

Recirculation potential of post-consumer /industrial bio-based plastics through mechanical recycling - Techno-economic sustainability criteria and indicators

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Abstract

The sustainable circular bioeconomy turns biogenic waste and residues into renewable resources to produce added value bio-based materials. According to the recent, Circular Economy Package EU legislation, mechanical recycling offers the best alternative EoL option for bio-based plastics in a complementary way with chemical recycling, with the latter taking over materials inadequate to be mechanically recycled. Techno-economic sustainability analysis (TESA) criteria and indicators to assure the feasibility and viability of mechanical recycling of post-consumer bio-based plastics, and the recirculation potential of the recovered material, are proposed based on the evaluation and synthesis of research results selected through a critical literature review. Organic recycling is considered as a preferred EoL option for post-consumer biodegradable bio-based plastics only when these products are found to be non-recyclable by the proposed TESA criteria. Environmental and social sustainability criteria, that constitute the other two pillars of sustainability, are not considered in the present work, but need to be included to complete the sustainability assessment of any End of Life (EoL) option. The proposed techno-economic sustainability criteria for mechanical recycling include: a) Mechanical recyclability, b) Economic viability, c) Common environmental /techno-economic criteria and also d) Recirculation potential of the materials recovered. Specific indicators are proposed as metrics for assessing the corresponding criteria.

Keywords: Techno-economic sustainability, bio-based plastics, biodegradable plastics, plastic waste streams, mechanical recycling, material recovery, recirculation, circular bioeconomy

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1 Introduction

1.1 Sustainable circular bioeconomy

The linear ‘*take, make, dispose*’ economic model that relies on the use and waste of large quantities of cheap, easily accessible materials and energy still dominates the global and EU economy, despite the improved rates of recycling (municipal waste generated in 2017 in EU-28: 249 Mt; landfilling: 23%, incineration: 28%; material recycling 30%; composting: 17%; other: 2%) [1]. On the opposite, the model of the circular economy is “*restorative and regenerative by design*” [2]. The basic principles on which the circular economy is based include optimization of resources and minimization of system risks [2]. The circular economy promotes re-use and stimulates the industrial symbiosis by turning the by-products of one industry into raw materials for another industry.

Despite the increasing recycling rates, the quantities of plastics disposed to the environment uncontrollably or in landfills, including single use plastics, represent a growing problem, and a major contributor to marine littering [3,4]. To cope with this problem, EU policies, action plans and regulations are developed, while the European Parliament and Council (EP&C) approved the EU directive aiming at banning or controlling the use of the most threatening 10 single-use plastic products and oxo-degradable plastics [5].

The Circular Economy Package (CEP) [6,7], that replaced the Waste Framework Directive (WFD) [8], aims at helping the transmission of businesses and consumers in Europe towards a more circular economy where resources are re-circulated and used in a more sustainable way. The proposed by the European Commission (EC) closing of the product lifecycles loop can be achieved for post-consumer plastics with higher rates of recycling (material recovery) following the EU Strategy for Plastics in the Circular Economy [9]. According to [9], by 2030, all plastics packaging should be recyclable (mechanically). Specific targets to reduce

waste streams and increase recovery rates of post-consumer products have been set by CEP [6,7]. A common EU target has been set for recycling of municipal waste at 65% by 2035 along with a maximum binding landfill target of 10% of municipal waste and a ban on landfilling of separately collected waste. Recycling targets of 70% and 55 % have been set for packaging and plastic packaging waste by 2030, respectively. Separate collection obligations are strengthened and extended to bio-waste by 2023 (Bio-waste is defined in WFD [8]: “*biodegradable garden and park waste, food and kitchen waste from households, restaurants, caterers and retail premises and comparable waste from food processing plants*”).

The terminology and hierarchy of the alternative EoL routes of bio-based products are defined in CEP [6,7]. The waste hierarchy sets waste prevention as first priority followed by: preparing for re-use, materials recovery, organic recycling, energy recovery, and disposal. The inventory of the available alternative EoL routes for post-consumer /post-industrial bio-based plastics is presented in [10].

The creation of a flow of materials recirculated back into the manufacturing of new products has not been established yet. The product design and manufacturing stages do not take into account the high collection and treatment costs resulting from the post-consumer product’s raw material recovery. This disconnection is attributed to the gap between product design, materials supply, marketing, production and the materials return flow through waste management and recycling [9].

The EU has set the goal [11] for a more innovative economy characterised by sustainable agriculture and fisheries, combined with sustainable use of renewable biological resources for industrial production of bio-based products, while ensuring food security, biodiversity and natural resources protection. Biomass is valorised in biorefineries to produce bio-based materials such as polymers, chemicals and bioenergy [12]. The Bioeconomy Strategy and

action plan [13] set by the European Commission focuses on developing “*new technologies and processes for the bioeconomy; markets and competitiveness in bioeconomy sectors*” and on “*pushing policymakers and stakeholders to work more closely together*”. Various programmes and instruments have been launched in support of the Bioeconomy development.

Using farmland and biomass for producing high value bio-based products that end up in landfilling or incineration, following the linear model of economy, is not in line with the circular economy principles. The bioeconomy represents a circular economy model by nature: consumption of resources may be observed only in effective bio-cycle through regeneration of biodegradable products by life processes. In the case of bio-based products cycle, resources may recirculate by means of material recovery and recreation. The sustainability analysis of the relationship between farmland, biomass and bio-based products must include the EoL options. All different EU policies converge to a common target: the development of a sustainable and circular bioeconomy in Europe, that is the renewable segment of the circular economy [11]. The sustainable circular bioeconomy turns post-consumer bio-based products and residues into secondary renewable resources to produce added value bio-based materials and chemicals through industrial symbiosis.

1.2 Techno-economic sustainability of bio-based products

The competitiveness of bio-based products, as compared to fossil feedstock-based products, requires optimization of resources and high efficiency combined with minimization of negative environmental impact [14]. A quantitative and qualitative analysis of the impact of technological advances on the financial viability of the conversion of biomass to materials and products including their use and EoL routes is provided by means of Techno-Economic Analysis (TEA). The Techno-Economic Sustainability Analysis (TESA) incorporates the dimension of sustainability on the TEA analysis of alternative feedstock conversion and materials, products processing routes and EoL routes. A comprehensive approach on the

definition of TESA is provided by Gargalo, et al [15] who describe the proposed framework for techno-economic and environmental sustainability analysis as “*a step-by-step framework whose purpose is to identify the best potential alternative(s) that would sustainably create value with the least potential risk of economic and environmental impact*”.

Therefore, the TESA of bio-based products evaluates the sustainability aspects related to technology and financial profitability of all the involved stakeholders. In interaction with the other two pillars of sustainable systems: the environmental and the social sustainability analysis, TESA completes the Sustainability Analysis.

1.3 Scope of the present work

The methodological approach for the definition of environmental, TESA and social sustainability criteria for resources, processing and alternative EoL routes of bio-based products was developed in the framework of the STAR-ProBio project [16,17]. The TESA of alternative EoL routes for the post-consumer /industrial bio-based plastics ensures optimal alternative routes to turn these products into valuable resources for the circular bioeconomy. One key factor is how mechanical recycling, as a preferred alternative EoL option for post-consumer plastics by CEP [6,7], can affect the design and resource efficiency of a new bio-based product complementing its functionality and/or its high bio-based content [18,19].

Techno-economic sustainability criteria and indicators are proposed in the present work to assure the feasibility and viability of mechanical recycling for post-consumer/industrial biodegradable and non-biodegradable bio-based plastics. Environmental and social sustainability criteria, that constitute the other two pillars to complete the sustainability assessment of any EoL option, are discussed in other works (e.g. [20]). The confirmation of mechanical recycling as a potentially optimal alternative EoL route based on the proposed TESA criteria for both, non-biodegradable and biodegradable plastics, will allow for

promoting recycling of bio-based post-consumer plastics. Organic recycling is considered as a preferred EoL option for post-consumer biodegradable bio-based plastics only when these products are shown to be mechanically/chemically non-recyclable. The application of the TESA criteria to illustrative case studies is the subject of research work in progress.

2 Methodological approach

2.1 Alternative EoL routes of the post-consumer/industrial bio-based plastics

The terminology concerning the biobased economy, used in the present work, was defined in detail in [10] following EU and international standards and regulations. Thus, according to EN 16575 [21], bio-based plastics are the plastics made in whole or partially from renewable biological resources (biomass). The bio-based plastic materials are classified in two major categories: bio-based, non-biodegradable and bio-based, biodegradable plastics [22]. The bio-based non-biodegradable mass commodity plastics with the same chemical structure as their fossil-based counterparts are called drop-ins (e.g. bio-polyethylene (bio-PE), bio-propylene (bio-PP), bio-polyethylene terephthalate (bio-PET)). Another category of bio-based non-biodegradable plastics are the technical performance polymers (e.g. polytrimethylene terephthalate (PTT) or thermoplastic copolyester elastomers (TPC-ET)). The bio-based biodegradable plastics are defined and certified as biodegradable in specific environment(s), in compliance to the definitions given by the relevant standard specifications. Some of them (e.g. polylactic acid (PLA)), are compostable under industrial composting conditions, while others (e.g. polyhydroxybutyrate (PHB), polybutylene succinate (PBS)), are biodegradable in several environments, including soil and water. Fossil based biodegradable plastics (e.g. polybutylene adipate terephthalate (PBAT)) are not considered in this work. The global bioplastics production capacity in 2019 was estimated

at 2.1 Mt, expected to reach 2.4 Mt in 2024, with the growth driven by innovative bio-based polymers such as bio-PP and polyhydroxyalkanoates (PHAs) [23].

The alternative EoL routes of the post-consumer/industrial bio-based plastics, analysed extensively in [10], follow the hierarchical principles set by CEP [6,7] and WFD [8]: The highest valorisation priority for all post-consumer/industrial bio-based plastics, both non-biodegradable and biodegradable, is given to the material recovery routes and recirculation (though mechanical recycling and possibly chemical recycling). The produced recyclates, and monomers/oligomers, respectively, are valorised by replacing virgin materials. The second priority, applicable only to bio-based biodegradable plastics, is given to organic recycling (aerobic industrial composting and anaerobic digestion (AD)). In this case, the bio-based plastic materials are decomposed by life processes. The evolved biogenic carbon is closing the carbon loop. If the material recovery (for all plastics) and organic recycling (for biodegradable plastics) routes are not feasible and/or viable, post-consumer/industrial bio-based plastics are routed to energy recovery for the production of renewable energy. Energy recovery is not one of the EoL preferred options anymore according to CEP [6,7], as it results in loss of valuable materials. The flow diagram presented in Fig. 1 shows the prioritisation of the alternative EoL options for post-consumer/industrial bio-based plastics [10], following the CEP hierarchy [6,7].

It is important at this point to clarify some ambiguous issues pertaining to the mechanical recyclability of bio-based plastics: Post-consumer bio-based plastic products labelled as biodegradable in specific environment(s), (e.g. labelled compostable), may, but should not necessarily, be routed to industrial composting or AD facilities as a first option. “Compostable” implies that this EoL option is possible but not mandatory. In contrast, compostable bio-based materials that are also labelled as “recyclable”, should be routed to mechanical/chemical recycling as a first priority in order to recover materials and chemicals

that can be reprocessed into new products. Mechanical recycling of bio-based materials supports the circular bioeconomy development. In contrast, their biodegradation simply closes the carbon loop, wasting high added value materials. Further on, the biodegradation of fossil-based components of biodegradable plastics results in loss of the fossil carbon. This carbon, along with the biogenic carbon, enter the biological carbon cycle in the form of CO₂.

