ciacci et al., 2010; Cossu and Lai, 2015). The developed LCA assumes, for all the analysed scenarios, the average shares reported by Eurostat Database (2020) for the separation of plastics for reuse, re-manufacturing, shredding or disposal by thermal treatment or landfill.

2.1.2. ELV shredding

During the shredding stage, the depolluted ELVs are represented by stripped car wrecks (stream F7 of Fig. 1), which are shredded in smaller pieces (ASR) and separated based on their composition. Three streams typically arise from this process: ferrous metals (approximately 65–75%_w of the shredder feed); shredder heavy fraction (SHF), mainly composed by non-ferrous metals but also con-

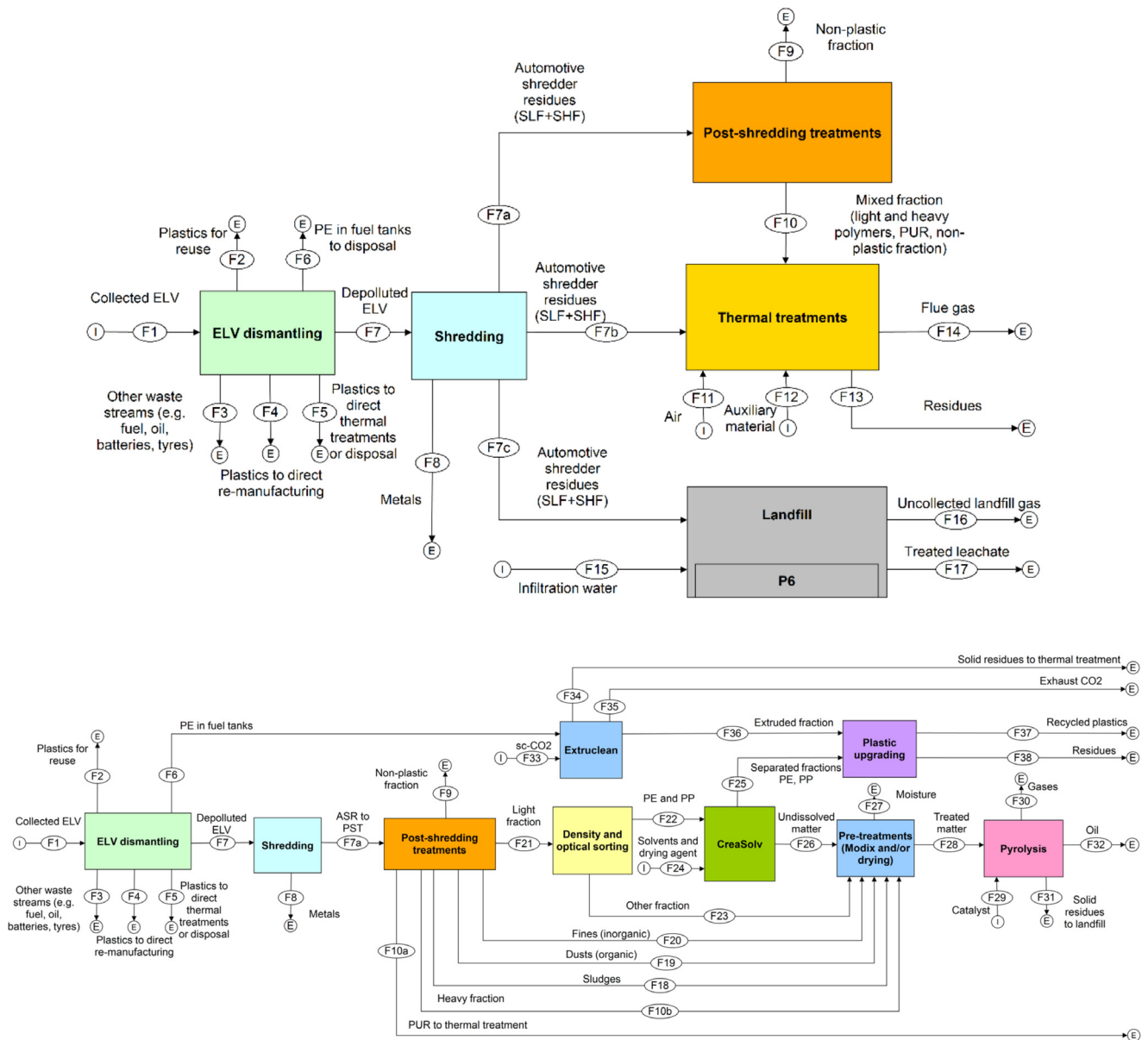


Fig. 1. Management pathways for ELV plastics in the Current (top) and Innovative (bottom) management schemes. I = import stream; E = export stream.

taining wires, high density plastics and glass (5–10%_w of the material supplied to the shredder); the remaining shredder light fraction (SLF, also known as fluff), which contains light metals, paper, plastic, foam, textile, wires, wood and others (Cossu and Lai, 2015; EC – JRC, 2018; Nontox Project 2020). They are sent to a post-shredding treatment (PST) to provide a further separation and recovery of the fractions that make up SHF and SLF, especially metals, which represent the stream with the higher added value and mass fraction (EC – JRC, 2018; Eurostat Database, 2020). Sometimes the PST stage is integrated in the shredding phase, otherwise ASR is sent to a specific PST plant. In most of EU countries, after the stage of metal recovering, almost all shredding residues are no further separated and directly sent to disposal (Mehlhart et al., 2018; Eurostat Database, 2020; UNEP, 2017). Accordingly, in the current management scheme, some of the fractions containing plastics are directly sent to thermal treatments (stream F7b of Fig. 1 top) or landfills (stream F7c of Fig. 1 top), with related annual mass flow rates quantified based on the percentages reported by Eurostat Database (2020).

2.1.3. Post-shredding treatment

The post-shredding treatment can occur in a specialised facility for ASR (as it happens in the Autorecycling Nederland (ARN) system (ARN, 2020)) or in a generic sorting facility, where wastes from ELV sector can be commingled with wastes from other sectors, such as those from electrical and electronic equipment (WEEE). This is aimed at complying with the recycling quotas established by European directives or landfill ban (in the countries where it is applied), or simply at increasing the economic incomes by selling materials, especially metals (EC-JRC, 2018). In the PST plants, the support of different processes - such as eddy currents/magnetic separation, gravimetric separation and others - allows a sorting mainly aimed at recovering of metals: in some cases, a plastics recovery stage is present, generally utilising a density-based separation (Buekens and Zhou, 2014; EC-JRC, 2018). This allows sending the different polymers to the most appropriate treatment process. Nonetheless, most of the companies are nowadays focused just on metals recovery, and plastic streams are usually not sent to mechanical recycling since they show a high rate

of contamination (wood, rubber, PUR and textiles), which requires the application of complex and costly sorting stages (Stena Metall AB, 2020). In the analysed current European management scheme, it is assumed that there is no plastics recovery from PST, the latter being used to obtain metals only. Then, a stream of mixed plastics commingled with other fractions (stream F10 of Fig. 1 top), is directed to conventional thermal treatments, while a stream of non-plastic fraction (stream F9 of Fig. 1) is separated and sent to the most appropriate treatment, in agreement with information received from important companies active in the sector, such as Stena Metall AB (2020).

