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Mechanical, chemical, biological: Moving towards closed-loop bio-based recycling in a circular economy of sustainable textiles

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ABSTRACT

The textile industry is facing increasing criticism because of its intensive use of resources –both natural and fossil derived– and the negative environmental and societal impacts associated with the manufacturing, use and disposal of clothes. This has led to a desire to move towards a circular economy for textiles that will implement recycling concepts and technologies to protect resources, the environment and people. So far, recycling processes have been focused on the chemical and mechanical reuse of textile fibres. In contrast, bio-based processes for textile production and recycling have received little attention, beyond end-of-life composting. However, the selectivity and benign processing conditions associated with bio-based technologies hold great promise for circularising the textile life cycle and reducing the environmental impacts of textile production and processing. Developing circular and sustainable systems for textile production requires a revolutionary system approach that encompasses the choice of material and finishes being designed for recycling at the end of life, and in this context bio-based processes can help provide the means to maintain materials in a closed loop. This paper reviews established methods in mechanical and chemical recycling processes in closed-loop textile recycling of all fibre types, as well as bio-based processes that demonstrate open-loop textile recycling. Fermentation and enzymatic processes have been demonstrated for the production of all types of textiles, which in combination with enzymatic deconstruction of end of life cellulosic textiles could allow them to be recycled indefinitely. Within the context of the circular economy, bio-based processes could extend mechanical and chemical textile recycling mechanisms in the technical cycle, enabling greater circularity of textiles in the biological cycle before composting takes place.

1. Introduction

The need for a circular economy of the textiles industry has been widely promoted as a response to the publication of damning reports on the negative impacts of fashion consumption (Ellen MacArthur Foundation, 2017). Globally, 53 million tonnes of fibres are consumed annually for making clothing responsible for 10% of greenhouse gas (GHG) emissions and 20% of wastewater generating large amounts of waste. In Europe, 180,000 tonnes of textile waste were generated in 2016 (Roos et al., 2019), of which only 15–20% were collected for recycling and less than 1% was recycled into new clothes (Textile Recycling Association, 2005). Textile consumption is set to continue to rise globally by 63% by 2030 (Boston Consulting Group and Global Fashion Agenda, 2017), and in the UK grew by 200,000 tonnes since 2012 (WRAP, 2017). The situation is greatly exacerbated by the high amount of low-value fast fashion items, which results in increased quantities of textiles with limited reuse capabilities in secondary markets, leading to increased levels of textile waste (WRAP, 2019). We know that textiles have a far-reaching global impact. Supply chain waste including preparation of fibres to make yarn and during clothes production was estimated to be 800,000 tonnes in 2016, of which 440,000 tonnes arose in China and India (WRAP, 2017). Solutions involve

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degrowth, reduced consumption, extended product lifetimes (reuse, repair, upgrade and remanufacture) and waste minimisation in order to retain and recover the materials, energy, water and chemicals that went into the production of textiles (Niinimäki et al., 2020). This will take time and will not solve the issue associated with the million tonnes of textile discarded now, hence the need to complement these with recycling. The realisation of a circular economy, in which materials are retained in a closed loop, requires the development of sustainable and scalable high-quality textile production associated with effective recycling technologies and systems (Ellen MacArthur Foundation, 2021). In this context, recycling is a necessary step in enabling a circular resource flow.

1.1. Models for a circular economy for textiles

The concept of the circular economy is built on several schools of thought including McDonough and Braungart's Cradle to Cradle model where, 'waste equals food' (2002, p.92). Materials in the circular economy model are divided into technical and biological nutrients. Circular economy research for textiles has so far mainly focused on the technical cycle and the biological cycle is less explored. Within the technical cycle of the circular economy model, textile recycling is structured into mechanical and chemical processes (Ellen MacArthur Foundation, 2017). A comprehensive review of mechanical and chemical textile recycling processes in the technical cycle has been conducted to map the environmental impacts of fast fashion (Niinimäki et al., 2020), the environmental impact of textile reuse and recycling (Sandin and Peters, 2018) or to compare existing technologies for fibre waste recycling (Hamouda and Lu, 2014), cellulose dissolution technologies (Määttänen et al., 2018), and textile recycling (Roos et al., 2019). Reviews of circular processes rarely evaluate bio-based processes for textile recycling or tend to focus on one fibre type (Piribauer and Bartl, 2019), which mostly demonstrates open-loop processes and separation of fibre blends such as cotton and polyester that give specific properties to textiles, hence this paper explores the wider applications of these processes for textiles.