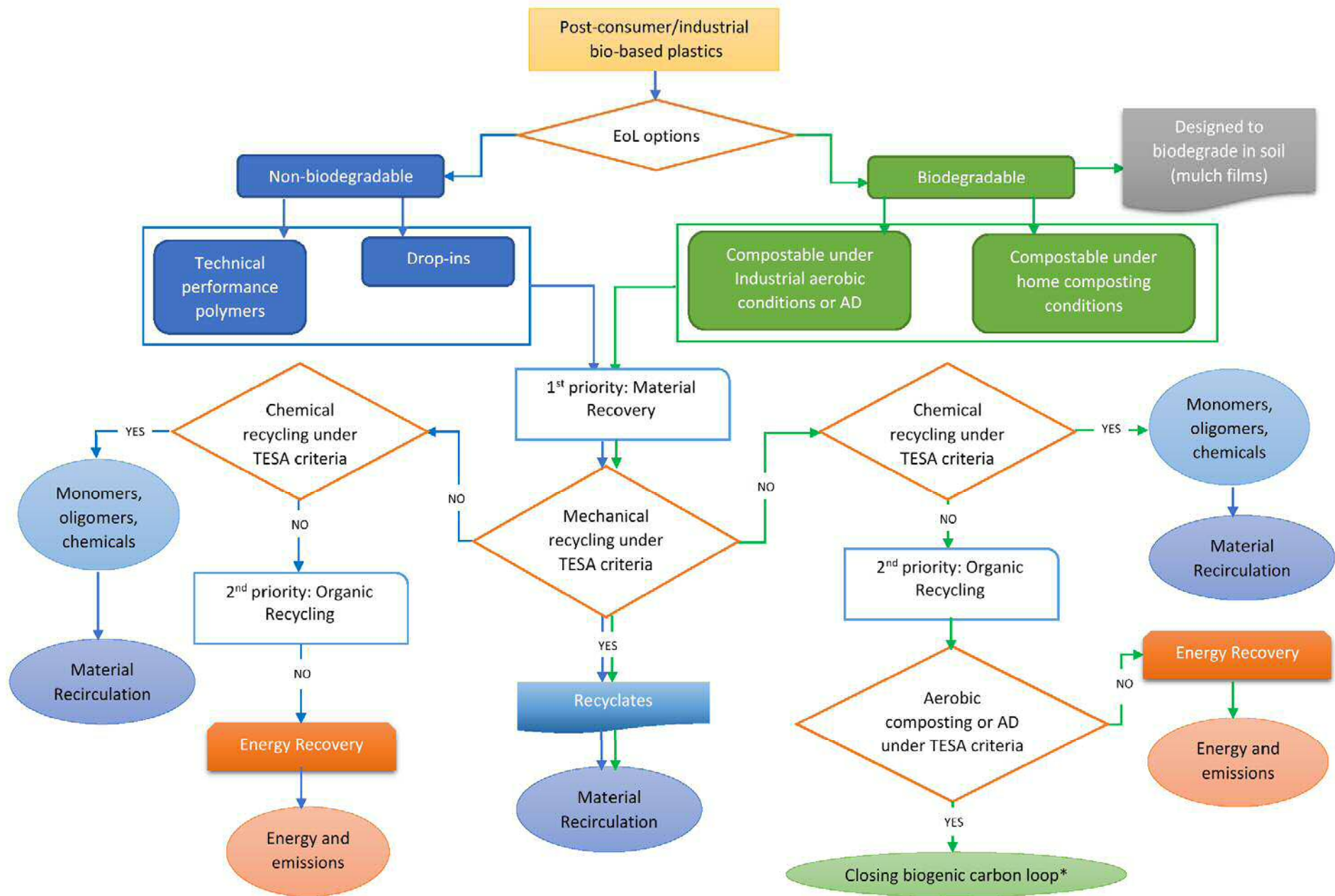


Fig. 1. Alternative EoL options for post-consumer/industrial bio-based plastics

(*) refers to the bio-based content of biodegradable products

The fast-growing market of bio-based plastics has led to an extensive number of research studies concerning the potential of mechanical recycling (as well as chemical recycling and organic recycling) of these materials [10]. Even though the majority of these studies concern lab-scale experiments, some review papers also include information from industrial trials.

A key question remains to be answered: Under which conditions is mechanical recycling of non-biodegradable and biodegradable bio-based plastics economically viable and technically feasible? The present work aims at investigating this crucial question systematically, proposing TESA criteria and indicators that ensure viability and feasibility.

2.2 The mechanical recycling EoL route

2.2.1 Background framework

The applicability of the mechanical recycling scenario for post-consumer/industrial bio-based plastics depends on the combination of several critical parameters. The provisions of CEP [6,7], the targets and objectives set by the EU Strategy for Plastics in the Circular Economy [9] and the EU environmental legislation, were used as drivers along with an extended literature review for defining the TESA criteria for the mechanical recycling route of post-consumer/industrial bio-based plastics. The inventory of the mechanical recycling of bio-based plastics analysed in [10] was used as background framework.

2.2.2 TESA Criteria

The proposed criteria are grouped into 4 integrated TESA Criteria: The first two concern “Technical feasibility” and “Economic viability”. “Common techno-economic – environmental” criteria are analysed as a third group. The “recirculation potential” of the recovered materials is analysed as a separate fourth criterion as it represents a challenging and complicated emerging process of the bioeconomy bridging the gap of the circular bioeconomy between the feedstock and manufacturing processing stage and the mechanical recycling EoL stage. Each TESA criterion is associated with a set of indicators, that are used as metrics for assessing the corresponding EoL option.

2.2.3 Boundaries of the present analysis

The boundaries defined for the processing of the feedstock raw material to the final product and the boundaries of the alternative EoL options of post-consumer/post-industrial bio-based products are shown in Fig. 2. The next stage is the stage of the post-consumer/industrial bio-based plastics waste management. The stages from the resources, feedstock processing to bio-based polymers and manufacturing of the final plastic products, the market and use stages are outside the scope of the present work. The waste management stage is not included in the present TESA since it is a major and very complicated stage that handles all kinds of municipal and industrial waste streams, including plastics for recycling, biowaste and many different waste management systems. Specific TESA criteria apply to the waste management systems considering their sustainability. For example, according to Gu et al. [24], much higher environmental benefits result from distributed recycling practices than from centralised recycling practices. These systems are analysed extensively in literature for conventional post-consumer products. Their applicability to also include bio-based plastics poses various challenges and limitations, such as organized separate collection.

It is beyond the scope of the present work to investigate the waste management stage. However, the requirements set for the sorted bio-based post-consumer plastics received at the entrance of the mechanical recycling facility are analysed.

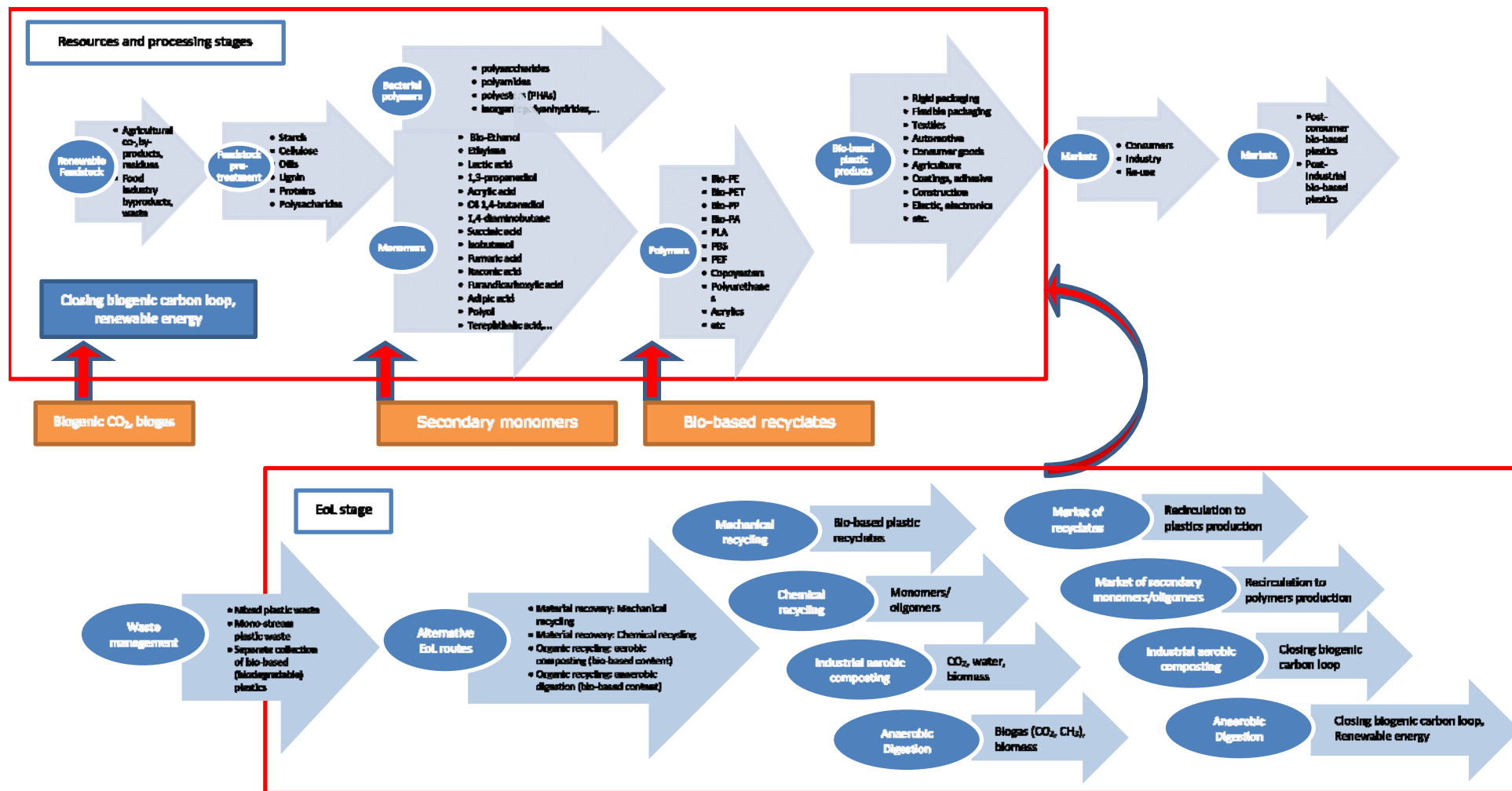


Fig. 2. Alternative recirculation routes from EoL options for post-consumer /industrial plastics

The boundaries for the present TESA are set at the entrance of the sorted post-consumer /industrial bio-based plastics to the recycling facility and ends at the exit of the facility with the final recovered materials (recyclates) that can be used as raw materials to produce new products through industrial symbiosis. The recirculation potential of the produced recyclates is included in the present analysis as it characterises their quality and valorisation potential. The recirculation and valorisation process of the recovered materials to produce new products is not included in the TESA analysis of mechanical recycling, as it represents a challenging and complicated issue of the bio-based plastics industrial sector.

2.2.4 Mechanical recycling processing

The current mechanical recycling processing technologies, described in this section, are applicable also to post-consumer bio-based plastics with proper adjustments. Understanding the critical parameters dominating the various processing steps, from the post-consumer plastics collection to the production of recyclates, is a prerequisite for proposing realistic TESA criteria and indicators for the mechanical recycling of bio-based plastics.

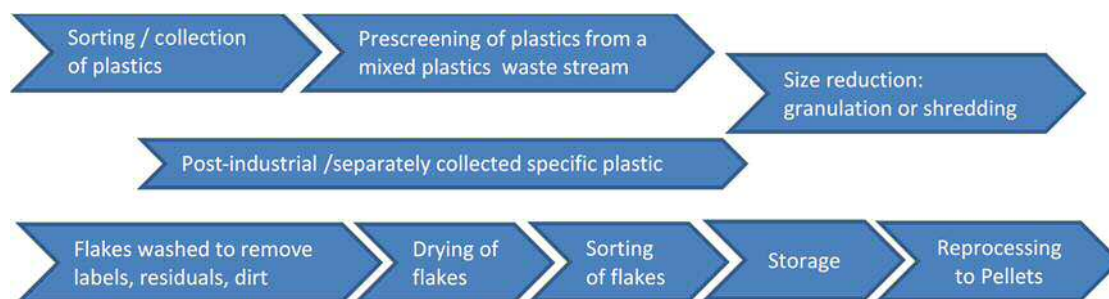


Fig. 3. Typical sequence of the basic steps of the plastics mechanical recycling

The mechanical recycling of post-consumer/industrial plastics into recyclates follows several basic steps. As shown in the schematic diagram of Fig. 3 large variations may exist

among different schemes and technologies. Mechanical recycling is a high cost, labour and energy, demanding process. These requirements are related to the different processing stages applied, briefly described as follows [^{25,26}]:

Collection: Various collection schemes have been introduced by the municipalities for the collection of all kinds of recyclable materials, including plastic articles, that may be divided into three main categories [²⁷]: a) Mixed Municipal Solid Waste (MSW): contaminated waste stream that needs intensive separation and cleaning treatments. b) Recyclable multi-material collection (co-mingled collection): all types of recyclable materials source separated and collected together (i.e. metals, plastics, paper etc.). Plastics are sorted out as a separate stream or directly into different plastics streams. c) Recyclable mono-material: source separated as one separate recyclable post-consumer plastics category. This stream may include conventional and bio-based plastic types together, difficult to be differentiated by the consumer, or targeted specific plastic types.