2.2. Innovative options for the management of ELV plastics

The proposed management scheme of ELVP in the bottom part of Fig. 1 includes the same stages of ELV dismantling, shredding and post-shredding of the Current scenario, but it also involves a series of novel processes (CreaSolv®, Extruclean, Modix, Pyrolysis, Plastic upgrading). In particular, this Innovative scheme implies that:

- Polyethylene (PE) of fuel tanks obtained from dismantling (stream F6 of Fig. 1 bottom) is directed to the process of extrusion with sc-CO₂ (Extruclean), after a preliminary shredding.
- All other plastics are directed to PST, where the following exit streams are obtained:
 - A light fraction, with a density <1.0 kg/L, which is sent to a further separation (stream F21 of Fig. 1 bottom) based on density and optical detection, where it is subdivided in: i) PE and polypropylene (PP) (stream F22 of Fig. 1 bottom), which are sent to the CreaSolv® process; ii) all other plastics (stream F23 of Fig. 1 bottom), which are sent to Modix to be pre-treated before pyrolysis.
 - A heavy fraction, with a density >1.1 kg/L (stream F10b of Fig. 1 bottom), which is sent to catalytic pyrolysis together with process residues, such as sludge, dust and fines (streams F18, F19 and F20 of Fig. 1 bottom), after the necessary pre-treatments.
 - Polyurethane (PUR) (stream F10a of Fig. 1 bottom), which is sent to thermal treatments.
 - A non-plastic fraction (stream F9 of Fig. 1), which is sent to appropriate treatments of recovery or disposal.

2.2.1. CreaSolv® process

CreaSolv® is a solvent-based recycling process, which selectively dissolves target polymers in a specific solvent. The undesired components (e.g. non-target polymers, BFRs, VOCs, hazardous substances, non-plastic fractions) – both dissolved or undissolved – are filtered, and the target polymer is recovered by a precipitation step adding an anti-solvent to the solution (CreaCycle GmbH, 2020). Even though any information about the specific solvents/anti-solvents utilised for the process is reasonably confidential, the Best Available Technologies Reference (BRef) document of European Community (EC-JRC, 2018) reports that, for a generic dissolution/precipitation process, the following solvents could be utilised, depending on the plastic feedstock composition: ketones, ethers, cycloalkanes or esters for the dissolution phase; water or alcohols for the precipitation phase. CreaSolv® process allows obtaining recycled plastics with low contaminants and with physicochemical properties comparable to those of virgin material (Wagner and Schlummer, 2020). The process has already been tested successfully in other projects (PolyStyreneLoop Cooperative, 2020) and is under investigation for its potential utilisation with other mixed plastic wastes (Circular Flooring Project 2021; Nontox Project, 2020). CreaSolv® treats the light fraction with a density <1.0 kg/L (streams F21 a of Fig. 1 bottom) after some fur-

ther sorting stages, since it is highly contaminated by other fractions (some light metals, wood, textiles, foam and others).

2.2.2. Extruclean process

Extruclean is a plastic re-manufacturing technology based on extrusion with simultaneous extraction by supercritical carbon dioxide. One of the main advantages of supercritical fluid extraction is its relatively rapid action, related to the low viscosities and high diffusivities associated with supercritical fluids (Manjare and Dhingra, 2019). The unit consists of two extruders connected in series where plastic waste is mixed with sc-CO₂. Temperature and pressure conditions inside the first extruder keep this gas under supercritical conditions to allow its diffusion into the polymeric matrix. The second extruder is connected to a forced degassing system like a vacuum pump, and the toxic contaminants are retained in a special filter (González et al., 2016). Extruclean process can extrude different polymers, such as styrenics and polyolefins, removing volatile compounds by simultaneous action of three factors: evaporation supported by extrusion temperature, well mixing of the polymers' melt, and addition of sc-CO₂ as stripping agent. Carbon dioxide is a good supercritical solvent for non-polar organic compounds: its utilisation allows extracting diverse undesired components, such as volatile compounds, odours, and, potentially, some hazardous contaminants from plastics, as claimed by Aimplas (2020). The process has been already evaluated in a European LIFE Project (LIFE Extruclean, 2017) for the decontamination of polyethylene jerrycans that had contained hazardous substances (e.g. fuel), proving the process efficiency for the removal of contaminants (e.g. VOCs) and the production of new jerrycans. The stream fed to Extruclean unit is that of PE from fuel tanks (stream F6 of Fig. 1 bottom), since its high VOCs content and its severe odour problems make it not recyclable with conventional processes. VOCs in the polymeric matrix are not removed during conventional extrusion, which then produces recycled plastics not properly decontaminated (Cabanes and Fullana, 2020).

2.2.3. Plastic upgrading process

The separated fractions of PE and PP coming from CreaSolv® and Extruclean (streams F25 and F36 of Fig. 1 bottom) are sent to an upgrading stage, where multipurpose recycled plastics are produced by means of compounding with virgin polymers, additives and masterbatch. This allows reaching high-quality secondary materials, which will have from 50% to 80% of post-consumer recycled plastics (Norner, 2021).

2.2.4. Modix extrusion

Modix is a modular extruder able to compact input feedstock and to reduce their particle size, which can be used as pre- or post-treatment of other processes. It is suitable for working with materials having different shapes and densities, thanks to large hollow screw diameter and wide feeding zone, obtaining compact and homogeneous outputs materials in the form of small fragments. The technology is still at the pilot scale (VTT, 2021), so that in the proposed new scheme, Modix has been utilised just as pre-treatment for non-target polymers, heavy fraction, and undissolved material from CreaSolv® (streams F23, F10b, F26 of Fig. 1 bottom), all directed to pyrolysis.