Given that established textile production is mostly constituted of non-biological processes, either via fibre blends, dyes or finishes for example, the focus in circularity of textiles has been mainly in the technical cycle. The technical cycle is based on non-renewable petroleum-based materials that can only be recovered and repurposed (Ellen MacArthur Foundation, 2017). Other recycling classifications are discussed in Sandin and Peters (2018):

- Closed-loop recycling: the material is recycled in a more or less identical product.
- Open-loop recycling: the material is recycled in another category of the product.
- Upcycling: the product from recycled materials is of higher value than the original product.
- Downcycling: the recycled material is of lower value compared to the original product.

In this paper, we use the categories of open and closed-loop processes in order to map the recycling processes reviewed. A circular economy is defined by closed-loop processes (Ellen MacArthur Foundation, 2017), in which waste textiles are transformed into textiles, for example through fibre-to-fibre recycling.

The biological cycle is based on resources that can decompose and build nutrients to transform into new renewable resources. Circularity in the biological cycle builds on 'biological nutrients' such as food waste to regenerate soil, through processes such as composting and anaerobic digestion (Ellen MacArthur Foundation, 2017). For textiles, composting is a poor use of resources and should not be part of a circular economy system, which keeps the value of textiles in the system. The Biomimicry Institute (2020) proposes to merge the technical and biological cycle within the biological cycle if solely biomaterials are produced which can be composted at the end of life. In this context, microbial processes are a transition technology towards dispersion of materials in the natural environment (Biomimicry Institute, 2020). In this paper, we argue that bio-based processes could establish closed-loop textile recycling processes within the biological cycle, and therefore provide a different strategy for merging technical and biological cycles.

1.2. Bio-based processes

Bio-based processes are the use of enzymes in biochemical reactions and microorganisms in biological transformations to convert feedstock into value-added products. When applied to textiles, these will be specific to each type of fibre and may be adapted from established methods for materials with a similar composition. Cotton, hemp or man-made viscose and rayon fibres are rich in cellulose which is a polymer of sugars and could be processed in biorefineries. By definition, biorefineries use bio-based feedstock instead of fossil feedstock used in oil refineries (Yu et al., 2020). They produce energy (biofuels) and platform chemicals (lactic acid) used in materials (bioplastics such as PLA), but this approach is less explored for textiles. Dedicated crops such as maize and wood are currently commercially used to produce biofuel and viscose respectively. Because of the negative impacts of using virgin resources (food vs fuel, land use and climate change), extensive research has been carried out to develop biofuel processes using second generation feedstock which are waste or by-products from the food and agricultural industry and include cereal straw and municipal waste. However, the deconstruction of these cellulose-rich feedstocks is difficult due to the recalcitrance of cellulose to depolymerisation (to sugars) and the presence of other plant-specific contaminants such as lignin and hemicellulose. In this context, waste textiles can be seen as a cellulose-rich feedstock for 2nd generation biorefinery in an open loop recycling process. Contaminants in this case are hardware, non-cellulosic textiles and the chemicals used for dyeing and finishing while recalcitrance of textile cellulose is due to the very high crystallinity of cellulose in the cotton fibres. Wool and silk consist of the structural protein keratin. Recycling keratin has been studied in the context of the poultry and sheep meat industry where they are considered an animal waste with strict associated costs and negative environmental impacts (Petek and Marinšek Logar, 2021). Synthetic fabrics are man-made polymers of not-seen-in-nature esters mostly polyester (60%) or polyamide (nylon). Polyester is a polymer of Polyethylene terephthalate (PET), the same material used to make clear plastic water bottles. The environmental issues associated with the use of disposable plastic has attracted much concern from the public. This has benefitted the development of research for bio-plastics produced from renewable resources (Hatti-Kaul et al., 2020) and the development of novel bioplastics that would be 'more' biodegradable (Salvador et al., 2019) but also for developing enzymatic processes able to depolymerise and recycle PET.

In this paper, we will explore the ways in which enzymatic and biological processes can be used to underpin a truly circular approach to closed-loop resource use in a textile economy. In this concept (depicted in Fig. 1), enzymes can be used to depolymerise end-of-life textiles into their monomeric building blocks, which can then be repolymerised to create virgin quality polymers and fibres for the next generation of textiles in the cycle. Clearly, these processes will never be 100% efficient and some material will be lost in the stages of textile regeneration. However, such losses can be made up for by incorporating other cellulosic waste, from crop residues and municipal waste, in the case of cellulosic fibres; and from other keratinous waste (poultry feathers etc.) in the keratinous fibre cycle. We will present a review of mechanical and chemical processes in textile recycling, as well as of bio-based processes in the context of textiles bio-manufacturing and recycling, to demonstrate how specific recycling processes can be applied to different textile fibre types, as well as agricultural waste feedstocks for the production of