Transportation: Collected mixed recyclable materials or collected mixed plastics or scrap plastic transported to “*recyclable materials collection centres*” where they are separated, if not already separated. Baled plastics transported to processing plants.

Sorting (pre-screening): If not cleaned before, the first stage of sorting is the removal of contaminants by separation of recyclable plastics from foreign materials. The manual sorting of collected mixed plastics represents the first decontamination process. It aims mainly at separation of the recyclable thermoplastics from non-recyclable thermosets and pre-selection of major plastic waste categories. The identification by the operators of foreign materials and decontamination and pre-selection of the plastic waste stream passing by a conveyor belt is based on specific separation codes (shape, colour, appearance-contamination, foreign materials, trademark etc.) [²⁸].

The main goal is the manual or automatic separation of recyclable from non-recyclable plastics, between different recyclable polymers and ensuring that recyclable plastic is free of foreign materials and contaminants. Several techniques and specially designed equipment are used to facilitate sorting of the collected plastics based on their polymer content. Manual pre-screening is a labour-intensive process done under stressful conditions and requires continuous attention [29]. The main selection of polymeric materials is applied at a later stage at the recycling facility, after shredding of the plastics.

Shredding: Shredding to flakes or chunks usually follows the first sorting (pre-screening). This process may include material size-reduced by wet type granulator or combined washing and size reduction.

Washing and drying of flakes: Improved latest generation wash plants are used allowing for using only 2-3 m³ water per tonne of material. Innovative technologies have also been developed for the removal of organics and surface contaminants by means of dry cleaning of surfaces e.g. friction methods [30].

Sorting of flakes: Various sorting techniques are used for the flakes' separation: a)

Mechanical: Separation process used to ensure that different plastics, of different density are not mixed up. The “float- and- sink” process separates the flakes that float and sink from tubs of water [28]. This method is applied to separate polyolefins from plastics like PET, PVC or PS [31]. Water solutions with chemicals are used to control the medium density for materials of targeted densities (e.g. PET separated from PS). Air elutriation and other techniques are applied to separate low-density films from denser ground plastics. *Spectroscopic:* For plastics with similar densities, methods such as X-ray fluorescence or mainly Fourier–transform near-infrared spectroscopy (FT-NIR) techniques render the separation of different materials possible [28,32]. The FT-NIR systems also use optical recognition cameras systems and other optical sorters to

distinguish coloured from clear streams [30]. *Laser*: Laser-sorting, still under development, improves the ability to separate complex mixtures and polymers by type or grade. This includes efficient removal of impurities (e.g. polyamide (PA), silicones) and sorting of plastics of different colours including black, and polymers of different grade and type (e.g. PP from HDPE) [28]. *Selective dissolution*: A new separation technology [28] is based on the selective dissolution of polymers. As this process requires high amount of energy and use of chemical solvents, is not considered a sustainable method.

Methods such as sorting techniques based on colour, NIR, fine sieving, etc. are more efficient than the sinking and floating flake sorting method, as they improve the purity and quality of the recyclates in terms of mechanical properties, but at a higher cost and lower recycling processing rates. The average mechanical recycling process efficiency is estimated at 60%, depending on the collection and sorting processes [33]. The remaining 40% is routed to other EoL options. The efficiency can be improved significantly if key collection and sorting processes are improved (e.g. separation at source, use of reliable detectors, advanced software systems for more precise and productive automated sorting, etc.). Flexible packaging cannot be separated efficiently by all recycling facilities equipment and so it may not be acceptable by the recyclers. Air-classification systems based on the materials density are used to remove films and labels. Other developments in the separation technologies allowing for recovering flexible packaging include ballistic separators and sophisticated hydrocyclones [34, 35]. The introduction of bio-based and biodegradable plastic categories, including packaging films, represents a new challenge.

Drying and Storage: Mechanical drying by dewatering and drying the flakes with a centrifuge. Subsequently, a stainless-steel cyclone operates as a thermal drying system discharging the dried flakes into bags. The dried flakes, if homogeneous, are directed to storage for extrusion processing, otherwise to sorting facilities for removing remaining

traces of foreign material. The washing effluent is treated for removing large contaminants by screening and smaller contaminants by sedimentation. Cleaned water is recirculated back into the system.

Purification: Decontamination is important especially for recyclates to be used in food industry applications, such as rPET. For post-consumer PET and HDPE bottles, for example, the shredding is followed by an optical sorting through a closed loop recycling [36]. This includes dry cleaning, automated optical sorting into three streams (clear PET, HDPE, coloured PET/HDPE), additional manual sorting, granulation of each stream separately into flakes, washing to remove labels etc., floatation and final optical sorting. The potential use of homogeneous clean PET or bio-PET flakes by the food industry requires decontamination. This is achieved with solid caustic soda at 200 °C, to remove the outer layer of the flakes and leave a pure r-PET polymer that can be used as virgin PET. Purification of HDPE or bio-HDPE flakes is achieved by treating the flakes at high temperatures and low pressure.

Melting: Dry flakes of cleaned homogeneous polymer stream are melted down by means of appropriate processing technologies and equipment. Melting processing may follow moulding technologies into new products or extrusion technologies into strands/strings and finally into pellets (recyclates) [37]. Usually the intrinsic properties of the recycled fossil- or bio-based polymeric materials associated with their functional performance are downgraded during reprocessing [38] rendering the recyclates suitable for lower end-use applications (downcycling). This may be avoided by applying technologies designed for reprocessing the targeted polymers according to their thermal properties under controlled temperatures and other processing parameters. Plasticizers, other additives and fillers may be used during extrusion to enhance specific properties of the recycle. The molten polymer degradation during reprocessing, can be prevented or eliminated by using

specially designed schemes of new formulations and compositions of stabilizers [39]. Physical treatments can also be applied, such as annealing to improve and upgrade the plastic products (upcycling) or blending of recycled polymers with other polymers acting as plasticizers or blending polymer matrixes with recycled polymers as value added modifiers [38,40]. In case of incompatible polymers, improved adhesion between the polymers can be achieved by means of compatibilizers. If the compatibilizer compound is miscible in the two phases, a physical linking is achieved with the creation of a bond between the two polymers. If the compound is miscible only in one phase, the functional group can react with some functional group of the other polymer creating a chemical linking between the two polymers. Chemical reactions can also take place between the macromolecules of the different phases during reprocessing (reactive blending) with a compatibilization polymer formed “in situ” [39]. Reprocessing of condensation thermoplastics is problematic because of fast degradation due to high processing temperatures, rendering necessary the use of special compounds such as impact modifiers or reactive chain lengtheners [41, 39].

Pelletizing: At the final stage of the extrusion process the regenerated plastic product in the form of strands/strings is cooled by water as to be granulated into pellets (recyclates).

Market: The bio-based plastic recyclates may be used in the market to produce new plastics or bio-based plastic products. Recyclates may be used in blends with virgin polymers and/or other polymers. The main technologies of moulding application for recyclates include [10]: extrusion, injection, blow, vacuum and inflation moulding. Recyclates may also be used with blown film extrusion processing. In some cases, recyclates especially from scrap, may be reused by the same manufacturer (recirculation inside the factory) to produce various products, usually of lower quality, in the form of original and recycled materials blends at various percentages.

3 TESA criteria and indicators

3.1 TESA criteria for mechanical recycling of bio-based plastics

Mechanical recycling follows the “reuse” option at the top of the hierarchy of the waste management alternatives of WFD [8] and CEP [6]. Four TESA criteria are proposed for the mechanical recycling of bio-based plastics.

3.2 Mechanical recycling feasibility TESA criterion

The first TESA criterion for the mechanical recycling of post-consumer/industrial bio-based plastics concerns feasibility and is assessed based on a combination of indicators related to key material characteristics and reprocessing factors:

Biodegradability: The nature and the intrinsic properties of the material, determine the possibility to use existing recycling processes for certain categories of bio-based products. The drop-in plastics (bio-based non-biodegradable) can be recycled through the existing recycling streams of their conventional counterparts [42,43]. These bio-based products can be sorted out from a mixed plastics stream with the existing technologies (e.g. NIR) and follow the corresponding mono streams of the conventional plastics [10]. A crucial issue, in the case of recycling bio-based non-biodegradable plastics together with their non-bio-based counterparts, is that the reprocessed bio-based plastics will lose their bio-based label (the bio-based content will change unpredictably and variably). Consequently, it would be preferable to collect/sort post-consumer bio-based non-biodegradable plastics in separate streams to retain their high bio-based added value. Biodegradable bio-based plastics can also be sorted out in separate mono streams by using existing techniques, as applicable (e.g. density separation, NIR technology etc.) [10]. The NIR technology, however, is not available in all cases [42]. As a result, bio-based biodegradable plastics are usually misdirected to incineration, a low value EoL route.

Provided that a specific bio-based biodegradable plastic reaches at least 5% of the collected mixed plastic waste, it would be possible in the future to organize its mono stream sorting, or even better, separate collection and mechanical recycling in a financially viable way [42].

In the specific case of bio-based plastic packaging, the separation of compostable from non-compostable bio-based plastic waste, at source is crucial for process efficiency. The separation of compostable bio-based plastic waste allows for alternative routing of this stream to mechanical, chemical or organic recycling, depending on the technical feasibility and the economic viability of each option.

Recyclers are currently reluctant, considering bio-based biodegradable plastics as possible contaminants for reasons of incompatibility with the conventional plastic waste streams. A qualitative indicator is proposed:

Bio-based biodegradable (Yes/No)

Sorting efficiency: In the case of mixed plastics waste streams, the bio-based plastics sorting efficiency is a critical point that can enhance the uniformity of the post-consumer bio-based plastic stream and reduce drastically contamination by other conventional polymers and foreign or organic materials. Bio-based biodegradable plastics can be sorted out by employing NIR technology. It has been demonstrated that PLA, for example, can be sorted out efficiently in a single pass through NIR with a minimum of 97.5% accuracy [44]. Some reports indicate that NIR based sorting is not always effective in separating black polymers, or plastics with special structures such as laminates [45].

Achieving high-quality separately collected and sorted post-consumer conventional plastics and bio-based non-biodegradable plastics, requires the collaboration of the national, regional and local authorities with the waste management companies. Financial

resources collected through the Extended Producer Responsibility schemes [46] and systematic public awareness activities can contribute towards this goal. A new guidance on separate collection and sorting of waste in the EU is under preparation by the Commission and European Parliament [9]. The CEP directive sets new rules to enhance the implementation of the separate collection of post-consumer plastics [6]. The European Strategy for Plastics [9], recommends expanding and improving separate collection of plastic waste and expanding, modernising and integrating the collection and sorting systems and the recycling capacity of the EU, to reduce the cost and enhance the inputs quality to the recycling industry.

Post-consumer bio-based biodegradable plastics may also be collected separately into mono-streams for mechanical recycling, provided that the investments for their installations would be covered by the commercial volumes and sales [47]. If the efficiency achieved is high, it may reveal the mechanical recycling as the most attractive EoL option for post-consumer/industrial bio-based biodegradable plastics [33]. The European Strategy for Plastics supports the establishment of a dedicated regulatory framework for separate collection of biodegradable plastics [9].