2.2.5. Catalytic pyrolysis process

Pyrolysis is a thermochemical process that provides a degradation of the feedstock under an inert atmosphere, producing oil, gas and solid char (Scheirs, 2006; Vollmer et al., 2020; Nanda and Berruti, 2020). Several companies are involved in the field of plastic waste pyrolysis, by proposing technical solutions that can differ with reference to the upstream feed processing equipment, pyrolyser design and operating conditions, and downstream processing of products (Haig et al., 2013; ORA, 2015). Thermal pyrolysis is

Table 1

Main features and annual flows amount for Current and Innovative scenarios of ELVP management in Europe (see also Figures A.1–A.4 of Annex A).

ELV plastics treatment, %		Current	Innovative
ELVP from dismantling directly to reuse/re-manufacturing/Extruclean/ thermal treatments/disposal		8%	8%
ELVP to shredding		92%	92%
<i>of which</i>			
ELVP from shredding to PST		31%	100%
ELVP from shredding to direct energy recovery		29%	-
ELVP from shredding to landfilling		40%	-
Data sources		(Eurostat Database, 2020)	(Nontox Project, 2020)
ANNUAL FLOWS			
Amount, t/y	ID	Current	Innovative
Total ELV plastics	F1	1,009,800	1,009,800
Plastics from dismantling to reuse	F2	12,200	12,200
Plastics from dismantling to direct re-manufacturing	F4	13,617	13,617
Plastics from dismantling to direct thermal treatments/disposal	F5	556	556
PE in fuel tanks from dismantling to thermal treatments	F6	53,856	-
PE in fuel tanks from dismantling to Extruclean	F6	-	53,856
Plastics to shredding	F7	929,571	929,571
ASR plastics to PST	F7a	286,235	929,571
Plastics to CreaSolv®	F22	-	432,577
Plastics to upgrading	F25 and F36	-	473,594
Plastics to pyrolysis	F28	-	335,636
ASR plastics to landfill	F7c	376,256	-
Total plastics to thermal treatments¹		553,315	188,864
<i>of which</i>			
ASR plastics from shredding to direct thermal treatments	F7b	267,080	-
Mixed plastics from PST to thermal treatments	F10	286,235	-
Separated PUR from PST to thermal treatments	F10a	-	161,023
Residues from Extruclean to thermal treatments	F34	-	4,161
Residues from upgrading to thermal treatments	F38	-	23,680

¹ Does not include plastics separated during dismantling and sent to thermal treatments (streams F5 and F6).

strongly dependent on waste composition as well as reactor temperature profile, heating rate and residence time (Arena and Mastellone, 2006). Catalytic pyrolysis utilises specific catalysts to reduce the process temperature and residence time and positively affect the yield and composition of final products (Kasar et al., 2020; Vollmer et al., 2020). In both processes, the gas fraction is usually exploited to provide energy necessary for degradation process. A catalytic pyrolysis has been selected for the Innovative scheme. The obtained liquid fraction can be refined on-site or sold to a refinery for further processing (ORA, 2015) and the char, composed of unreacted materials, can be disposed of in landfills for inert materials (Haig et al., 2013). The streams sent to catalytic pyrolysis in the proposed new scheme are residues from CreaSolv® (stream F26 of Fig. 1 bottom), non-target polymers from sorting stage (streams F10b and F23 of Fig. 1 bottom), both pre-treated by Modix, together with preliminary dried PST residues (streams F18, F19 and F20).

2.3. Management scenarios of ELV plastics

Two management scenarios have been considered in the LCA study (Table 1 and Table A.1 in Annex A). The current ELVP management scenario in Europe, which includes (Fig. 1, top), after the dismantling/depollution and shredding phases, a post-shredding stage (PST), thermal treatment of combustion, and landfill. The innovative ELVP management scheme (Fig. 1, bottom), which includes the treatments of sorting (based on density and optical differences), CreaSolv®, Extruclean, Modix, catalytic pyrolysis and plastic upgrading.

As already done in a similar study focused on WEEE plastics (Cardamone et al., 2021), the mass flow rates of all the streams in input and output have been quantified through a MFA, reported in detail in Annex A (Figures A.1–A.4), which provides the data reported in Table 1. The MFAs of current and alternative scenarios allow quantifying the amount of ELV polymers annually recovered (i.e. re-used or sent to recycling processes of conventional re-

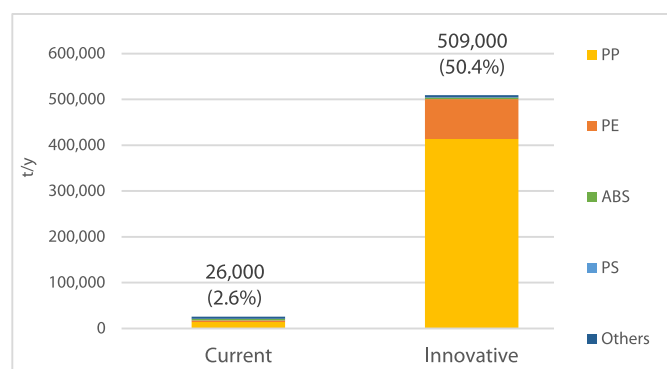


Fig. 2. Annual amount (and percentage) of ELVP sent to reuse and recycling processes in the Current and Innovative scenarios, as obtained by comparative MFA results.

manufacturing, CreaSolv® and Extruclean) and the percentage of recovered ELV plastics, as the ratio between the annual mass flow rate of ELVP sent to recycling processes or reuse and that of total collected ELVP. The results, reported in Fig. 2, indicate that the current scenario shows remarkably lower recovery rates, obtained only from reuse and re-manufacturing of plastics separated during the dismantling stage. The overall recovery percentage for this scenario is just 2.6%, which corresponds to less than 26,000 t/y of polymers sent to reuse or conventional plastic re-manufacturing. Alternative scenario allows an increase of ELVP sent to recycling processes, both in terms of annual amount (up to 509,000 t/y) and overall percentages (up to 50.4%). These improvements relate to the recovery of PE and PP, thanks to the introduction of CreaSolv® recycling process and, to a smaller extent, to the recovery of PE in fuel tanks by Extruclean process. Fig. 3 quantifies the huge amount of ELVP and residues that are sent to combustion process or landfilling in the Current scenario, compared with that of the

