Bio-based plastics in a circular economy

A review of recovery pathways and implications for product design

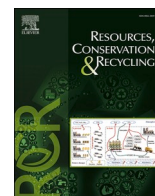Ritzen, Linda; Sprecher, Benjamin; Bakker, Conny; Balkenende, Ruud

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Review

Bio-based plastics in a circular economy: A review of recovery pathways and implications for product design

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A B S T R A C T

Bio-based plastics are attracting increasing attention due to their perceived sustainability and circularity. While enabling circularity by using renewable feedstocks, they still contribute to plastic pollution. Furthermore, their rapidly growing market will cause bio-based plastics to constitute significant fractions of plastic waste, necessitating efficient recovery at end-of-life. Technical overviews of potential recovery pathways for bio-based plastics exist, although these have not yet been translated into product design recommendations. In this article, we assess the impact of material composition and product design on the feasibility of eight recovery pathways for bio-based plastics. The ability to recover a plastic not only depends on the plastic composition, but also on the way a product is designed. The alterations made to tailor plastics to be applied in products, and the product architecture, can enable or prohibit some recovery pathways. The outcomes highlight the importance of establishing a wider range of recovery pathways for plastics, and the crucial role of product design in enabling a circular economy for bio-based plastics. We also present a first guidance for product design to enhance the recovery of bio-based plastics.

1. Introduction

Plastics have become vital for modern life, owing to their low costs and wide range of properties. In recent years, environmental concerns regarding fossil-fuel consumption and pollution in the linear plastics economy have emerged. Plastic production consumes up to 8 % of fossil-fuels extracted annually (Lambert and Wagner, 2017), and it is estimated that 79 % of all plastic ever produced has accumulated in landfills and the natural environment (Geyer et al., 2017), causing irreversible harm (Barnes et al., 2009). A transition to a circular economy has gained traction as a response to these challenges. A circular economy is restorative or regenerative by design, with the aim of eliminating waste (Ellen MacArthur Foundation, 2013).

Bio-based plastics are considered a key component of the circular economy (Ellen MacArthur Foundation, 2015; European Commission, 2019), since they are based on polymers produced (at least partially) from biomass (International Organization for Standardization [ISO], 2015). The renewable nature of bio-based plastics enables circularity at the polymer production level. Nevertheless, using bio-based plastics does not solve the environmental issues of plastics. The environmental impact of bio-based plastic production and end-of-life is still a debated

topic with little data available (Walker and Rothman, 2020). Furthermore, bio-based plastics can still contribute to plastic pollution, as the term 'bio-based' only refers to the sourcing of a polymer and not biodegradability in nature. Therefore, recovery at end-of-life will play a vital role in sustainability and circularity for bio-based plastics. In this article, we limit ourselves to the technical feasibility of bio-based plastics recovery.

In a circular economy, products need to be recovered at their highest possible value. As such, recovery strategies are categorised into a waste hierarchy. At the top of this hierarchy are recovery pathways that focus on product integrity (i.e., product-level recovery pathways), such as maintenance, repair, re-use, and remanufacturing (Ellen MacArthur Foundation, 2013). Bio-based plastics do not perform differently from petrochemical-based plastics in these pathways (Badia et al., 2017). Lower in the hierarchy are material-level recovery pathways, such as recycling. In material focused recovery pathways, the chemical composition of a plastic affects the feasibility and efficiency of recovery. Here, bio-based plastics may perform differently from petrochemical-based plastics. Material-level recovery includes molecular decomposition pathways, where bio-based plastics occupy a special position. Since bio-based plastics are based on renewable resources, if

the plastic is returned to simple molecules through processes such as incineration and biodegradation, they do not contribute fossil carbon dioxide (CO_2) to the atmosphere. Therefore, molecular decomposition of bio-based plastics can be considered a circular loop (Bakker and Balkenende, 2021).

Bio-based plastics are commonly divided into two categories: drop-in and dedicated. Drop-in bio-based polymers are chemically identical to petrochemical-based polymers, whereas dedicated bio-based polymers do not have a petrochemical-based equivalent. Drop-in bio-based polymers can be integrated into existing recycling streams for petrochemical-based equivalents. Dedicated bio-based polymers (Carus et al., 2017) are currently considered a contaminant in plastic waste streams due to their small volumes (Alaerts et al., 2018; Briassoulis et al., 2019). Although bio-based plastics only account for 1 % of annual plastics production, their market is growing at twice the rate of petrochemical-based plastics (Skoczinski et al., 2021). Dedicated bio-based plastics make up roughly 60 % of the bio-based plastics market today, and their share is expected to grow in the near future. New recovery systems need to be established when dedicated bio-based plastics grow into significant fractions of generic plastic waste. The development of the bio-based plastics market further necessitates research into end-of-life management.

Plastic recovery depends not only on recovery infrastructure, but also on product design. Product design covers the entire development process of a product or system to optimise function, value and appearance to benefit users and manufacturers (Industrial Designers Society of America 2023). One of the core principles of the circular economy is that the value of products and the materials they are made of can be preserved by keeping them in the economic system, either by lengthening the life of the products formed from them, or ''looping'' them back in the system to be reused (den Hollander et al., 2017). Decisions made during product design and development affect the ability to recover a product at end-of-life. For example, in material-level recovery, product design influences the ability to separate plastic parts from a product, which is important for material-level recovery.

The importance of product design in the transition to a circular economy with bio-based plastics has already been highlighted (Awasthi et al., 2021; Badia et al., 2017; Bakker and Balkenende, 2021; Briassoulis et al., 2019; Hatti-Kaul et al., 2020; Hildebrandt et al., 2017; Sauerwein et al., 2020). There are technical overviews of possible recovery pathways for bio-based plastics (Badia et al., 2017; Briassoulis et al., 2019; Hatti-Kaul et al., 2020; Hildebrandt et al., 2017; Kawashima et al., 2019; Lamberti et al., 2020; RameshKumar et al., 2020), but, to our knowledge, these reviews have not yet been translated into recommendations for product design.

In this article, we assess how existing and future recovery pathways influence product design with bio-based plastics. An overview and terminology of 8 recovery pathways is established, and detailed descriptions of the state-of-the-art of recovery pathways for bio-based plastics are presented. This allows us to highlight the relevant technical characteristics of specific recovery pathways and assess their implications on material selection and product design.

2. Methodology

The methodology employed in this study is divided into three steps, displayed in the three boxes in Fig. 1.

2.1. Establishing a framework for recovery pathways of bio-based plastics

A rigorous literature review was conducted in Scopus in May 2022 to map the existing recovery pathways for bio-based plastics. The search terms consisted of a combination of the following: (1) synonyms for recovery, namely "end-of-life", "recycling", and "recovery", (2) synonyms for bio-based plastics, including "bio-based polymer", "biopolymer", and "bio-based plastics", yielding 252 articles. Articles presenting an overview of recovery pathways for bio-based plastics were selected, resulting in 7 articles and reports. Snowballing yielded three additional articles. The recovery pathways discussed in these articles were categorised based on their reported definition and combined with ISO standards, resulting in a comprehensive set of 8 recovery pathways and definitions. Finally, a framework was created with these recovery pathways based on how their products feed back into the circular economy.