The goal that all plastic packaging products should be recyclable by 2030 [9], requires improved and innovative designs (including additives, colours, labels, etc.) allowing for easier recycling of plastic products [9]. Furthermore, post-consumer bio-based plastic materials used for food packaging, meeting the strict food contact requirements, whether biodegradable or not, should be sorted separately if the produced recyclates are to be reused for food packaging through recirculation. Alternatively, special processes should be employed. As an example, a special process '*General Plastic*' based on the Starlinger Decon technology has been proposed by the '*Panel on Food Contact Materials, Enzymes and Processing Aids*' (CEP Panel) of the European Food Safety Authority (EFSA) [48] for

the case of mechanical recycling of post-consumer polyethylene terephthalate (PET) used in food contact applications. The proposed process at pilot scale, applicable also to bio-PET, used as input hot caustic soda (e.g. 2% solution) washed and dried PET flakes coming from post-consumer containers (less than 5% PET from non-food market applications). The preheated PET flakes were submitted to solid state polycondensation (SSP) at high temperature under vacuum and gas flow in a continuous reactor. This process was shown to limit potential contaminants migration into food to 0.1 µg/kg food. This is considered safe for manufacturing food contact materials and articles even with 100% rPET content [48]. A key question for this process, or analogous processes for other bio-based plastics, is the installations and processing cost.

A case of commercial interest concerns the sorting efficiency of PET and PLA bottles. PLA and PET cannot be easily sorted by sight or based on density while even a level of PLA >0.1% can compromise the recyclability of PET. However, it has been reported that PLA can be effectively sorted out by using NIR, at accuracy 98 % [49, 44].

Sorting efficiency is a quantitative measure of the quantity of the post-consumer bio-based plastic mono stream (% dry weight) sorted out from the initial quantity of the collected post-consumer bio-based plastic. The quantitative indicator η_{sort} is a measure of the sorting efficiency process for the targeted post-consumer bio-based plastic. The indicator η_{sort} is calculated by combining the sorting yield with the purity (e.g. sorting yield 80% and purity 95% may be obtained by NIR and so η_{sort} is 0.76); while purity may be raised to 99% if complemented by manual sorting [50, 27]. The following sorting efficiency quantitative measure is proposed:

$$\eta_{\text{sort}} (\%) = \text{mass of sorted (kg)} \times 100 / \text{mass of the collected post-consumer specific bio-based plastic (kg)}$$

Note: all measures of quantities /mass (kg) in the present work are defined on a dry weight basis.

Compatibility: Another critical question concerning the processing through mechanical recycling is the plastics waste stream uniformity (mono-stream, industrial or sorted post-consumer bio-based plastic). Presence of non-compatible polymers with the main polymer under reprocessing and/or foreign materials result in contamination introducing processability problems and degraded quality of the recycle [51]. Although bio-based biodegradable plastics are considered as contaminants in the streams of conventional plastic wastes, some relevant research works with LDPE streams, briefly resented in the meta-study of [52], showed that possible contamination up to 10% by bio-based compostable plastics or mixed bio-based plastics waste did not introduce any problems to the recycling processing or the quality of the produced polyethylene recycle. In particular, contamination of LDPE streams by 0.5 % to 10 % impurities of PLA/PBAT blend and pure PBAT showed no significant change in the viscosity behaviour, elasticity, tensile strength, processing and optical properties of the mixtures of LDPE with PLA/PBA and PBAT as compared to pure LDPE. A first slight decrease was observed in the melt flow rate at 10 % impurity [53,54]. In the case of mixtures of LDPE with starch blends at the highest percentage of 10 %, a marginal influence was found on the viscosity of the LDPE and flow characteristics. The colour and water uptake were observed to change with increasing ratios of starch blends [53].

The compatibility between individual bio-based polymers or bio-based and conventional polymers, is a major issue for the recycling industry. The compatibility factor impacts the recycling process feasibility and efficiency. Sorting efficiency and cleaning treatment prevent even small percentages of non-compatible polymers to be present in the sorted (clean) plastic waste stream. The compatibility characteristics between different

conventional and bio-based polymers have been studied and presented in the literature [10]. Special compatibilizers and other additives may be used to enable processability of selected incompatible polymers [25,55,56].

Bio-based polymers often contain different reactive groups in their structure allowing for convenient application of compatibilization methods [57]. Among the alternative compatibilization methods under development, the reactive techniques represent a flexible and effective way to compatibilize bio-based polymer blends during reprocessing. Research is ongoing on investigating the effect of the molecular processes and the reaction mechanisms on the functionalities of the bio-based blends [57]. Anhydrides, diisocyanates, epoxides and other compounds have been reported as compatibilizers. The one-step blending and compatibilization offers the advantages of eliminating the need for multiple processing steps allowing for a shortened processing time representing a more economical and environmentally friendly route for the improved reprocessing of post-consumer bio-based polymers. The efficient reactive reprocessing requires the use of lower MW polymers or small molecules as compatibilizers that can be distributed in the polymer molten mixture easily at high rates and can react with the two components that constitute the two phases to be coupled [57].

Compatibilizers and some new methods, such as electron irradiation, allow for improved interfacial adhesion in the case of composites [58,59]. Fibre reinforced composites (FRP) generally made of glass (GFRP), carbon (CFRP) or aramid (AFRP) reinforcing fibres dispersed in an organic matrix, as well as heavily contaminated plastics can under specific circumstances be recycled into plastic lumber [60,61].

The limiting amount of an incompatible polymer present in the mainstream of a reprocessed polymer (%), may be defined by the proposed quantitative compatibility indicator:

$\eta_{cont} (\%) = \text{mass of contaminants (kg)} \times 100 / \text{mass of the sorted post-consumer specific bio-based plastic (kg)}$.

A second indicator defines the percent presence of other compatible polymers (conventional or bio-based) in the sorted bio-based plastic mono-stream:

$\eta_{comp} (\%) = \text{mass of compatible polymers (kg)} \times 100 / \text{mass of the sorted post-consumer specific bio-based plastic (kg)}$.

Thermal degradation characteristics: The most important limiting factor for the mechanical recyclability of post-consumer bio-based plastics is the thermal degradation characteristics of the polymer during recycling. Thermal stability is a first prerequisite for any polymer to be recyclable [62]. For example, (bio)PET deteriorates appreciably (down cycling) during mechanical recycling as a result of thermal and thermo-oxidative degradation, hydrolysis and side reactions [63].

The biodegradable bio-based polymers are usually more susceptible to hydrolysis than the conventional ones. They degrade to a stream exhibiting high MFR, associated with decreased molecular weight (MW) leading to low quality recyclates [64]. The temperature and humidity impact their hydrolysis rate [64, 65].

PLA, for example, encounters difficulties with mechanical recycling due to its tendency to undergo thermal degradation [66, 67]. PLA's thermal degradation at the molten stage is related to processing temperature, residence time in the extruder and the moisture content of its granules during recycling [68]. However, according to [42] it has been proven that homogeneous clean streams of PLA waste can be recycled without degradation problems (e.g. PLA drinking cups) [69]. Various methods have also been applied to improve the quality of the PLA recyclates. Blending of post-consumer PLA plastic with virgin PLA and a chain extender led to PLA recyclates with increased intrinsic viscosity (by 9%) and

improved performance (upcycling) [70]. The poor thermal stability of PLA was shown to be significantly improved in blends of PLA (98% L-lactic acid) with natural rubber (NR) and liquid natural rubber (LNR) as a compatibilizer, prepared using a melt blending process [71]. Bio-polyurethanes are also known to be recyclable by mechanical reprocessing through regrinding (or alternatively by chemical recycling). The reground post-consumer polyurethanes may be used in various applications (e.g. fillers, mouldings, blends etc.) [72, 73]. A synoptic review of the effect of blends of recycled with virgin bio-based biodegradable plastics, is shown in Table 1.

In the case of bio-composites, the bio-based polymeric matrix and the reinforcements can undergo thermo-mechanical degradation during thermal processing. Bio-based polymer-reinforced composites are expected to have lower recyclability potential than their synthetic thermoplastic matrix counterparts [74]. Studies on mechanical recycling of bio-composites have reported that the advantage of the mechanical recycling technique is the speed of the process into new materials while decreasing the environmental burdens related to the separation and processing of the composites' components [75].

The mechanical recycling processing of post-consumer bio-based plastics may be improved in terms of feasibility and efficiency by means of various additives, such as plasticizers, compatibilizers, reactive chain lengtheners, impact modifiers etc. that can facilitate mechanical recycling processability, reducing the cost and the environmental impact, act as reinforcements, increase thermal resistance and decrease the degradation risk, rendering the recycled materials suitable for advanced applications (upcycling).

Improvement of the processability of post-consumer bio-based polymers through mechanical recycling can be achieved by a plasticization process. Plasticizers allow the exchange of the bonds between the polymer macromolecules and the low molecular weight compound. This results in increased deformability of the recyclate and decrease of

the glass transition temperature of the polymer blend. This is especially important for bio-based polymers sensitive to thermal degradation such as PHAs [76].

Table 1. Effect of extrusion of blends of recycled with virgin bio-based polymers

Material	Processing conditions	Mechanical properties	Physical properties	Polymer Degradation	Reference
Virgin PLA (Ingeo™ 2003D) / Recycled – degraded PLA blend at 50/50	<i>Extrusion processing of aged and washed samples with virgin PLA</i>		Viscosity: (recycled PLA): -17%; (blend with 50% virgin): -13% T _{cc} : (reprocessing): Decr; (blend with 50% virgin): Incr Ageing and washing processes: <i>crystallization of the polymer</i> Reprocessing step: <i>crystalline structure destroyed</i>	Chain scission during melt processing MW: (blend with 50% virgin): Incr Alternative for upgrading the properties of recycled PLA	[77]
Virgin PLA (Ingeo™ o 2003D) / reprocessed PLA blend: up to 60 % recycled blend	<i>Extrusion processing of aged and washed samples with virgin PLA</i>	Mechanical properties: (up to 50% recycled): <i>Acceptable</i>	Viscosity: (up to 50% recycled): -15% for amorphous or -13% for semi-crystalline; (100% recycled PLA): -24% and -26% Optical properties: (up to 50% recycled): <i>Acceptable</i> Thermal properties: (up to 50% recycled): <i>Acceptable</i>	MW: (up to 50% recycled): negligible <i>Internal recycling is highly applicable for PLA</i>	[78]
Virgin PLA (Ingeo™ PLLA resin PLA 2002D) / recycled postconsumer PLLA bottle flakes blend (20, 40, 60, 80 wt.%)	<i>Extrusion processing of post-consumer with virgin PLA</i>	σ_u : (recycled content $\geq 40\%$): Decr Modulus E: (recycled content $\geq 80\%$): Decr	Colour: (recycled content $\geq 20\%$): Darker recyclates; (40% recycled content): blue and red tones T _m : (recycled content $\geq 80\%$): Decr	MW: (recycled content $\geq 60\%$): Decr All sheet samples were successfully thermoformed	[79]
Virgin PHB: / regrind PHB: 50/50 (wt%); seven regrind ratios	<i>Extrusion processing</i>	σ_u : -5%	Viscosity: -5%		[66, 80]

Stabilisers used in reprocessing are selected to control the thermal degradation and viscosity of polyolefin recyclates. Reactive molecules are used to repair broken chains and restore chain lengths and melt strengths, increasing the tensile strength and the molecular weight of the recyclates. The mechanical properties of recycled polymers may be enhanced also by means of (nano)fillers (bio-composites) like carbon black (e.g. carbon nanotubes (CNT), sepiolite (1D) and graphene (2D)) [81].

Several measures can be used as qualitative indicators of thermal degradation of a bio-based plastic. Among them, Melt Flow Rate (MFR) (EN ISO 1133) and melting point, T_m , that depend on the polymer's MW and its thermal history and crystallinity measured by means of DSC (Table 1). Decrease of T_g and MW or increase of crystallinity and MFR during the recycling process indicates degradation. A sensitive indicator for the degradation of plastics is the decrease of their elongation at break value (ϵ_{br}).