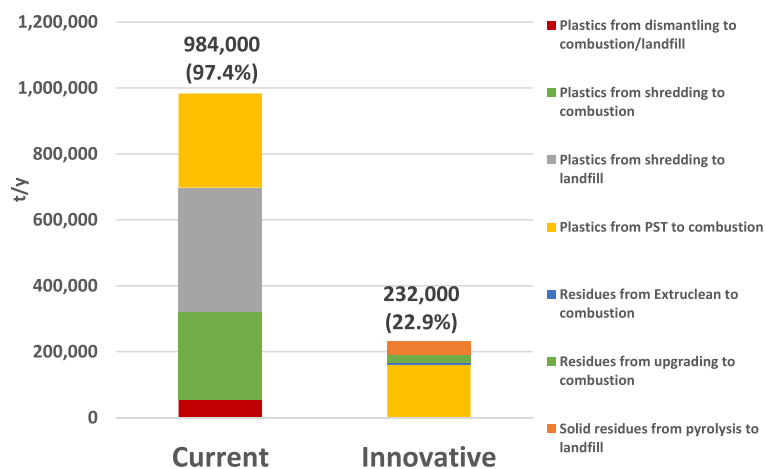


Fig. 3. Annual amount (and percentage) of ELVP sent to combustion or landfill in the Current and Innovative scenarios, as obtained by comparative MFA results.

Innovative scenario (from 980 kt/y to 230 kt/y). This is again due to CreaSolv®, whose adoption makes possible recovering large part of light polymers (PE and PP), and, to a smaller extent, to the catalytic pyrolysis process, which treats heavy fraction, residues and non-target polymers, and Extruclean, which recovers PE of the fuel tanks. A SFA related to the brominated flame retardants has been also carried out in order to evaluate environmental burdens deriving from their content along ELVP management schemes. SFA is based on the bromine content measured in samples provided by plastics recycling companies or reported in the scientific literature (Table A.2 of Annex A). The results, reported in Annex A (Figures A.5–A.6), allow quantifying a BFR content of ELV plastics of about 1980 ppm (i.e. 1980 g/t_{ELV plastics}), and identifying its share in each output stream. According to these analyses, the BFR content has been assumed to be composed of Tetrabromobisphenol A, TBBPA (24%), Decabromodiphenyl ether, DecaBDE (52%) and 1,2-bis(tribromophenoxy)ethane, TBPE (24%). Some of the analysed samples, especially the lightest fractions, presented similar concentrations for each analysed BFR, showing no prevalence of one over the others. On the other hand, when analysing heavy fraction samples (the richest in BFR), the amount of DecaBDE overwhelmed the others, being always around 90% of the total. DecaBDE has been first listed as PBT (Persistent, Bioaccumulative, and Toxic) substance and vPvB (very Persistent, very Bioaccumulative) substance by European Commission, and subsequently its production, use and placing on the market have been restricted by a specific regulation (EC, 2017), which includes that of DecaBDE in the REACH regulation (EC – European Commission 2006). However, the restrictions adopted by EU are effective only since March 2019: being the life cycle of motor vehicles equals many years, several decades will be necessary to benefit from these regulations.

3. Environmental life cycle assessment

3.1. Goal and scope definition

A process-based Life Cycle Assessment has been carried out following the ISO international standards (ISO, 2006) and utilising an attributional approach to assess environmental impacts of the system under analysis (Royal Academy of Engineering, 2017). The “system to be studied” is the management scheme(s) of mixed ELV plastic wastes, starting from ELV dismantling and shredding, followed by plastics sorting and different processes to remove contaminants, until the recovery of secondary resources and the proper final management of residues. The flow diagram in Fig. 4 summarises the system boundaries. They start from mixed plastics

Table 2

Amount and composition of ELV plastics. Sources: (MGG Polymers – Müller-Guttenbrunn Group, 2018; Mehlhart et al., 2018).

Collected ELV (t/y)	6,732,000
Mixed ELV plastics (t/y)	1,009,800
Other waste (t/y)	5,722,200
Mixed plastics composition (%)	
PE	11.0
PP	46.0
ABS	2.6
PS	2.6
PC	2.2
PUR	17.0
PVC	3.0
PA	8.0
Other plastics	7.7

obtained by ELVs collection, dismantling and depollution, which are sent to shredding and post-shredding treatments, and end when energy (in Current scenario) or decontaminated recycled plastics and oil (in the proposed alternative scenario) are obtained. Official reports prepared by United Nations and European Community (EC – European Commission 2018b; UNEP – United Nations Environment Programme 2020) indicate that a large share of ELVs (and thus their plastic content) is not being collected within official management schemes, having an uncertain fate that can involve their disposal in car cemeteries in Eastern Europe or Asia (Leslie et al., 2013; Bobba et al., 2020). However, ELVP exported outside Europe are not considered in this study, since the exportation occurs before the dismantling (i.e. outside the defined system boundaries), and it refers to the entire vehicle, and not only to its plastic fraction.

The “function of the systems under analysis” is the management of plastics in ELVs, regulated by the ELV Directive (EC, 2000). Table 2 indicates an amount of 1,009,800 t/y mixed plastics obtained from ELV collected in Europe in 2018, with the reported plastics composition, which corresponds to the “functional unit”. Different factors affect the definition of a reliable ELVP composition, as the lack of exhaustive and official information and the huge amount of different types of plastics (up to 39) used for vehicles manufacturing (The Plastics Industry Trade Association, 2016). Moreover, it is not possible to correlate European plastics demand in the automotive sector with the expected ELVP composition, for various reasons: cars have a rather long life cycle, implying that an ELV today is a car manufactured 15 to 20 years ago, when

ing recovery and recycling of plastics (Buekens and Letcher, 2020; EC-JRC, 2018). The operations of dismantling and shredding imply the same sets of direct, indirect and avoided burdens for both the analysed scenarios, so it is convenient to exclude them from the analysis.

PST plants represent the further reprocessing stage of shredder residues, aimed at recovering materials from SLF and SHF. This study assumes, for Current scenario, that 31% of ASR plastics are sent to PST (Eurostat Database, 2020), and, for the Innovative scenario, that all ASR plastics are sent to PST (Table 1). Usually, plastics outputs from PST are strongly commingled with other wastes and contain high amounts of additives and contaminants (BFR, fuel traces, VOCs), and are not sent to mechanical recycling. In the Innovative scheme, PST has been considered able to recover some valuable plastic materials, while in the Current scheme, plastic fractions (both light and heavy) are sent to thermal treatments, as shown in Figures A.1-A.4 of Annex A. The composition of plastics to PST (ASR plastics) is reported in Table A.3 of Annex A. A loss of 2.5% in sorting residues (dust, sludge and fines) has been evaluated for ELV plastics during this phase, together with an electricity consumption of 40 kWh for each tonne of plastics in input (Coolrec, 2020).