2.2. Scope

The scope of this research is limited to polymers that are either already produced commercially or expected to become commercially available in the near future (Skoczinski et al., 2021). This resulted in 13 polymers: high-density polyethylene (HDPE), low-density polyethylene
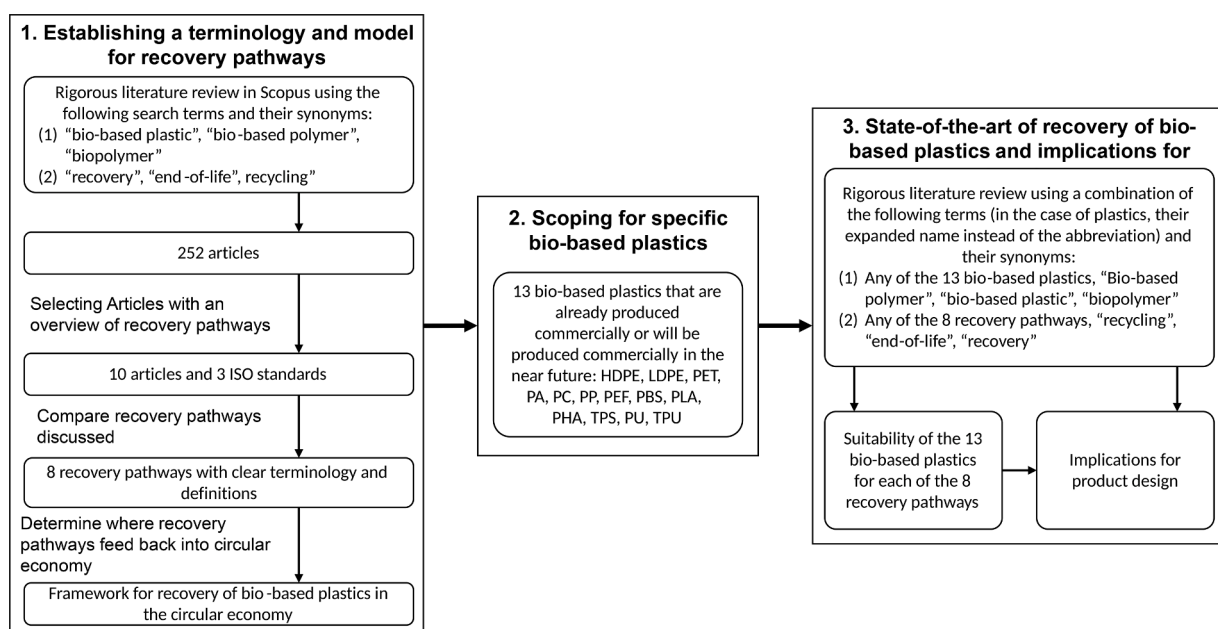

Fig. 1. Visualisation of the methodology employed in this study.

Table 1

Terminology for recovery pathways used in this article.

Selected terminology for specific recovery pathways	Definition	Alternative terms	Umbrella terms covering multiple recovery pathways
Mechanical recycling [a-l, y]	"*Processing of plastic waste into secondary raw material or products without significantly changing the chemical structure and composition of the material*" [o].	Recycling [g]	
Dissolution [c]	The recovery of a polymer through its dissolution and precipitation in a suitable solvent/non-solvent, without any alteration to its molecular structure.	Solvent-based purification [e,l]	Chemical recycling [a-l] Feedstock recycling [a,c]
Solvolysis [c,i]	The cleavage of a polymer by a solvent such as water or alcohol, often in the presence of a catalyst.	Chemolysis [c] Feedstock recycling [a] Depolymerisation [e,l]	
Thermochemical recycling	The dissociation of polymers through high temperatures.	Feedstock recycling [b,e,l] Thermolysis [a] Plastic-to-fuel recycling [b]	
Anaerobic digestion [a,b,c,d,e,j,l]	"*The breakdown of an organic compound by microorganisms in the absence of oxygen to carbon dioxide, methane, water and mineral salts of any other elements present (mineralisation) plus new biomass*" [m].	Anaerobic respiration [a] Anaerobic biodegradation [a,h]	Biodegradation [a,e,g] Composting [g,h] Organic recycling [c] Microbial degradation [f]
Aerobic digestion [b]	"*The breakdown of an organic compound by microorganisms in the presence of oxygen to carbon dioxide, methane, water and mineral salts of any other element present (mineralisation) plus new biomass*" [n].	Aerobic respiration [a] Aerobic biodegradation [a,c,h] Aerobic composting [d,e] Composting [j,l]	
Incineration [a,b,c,d,e,g,h,l]	The combustion of the plastic into mainly carbon dioxide, water and ash.	Energy recovery [d,g,n,i]	

[a] (Badia et al., 2017).
[b] (Bakker and Balkenende, 2021).
[c] (Briassoulis et al., 2019).
[d] (D'Adamo et al., 2020).
[e] (European Commission, 2019).
[f] (Hatti-kaul et al., 2020).
[g] (Hildebrandt et al., 2017).
[h] (Kawashima et al., 2019).
[i] (Lamberti et al., 2020).
[j] (RameshKumar et al., 2020).
[k] (Soroudi and Jakubowicz, 2013).
[l] (Spierling et al., 2020).
[m] (ISO, 2014).
[n] (ISO, 2018).
[o] (ISO, 2013).

(LDPE), polyethylene terephthalate (PET), polyamide (PA), polycarbonate (PC), polypropylene (PP), polyethylene furanoate (PEF), polybutylene succinate (PBS), polylactic acid (PLA), polyhydroxyalkanoate (PHA), thermoplastic starch (TPS), polyurethane (PU), and thermoplastic polyurethane (TPU).

The recovery pathways were limited to material-level recovery pathways, excluding product-level recovery pathways.

2.3. State-of-the-art of recovery of bio-based polymers and plastics

In order to understand how specific recovery pathways are suitable for specific bio-based plastics, a rigorous literature review was conducted in Scopus in June 2022. Search terms were combinations of (1) the recovery pathways and their synonyms, and (2) "bio-based polymer", "bio-based plastic", or any of the 13 bio-based polymers established in Section 2.2. Polymer blends and composites were excluded from the results. Commercial plastics (including additives such as stabilisers and colourants) were included. For drop-in bio-based polymers, the results of (chemically identical) petrochemical-based counterparts were also included.

For product design, it is essential to know which recovery pathways are suitable for specific bio-based plastics and also how these recovery pathways are influenced by product design. Based on the discussed literature, the suitability of specific bio-based plastics for specific recovery pathways was established. Furthermore, specific technical aspects of recovery pathways were highlighted to define implications for product design.

3. Results

3.1. An overview of recovery pathways for bio-based plastics

Table 1 presents an overview of the terminology used for the recovery of bio-based plastics as obtained from existing overviews and frameworks. Terminology in existing literature is often inconsistent for novel recovery pathways, such as chemical recycling and biodegradation. The terminology used throughout this article was established based on the terms used in existing overviews and active ISO standards, as displayed in Table 1.