A quantitative indicator for the thermal degradation of bio-based plastics is the decrease of the recycle's ϵ_{br} (alternatively MFR or T_g or T_m) as compared to the corresponding value of the virgin polymer:

$$\eta_{TD} (\%) = \epsilon_{br} \text{ of recycle} \times 100 / \epsilon_{br} \text{ of virgin polymer}$$

An indicative ϵ_{br} value of the incoming post-consumer bio-based plastics will allow for the evaluation of a possible serious degradation of the post-consumer plastics during the use stage and before reprocessing. This will allow for determining the thermal degradation share exclusively attributed to reprocessing.

Physical and chemical characteristics limiting recyclability: Physical and chemical limiting factors for the mechanical recycling of conventional plastics also apply to the recyclability of bio-based plastics [66]. The degree to which the mechanical recycling processing of the conventional and bio-based plastics is influenced by various

physical limiting factors depends on the processing technologies applied, the equipment used and specific plastic waste stream characteristics [30, 82].

The limiting recyclability factors are evaluated case by case. For example, the contamination of the plastic waste stream by organic materials, degradation during use, thin films, etc., are common limiting factors for the recyclability of most conventional and bio-based plastics [61, 83, 62]. The more complex and contaminated the waste is, the more difficult is to recycle it. The polymer degradation that may occur during its useful life as well as the heterogeneity and nature of waste, are among the major problems the mechanical recyclers face [62].

The long-term exposure of polymers to UV or chemicals renders them non-recyclable [84]. The biodegradable bio-based polymers are more sensitive to ageing [85, 86]. Long-term storage of bio-based biodegradable polymers or inappropriate storage conditions can render them brittle and non-recyclable [87].

All biobased plastics are recyclable under specific conditions. Some biodegradable biobased plastics may be more sensitive than the non-biodegradable ones to hydrolysis (if exposed to humidity and high temperature) and/or thermal degradation (e.g. they tolerate a narrow range of processing temperatures), but this does not mean that they are not recyclable or that the resulting recyclate is not commercially valorised.

Complex and multilayer products require special techniques for the separation of the different components and sorting of the different polymers. The main techniques are classified into: a) recycling of a target polymer by dissolution and re-precipitation; b) mechanical, chemical or physical delamination [88]. Laminated products and labels with binders and inks require special removal/washing techniques. Concerning composite plastics, especially for fibre reinforced plastics, mechanical recycling is considered as

the most mature, economic and environmentally friendly recovery option as compared to incineration or chemical recycling. It involves size reduction to a mix of fibrous and powdered materials by shredding or crushing or milling processes. The recyclates, sorted into fractions of different sizes, are of lower value as compared to virgin materials and may be used as fillers in new composite products or in a closed loop recycling process [60].

A qualitative indicator is proposed:

Are any physical or chemical characteristics present in the post-consumer bio-based stream limiting recyclability? (Y/N)*

() thin films, seriously degraded or multilayer plastics etc.*

Mechanical recyclability TESA criterion indicators: A brief overview of proposed indicators for the “mechanical recyclability” TESA criterion of post-consumer/industrial bio-based plastics is shown in Table 2.

Table 2. Proposed indicators for the mechanical recyclability TESA criterion

Indicators	Metrics
Mechanical Recyclability TESA Criterion	
Biodegradability	<i>Bio-based biodegradable (Yes/No)</i>
Sorting efficiency	$\eta_{\text{sort}} (\%) = \text{mass of sorted (kg)} \times 100 / \text{mass of the collected post-consumer specific bio-based plastic (kg)}$
Compatibility	$\eta_{\text{cont}} (\%) = \text{mass of contaminants (kg)} \times 100 / \text{mass of the sorted post-consumer specific bio-based plastic (kg)}$. $\eta_{\text{comp}} (\%) = \text{mass of compatible polymers (kg)} \times 100 / \text{mass of sorted post-consumer bio-based plastic (kg)}$
Thermal degradation characteristics	$\eta_{TD} (\%) = \epsilon_{br} \text{ of recyclate} \times 100 / \epsilon_{br} \text{ of virgin polymer}$
Physical characteristics limiting recyclability	<i>Are any physical or chemical characteristics present in the post-consumer bio-based stream limiting recyclability? (Yes/No)</i>

3.3 Economic viability criterion

Availability of mechanical recycling facilities: The prerequisite for considering mechanical recycling as a priority alternative EoL option for post-consumer/industrial bio-based plastics is the availability and/or the distance of available infrastructures that accept bio-based plastics [89], especially bio-based biodegradable plastics (under certain conditions) [42,90,10].

The operation of the mechanical recycling facilities is evaluated in terms of techno-economic sustainability considerations as well as environmental and social considerations [42]. Operating permits are required under the Directive on industrial emissions 2010/75/EU (IED) [91], based on Best Available Techniques Reference Documents (BREFs), to ensure that environmental protection practices are adopted. The application for a permit requires full description of [91]: “(a) *installation and its activities; (b) raw and auxiliary materials, other substances and energy used in or generated by the installation; (c) sources of emissions from the installation; (d) conditions of the site of the installation; (e) where applicable, a baseline report in accordance with Article 22 (hazardous substances); (f) nature and quantities of foreseeable emissions from the installation into each medium as well as identification of significant effects of the emissions on the environment; (g) proposed technology and other techniques for preventing or, where this is not possible, reducing emissions from the installation; (h) measures for the prevention, preparation for re-use, recycling and recovery of waste generated by the installation; (i) further measures planned to comply with the general principles of the basic obligations of the operator as provided for in Article 11; (j) measures planned to monitor emissions into the environment; (k) main alternatives to the proposed technology, techniques and measures studied by the applicant in outline*”. Accordingly, a set of important data needed for the TESA and

Environmental sustainability analysis of a facility, should be available with the permit application process of the specific facility. Proposed indicator qualitative:

Availability of mechanical recycling facilities with operating permits (Yes/No)

Availability of collected and sorted bio-based plastic waste: The economic viability of the mechanical recycling of post-consumer bio-based plastics, requires the availability of sufficient commercial mono stream bio-based plastic volumes (quantities) at a rather steady state of supply [28].

The quantities of the bio-based plastic waste entering the recycling facility are those collected and sorted under the operating waste management system (e.g. municipal and/or dedicated post-industrial waste management system) and/or transported from remote areas. According to [2,9], the economics of the mechanical recycling process of plastic waste, and so also of post-consumer bio-based plastics, can be improved significantly if the collection and sorting systems fragmentation and disparities are improved. If the volume of one type of bio-based plastic would reach 5-10 % of the collected plastic waste, the sorting of this bio-based plastic in separate streams would be feasible, in economic terms [42]. The currently collected and sorted volumes of bio-based polymers are insufficient. It is estimated that new separate streams of post-consumer PLA plastics, technically easily sorted and mechanically recyclable, will be established in the near future, once sufficient volumes enter the market [42,47].

In addition to quantity, the post-consumer bio-based plastics quality is important. The EN 15347:2007 [92] standard, provides a characterisation scheme for plastic wastes by laying out selected properties and test methods that the supplier of the waste makes available to purchaser (e.g. recycler). The requirements for a packaging to be classified as recoverable by means of material recycling are specified by ISO 18604:2013 [93].

Guidelines and assessment procedures on the further development of both packaging and recovery technologies are offered. The assessment of substances and materials constituting a sustained impediment to recycling activities are presented in a non-exhaustive overview by ISO/TR 17098:2013 [94].

A qualitative indicator for the quality of the material supply is proposed:

Is collected and sorted post-consumer bio-based plastic supply characterised according to EN 15347? (Yes/No)

A second qualitative indicator is proposed specifically for bio-based plastic packaging:

Is collected and sorted post-consumer bio-based plastic packaging classified as recoverable according to ISO 18604? (2013) (Yes/No)

The recycling capability of bio-based plastics in a region is described through the indicator:

Material recycling capability of the region (%) = Annual supply of collected and sorted used bio-based plastics (kt/an) x100 / capacity of the mechanical recycling facility in the region to process the specific bio-based plastic stream (kt/an)

The quantities of collected and sorted post-consumer specific bio-based plastic, available per year, are described through the indicator:

Material supply availability (kt/an) meets a critical mass of bio-based plastic waste available at a steady state (kt/an) (Yes/No)

Recyclates quality: Existing standards define required general characteristics and some optional degradation characteristics of conventional plastic recyclates, applicable also to their bio-based non-biodegradable counterparts (e.g. bioPS: [95]; bioPE: [96];

bioPP: [97]; bioPET: [98]), thus imposing analogous quality specifications for the acceptable bio-based plastic waste streams, (EN 15347:2007 [92]).

The characterization of the recyclates of conventional plastics and drop-ins includes the following characteristics [95, 96, 97, 98]:

- a) *Required characteristics: Colour (visual); Density (EN ISO 1183-1-A); Impact Strength (EN ISO 179-1,2 or EN ISO 180); Melt Flow Rate (EN ISO 1133-M); Shape (visual)*
- b) *Optional characteristics: Mechanical tests: Stress at yield, Elongation at break (EN ISO 527-1,2), Flexural properties (EN ISO 178); Bulk density; Ash content (EN ISO 3451-1); Volatile content (EN 12099); Recycled content (EN 15343:2007 [99]); Extraneous polymers (Thermal/ Infrared analyses with Differential Scanning Calorimetry (DSC) and Fourier-transform infrared spectroscopy (FTIR)); Filtration level (Mesh size).*

The quality specifications of EN 15347:2007 [92] may also be adopted for the acceptable bio-based biodegradable plastic waste streams. A major issue is the lack of standard test methods for the evaluation of the quality of the recyclates of bio-based biodegradable plastics. Guidelines for the development of standards and standard specifications for plastic waste recovery, including mechanical recycling, are provided by EN 15343:2007 [99] and ISO 15270:2008 [100]. Concerning the plastics recycle characterisation, EN 15343:2007 [99] refers to the relevant standards [95, 96, 97, 98] in combination with traceability and quality control. ISO 15270:2008 [100] standard establishes recommendations of general applicability for recycled material including criteria for acceptance: identification, data on additives, fillers, reinforcements and composition, e.g. nature and concentration of contaminants and the content of identified polymers and recyclates, mechanical physical and chemical properties and packaging

requirements. In addition, it is recommended that a) the recyclate meets at least the minimum material and end-use performance criteria; b) the specifications of products and the standards of plastic materials should not be compromised to facilitate, and also should not inhibit, the use of recyclate. The technical report CEN/TR 15353:2007 [¹⁰¹], also provides guidelines for drafting standards relating to the proper use of recycled plastics. These recommendations should be followed to develop standards for the quality of recyclates produced from post-consumer/industrial bio-based biodegradable plastics.

The European Parliament, in a latest resolution [⁹], calls for quality standards that will allow for building trust and for providing incentives to support the secondary plastics market. Concerning the development of quality standards, it recommends considering the various grades of recyclates to be compatible with the different products functionalities, while they meet safety requirements for the public health, food contact and the environment, by promoting innovations. Another important issue raised by the European Parliament [⁹] concerns the need to encourage the certification of recycled materials by independent third parties, aiming at their quality characteristics verification and at boosting the confidence of the market, industry and consumers, in the recycled materials.

A proposed qualitative indicator for the quality of recyclates of bio-based non-biodegradable waste plastics is that the recyclates are characterized according to the standards of their conventional counter parts:

Are bio-based non-biodegradable recyclates characterized according to the standards of their conventional counter parts? (Yes/No)

For the recyclates of bio-based biodegradable plastics comparison of their properties to the virgin materials may be adopted as a qualitative indicator until more specific standards develop:

Are bio-based biodegradable recyclates characterized in comparison to their virgin materials? (Yes/No)

The quality specifications of EN 15347 [92] represent an additional qualitative indicator for the acceptable bio-based plastic waste streams:

Are accepted bio-based plastic waste streams characterized according to EN 15347? (Yes/No)

Market of bio-based recyclates: The market price for a specific quality of bio-based recyclates is a crucial factor of the economic viability criterion. Does a market exist for recyclates of bio-based products? If a market exists, then besides the quantity of the produced recyclate, its quality is also crucial. The recyclate's market depends on the recyclates' quantity and quality. In case of a limited market for recyclates and/or deteriorated properties of the recyclate, the prices obtained do not support the mechanical recycling of bio-based plastics. In that case, mechanical recycling may be feasible but not economically viable and so not an optimum EoL option for bio-based plastics [55, 52]. According to Nature Works [44], it is often cheaper to make new virgin plastics from fossil-based materials than to recycle and use the recyclates for production of plastics. This statement does not consider the environmental sustainability of the two options. However, Nature Works' statement does not consider the environmental sustainability between the two options: a) producing fossil-based virgin plastic, versus b) recycling the used plastic and using it for production of second-generation plastics.