3.2.2. Current options: thermal treatments and landfilling

Thermal treatments such as combustion in waste-to-energy plant, in cement kilns or in metallurgical processes are widely used for ELVP (Buekens and Zhou, 2014; Vermeulen et al., 2011), especially when the commingling with other fractions is so strong that an appropriate material recovery is unfeasible. Here, it has been conservatively assumed that some streams are treated in a moving grate with energy recovery, which can be considered the best available current option: i) in the Current scenario, part of the ASR separated from shredding (stream F7b of Fig. 1 top) and the mixed plastics from PST process (stream F10 of Fig. 1 top); ii) in the Innovative scenario, PUR (stream F10a of Fig. 1 bottom), residues from Extruclean (stream F34 of Fig. 1 bottom) and residues from upgrading process (stream F38 of Fig. 1 bottom). The related environmental burdens have been quantified based on the average values

of BAT-Associated Environmental Performance Limits (BAT-AEPLs), reported in the BREF document (EC-JRC, 2019), and with reference to the composition of treated ELVP, by identifying and quantifying waste-specific and process-specific burdens, as suggested by Ardolino et al. (2020a). Table B.1 of Annex B reports the values of the main direct and avoided burdens related to thermal treatments of ELVP and recycling residues.

Landfilling of different ELV fractions, plastics included, is widespread in the European Community (Mehlhart et al., 2018; Eurostat Database, 2020; UNEP, 2017), especially for ASRs, which are classified as hazardous waste (Buekens and Zhou, 2014). Landfills can be a major source of emissions to different environmental matrices, particularly when waste with high-BFRs content are disposed of (Levis et al., 2017), besides causing other environmental impacts such as material loss and soil consumption. The landfill modelling developed for this study utilises the degradation mechanisms of DecaBDE in its congeners with low bromine content, which has been proposed by Cardamone et al. (2021), by assuming the same emission factors. Table B.2 of Annex B reports the direct burdens, while Figure B.1 of Annex B shows the details of PBDEs released to biogas and stocked in landfill.

3.2.3. Innovative options

In the proposed new management scheme, the light fraction from PST, with a density <1.0 kg/L, is sent to a further separation, based on density and optical detection, prior to be fed to CreaSolv® (stream F21 of Fig. 1 bottom). These plastics are highly contaminated by other fractions, such as light metals, wood, textiles, foam and others. The obtained input stream to CreaSolv® is made of 92.8% of target polymers PE and PP (stream F22 of Fig. 1 bottom), whose amount and composition have been quantified in Figure A.4 of Annex A and Table B.3 of Annex B. Fig. 5 shows a quantitative flowsheet of CreaSolv® process, with the coupled optical and density separation unit, which has a recovery efficiency of 94% for ELV target polymers, and a removal efficiency of 91% and 76% for non-target polymers and non-plastic fraction, respectively (Fraunhofer, 2021). CreaSolv® process has a recovery efficiency for target polymers of 97%. Other fractions are collected

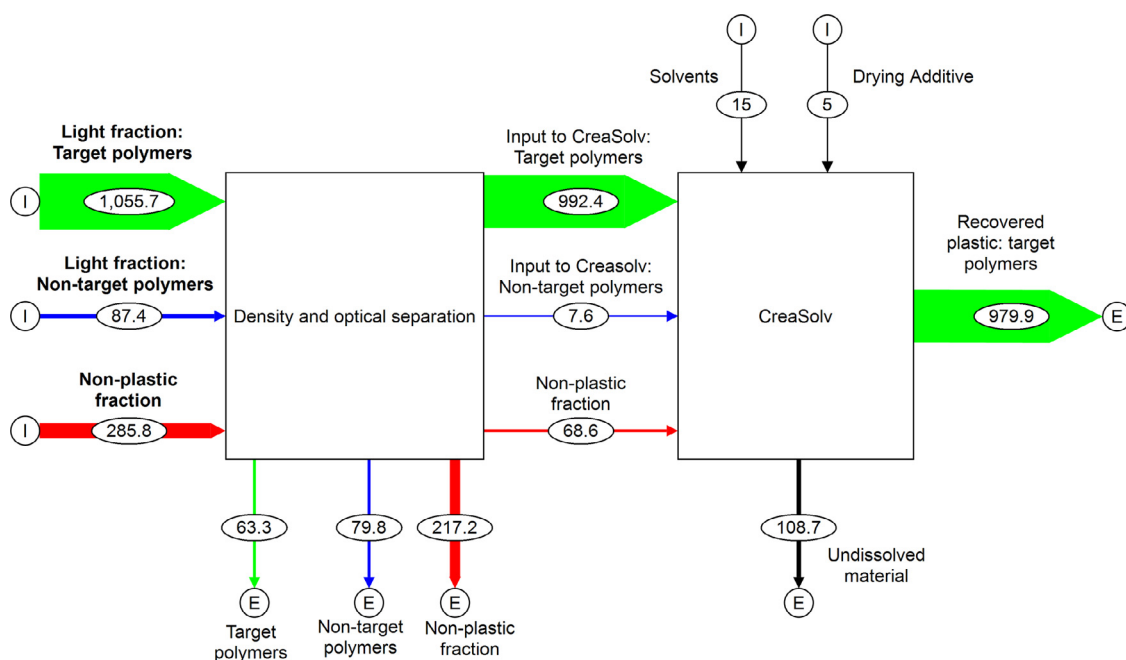


Fig. 5. Quantitative flowsheet of CreaSolv® process with prior sorting process based on optical and density separation. Each flow (light fraction, input to CreaSolv®, and recovered polymers) is reported by indicating the related amount in terms of target polymers, non-target polymers and non-plastic fraction. Data are reported in kg and refer to 1000 kg of plastics in input to CreaSolv®. I = import stream; E = export stream.