Fig. 2 presents a framework of the circular economy for bio-based plastics incorporated into products, considering the re-entry points of the recovery pathways established in Table 1. Bio-based plastic product manufacturing was divided into six steps. Simple molecules (e.g. CO_2 and water) in the atmosphere are absorbed by plants and converted into biomass. Specific molecules, such as glucose, are isolated and used as feedstock to produce monomers for polymers. The polymer is often compounded with, for instance, additives or other polymers through blending to yield a plastic that is further manufactured into a product. After the use-phase of the product, the product lifetime is extended through product-level recovery. At the material level, recovery can

occur through the eight established recovery pathways. The re-entry points of recovery pathways are based on their products. For instance, by definition, the main products of aerobic digestion of polymers are CO_2 and H_2O, which are categorised as simple molecules.

Anaerobic digestion of polymers, by definition, yields large amounts of methane, which can be captured as a feedstock for the production of new plastics in an industrial environment. If anaerobic digestion of bio-based polymers occurs in nature, the product could be considered a "simple molecule"; however, plants do not absorb significant amounts of methane during their growth (Cicerone and Oremland, 1988), and therefore, this cannot be considered a circular loop.

Table 2 summarises the result of the literature review and provides a first indication of recovery pathways that may be considered when using a specific bio-based polymer in a product. The selected literature has been categorised into the specific recovery pathway and bio-based polymer each article presents. Details can be found in the supplementary information (Tables S1–S6). Recovery pathways for polymers that currently represent a large part of the (petrochemical-based) plastic waste composition, such as bio-HDPE, bio-LDPE, bio-PET and bio-PP, have already been extensively studied. Furthermore, the recovery of commodity-grade dedicated bio-based polymers is well understood, with the exception of PEF.

However, for bio-based polymers classified as engineering-grade (bio-PC, bio-PA, bio-PU and bio-TPU), the opportunities and effects of recovery have not yet been studied. Recovery of these polymers through various technologies is theoretically possible, but the rigorous literature review did not yield any articles in which the possibilities have been reported. It should be noted that although the engineering-grade bio-based polymers presented here share a name with petrochemical-based polymers, they are not chemically identical (Cywar et al., 2022).

3.2. State-of-the-art of recovery pathways for bio-based plastics

3.2.1. Mechanical recycling

Mechanical recycling is the "*processing of plastic waste into secondary raw material or products without significantly changing the chemical structure of the material*" (ISO, 2013). In order to be suitable for mechanical recycling, a plastic must be melt-processable, i.e. a thermoplastic, and withstand the conditions under which mechanical recycling occurs. Mechanical recycling generally consists of the following steps: sorting, shredding, washing and drying, and reprocessing (Worrell and Reuter,

2014). During sorting, different types of plastic are separated, and impurities are removed. Next, the plastic is washed and dried before reprocessing, where granulate or new products are produced from recycled plastics using conventional melt-processing techniques. Thermomechanical stresses during reprocessing can change the molecular structure of a polymer through chain scission, oxidation or a reaction with contaminants, additives or dissociated pieces of the polymer itself. These changes can result in a recycled plastic with different properties than virgin plastic, which cannot directly replace virgin plastics (Bakker and Balkenende, 2021; Roosenboom et al., 2022). Contaminants and impurities cause degradation in plastic processing during reprocessing, and thus sorting accuracy affects the quality of recycled plastics. Currently, the most used separation techniques are air and float-sink separation, which are inaccurate when different plastics have a similar density; other techniques, such as near-infrared (NIR) sorting, can improve sorting accuracy in the future (Serranti and Bonifazi, 2019).

Table 3 summarises the effects of mechanical recycling, specifically reprocessing, on drop-in and dedicated bio-based polymers. Some drop-in bio-based plastics, namely bio-HDPE, bio-LDPE, bio-PET and bio-PP, are known to be good candidates for mechanical recycling due to extensive research into their petrochemical-based counterparts. Since bio-HDPE, bio-LDPE, bio-Pet and bio-PP are chemically identical to their petrochemical-based counterparts, the results of this research also apply to the bio-based versions. Mechanical recycling of PLA has been investigated extensively and is applied on a small scale through at-home recyclers for 3D printed parts (Beltrán et al., 2021). The thermal properties and processing window of recycled PLA do not differ from its virgin form, but barrier, tensile and impact properties degrade rapidly upon reprocessing (Badia et al., 2012b; Żenkiewicz et al., 2009). PHAs show a sharp decline in many properties after mechanical recycling (Vu et al., 2020). (Bio-)PBS withstands mechanical recycling without a significant change in the molecular structure (Georgousopoulou et al., 2016) in bending strength or bending modulus (Kanemura et al., 2012), although the effect on impact properties has not yet been reported. The degree of degradation due to mechanical recycling is affected by the processing conditions. The processing conditions used to study the effects of mechanical recycling on a plastic in the studies in Table 3 varied, which also affected the outcomes. A detailed overview of these results and the processing conditions can be found in the supplementary information (table S1).
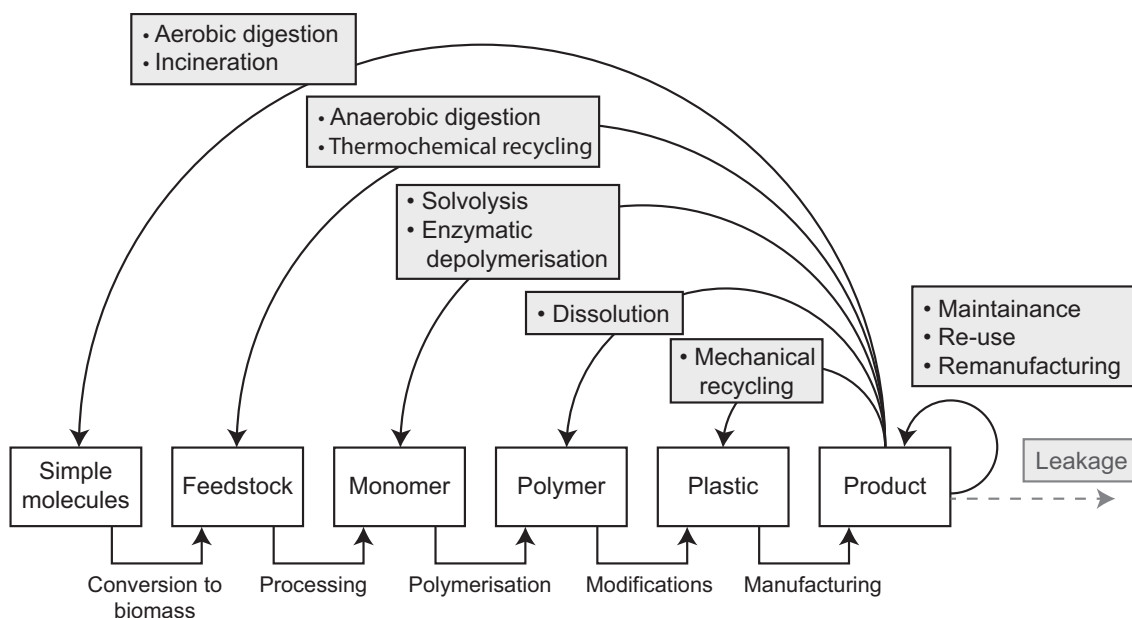
Fig. 2. Framework for the circular economy of bio-based plastics incorporated into products.