The demand for recycled plastics remains weak in the EU because of export restrictions, and the ability of a sufficient and reliable supply of recyclable materials meeting quality specifications. Sorting improvement and recycling capacity increase in the EU, are set as high priorities [9], aiming at enhancing the quality of the post-consumer plastic inputs and subsequently, at creating viable markets for high quality recyclates and recycled plastics. A second action is improving the plastics packaging design for recyclability, aimed at increasing the recycling levels and therefore decreasing the cost of recycling plastic packaging waste by half [9]. These actions also apply to bio-based plastics.

The variability of price of conventional (oil based) plastics and the plastic waste stream correlates well with the variability of the oil price [102]. This suggests to adopt as indicator for the valuation of the recyclate stream, the ratio of the price of the recyclate over that of the virgin polymer. A proposed quantitative indicator refers to the market price for a specific quality of bio-based recyclates:

Relative value (%) = price of bio-based plastic recyclate (€/kg recyclate)/price of virgin polymer (€/kg Virgin plastic)

Financial feasibility: The financial feasibility encompasses the profitability of all stakeholders involved in the operation: The mechanical recycler, the local community it serves and the industry using the produced recyclates as secondary raw materials.

The profitability of the mechanical recycler is necessary for his viability. Limited data are available in the literature for the economic profitability of mechanical recycling of conventional plastics and drop-ins [103, 104, 102] and no data are available for the mechanical recycling of bio-based biodegradable plastics.

The financial feasibility of mechanical recycling of conventional and bio-based plastics may be described in terms of profitability. Two indicators are proposed: The return on

investment (ROI) measured as a percent (net investment/ initial investment) and the Net Present Value (NPV) which is the present value of benefits minus costs [105].

$$ROI (\%) = \text{net investment } (\text{€}) \times 100 / \text{initial investment } (\text{€})$$

$$NPV (\text{€}) = \text{present value of benefits } (\text{€}) - \text{costs } (\text{€})$$

The profitability of the local community can be estimated in a similar way (by similar indicators like NPV) by monetizing the social and environmental benefits and costs associated with the presence of the mechanical recycling operation. This approach should be consistent with the life cycle costing methodology (LCC) with an effort to quantify parameters using subjective values that implicitly reflect the priorities of the community.

Economic viability TESA criterion indicators: A brief overview of proposed indicators for the “Economic Viability” TESA criterion of mechanical recycling of post-consumer/industrial bio-based plastics is shown in Table 3.

Table 3. Proposed indicators for the economic viability TESA criterion

Indicators	Metrics
Economic Viability TESA Criterion	
Availability of mechanical recycling facilities	<i>Availability of mechanical recycling facilities with operating permits (Yes/No)</i>
Availability of collected and sorted industrial/post-consumer bio-based plastics	<p><i>Is collected and sorted post-consumer bio-based plastic supply characterised according to EN 15347? (Yes/No)</i></p> <p><i>Is collected and sorted post-consumer bio-based plastic packaging classified as recoverable according to ISO 18604 (2013)? (Yes/No)</i></p> <p>Material recycling capability of the region (%) = Annual supply of collected and sorted used bio-based plastics (kt/an) x100 / capacity of the mechanical recycling facility in the region to process the specific bio-based plastic stream (kt/an)</p> <p><i>Material supply availability (kt/an) meets a critical mass of bio-based plastic waste available at a steady state (kt/an) (Yes/No)</i></p>
Recyclates quality characterization	<p><i>Are bio-based non-biodegradable recyclates characterized according to the standards of their conventional counter parts? (Yes/No)</i></p> <p><i>Are bio-based biodegradable recyclates characterized in comparison to their virgin materials? (Yes/No)</i></p> <p><i>Are accepted bio-based plastic waste streams characterized according to</i></p>

	<i>EN 15347? (Yes/No)</i>
Market of bio-based recyclates	Relative value (%) = price of bio-based plastic recyclate (€/kg recyclate)/price of virgin polymer (€/kg Virgin plastic)
Financial feasibility	Return on Investment (ROI) Net Present Value (NPV)

3.4 Common environmental and techno-economic criterion

Recyclate mass recovery efficiency: As far as the main output (recyclate) is concerned, a critical point is if the recycling process of bio-based plastics is efficient in terms of recyclate yield. The mass efficiency of the mechanical recycling process of post-consumer/industrial bio-based plastics may be measured by the ratio of the recyclate mass produced to the initial amount of the bio-based plastic mass entering the recycling facility. The higher the recyclate mass recovery efficiency is, the higher the feedstock recovery and recirculation potential achieved. Proposed quantitative indicator for the reprocessing stage efficiency:

$$\eta_{mr} (\%) = \text{mass recyclate (kg/an)} \times 100 / \text{mass of sorted post-consumer/industrial product (kg/an)}$$

This indicator should be combined with the sorting process efficiency indicator of the collected quantity to determine the overall efficiency of recycling of a post-consumer waste stream. For example, in the case of municipal mixed plastic waste, the characteristic sorting efficiency is $\eta_{\text{sort}}=0.75$ and the reprocessing stage efficiency is $\eta_{\text{rep}}=0.88$ for sorted (bio)PE and $\eta_{\text{rep}}=0.76$ for sorted (bio)PET [106]. The overall efficiency for recycling of (bio)PE and (bio)PET waste is 0.66 and 0.57, respectively.

Additives impact on sustainability: Several types of additives may be used to enhance the processability and stabilize the polymers against thermal degradation. The additives used by the plastics industry may be categorized into functional additives (e.g. stabilisers, plasticizers, lubricants, biocides, etc.), colorants (e.g. pigments, etc.), fillers

(e.g. clay, kaolin, calcium carbonate, etc.), and reinforcements (e.g. glass fibres, carbon fibres etc.) [107, 108]. A major concern for the mechanical recycling of plastic products, including bio-based plastics, is the possible transfer of additives containing potentially toxic substances (PoTSs) (e.g. persistent organic pollutants (POPs), brominated flame retardants (BFRs), phthalates, etc.) into the recyclates and eventually into recycled products that may be used in various sensitive applications including toys, food contact materials and household items [109, 110, 111]. Guidance on the global monitoring plan for persistent organic pollutants has been developed within the Stockholm Convention process [112]. The guidance addresses the recycling sorting, separation and management of plastics that contain POPs and BFRs based on the best available technique/best environmental practice (BAT/BEP).

Another possible complication concerns the presence of metal-containing additives. The metal salts or oxides may result in the formation of pro-oxidants that trigger the degradation of plastics during reprocessing and recycled products during use [113]. Hazardous materials present in plastic products vary depending on the product [110]. According to the latest version of the restriction of the use of certain hazardous substances in electrical and electronic equipment, RoHS 3 directive (EU) 2017/2102 [114, 115], maximum concentration values tolerated by weight in homogeneous materials were set for 10 Hazardous substances: Pb, Hg and Cr⁶⁺: 1000 ppm; Cd: 100 ppm; Polybrominated biphenyls (PBB), Polybrominated diphenyl ethers (PBDE), Bis(2-ethylhexyl), phthalate (DEHP), Butyl benzyl phthalate (BBP), Dibutyl phthalate (DBP) and Diisobutyl phthalate (DIBP): 1000 ppm. Four of these substances are phthalates used as plasticizers (DEHP accounts for more than 50% of plasticizers). Some of these restrictions are applicable in 2021 to other areas (e.g. medical devices, monitoring

instruments) as well as to controlled treatment and recycling of plastics and bio-based plastics.

Challenges are faced with recycling of products containing flame retardants (e.g. various types of brominated flame retardants (BFR)) [116]. Several antioxidants (e.g. phenolic or phosphite) used with food packaging have been found to contain heavy metals, labelled as R53 or S61 [117], while others are reported for carcinogenic effect [118]. Molecules missing from the additive data sheets that are included on the positive list of substances that may be used in food contact material have also been identified (possible stabilisers or intermediates). From the large variety of plasticizers used, substances were identified that are not included in the positive list of allowed substances (e.g. some types of phtalates) and some of them are toxic [118]. Compounds that are characterized as non-intentionally added substances (NIAS) have been found in plastic packaging coming from impurities, by-products etc.

It has been shown that some additives used in the plastics manufacturing are not very stable, and they are degraded during use (e.g. antioxidants) leading to post-consumer plastics with different chemicals than the original material.

Various additives are also added during the mechanical recycling processing of plastics (including bio-based plastics). The main additives used are stabilisers, reactive molecules (e.g. chain extenders, repair systems) and compatibilisers [108].

For bio-based plastics, the use of environmentally benign additives would contribute to the overall sustainability of the mechanical recycling process [119]. The additives should be preferably bio-based and in the case of biodegradable polymers, also biodegradable. For the case of recycled bio-based plastic intended to be used as food contact material

(FCM) the restrictions of the relevant directives apply, including the additives used [120, 121]. A relevant indicator on the use of additives is:

$$\eta_{add} (\%) = \text{mass of bio-based or natural environmentally benign (biodegradable) additives (kg)} \times 100 / \text{mass of total additives (kg)}$$

Potential use of environmentally non benign or toxic additives should be reported separately.

Resources utilisation efficiency: The resources used efficiency contributes to the overall environmental and techno-economic sustainability of the mechanical recycling process. The energy used, may be derived from renewable and/or conventional sources while the water used may partially come from a closed recycling loop of the wastewater [122]. Proposed indicators for the utilization efficiency of resources:

- *Total water consumption efficiency:* Total water used in the reprocessing of the post-consumer/industrial bio-based plastics, with respect to the recyclate mass produced over a specific period:

$$\eta_{water} (\text{kg/kg}) = \text{Total water consumption for the recycling process (kg)} / \text{mass of recyclates (kg)}$$

- *Water use efficiency:* The mass of water recovered through water recycling and collected rain water to the total mass of water used. This provides a quantitative measure of water conservation within the process:

$$\eta_{Rwater} (\%) = \text{quantity of water recycled and collected rain water (kg)} \times 100 / \text{quantity of total water usage (kg)}$$

- *Total energy consumption efficiency:* Total energy used, including fossil-derived plus renewable - internally derived energy, in the recycling of post-consumer /industrial bio-based plastics with respect to the recyclate mass produced over a specific period:

$\eta_{energy} (kWh/kg) = \text{Total energy consumption (kWh) for the recycling process} / \text{mass of recyclates produced (kg)}$

- *Renewable energy use*: Provides evidence on renewable energy (internally produced or externally provided) usage as related to the total energy consumption:

$\eta_{Renewable} (\%) = \text{Energy derived from renewable sources (kWh)} \times 100 / \text{total of energy consumption (kWh)}$

Waste – Emissions impact on sustainability: Apart from the main output (recyclate), emissions and the management of the waste streams (e.g. wastewater and solids) are important common criteria [122]. According to directive 2010/75/EU (IED) [91], the physical and chemical characteristics and the polluting potential of the industrial residues shall be determined by measuring the total soluble fraction and heavy metals soluble fraction, before deciding about their possible recycling or disposal routes. The possibility of hazardous substances released to the environment, which may contribute to chronic health effects or cancer risks, depends on the polymers and additive pyrolysis at the reprocessing operating temperatures. Among the substances that may be released included are: toxic metals, volatile organic compounds (VOCs), polycyclic aromatic hydrocarbons (PAHs), phthalates, phthalate esters (PAEs), PAHs polybrominated dibenzo-p-dioxins and furans (PBDD/F), polybrominated diphenyl ethers (PBDEs), etc. [108, 111, 123]. Possible health effects (e.g. due to long exposure to PAEs inside the industry) and the environmental impact should be evaluated [124].