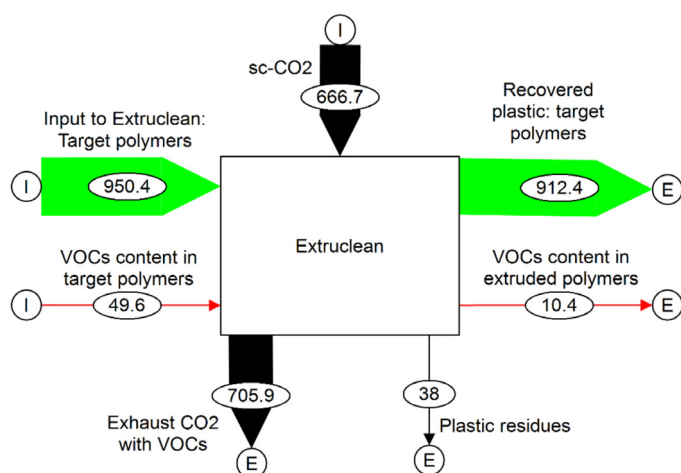


Fig. 6. Quantitative flowsheet of Extruclean process. Data are reported in kg and refer to 1000 kg of input to the process. I = import stream; E = export stream.

separately as undissolved material, together with contaminants (BFRs, VOCs), with a removal efficiency of 98% (Strobl et al., 2021; Fraunhofer, 2021). The process utilises 5 kg of drying agent and 15 kg of solvents' make-up, for each ton fed, which are transferred for large part (up to 90%) to the recovered polymers. The type of solvents is confidential, thus the LCA conservatively considers tetrahydrofuran for the dissolution and methanol for the re-precipitation, since they are the solvents with highest environmental impacts among those commonly utilised for these kinds of processes (EC-JRC, 2018). Tables B.4 and B.6 of Annex B are the LCI tables for CreaSolv® process, respectively without and with the upgrading process. Tables B.5 and B.7 of Annex B report data utilised to estimate avoided burdens.

Extruclean treats PE from fuel tanks obtained from dismantling phase (stream F6 of Fig. 1 bottom, as quantified in Figure A.4 of Annex A). Three main assumptions have been applied for the quantification of the flowsheet of Fig. 6, and estimation of the environmental burdens: i) the process has a recovery efficiency of 96.0% for target plastics; ii) the average VOCs content in target polymers is 49,600 ppm and the related removal efficiency is 79%, as measured by trials conducted in the framework of Nontox activities; iii) the ratio between contaminated plastic fed to the extruder and sc-CO₂ is about 60%_w/40%_w, with exhaust CO₂ directly discarded into the atmosphere. The latter is a conservative assumption, since CO₂ recovery for this process has not been investigated yet, even though it could be applied at industrial scale. The LCI for Extruclean process is reported as Tables B.8 and B.10 of Annex B (without and with upgrading, respectively), while Tables B.9 and B.11 of Annex B report the assumed values of parameters for the quantification of the avoided burdens. Plastic upgrading stage receives recovered polymers from CreaSolv® and Extruclean processes (streams F25 and F36 of Fig. 1 bottom) and allows increasing their quality, and then their substitutability factor up to 0.93, which in turn implies higher avoided burdens. On the other hand, the process requires some compatibilizers (5% of input stream) as well as electric energy (356 kWh/t_{IN}) and residues disposal (5% of input stream).

Modix extruder pre-treats the streams directed to the catalytic pyrolysis process. Table B.12 of Annex B is its LCI table. The polymeric and elementary compositions of input stream to the pyrolysis process are reported in Table B.13 of Annex B. The LCA study refers to an industrial scale catalytic pyrolysis unit (Figures B.2 and B.3 of Annex B), which treats different kind of waste streams, and aims at producing crude oil to be further refined off-site. It has been assumed that the input ELVP has a low heating value of

34.6 MJ/kg, based on its composition. Fe₂O₃ is used as the process catalyst (Imdea, 2021), with a feeding mass flow rate of 5% of that of pre-treated plastics, i.e. about 50 kg (Scheirs, 2006). The dry-ash free (daf) input is obtained as gas/liquid/residues, with a relative distribution of 12/85/3 on gross basis, and 0/80/3 on net basis (Haig et al., 2013). The latter includes the consumption of gas and liquid for the energy required to the plastics pre-treatment, to reach the process operating temperature and for the heat requirement for hydrocarbons condensation phase. Low heating values of obtained gas, oil and residues are 35.4 MJ/kg, 40.7 MJ/kg and 15.1 MJ/kg, respectively (Imdea, 2021). Unrefined crude oil is assumed to be the final gate of the system under analysis, then possible further refinery activities are not considered. The energy requirement for pyrolysis process has been quantified as 4390 kJ for each kg of pre-treated waste, meaning an absolute value of 4580 kJ for kg of daf matter entering the reactor, and 13% of the input energy, in agreement with Evangelopoulos et al. (2020). Total gas flow rate and a part of unrefined oil (3.7%) are sent to combined heat and power (CHP) units, which have an electrical conversion efficiency of 38% and thermal conversion efficiency of 45% (Ardolino et al., 2020b) and allow covering total electrical requirement (100%) and part of thermal requirement (49%). Finally, an air pollution control (APC) system has been included to treat flue gases generated by gas combustion, with a consumption of 1 kg of activated carbon for each t of input waste.

3.3. Life cycle impact assessment

Fig. 7 shows the normalised results of impact assessment for the analysed scenarios, with the contribution of each single life cycle stage and with reference to the functional unit. The results are reported only for impact categories that have a major role in the reported analysis. Numerical values can be found in Table C.1 of Annex C. The figure refers to six scenarios that gradually move from the Current ELVP management scheme to the proposed Innovative scheme. The histograms referring to the current scenario clearly show the overwhelming detrimental effects of plastics landfilling in terms of Non-Carcinogens (2.51·10⁶ person-year). This contribution is mainly due to the huge amount of ELVP that annually ends up in landfill (376 kt/y) as well as to their BFR content (2133 ppm), which implies relevant releases of PBDEs into the atmosphere (25 t of higher PBDEs and 114 t of lower PBDEs, along the 100 years of landfill lifetime). The results confirm the awful effects of landfilling option, especially when applied to contaminated plastics, as already assessed by Cardamone et al. (2021) with reference to WEEE plastics (whose content of BFR, 8840 ppm, is anyway much higher than that of ELVP). The “Current no landfill” scenario assumes the implementation of a landfill ban for ELVP in the European Community, so that all the non-recovered plastics in the current scenario would be treated by thermal treatments. It indicates that avoiding landfilling is a crucial step, sufficient to remarkably enhance the overall environmental performances in terms of Carcinogens (for 101%), Non-Carcinogens (for 100%), Respiratory Inorganics (for 59%), and Non-Renewable Energy (for 59%). On the other hand, a higher impact on Global Warming (for 56%) is expected, due to plastic residues sent to Waste-to-Energy. The histograms “Ext only” relate to the introduction of the Extruclean process to treat PE from fuel tanks, indicate a further improving (i.e. with reference to the “Current no landfill” scenario) of the potential impact for Carcinogens (for an additional 781%) and, to a smaller extent, those of Non-Carcinogens (for an additional 9%), Global Warming (6%) and Non-Renewable Energy (8%), as a consequence of the lower amount of ELVP sent to WtE and the avoided production of virgin PE. These positive effects are also highlighted by the contributinal analysis reported in Figure C.1 of Annex C. The “Ext+CrS” histograms refer to the introduction