Table 2

Summary of possible recovery pathways for commodity-grade, drop-in, dedicated, and engineering-grade, bio-based polymers. Green: research proving that this is a viable option could be found. Yellow: existing research suggests that using this recovery pathway for this specific polymer yields poor results. In the case of aerobic and anaerobic digestion, this means that digestion does not occur in a timeframe that corresponds with current industry practice, or that may cause harm to the natural environment In the case of mechanical recycling, it means a rapid decline in properties. In the case of thermochemical recycling, the products of the process were only suitable as energy resources and not for the production of new polymers. Red: theoretically impossible based on the characteristics of the polymer combined with the targeted recovery pathway. For example, solvolysis of HDPE is theoretically impossible due to the chemical structure of PE. Grey: theoretically possible, but no studies could be found. For example, based on the chemical structure, bio-PA can undergo mechanical recycling, but this has not been demonstrated yet.

Commodity grade, drop-in bio-based polymers					
	Bio-PET	Bio-HDPE	Bio-LDPE	Bio-PP	Bio-PBS
Mechanical recycling	**Viable option** (Oromiehie & Mamizadeh, 2004; Paci & La Mantia, 1998)	**Viable option** (Kanemura et al., 2012; Kostadinova Loultcheva et al., 1997; Meran et al., 2008; Oblak et al., 2015; Vidakis et al., 2021a)	**Viable option** (Jin et al., 2012; Kabdi & Belhanechebensemra, 2008; Meran et al., 2008; Waldman & De Paoli, 1998)	**Viable option** (Aurrekoetxea et al., 2001; Liang & Peng, 2009; Meran et al., 2008; Vidakis et al., 2021b; Waldman & De Paoli, 1998)	**Viable option** (Georgousopoulo et al., 2016; Kanemura et al., 2012)
Dissolution	**Viable option** (Achilias et al., 2009; Poulakis & Papaspyrides, 2001)	**Viable option** (Achilias et al., 2007; 2009; Kannan et al., 2017; Pappa et al., 2001; Poulakis & Papaspyrides, 1995)	**Viable option** (Achilias et al., 2007; 2009; Kannan et al., 2017; Papaspyrides et al., 1994; Pappa et al., 2001)	**Viable option** (Achilias et al., 2007; 2009; Drain et al., 1983; Murphy et al., 1979; Pappa et al., 2001; Poulakis & Papaspyrides, 1997)	**Theoretically possible, but not researched yet**
Solvolysis	**Viable option** (Carniel et al., 2021; Carta et al., 2003; Fukushima et al., 2013; Goje et al., 2004; Kaabel et al., 2021; Kosmidis et al., 2001; Liu et al., 2018; Neves Ricarte et al., 2021; Quartinello et al., 2017; Ragaert et al., 2017; Sánchez & Col-linson, 2011; Tournier et al., 2020)	**Theoretically impossible**	**Theoretically impossible**	**Theoretically impossible**	**Theoretically possible, but not researched yet**
Thermochemical recycling	**Viable option** (Brems et al., 2011; Sharuddin et al., 2016)	**Viable option** (Achilias et al., 2007; Donaj et al., 2012; Sharuddin et al., 2016; Simon et al., 1996; Thunman et al., 2019; Wilk & Hofbauer, 2013)			**Theoretically possible, but not researched yet**
Anaerobic digestion	**Theoretically impossible**	**Theoretically impossible**	**Theoretically impossible**	**Theoretically impossible**	**Poor outcome** (Cho et al., 2011; Yagi et al., 2013; Yagi et al., 2014)
Aerobic digestion (industrial)	**Theoretically impossible**	**Theoretically impossible**	**Theoretically impossible**	**Theoretically impossible**	**Poor outcome** (Kim et al., 2006; Rafiqah et al., 2021; Zhao et al., 2005)
Aerobic digestion (nature)	**Theoretically impossible**	**Theoretically impossible**	**Theoretically impossible**	**Theoretically impossible**	**Poor outcome** (Kim et al., 2006; Nishide et al., 1999; Rafiqah et al., 2021)

Table 2 (*continued*)

Commodity grade, dedicated bio-based polymers			
	PLA	PHA	TPS	PEF
Mechanical recycling	**Poor outcome** (Badia et al., 2012a; Badia et al., 2012b; Badia et al., 2012c; Badia & Ribes-Greus, 2016; Beltrán et al., 2021; Żenkiewicz et al., 2009)	**Poor outcome** (Vu et al., 2020)	**Theoretically possible, but not researched yet**	**Theoretically possible, but not researched yet**
Dissolution	**Theoretically possible, but not researched yet**	**Theoretically possible, but not researched yet**	**Theoretically possible, but not researched yet**	**Theoretically possible, but not researched yet**
Solvolysis	**Viable option** (Alberti et al., 2019; Fliedel et al., 2014; Hajighasemi et al., 2016; Hirao et al., 2010; Leibfarth et al., 2012; Liu et al., 2015; 2017; 2018; Petrus et al., 2016; Román-Ramirez et al., 2019; Sánchez & Collinson, 2011; Song et al., 2013; 2014; Tsuji et al., 2003; Whitelaw et al., 2011)	**Viable option** (Liu et al., 2018; Song et al., 2018; 2019; Spekreijse et al., 2015; Tang & Chen, 2019)	**Theoretically possible, but not researched yet**	**Viable option** (Austin et al., 2018; Pellis et al., 2016; Weinberger et al., 2017a; 2017b)
Thermochemical recycling	**Poor outcome** (Dai et al., 2018)	**Poor outcome** (Ariffin et al., 2010)	**Theoretically possible, but not researched yet**	**Theoretically possible, but not researched yet**
Anaerobic digestion	**Poor outcome** (Itävaara et al., 2002; Yagi et al., 2009; 2010; 2014; Zhang et al., 2018)	**Poor outcome** (Abou-Zeid et al., 2001; Yagi et al., 2013; 2014)	**Poor outcome** (Massadier-Nageotte et al., 2006; Mohee et al., 2008)	**Theoretically possible, but not researched yet**
Aerobic digestion (industrial)	**Poor outcome** (Castro-Aguirre et al., 2017; Itävaara et al., 2002; Kale et al., 2007; Longieras et al., 2007; Luo et al., 2019; Saadi et al., 2012; Sarasa et al., 2009)	**Poor outcome** (Weng et al., 2010; 2011)	**Poor outcome** (Accinelli et al., 2012; Mohee et al., 2008)	**Theoretically possible, but not researched yet**
Aerobic digestion (nature)	**Poor outcome** (Lambert & Wagner, 2017; Mulbry et al., 2012; Ohkita & Lee, 2006; Pelegrini et al., 2016; Song et al., 2009)	**Poor outcome** (Boyandin et al., 2013; Fernandes et al., 2020; Kunioka et al., 1989; Voinova et al., 2008; Webb et al., 2012; Wool-nough et al., 2010)	**Poor outcome** (Accinelli et al., 2012)	**Theoretically possible, but not researched yet**
Engineering-grade bio-based polymers			
	Bio-PA	Bio-PC	Bio-PU	Bio-TPU
Mechanical recycling	**Theoretically possible, but not researched yet**	**Theoretically possible, but not researched yet**	**Theoretically impossible**	**Theoretically possible, but not researched yet**
Dissolution	**Theoretically possible, but not researched yet**	**Theoretically possible, but not researched yet**	**Theoretically impossible**	**Theoretically impossible**
Solvolysis	**Theoretically possible, but not researched yet**	**Theoretically possible, but not researched yet**	**Theoretically possible, but not researched yet**	**Theoretically possible, but not researched yet**
Thermochemical recycling	**Theoretically possible, but not researched yet**	**Theoretically possible, but not researched yet**	**Theoretically possible, but not researched yet**	**Theoretically possible, but not researched yet**
Anaerobic digestion	**Theoretically possible, but not researched yet**	**Theoretically possible, but not researched yet**	**Theoretically impossible**	**Theoretically impossible**
Aerobic digestion (industrial)	**Theoretically possible, but not researched yet**	**Theoretically possible, but not researched yet**	**Theoretically impossible**	**Theoretically impossible**
Aerobic digestion (nature)	**Theoretically impossible**	**Theoretically impossible**	**Theoretically impossible**	**Theoretically impossible**

Table 3
Summary of mechanical recycling of bio-based polymers (without additives).