During the melting process of polymers, hazardous substances may be emitted which are not only attributed to the polymer degradation but can be derived also from the additives. Hazardous compounds can be, among others, VOCs. VOC emissions may

occur even at low temperatures from volatile additives such as antioxidants, UV stabilizers, and low-molecular-weight plasticizers.

It has been found that the higher the temperatures used for the polymers processing, the higher is the quantity of emitted VOCs. Lower temperatures and lower oxygen level can reduce the emissions of these compounds during the melting stage of the recycling process. During the melting process of LDPE, for example, butylated hydroxytoluene (BHT) is emitted, which is a kind of most used antioxidants in polymers [125, 126]. Reprocessing bio-based biodegradable plastics at lower temperatures is also associated with reduced emissions. Proposed indicators:

- *Hazardous VOCs emitted*: The ratio of hazardous VOCs, defined according to relevant regulations and standards for specific applications, to the recyclates mass:

$$\text{VOCs (kg/kg)} = \text{quantity of hazardous VOCs (kg)} / \text{mass of recyclate produced (kg)}$$

- *Hazardous non-volatile compounds released to the environment*: The ratio of hazardous non-VOCs defined according to relevant regulations and standards for specific applications, to the recyclates mass:

$$\text{Hazardous non-VOCs (kg/kg)} = \text{quantity of hazardous non-volatile substances (kg)} / \text{mass of recyclate produced (kg)}$$

Other LCA related emissions attributed to the recyclates mass produced shall be taken into consideration through the environmental sustainability criteria and indicators.

Common environmental and techno-economic TESA criterion indicators: A brief overview of the proposed indicators for the “Common Environmental and Techno-economic” TESA criterion of mechanical recycling of post-consumer/industrial bio-based plastics is shown in Table 4.

Table 4. Proposed indicators for common environmental and techno-economic TESA criterion

Indicators	Metrics
Common environmental and techno-economic TESA Criterion	
Recyclate mass recovery efficiency	$\eta_{mr} (\%) = \text{mass recyclate (kg/an)} \times 100 / \text{mass of sorted post-consumer, industrial product (kg/an)}$
Additives impact on sustainability	$\eta_{add} (\%) = \text{mass of bio-based or natural environmentally benign (biodegradable) additives (kg)} \times 100 / \text{mass of total additives (kg)}$
Resources utilisation efficiency	$\eta_{water} (\mathbf{kg/kg}) = \text{Total water consumption for the recycling process (kg)} / \text{mass of recyclates (kg)}$ <hr/> $\eta_{Rwater} (\%) = \text{quantity of water recycled and collected rain water (kg)} \times 100 / \text{quantity of total water usage (kg)}$ $\eta_{energy} (\mathbf{kWh/kg}) = \text{Total energy consumption (kWh) for the recycling process} / \text{mass of recyclates produced (kg)}$ $\eta_{Renergy} (\%) = \text{Energy derived from renewable sources (kWh)} \times 100 / \text{total of energy consumption (kWh)}$
Waste – Emissions impact on sustainability	$\mathbf{VOCs (kg/kg)} = \text{quantity of hazardous VOCs (kg)} / \text{mass of recyclate produced (kg)}$ $\mathbf{Hazardous non-VOCs (kg/kg)} = \text{quantity of hazardous non-volatile substances (kg)} / \text{mass of recyclate produced (kg)}$

3.5 Recirculation potential criterion

A gap exists between product design, materials supply, marketing and manufacturing and the return flow of recycled/recovered materials [10]. This fragmentation has been recognized as a major missing link in the circular economy [9]. To integrate the fragmented cycle and allow for the circular economy to develop, new rules have been proposed by the European Commission including “*more closely harmonised rules on the use of extended producer responsibility (EPR)*”. This requires a new approach for the recycling of post-consumer bio-based products, where the currently fragmented EoL routes, disconnected from the manufacturing industry, are integrated together into a holistic materials recovery, recirculation and valorisation system through industrial symbiosis.

The recirculation through industrial synergies requires the implementation of the concept of “eco-design” that aims at ensuring the bio-based plastic products sustainability throughout their life cycle and beyond. The eco-design concept is an integral design, conceived at the development phase, which integrates functionalities and aesthetics of a product, considered already by the contemporary design concepts, with social and safety issues, economic value and environmental impact for the whole life cycle, including the post-consumer phase [127]. Concerning the EoL priorities of CEP [6], a special emphasis should be placed on the product design for easy and quick disassembly into the product components. The EU Directive 2009/125/EC [128], amended in 2012 and 2019, has established a framework of eco-design requirements for energy-related products. Analogous EU legislation is urgently needed for the eco-design of bio-based plastic products.

The so-called eco-effectiveness and cradle-to-cradle design concept has also been proposed as an alternative design and production concept to the strategies of zero emission and ecoefficiency [129]. In fact, eco-effectiveness, like the eco-design concept, focuses on developing products and industrial systems with enhanced quality and productivity of materials through recirculation in new life cycles. The eco-effectiveness process introduces an eco-effective management system through the intelligent materials pooling concept to coordinate the recovered material flows amongst actors in the manufacturing system.

A special recirculation category is the “closed-loop mechanical recycling” where the recycle of the post-consumer conventional and bio-based plastic product is processed back into the same type of final plastic product (e.g. PLA bottle) independently of manufacturing facility [130].

Factors affecting the recirculation potential of recovered materials: The recyclability properties of a material are deteriorated during recirculation through multiple mechanical recycling reprocessing cycles [10,131]. The polymer deterioration (degradation) in most cases is gradual until reaching a maximum number of possible reprocessing cycles before becoming mechanically non-recyclable anymore. The limiting reprocessing number of cycles depends on the specific material and processing conditions. The degree of deterioration of the bio-based materials' properties as a function of the reprocessing cycles is an important techno-economic criterion that defines the recirculation potential of each material through mechanical recycling [55, 132, 10]. The recirculation potential of recycled bio-based plastics can be enhanced by means of blending with virgin or other bio-based polymers, additives, fillers and nanoparticles [55, 66, 10, 40]. Taking into consideration the fact that the mechanical and physical properties of plastics, including bio-based plastics, are degraded with the number of reprocessing cycles the question of recirculation of seriously degraded recyclates needs to be addressed (e.g. the (bio)PET ductility decreases from 310% to 218% and 2.9% after 1 and 3 cycles [133]). Degraded (bio)PET recyclates with elongation at break 3% can only be used in low-value products (e.g. carpets) [133] while they cannot be mechanically recycled anymore because of the complex structure of these products due to the presence of fibres and rubber backing. A synoptic literature review of the degradation of post-consumer bio-based biodegradable plastics due to repeated processing is presented in Table 5.

Non-recyclable reprocessed polymers used in various post-consumer products, including composite products, can be further recovered through chemical recycling and recirculated in the form of monomers or oligomers [10].

Proposed qualitative indicators:

Recirculation potential of the recyclate (Y/N)

Proposed quantitative indicator:

*Maximum number of possible reprocessing cycles defined according to literature
or research*

Table 5. Synoptic review of literature on the degradation characteristics of bio-based biodegradable plastics under multiple reprocessing

Multi-extruded PLA					
Material	Reprocessing cycles / conditions	Mechanical properties	Physical properties	Polymer Degradation	Reference
Repeated extrusion of bio-based polymers – PLA grades					
PLA (Ingeo™ 2002D (L: D-Lactide 95.75: 4.25%))	10 (<i>repeated extrusions</i>)	σ_u : -5.2% ϵ_{br} : -2.2 to -2.4% Charpy: -20.2%	MFR: +236% WVTR: +39%; OTR: +18% T_{cc} : -7,4%; T_m : -1,6%; T_g : -		[65]
PLA (Ingeo™ 2002D)	3 (<i>reprocessing cycles under hygrothermal ageing conditions</i>)	Mechanical performance: <i>general loss</i>	WVTR: <i>Decr</i> WVTR (<i>hygrothermal ageing</i>): <i>Incr</i> Thermal performance: <i>general loss</i> T_g : -7,5% to -7,8%	Poor recyclate quality (hydrolytic chain scission increase with reprocessing, higher temperature; MW: <i>Decr</i>) Crystallites formed faster at higher reprocessing cycles, lower hygrothermal ageing temperatures	[134, 135]
PLA (Ingeo™ 2002D)	5 (<i>injection reprocessing cycles</i>)	σ_u : -7% Modulus E: -28% Charpy: -19%	Cold-crystallization and significant loss of PLA performance after 2 nd cycle	Chain scission, M_w : -50% Remained amorphous	[136]
PLA (Ingeo™ 3051D) and PLA/PC blend	<i>Multiple reprocessing of artificially aged PLA via injection moulding</i> <i>Post-consumer recycling of neat PLA and PLA/PC blend was simulated by accelerated ageing in humid air followed by reprocessing</i>	No effect on Izod impact after 6 cycles Mechanical properties: (<i>not possible to measure after ageing in moist air; PLA severely degraded after only one ageing cycle</i>) Mechanical properties: <i>improved (for PLA/PC blend)</i>	MFR (6 cycles): +5%; (<i>after ageing of PLA in moist air</i>): 100%; (<i>PLA after a second cycle</i>): <i>not possible to handle the degraded PLA</i> T_g : (<i>PLA and PLA/PC</i>): <i>no effect</i> T_m : (<i>PLA</i>): <i>no effect</i> ; (<i>blend PLA/PC</i>): <i>significant increase</i> T_{cc} : (<i>2-6 cycles for blend PLA/PC</i>): <i>Decr</i> Crystallization ability: <i>increase in cold crystallization with extrusion cycle due to shorter chains</i>	Degradation: (<i>pure PLA completely degraded after only one ageing cycle corresponding to 1 year of service at ambient conditions; PLA/PC blend showed some improved resistance to degradation: one ageing cycle still caused severe degradation of the PLA part and even the PC part</i>) MW: <i>Major reduction</i>	[137]

			T_g , T_g , T_{cc} : major decrease after ageing with 0, 1, 6 extrusions Miscibility: not miscible components; PC acted as nucleating agent		
PLA (Ingeo™ 2003D)	Recycled and reprocessed following accelerated ageing and then reprocessed following a washing step at 85 °C.	Hardness of reprocessed PLA (virgin): +5%; (aged): +15%; (aged and washed): +13%	Water absorption: <i>Incr with time (90 days)</i> : +1,2% at 37 °C; +2.1% after washing cycle; smaller at 58 °C Intrinsic viscosity after 84 days immersion: (virgin and reprocessed): -30% T_{cc} = -6.0% to - 8.0% Slight crystallization of PLA at 37 °C	Water absorption and hydrolytic degradation at 58 °C faster than at 37 °C: Crystallization of reprocessed PLA: (4–8%) after 56 days at 37 °C	[138]
PLA (Ingeo™ 2003D)	Recycled and reprocessed following accelerated ageing, then reprocessed following a washing step at 85 °C.	Hardness of reprocessed PLA: (aged): +7%; (aged and washed): +11%	Barrier properties: insignificant effect Intrinsic viscosity after 84 days immersion: -16% following ageing; -20% following also washing step T_g : insignificant T_{cc} : -5°C	PLA can withstand a single recycling process: insignificant loss of molecular weight The introduction of a washing step weakens the polymer structure: degradation during reprocessing step	[139]
PLA (Ingeo™ 4042D (4.2% D-lactide))	Multiple reprocessing of PLA and plasticized pPLA (PLA/acryl-PEG/L101 at 79/20/1)	pPLA (5 cycles): σ_u : -117% at 50 °C, -49% at 80 °C ϵ_{br} : -96% at 50 °C, -99% at 80 °C Izod Impact (3 cycles): -55% PLA (5 cycles): σ_u : -34% at 50 °C ϵ_{br} : -14% at 50 °C Izod Impact (3 cycles): -33%	T_g : stable (up to 5 cycles) T_m : stable (up to 5 cycles) T_{cc} : weak decrease pPLA Crystallinity: +14% PLA Crystallinity: remained amorphous	Main degradation mechanism: PLA: simple random chain scission pPLA: transformation of poly(acrylpoly(acryl-PEG) inclusions, formation of porosity and fibrillation; formation of cracks in the polymer matrix pPLA MW: strong >-97% PLA MW: -47%	[140]
PLA (Ingeo™ 2002D)	5 (repeated extrusions)		T_g : -, T_m : small decrease MFR: +14%	Small degree of thermal degradation with reprocessing	[141]
PLLA (Biomer L9000 (L:D-Lactide 92:8);	7 (repeated injection moulding cycles)	σ_u : -62% ϵ_{br} : -87%	Viscosity: --82% in one cycle; -99% after 7 cycles	Poor recycle quality Chain scissions MW: -50% after 3	[142] -