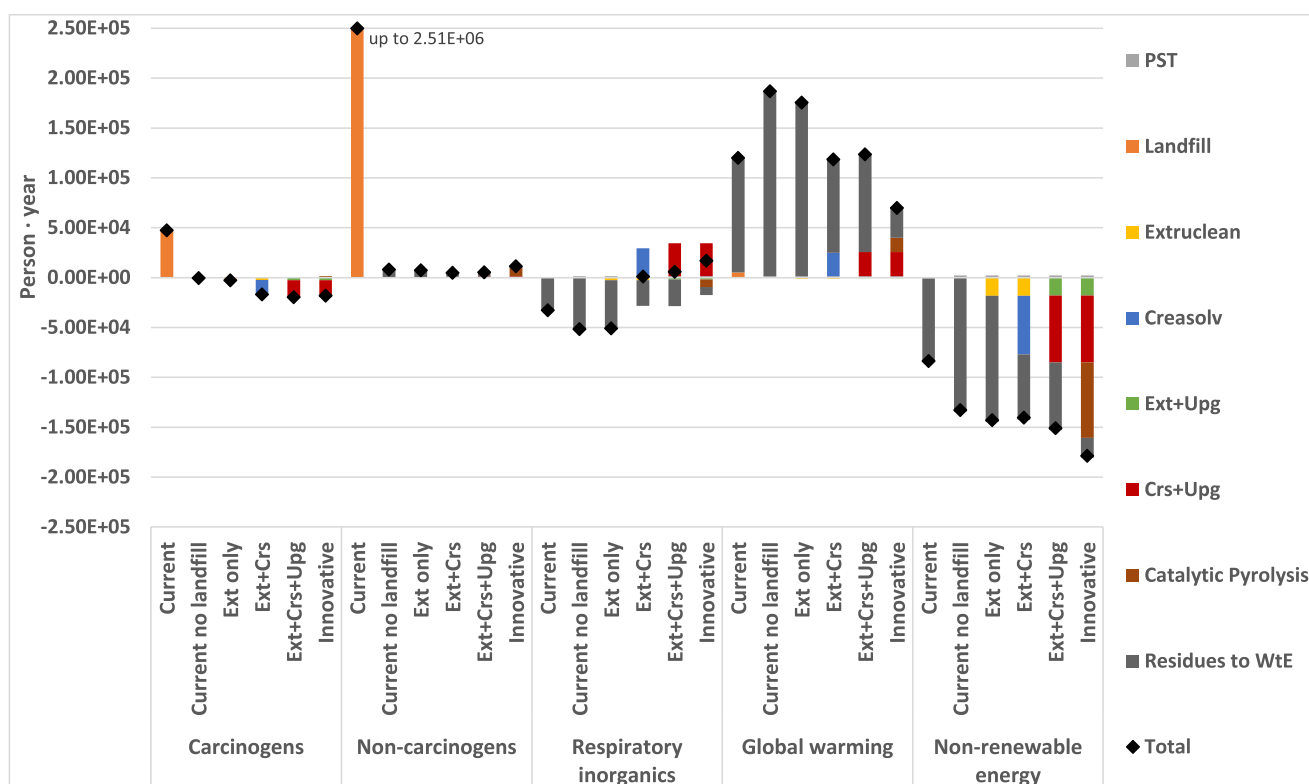


Fig. 7. Normalised results of impact assessment for the Current and Innovative scenarios, with a series of intermediate stages. Results refer to the functional unit and the main midpoint impact categories, and with the contribution of each single stage of the life cycle. Legend: PST = Post-Shredding Treatment; WtE = Waste-to-Energy; Ext = Extruclean; Crs = CreaSolv®; Upg = Plastic Upgrading. The shaded rhombus indicates the total value for each impact category. Results are normalised in “Person · year”, i.e. the average impact in a specific category caused by a person during one year in Europe.

of CreaSolv® in the management scheme to process polyolefins in the light fraction from PST, being all the remaining residues treated by combustion. This new scheme allows recovering high amount of polymers (PP and PE, as reported in Table B.4 of Annex B), so providing a further improvement of the potential impact for Carcinogens (for an additional 534%) and Global Warming (33%) categories. On the other hand, the required energy of the CreaSolv® process worsens the impacts for Respiratory Inorganics (for 102%), as it can be also deduced from the contributonal analysis of Figure C.2 of Annex C. The further addition of the upgrading process for plastics coming from Extruclean and CreaSolv® (“Ext+CrS+Upg”) leads to a worsening of Respiratory Inorganics (382%) due to the consumption of electric energy and additives, but also to an improvement of Carcinogens (16%), thanks to the better quality of recovered polymers, that are characterised by higher values of the substitutability factor (Tables B.7 and B.11 of Annex B).

Finally, the implementation of the whole Innovative scheme leads to a great improvement of expected impacts of the Global Warming and Non-Renewable Energy midpoint categories. This is explained by the introduction of catalytic pyrolysis, which permits a lower utilisation of combustion process and therefore a reduction of fossil CO₂ emissions. All the other categories show instead a (generally limited) worsening, due to direct burdens related to the catalytic pyrolysis process, mainly for its thermal energy requirement (Figure C.3 of Annex C). Overall, the Innovative scheme implies a remarkable improvement of the environmental performance of the ELVP management with reference to the Current situation, in terms of the main impact categories of Carcinogens (for a total 138%), Non-Carcinogens (100%), Global Warming (42%) and Non-Renewable Energy (114%). The estimated worsening of poten-

tial impacts of Respiratory Inorganics (152%) is mainly related to the higher energy consumptions, and thus it can be in a next future largely mitigated by the utilisation of renewable and less pollutant energy sources.