	Polymer	Thermal and processing properties	Mechanical properties	Impact properties	References
Drop-in	**Bio-HDPE**	Reduced melt flow index after 5 cycles	Yield stress, tensile stress and elastic modulus Remain constant for 10 cycles, then decrease	Slight increase in impact strength after one cycle	(Kanemura et al., 2012; Kostadinova Loultcheva et al., 1997; Meran et al., 2008; Oblak et al., 2015; Vidakis et al., 2021a)
	Bio-LDPE	Unchanged until 40 cycles			(Jin et al., 2012; Kabdi and Belhane-che-bensemra, 2008; Meran et al., 2008; Waldman and De Paoli, 1998)
	Bio-PP	Reduced melt viscosity after 4 cycles	Strength and stiffness increase; strain at break decreases after 4 cycles	Impact strength remains unchanged for up to 6 cycles, then decreases	(Aurrekoetxea et al., 2001; Liang and Peng, 2009; Meran et al., 2008; Vida-kis et al., 2021b; Waldman and De Paoli, 1998)
	Bio-PET	Reduced melt viscosity after one cycle	Yield stress, tensile stress and elastic modulus reduced after one cycle	Impact strength reduced after one cycle	(Oromiehie and Mamizadeh, 2004; Paci and La Mantia, 1998)
	Bio-PBS	Reduced melt flow index after one cycle	Unknown	Unknown	(Georgousopoulo et al., 2016; Kanemura et al., 2012)
Dedicated	**PLA**	Unchanged after one cycle	Young's modulus and hardness decreased after one cycle	Impact strength decrased after one cycle	(Badia et al., 2012a, 2012b, 2012c; Badia and Ribes-Greus, 2016; Beltrán et al., 2021; Zenkiewicz et al., 2009)
	PHA	Viscosity reduced after one cycle	Tensile strength, tensile modulus remain unchanged until 6 cycles	Impact strength unchanged until 6 cycles	(Vu et al., 2020)

3.3. Dissolution

Dissolution is the recovery of a polymer by dissolving it in a solvent, followed by precipitation in a non-solvent, without any alteration to its molecular structure. Dissolution is applied in numerous chemical processes, such as coating deposition, and its molecular transport phenomena have been studied extensively (Miller-Chou and Koenig, 2003). During dissolution, any additives or contaminants in a plastic can be removed, achieving high recovery rates with homogeneous products while being able to selectively recover specific polymers from mixed plastic waste (Kannan et al., 2017; Pappa et al., 2001). However, dissolution requires large amounts of solvents and non-solvents, which are currently not bio-based. The solvent-to-polymer volume ratio is usually above 7:1, while solvent recovery is never 100 %. Dissolution is also energy intensive as it often requires temperatures above 100 °C.

Dissolution for polymer recovery is currently only applied at a pilot scale for polystyrene (Fraunhofer IVV, n.d.), and no research has been reported for bio-based polymers. Nevertheless, the dissolution of commodity polymers has been studied with good results. These results can be extended to chemically-identical drop-ins: specifically bio-LDPE, bio-HDPE, bio-PET and bio-PP (Achilias et al., 2009; Kannan et al., 2017; Pappa et al., 2001). A detailed overview of articles reporting the dissolution of polymers for which bio-based drop-ins exist can be found in the supplementary information (table S2).

3.3.1. Solvolysis

Solvolysis is the cleavage of a polymer by a solvent, often in the presence of a catalyst. Polymers containing ether, ester and amine bonds (synthesised through condensation polymerisation) can be recovered through solvolysis (Bakker and Balkenende, 2021). In contrast with dissolution and mechanical recycling, solvolysis can also recover thermoset polymers. Solvolysis processes are subdivided based on the solvent used, i.e. hydrolysis for water or alcoholysis for alcohols. The products of solvolysis may be fuels, useful chemicals or molecules that can be used directly in the production of new polymers. In most cases, solvolysis products require extra conversion steps to be useful for polymers. Additives can be filtered out. Polymer-to-solvent ratios during solvolysis are typically low, requiring elevated temperatures and catalysts.

In theory, (bio-based) polymers and plastics suitable for solvolysis include bio-PET, PLA, PHA, TPS, bio-PBS, bio-PA and PEF. Solvolysis of (bio-)PET has been demonstrated through hydrolysis (Goje et al., 2004), glycolysis (Carta et al., 2003; Sánchez and Collinson, 2011), aminolysis

(Fukushima et al., 2013) and alcoholysis (Ragaert et al., 2017). Solvolysis of PLA can occur through alcoholysis (Alberti et al., 2019; Fliedel et al., 2014; Hirao et al., 2010), hydrolysis (Tsuji et al., 2003) or alcoholised. Hydrolysis (Tang and Chen, 2019) and alcoholysis (Song et al., 2019; Spekreijse et al., 2015) of PHA have been demonstrated to yield valuable chemicals.

Enzymatic depolymerisation is a specific type of solvolysis that uses enzymes as biological catalysts. Enzymatic depolymerisation occurs at temperatures between 50 °C and 75 °C, which is lower than those used with synthetic catalysts. However, the polymer chains need to have a certain mobility at the process temperature. For example, consumer-grade PET has a high crystallinity that reduces polymer mobility, making it difficult to degrade by enzymatic depolymerisation (Neves Ricarte et al., 2021; Quartinello et al., 2017). Enzymatic depolymerisation of polymers into monomers has been successfully demonstrated for PLA (Hajighasemi et al., 2016), PEF (Pellis et al., 2016; Weinberger et al., 2017a) and PET (Quartinello et al., 2017; Tournier et al., 2020). Depending on the enzymes present and the reaction conditions, enzymatic depolymerisation can also yield chemical intermediates that can be processed into monomers or used in other applications (Carniel et al., 2021; Kaabel et al., 2021; Neves Ricarte et al., 2021). A detailed overview of different solvolysis processes, including enzymatic depolymerisation and its products for bio-based polymers be found in the supplementary information (table S3).

3.3.2. Thermochemical recycling

Thermochemical recycling is the dissociation of polymers through high temperatures. Polymers are dissociated into gases (e.g. CO_2, methane), liquids (longer hydrocarbons) and solids (tar). Thermochemical recycling processes are differentiated by process temperature, pressure and atmosphere (Al-Salem et al., 2009). The most common thermochemical recycling processes are listed below.