<i>amorphous, no stabilisers</i>)		Hardness: -15% Modulus: -7%	T _g : -15% T _m : -2.5%	<i>cycles; -63% after 7 cycles</i> Stabilizers suppress Crystallization is suppressed with stabilizers that trap free radicals	
PLLA (Biomer L9000)	5-8 (<i>reprocessing cycles via injection moulding</i>)	Acceptable loss after 5 cycles: σ_u : -8.4%, ϵ_{br} : -11.8%, Izod : -14.4% total loss after 8 cycles: σ_u : -85.8%, Izod: -56.4%	Acceptable loss up to 4-5 cycles MFR after 8 cycles: +1417%	Reprocessing beyond 4–5 cycles is not recommended: higher <i>fluidity and lower melting temperature</i> MW: -87% after 8 cycles	[64]
Repeated extrusion of bio-based polymers – Various grades of bio-based polymers other than PLA grades					
Mater-Bi (TF01U/095R; based on aliphatic polyester; Novamont)	10 (<i>multiple reprocessing</i>)	No significant loss up to 10 cycles	No significant loss up to 10 cycles		[64]
Mater-Bi (YI014U/C; thermoplastic formulation based on starch; Novamont)	2 (<i>multiple reprocessing</i>)	Total loss	Total loss	Serious degradation of ductility (2 cycles); Should be destined to organic recycling May be reprocessed as a blend with virgin polymer at high rate	[64]
PHBV Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (Biopol, 4% valerate; Zeneca Bioproducts)	5 (<i>multiple melt reprocessing</i>)	σ_u : slight decrease of -7.1% after 5 cycles Izod impact: No effect after 5 cycles	Crystallinity: -21% after 5 cycles T _g : no change T _m : no change	Thermal decomposition: slight MW: -8.7%, -13.5% and -16.6%, after 3,4,5 cycles, respectively	[143]
PHB copolymer *	10 (<i>multiple regrind reprocessing</i>)	σ_u : -10% for the 10 th regrind generation	Viscosity: -79% for the 10 th regrind generation		[66, 80]
PHB powder (BIOCYCLE®, weight-average (Mw) of 167223 g.mol ⁻¹)	3 (<i>multiple reprocessing</i>)	Mechanical properties: (significant reduction at 2 nd cycle; > -50% at 3 rd cycle	Crystallinity: Increase (due to crystallization process) No significant changes in the chemical structure or in thermal stability	Degradation by chain scission	[144]

Repeated extrusion of PLA composites and blends – PLA grades-based composites and blends					
PLA (Ingeo™ 2003D); natural and reinforced with melt strength enhancer (PLA BS), silicate nanoclays (PLA nano)	20 (repeated extrusions, extracting one sample each four extrusions)	Mechanical properties: No remarkable loss for the natural PLA, PLA BS Flexural and Tensile Moduli on PLA nano increase with the number of extrusions	Viscosity: (higher decrease at 190 °C): PLA - 70%, PLA BS -42%, PLA nano -56% For less than 8 extrusions better results for PLA nano than PLA BS Great decrease on viscosity after 20 extrusions Mostly linear decrease that increases with temperature (170-190 °C)	Polymer chain degradation due to reprocessing of PLA Reprocessing of PLA BS is significantly enhanced	[145]
PLA (Ingeo™ 2003D) compound with 30% chalk (Schämmkreide 32), 5% of a bio-based plasticiser	Multiple extrusions	Tensile (σ_u , ϵ_{br} , E) and flexural properties: no major degradation after 6 cycles	Thermal properties T_{cc} , T_g : no significant changes after 6 cycles T_m : +6% Crystallinity: no significant change	MFR: + 300% NMR: reduction of chain length Degradation: PLA compounded with chalk can be recycled by repeated extrusion for up to 6 cycles, without severe degradation.	[146]
Recycled PLA (Ingeo™ 3251D) compound with organically-modified nanoclays	First extrusion and compression molding process and accelerated aging of PLA, a demanding washing step and, finally, a second melt processing step of nanocomposites	Mechanical properties: important improvement of the mechanical properties of PLA, exceeding those of PLAV. Hardness: Decr		Degradation: modified clays reduce the degradation of PLA during the reprocessing step and that the clay nanotubes reinforce the recycled PLA matrix utilization of functionalized nanoclays could lead to increasing its recyclability	[40,147]
PLLA (Biomer L9000) compound with flax fibre (20% and 30%)	6 (injection moulding cycles)	σ_u , ϵ_{br} : large drop due to significant reduction in failure properties of the PLLA matrix after multiple injection Modulus E: Slight decrease	T_g : Decrease with injection cycles Crystallinity: Increase with injection cycles Viscosity: Decrease as a function of injection cycles	MW: Decrease as fibre content and number of injection cycles increase Degradation: chain scission mechanisms during recycling Higher fibre content accelerates PLLA degradation during recycling)	[148]
PLA* compound with 30% cellulose fibres (Fibrolon F)	7 (multiple extrusions)	Mechanical properties: no significant loss up to 5 cycles σ_u : - 23% after 7 cycles	T_g : Decrease (degradation of PLA)	Hydrothermal ageing between each extrusion cycle: Samples were aged at 50°C in distilled water for 5 days: The	[149]

8530) (compounding by Fkur)				<i>thermal and mechanical testing showed that the material survived the ageing test fairly well.</i>	
PHBV/PLA (Ingeo™ 7001D): 50/50 (wt%) blends	<i>6 (melt moulding reprocessing process)</i>	Mechanical properties: relatively stable after 6 cycles; Modulus E: (PHBV): -11%; (PLA): -10.8; (PHBV/PLA): -9.6% ϵ_{br} : -24% Izod: -22%	Crystallinity: <i>Incr</i> (PHBV crystallinity enhancement due to PHBV chain mobility; significantly smaller for neat PLA and PHBV-PLA blends) T_g , T_m , T_c and T_{cc} : stable Complex viscosity: <i>Decrease</i> (PHBV drastically reduced, after 6 cycles becomes fluid; PLA: slight decrease; PHBV/PLA better stability than PHBV)	MW: (Decrease after 3 and 6 cycles): PHBV: -13% and -27 %; PLA: -1.2% and 4.2%; PHBV/PLA: -1.1% and 4.8 % Degradation by chain scission (mainly PHBV) Great recyclability of PHBV/PLA blends	[¹⁵⁰]

(* No details for the grade of the polymer are available

Reprocessed materials characterization: Mechanically reprocessed materials differ in quantities and qualities available in the market. The consumers hardly rely on constant high-quality products supplies. The Standards available for characterising recovered post-consumer/industrial conventional or bio-based non-biodegradable plastics [[^{95, 96, 97, 98}] may be used for recyclates produced through several reprocessing cycles. A new standardization process, possibly through ISO 15270:2008 [¹⁰⁰] is needed for the development of standards for the quality of bio-based biodegradable recyclates produced through several reprocessing cycles. The European plastics recycling industry needs uniform standards and certification schemes to strengthen the secondary raw materials market. Proposed qualitative indicator:

Characterization according to standards for recyclates after each cycle (Y/N)

Traceability schemes of secondary materials market: It is necessary for the quality characterization of the recyclates to be traced through a general traceability scheme for the whole life cycle of conventional and bio-based plastics, especially for the recirculation potential of the specific materials. The traceability scheme for secondary materials will allow for a better product control or tracking and withdrawal of unwanted material and/or defective products from the secondary market. It will also allow for the enhancement of confidence between producers and end users for reprocessed /recovered materials, ensuring their recirculation back to the useful life cycle stage.

A traceability scheme for the recyclates can be built on the existing traceability of the products' content, extended throughout their life cycle and reprocessing. The REACH (Registration, Evaluation, Authorisation and Restriction of Chemicals) regulation [¹⁵¹] applies to all chemical substances contained in any product, including bio-based plastics. However, this information may not be available in the sorting and recycling

stages, while plastics sorted out from mixed plastics and aged plastics pose additional complications [130]. Elimination of these problems could be based on advanced methods of the recyclates screening. A key incentive to increase traceability, is labelling of the content of the original plastic products all the way to the entrance of the recycling facility [130].

Specifications for a traceability scheme for bio-based recyclates can be developed following the EN 15343:2007 [99] provisions, applicable to recycled plastics in general. EN 15343:2007 includes procedures for calculating the recycled content of plastic products.

Proposed qualitative indicator:

Post-consumer bio-based plastics are traced through a traceability scheme for the product cycles (Y/N)

Recirculation potential TESA criterion indicators: A brief overview of proposed indicators for the “Recirculation Potential” TESA criterion of mechanical recycling of post-consumer/industrial bio-based plastics is shown in **Table 6**.

Table 6. Proposed indicators for the recirculation potential TESA criterion

Indicators	Metrics
Recirculation potential TESA Criterion	
Factors affecting the recirculation potential of recovered materials	<i>Recirculation potential of the recyclate (Y/N)</i> <i>Maximum number of possible reprocessing cycles defined according to literature or research</i>
Reprocessed materials characterization	<i>Characterization according to standards for recyclates after each cycle (Y/N)</i>
Traceability schemes of secondary materials market	<i>Post-consumer bio-based plastics are traced through a traceability scheme for the product cycles (Y/N)</i>

4 Conclusions

The recirculation potential of post-consumer bio-based plastics through mechanical recycling represents a new challenge.

Non-biodegradable biobased plastics (drop-ins) that have the same chemical characteristics as their fossil-based counterparts, can be recycled through the same conventional recycling schemes. Existing standards defining quality specifications for conventional plastic recyclates are applicable also to the drop-ins.

Bio-based biodegradable plastics can be recycled through established recycling schemes for conventional plastics under certain conditions. Recyclers are currently reluctant, considering them as contaminants for reasons of incompatibility. However, provided that the bio-based polymer waste stream reaches a critical quantity and if the sorting efficiency achieved is high, it may turn the mechanical recycling as the most attractive EoL option for bio-based biodegradable plastics. The separation of compostable from non-compostable bio-based plastic waste, at the source is crucial.

The biodegradable bio-based products can be sorted out in separate mono streams by using existing techniques (e.g. NIR technology). A major issue is the lack of standard test methods for the evaluation of the quality of the recyclates produced from bio-based biodegradable plastics. The recommendations provided by ISO 15270:2008 [100] and CEN/TR 15353:2007 [101], should be followed to develop standards for the quality of recyclates produced from post-consumer/industrial bio-based biodegradable plastics. A new standardization process, possibly through ISO 15270:2008 [100] is needed for characterising recyclates produced from bio-based biodegradable plastics through several reprocessing cycles. A traceability scheme for bio-based recyclates can be developed following the provisions of EN 15343:2007 [99].

The new design approaches should take full advantage of the unique characteristics of the bio-based polymers, incorporating the targeted valorisation options of the post-consumer product at the design stage, so as to maximize its recirculation potential. Even though the post-consumer quantities of bio-based biodegradable waste streams are rather low, the bio-based plastics sector develops dynamically, and the need to investigate and integrate the preferred EoL material recovery routes through mechanical, and possibly chemical, recycling to the products design is imminent. The possibility to support the development of the circular bioeconomy depends very much on the techno-economic (and environmental and social) sustainability of the added value material recovery EoL options and the integration of their recirculation potential into the design of the final bio-based products.

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