3.4. Sensitivity analysis

A sensitivity analysis has been implemented by varying some significant parameters in a reasonable range (Cardamone et al., 2021; Clavreul et al., 2012). These parameters have been selected with reference to the main novel options: consumption of sc-CO₂ in the Extruclean process; energy requirements in the CreaSolv® process, yield of oils in the catalytic pyrolysis, and substitutability factor of recycled plastics from CreaSolv®+Upgrading processes. Table D.1 of Annex D reports the assumed ranges of variation. Fig. 8 shows the results in terms of variation factor, VF, defined as the ratio between the result obtained in the sensitivity scenario for the changed parameter and that quantified in the base case scenario (Ardolino et al., 2018). All the estimated variation factors appear to be in a limited range. The VF obtained by reducing the pyrolysis light crude oil yield (with a gas/liquid/solid distribution changed from 12/85/3 to 12/75/13) reaches 1.6 for the Non-Carcinogens category; this is related to the lower avoided burdens (being the light sweet synthetic crude oil reduced from 784 kg to 692 kg) but also to the higher amount of solid residues sent to landfill. The electric energy requirement for CreaSolv® process (+/- 25% of the base case value) leads to appreciable VFs for the categories Respiratory Inorganics (in the range of +/- 0.8, i.e. 80%), Global Warming (in the range of +/- 0.2) and Non-Renewable Energy (+/- 0.10), since this represents the most relevant direct burdens, as it can be deduced by the contributonal analysis reported

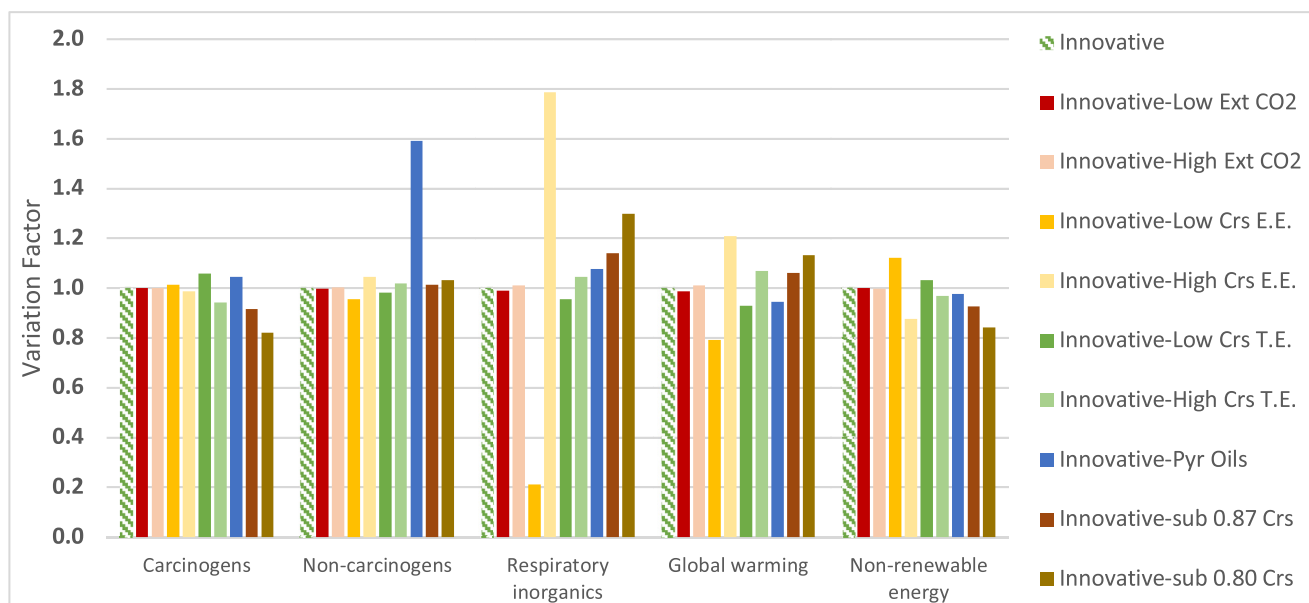


Fig. 8. Sensitivity results in terms of variation factor. VF = 1 indicates no variation; some variations occur when VF is < 1 or VF > 1; and a negative value of VF changes the potential impact from positive to negative or vice-versa.

in the Figure C.2 of Annex C. The effect is mainly due to the high shares of fossil and not renewable sources which still characterise the current European electricity mix. This result highlights the crucial role of renewable and not-fossil energy sources and their implementation in the future energy mixes. The substitutability factor α of plastics recovered by means of CreaSolv® process has been modified from the value 0.93 of the base case to 0.87 and 0.80. Its variation affects the results of almost all the categories, even though in the limited range +/- 0.30. For all other changed parameters, the effects related to the assumed variation are negligible for all the midpoint impact categories.

4. Conclusions

The study analysed and quantified the environmental performances of novel treatments of sorting, dissolution/precipitation (CreaSolv®), extrusion (Extruclean and Modix), catalytic pyrolysis, and plastic upgrading, which could be combined to define a new ELV plastics management scheme.

The novel options and the resulting new scheme have been compared to the options and the overall scheme of current management of ELVP in Europe, by implementing a Life Cycle Assessment with the support of Material and Substance Flow Analyses. The study refers to the annual amount and average composition of ELVP collected in Europe and utilises high-quality data for the innovative processes, acquired within a H2020 project consortium, which includes 12 research centres and industrial companies active in the plastics recycling sector.

The comparison between the two analysed scenarios allows quantifying the advantages of the proposed scheme. Its adoption in Europe may provide a great increase of annual amount of ELV plastics sent to recycling (from 26 kt/y up to 509 kt/y, i.e. from 3% to 50%), and a corresponding decrease in that of residues sent to combustion or landfilling (from 984 kt/y down to 232 kt/y). These promising results are mainly related to the utilisation of the dissolution/precipitation process (CreaSolv®), whose introduction allows recovering large part of light polymers (PE and PP).

The Life Cycle Impact Assessment indicates that the proposed new recycling scheme could strongly improve the environmental

performances of ELV plastics management, mainly in terms of Carcinogens, Non-Carcinogens, Global Warming and Non-Renewable Energy midpoint categories. Their potential impacts improved of 138%, 100%, 42% and 114%, respectively. This means that plastics from end-of-life vehicles could be managed in a sustainable way, by using the analysed novel options, appropriately combined in an efficient management scheme.

It is also noteworthy that a banning of landfill option applied to the current management scenario could imply a fast and significant improvement in all the impact categories, with the exception of that of Global Warming. This could be a first, even though not definitive, step to be adopted in Europe.

The sensitivity analysis indicates that the results could be affected by the variations values of the energy consumption of CreaSolv® process and by the yield of oils recovered by the catalytic pyrolysis. There is also an important role of the actual substitutability factor of recycled plastics obtained from CreaSolv® process, i.e. their capability to match the required properties of the virgin polymers. This latter aspect needs further and deeper investigations.

Annexes

Supplementary data associated with this article can be found in the online version of the journal.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at [doi:10.1016/j.spc.2021.09.025](https://doi.org/10.1016/j.spc.2021.09.025).

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