- **Pyrolysis** occurs in an inert atmosphere at around 500 °C and 1–2 standard atmosphere (atm), with or without a catalyst, resulting in either depolymerisation or random fragmentation, depending on the plastic composition and other materials present. The products of pyrolysis can be gases, liquids, solids or any mixture of these (Sharuddin et al., 2016; Al-Salem et al., 2009).
- **Gasification** occurs in an atmosphere of air or pure oxygen at 700–1200 °C at standard pressure. Plastics break down into so-called syngas (consisting primarily of nitrogen, carbon monoxide, hydrogen and CO_2) (Al-Salem et al., 2009).

- **Hydrogenation** occurs in the presence of hydrogen (H_2) and a catalyst at 350–400 °C at roughly 70 atm. The plastic is liquefied during the process and can be filtered to yield naphtha or oil. The presence of hydrogen improves the quality of the resulting feedstock (Al-Salem et al., 2009).

The decomposition behaviour of plastics in thermochemical recycling is complex, yielding a wide range of compounds (Al-Salem et al., 2009). Plastics can be studied in isolation, but thermochemical recycling usually uses mixtures, influencing the reactions that occur. For example, pyrolysis of PP in isolation at 760 °C produces benzene (Sharuddin et al., 2016), but under different conditions, no benzene is produced (Donaj et al., 2012; Kaminsky et al., 2004). Products of thermochemical recycling are primarily used as fuels due to the wide and poorly defined range of feedstocks produced (Al-Salem et al., 2009). Specific chemicals could be removed from the mixture and used to produce new polymers (either directly or after conversion) (Solis and Silveira, 2020).

3.3.3. Anaerobic digestion

Anaerobic digestion is "*the breakdown of an organic compound by microorganisms in the absence of oxygen to carbon dioxide, methane, water and mineral salts of any other elements present (mineralisation) plus new biomass*" (ISO, 2014). Anaerobic digestion can occur in industrial composting facilities and landfills (with methane capturing), but also in uncontrolled environments, such as underground or in the bottom layers of a home compost bin (Quecholac-Piña et al., 2020). Industrial anaerobic digestion focuses on producing biogas (methane) as an energy source, which can also be a precursor for polymers (Strong et al., 2016). Another product of anaerobic digestion is the so-called digestate consisting of residual materials. This digestate is often used as a fertiliser (Xu et al., 2018). However, synthetic biodegradable polymers degrade fully into methane, water and other gases and do therefore not add to this fertiliser. When anaerobic digestion occurs in natural environments, the highly potent greenhouse gas methane leaks into the atmosphere (Shine and Sturges, 2007).

Before industrial anaerobic digestion, physical contaminants (e.g. glass or metals) are removed from bio-waste and its composition is optimised, commonly by adding carbon-rich waste, since bio-waste is often too high in nitrogen (Briassoulis et al., 2021). Since biodegradable plastics are relatively high in carbon, they could be used to optimise the waste composition. To be compatible with industrial anerobic digestion, the degradation time for plastics under typical industrial anaerobic digestion conditions needs to be similar to that of the bio-waste. A typical industrial anaerobic digestion process occurs at 30–60 °C for up to 60 days (Xu et al., 2018; Zhang et al., 2014). For example, PLA can degrade up to 95 % in 40 days at 55 °C (Yagi et al., 2010) but only reached 20 % biodegradation after 65 days at 37 °C (Zhang et al., 2018). PHA degrades fully within 42 days at 37 °C (Abou-Zeid et al., 2001). TPS degraded by 23 % after 28 days at 35 °C (Massardier-Nageotte et al.,

2006). This implies that bio-based plastics are only candidates for anaerobic digestion if additional heat is applied. An overview of experiments with biodegradable plastics under anaerobic conditions is provided in the supplementary information (table S4).

3.4. Aerobic digestion

Aerobic digestion is "*the breakdown of an organic compound by microorganisms in the presence of oxygen to carbon dioxide, methane, water and mineral salts of any other element present (mineralisation) plus new biomass)*" (ISO, 2012). Composting facilities utilise industrial aerobic digestion to produce compost from organic waste (Briassoulis et al., 2019). Most polymers degrade into CO_2 and water under aerobic digestion conditions and do not contribute significant mass to the compost (Lambert and Wagner, 2017; Mueller, 2006). Similar to anaerobic digestion, aerobic digestion occurs at an optimal ratio of carbon and nitrogen atoms to which plastics can contribute (Briassoulis et al., 2019). Aerobic digestion also occurs in home composting bins and in the top layers of soil and agricultural fields (ISO, 2021a).

Bio-based plastics should be compatible with the industrial composting cycle, which ISO standards describe as taking up to 56 days at temperatures above 50 °C (ISO, 2021b). Table 4 contains a summary of experiments on aerobic digestion of pure polymers of different shapes under specified composting conditions, demonstrating the differences in the degree of degradation due to different product thicknesses and composting conditions. This highlights the importance of understanding the degradation of not just the material, but also the product in the targeted composting conditions. A table with further details of aerobic digestion experiments, including blends and additives, can be found in table S5 in the supplementary information. If biodegradation of a plastic is not completed during industrial composting, the partially decomposed plastic will end up as fragments in the compost that is often used as a fertiliser. Aerobic digestion conditions in nature are different from industrial composting, and the plastic fragments may not fully decompose (de Wilde and Boelens, 1998). This may result in the introduction of more micro- and nanoplastics formation but the effects of this are still unknown (Lambert and Wagner, 2017).

Aerobic digestion in nature could be an attractive recovery pathway for plastic products, as it can prevent plastic pollution. Understanding biodegradation in nature remains challenging, with little research in realistic conditions. Moreover, soil microbiomes vary per region and soil type, influencing the effectiveness of biodegradation (Boyandin et al., 2013). (Bio-)PBS only degrades slowly (<5 % weight loss in 80 days) in natural soil and requires additives to enhance aerobic digestion under natural conditions (Kim et al., 2006). PLA does not biodegrade under natural conditions (Lambert and Wagner, 2017). Biodegradation of PHAs depends mainly on the molecular structure of the type of PHA and the soil microbiome (Fernandes et al., 2020). PHAs are often blended with other polymers in order to enhance their properties, but how this

Table 4
Summary of industrial aerobic digestion experiments with pure polymers.

Polymer	Inoculum type, weight ratio plastic:inoculum	Product/ shape	Time (days)	T	Degradation degree	Reference
PLA	Compost, unknown ratio	Pellets	60	58 °C	34 %–45 %	(Castro-Aguirre et al., 2017)
		Film			35 %–100 %	
	Municipal organic solid waste, unknown ratio	Bottle	58	65 °C	78 %–84 %	(Kale et al., 2007)
	Agricultural and tree waste, unknown ratio	Powder (0.5 mm)	45	58 °C	55 %–75 %	(Longieras et al., 2007)
PLLA	Vegetable waste, 1:500	Film	20	70 °C	99 %	(Itävaara et al., 2002)
		Fabric	40		73 %	
	Inoculated compost, 1:500	Powder (0.5 mm)	90	58 °C	90 %	(Saadi et al., 2012)
PHA	Synthetic compost, unknown ratio	Film	39	58 °C	100 %	(Weng et al., 2010)
	Mature compost, unknown ratio	Film	110	58 °C	80 % – 91 %	(Weng et al., 2011)
PBS	Mature compost, 1:6	Pellets (3 mm)	90	58 °C	14.1 %	(Zhao et al., 2005)
		Powder (0.042 mm)			71.9 %	
		Film			71.9 %	
TPS	Municipal biowaste, 1:100	Carrier bags	72	60 °C	100 %	(Mohee et al., 2008)

Table 5

Identification of product design implications when targeting specific recovery pathways, based on plastic suitability and technical properties.

Recovery pathway	Technical characteristic	Product design implication(s)
Mechanical recycling	Recycled plastic becomes a mix of the input plastics. Additives, blends and different molecular weights are not separated.	**Avoid** or minimise using additives that reduce the value of recycled plastics. **Avoid** blending different polymers unless a closed-loop system can be set-up for a product to recycle the blend directly. **Ensure** that different plastics in a product can be mechanically separated, for instance, during shredding. Products should not be produced using multi-material manufacturing, such as 2 K or 3 K, and products should not have coatings.
	Sorting effectivity determines the quality of recycled plastics	**Avoid** using additives that hinder sorting. **Avoid** mechanical recycling for products that can easily be contaminated by substances that cannot be removed in the targeted mechanical recycling process.
Dissolution and Solvolysis	Dissolution and solvolysis occur at the surface of a plastic	**Optimise** surface area to volume ratio.
	Dissolution and solvolysis can sort out additives and separate different molecular weights and blends	**Allows** for the incorporation of additives and different molecular weights and blends.
Solvolysis (enzymatic depolymerisation)	In the case of enzymatic depolymerisation: the process occurs at a lower temperature (below 60 °C).	**Ensure** solvolysis can occur at moderate temperatures.
Thermochemical recycling	A change in waste composition due to a growing market share of bio-based plastics may affect the feedstock produced from thermochemical recycling	**Consider** the current plastic waste composition and if introducing the targeted plastic will not yield harmful products.
Industrial biodegradation (anaerobic digestion and aerobic digestion)	Anaerobic and aerobic digestion occur at the surface of a plastic	**Optimise** for surface area to volume ratio.
	After the industrial process, anything that does not degrade may be used as fertiliser, eventually ending-up in nature	**Ensure** all components (such as the polymer and additives) will degrade fully during industrial biodegradation. If they do not, they should degrade aerobically in natural conditions. **Avoid** for products that get contaminated with non-biodegradable substances that are harmful to nature.
Biodegradation in nature (only aerobic)	Any value of the plastic is completely lost as it returns to CO_2 and H_2O while no energy is recovered	**Use** for products that inevitably end up in nature (e.g. products that wear such as shoe soles or agricultural films).
	Anaerobic digestion in uncontrolled environments is highly undesirable as the methane produced will leak into the atmosphere.	**Avoid** anaerobic digestion for products that will end up in anaerobic environments, such as the bottoms of compost bins.

affects biodegradation is not yet understood (Fernandes et al., 2020). TPS showed aerobic digestion under simulated natural conditions but showed little deterioration in actual natural conditions (Accinelli et al., 2012). A more detailed overview of these experiments can be found in table S6 in the supplementary information.

3.4.1. Incineration

Incineration, or energy recovery, is the "*production of useful energy through direct and controlled combustion*" (ISO, 2013). During incineration, the plastic is returned to simple molecules and bottom ash. Because bio-based plastics contain carbon originating from CO_2 from the atmosphere, their incineration could be considered carbon neutral (Bakker and Balkenende, 2021). The amount of energy released during the incineration of specific polymers can be estimated according to their chemical structure (Rhyner et al., 1995). Incineration is currently the most viable recovery pathway for bio-based plastics without dedicated other dedicated recovery pathways. Incineration of plastics is accepted as a total conversion of plastic waste (Geyer et al., 2017). However, the incineration of plastics can produce toxic by-products (Li et al., 2001). Furthermore, microplastics have been found in bottom ash from municipal waste incinerators: 1.9–565 pieces of microplastic per kilogram of bottom ash (Yang et al., 2021).

4. Discussion – defining implications for product design

We presented a framework for the material-level recovery of bio-based plastics in a circular economy. This demonstrated that bio-based polymers operate differently in the circular economy compared to petrochemical-based polymers when material-level recovery is considered. The circular economy typically distinguishes between a biocycle and a technocycle, where recovery of petrochemical-based plastics occurs only in the technocycle. For petrochemical-based plastics, recovery pathways like incineration and aerobic digestion cannot be considered circular loops, as they introduce fossil greenhouse gas emissions to the atmosphere. Bio-based plastics, on the other hand, can also flow through

the biocycle because they are produced from biomass (Bakker and Balkenende, 2021).

Section 3 has provided an overview of the state-of-the-art of different recovery pathways for bio-based plastics from a technical perspective, indicating which recovery pathways can be considered for specific bio-based polymers. Merely choosing a suitable polymer and recovery pathway is insufficient to guarantee efficient recovery. Additives and blending need to be considered, as well as how a product is constructed and manufactured since these aspects can make a product unsuitable for specific recovery pathways. This implies that the recovery of bio-based plastics has implications for product design. In this section, we describe some of these implications based on the technical characteristics of recovery pathways. The product design implications are summarised in Table 5. Fig. 3 displays how these product design implications may be applied in the design process. Note: in a CE, higher-value recovery options are preferred - this figure and Table 5 focus solely on material-level recovery.

In the hierarchy of material-level recovery pathways, mechanical recycling is usually at the top (Briassoulis et al., 2019). However, mechanical recycling is not always preferred or possible as it may result in reduced properties. Kriwet et al. (1995) developed some design for mechanical recycling recommendations, focussing on material composition and ease of separation of plastic components in the product. Since its publication in 1995, separation and sorting technologies have improved, and they will become more accurate (Serranti and Bonifazi, 2019). This renders some recommendations obsolete, for example, avoiding different plastics in a single product or not using certain additives that change the density of the plastic. Multi-material manufacturing, where different plastics are fused and cannot be mechanically separated, should still be avoided when designing for mechanical recycling. Furthermore, plastics with different molecular weights and additives are not separated. Therefore, additives that reduce the value of recycled plastics should be avoided, such as colourants and additives that induce molecular damage during reprocessing. Blends of different polymers are typically not sorted and should also
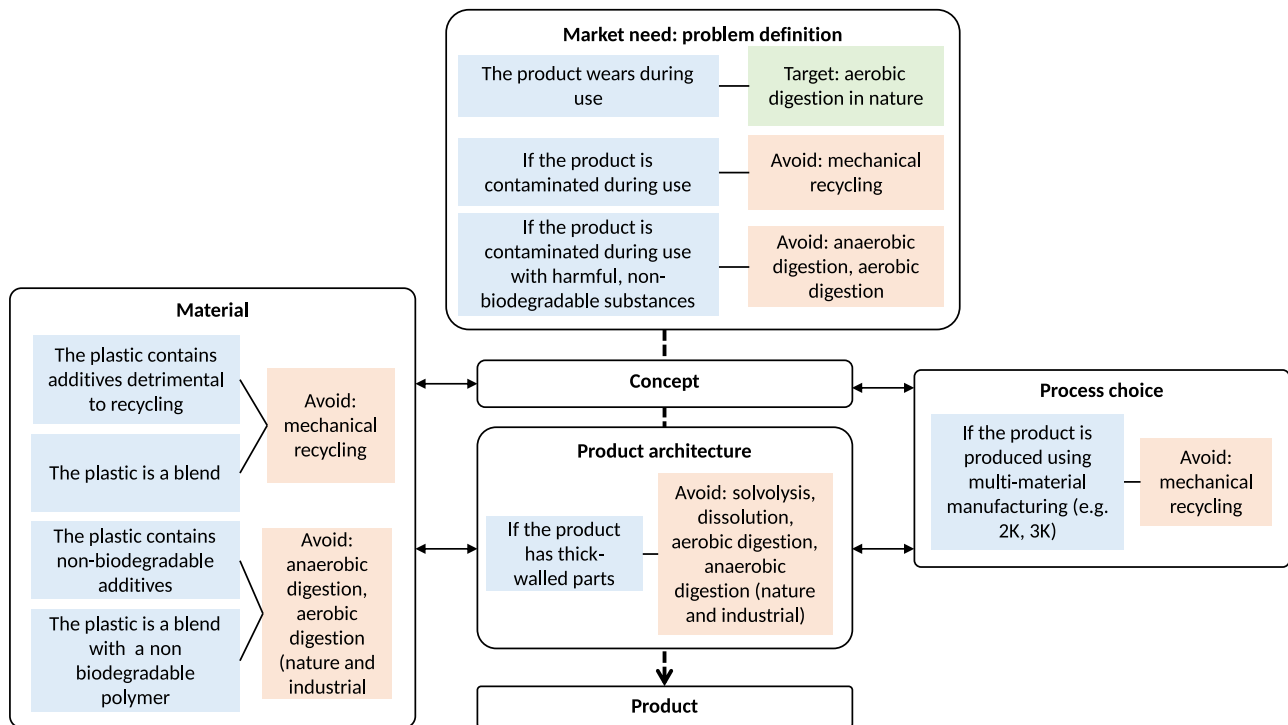
Fig. 3. First iteration of the implementation of material-level recovery in a product design process using bio-based plastics. Figure adapted from Ashby et al. (2007).

be avoided.

Dissolution and solvolysis occur at the surface of a plastic, and their reaction rate is affected by the surface area to volume ratio (Miller-Chou and Koenig, 2003). Both dissolution and solvolysis enable the separation of additives and blends (Kannan et al., 2017; Pappa et al., 2001), implying that products can contain additives and blends without compromising the value of recovered polymers and monomers: an advantage over mechanical recycling. Thermochemical recycling can process essentially any plastic, but thermochemical recycling processes are sensitive to the input composition (Al-Salem et al., 2009). The entire plastic waste composition will likely not be affected significantly by a single product range, but plastics can yield harmful products in combination with the current plastic waste composition.

Biodegradation processes (aerobic and anaerobic) occur at the surface of a plastic, and the surface-to-volume ratio should be optimised to increase the reaction rate (Castro-Aguirre et al., 2017). Any components not fully degraded during industrial biodegradation may be used as fertiliser, and the product used should either fully degrade within the industrial process or be able to degrade in soil. This also applies to any contaminants from the use-phase. In aerobic conditions, polymers biodegrade into CO_2 and H_2O, not adding mass or nutrients to compost (Lambert and Wagner, 2017; Mueller, 2006) and any functional value in the plastic is completely lost. Therefore, aerobic biodegradation should primarily be considered as a recovery pathway to prevent plastic waste; if the product is bound to end up in nature and if the plastic is shown to disintegrate completely. In uncontrolled anaerobic conditions, polymers produce methane, which can be released into the atmosphere, where it is a potent greenhouse gas (Shine and Sturges, 2007). Anaerobic biodegradation should therefore be avoided in natural environments.

5. Conclusions

Bio-based plastics have attracted attention due to their perceived sustainability and circularity, evidenced by a rapidly growing market share. In order to avoid contributing to plastic pollution, efficient recovery of bio-based plastics at end-of-life needs to be facilitated. Understanding and enabling the recovery will become increasingly

relevant as bio-based plastics grow into a larger fraction of plastic waste. Although bio-based plastics do not necessarily perform differently from petrochemical-based plastics in higher levels of the waste hierarchy (product-focused recovery pathways), they do perform differently in material-level recovery. The existing body of scientific knowledge does not sufficiently support circularity for bio-based plastics. The potential recovery pathways for many bio-based plastics have not yet been studied. This article contributes to this body of research by describing the available recovery pathways, how they work for specific bio-based polymers and addressing the role of product design in improving the circularity of bio-based plastics.

Dissolution, solvolysis and thermochemical recycling can deal with plastics containing additives and blends of different polymers, but often at a high environmental and economic cost. Moreover, the application of these novel recovery pathways to most bio-based plastics is not yet understood, especially in the case of dedicated bio-based plastics. Therefore, further development of novel recovery pathways will be required, as well as further development of waste collection and sorting systems.

Biodegradation in nature is often seen as a recovery pathway to reduce plastic pollution. However, there is insufficient evidence that most of these biodegradable bio-based plastics fully degrade in nature into CO_2 and water, avoiding methane emissions. More research in realistic natural conditions over longer periods is needed to justify the use of biodegradable plastics in nature. It is also unknown if partially biodegraded plastics may become a source of micro- and nanoplastics. However, some plastic products will inevitably end up in nature, specifically products that wear during use (such as car tires or elastomer shoe soles). For these applications, biodegradation in nature may be a valid choice.

Product design plays an essential but often overlooked role in improving the circularity of bio-based plastics. Decisions made during the product design process determine the range of recovery pathways for a product, along with the presence of associated services. Therefore, the recovery must be considered starting early in the design process. The implications discussed in this paper can be expanded by including different perspectives, such as those of legislation, business development

and economy. The environmental impact of different recovery pathways has not yet been sufficiently quantified and presents an important area for future research.

Bio-based plastics offer an opportunity to accelerate the transition to a circular economy, but this requires a concerted effort to consider recovery at end-of-life carefully. The results presented in this article can be used by product designers, recyclers, and plastic producers. Product designers may use the outcomes when selecting a bio-based plastic and a targeted recovery pathway while ensuring that this recovery pathway is encouraged though the product design. The results should help recyclers and plastic manufacturers facilitate efficient recovery of bio-based plastics at end-of-life. Recyclers are encouraged to consider which recovery infrastructure may become relevant for the future end-of-life plastics composition. Finally, plastic producers are stimulated to consider the potential recovery of plastics under development.

CRediT authorship contribution statement

Linda Ritzen: Conceptualization, Investigation, Writing – original draft, Visualization. **Benjamin Sprecher:** Writing – review & editing, Supervision. **Conny Bakker:** Writing – review & editing, Supervision. **Ruud Balkenende:** Writing – review & editing, Supervision, Funding acquisition.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Linda Ritzen reports financial support was provided by The KAITEKI institute, inc.

Data availability

No data was used for the research described in the article.

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Supplementary materials

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