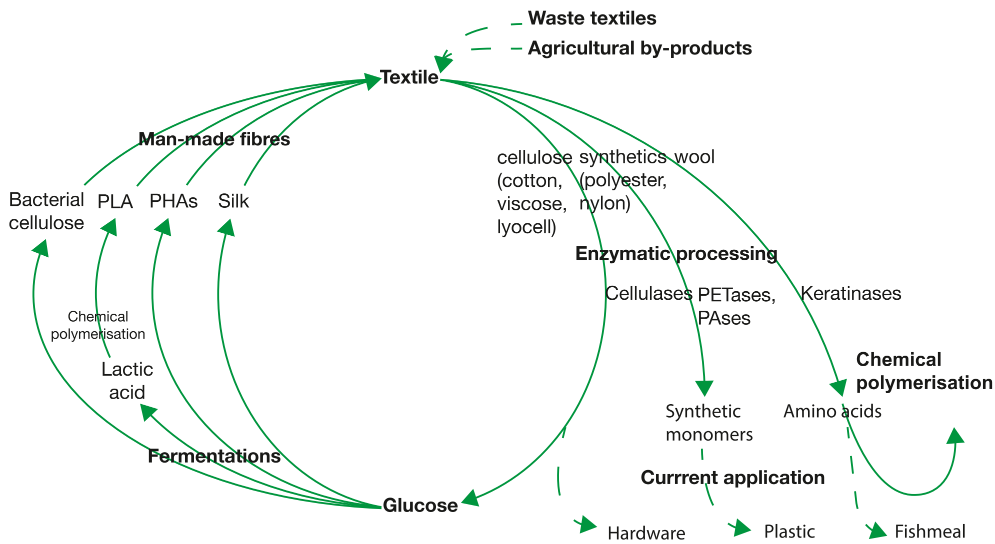

Fig. 1. Establishing a closed-loop bio-based process for cellulose-based textile recycling routes.

textiles.

2. Method

The research was conducted with a review of literature to establish methods in mechanical, chemical and bio-based processes for textiles recycling. We established using open-and closed-loop process categories to evaluate bio-based processes in the context of a circular economy, which are described in the introduction. This was followed by mapping of textile recycling case studies into closed-loop (textile to textile) and open-loop (a waste feedstock into a textile, or a textile into another product) processes.

2.1. Review of literature

The literature review consists of two parts. The first part applied an ‘umbrella review’ (Grant and Booth, 2009) to determine key components of textile recycling in existing reviews, described in sections 3.1 and 3.2, exploring established circular mechanical and chemical textile recycling processes. In the second part, we have applied a ‘mapping review’ (Grant and Booth, 2009) to contextualise bio-based processes that have been applied to textiles for future research recommendations, described in section 3.3. The review of literature was conducted in Science Direct, Scopus, Web of Knowledge and Google Scholar with search terms for ‘mechanical recycling’ and ‘chemical recycling’ such as ‘monomer recycling’, ‘polymer recycling’, and ‘cotton’, ‘polyester’, ’synthetics’, ‘wool’ or ‘agricultural waste’, and ‘closed-loop’, ‘textiles’ or ‘fibres’; for bio-based processes ‘textile wastes’ and ‘ethanol, compost, anaerobic digestion, fermentation, cellulase’, ‘recycling’, ‘cotton, wool or PET and depolymerisation’, ‘polyhydroxynolates’, ‘silk’, bacterial cellulose’, ‘biopolymer’ and ‘biorefinery’. For the purpose of this research, the review excluded the analysis of enzymatic or biological processes where neither their input nor output were related to textiles. To delineate the scope of the review, we have included studies that either consider recycling of a textile into a textile or into another circular feedstock, or that recycle a waste feedstock into a textile. Given that mechanical and chemical recycling have achieved substantial developments in closed-loop processes, we exclude the review of methods that do not aim for closed-loop textile to textile recycling in these two categories, and where textile waste is the input of open-loop processes. However, we have limited the review of open-loop recycling in mechanical and chemical processes to textile fibres or textiles that are produced from agricultural by-products.

2.2. Mapping of case studies for textile recycling in mechanical, chemical and bio-based processes

We use the mechanical, chemical and bio-based process categories from the literature review to map case studies of textile recycling. The literature review established the key terminology of mechanical and chemical recycling processes, which we structured into the following categories: (i) recycling process; (ii) method; (iii) waste feedstock; (iv) open-loop or closed-loop process; and (v) output. We have applied these categories to the mapping of enzymatic and biological processes with the exception of the open or closed-loop categories, as no evidence of closed-loop processes for textile recycling was found: (i) method; (ii) waste feedstock; (iii) pretreatment; (iv) product; and (v) value added product/textile (if demonstrated). The maps support the analysis of the types of waste feedstock introduced, as well as identify where open and closed-loop processes have been achieved (see Figs. 2–4 in section 3).

3. Results and discussion

Fig. 2 shows the review of mechanical and chemical recycling processes with established textile fibre types. The maps in Figs. 3 and 4 illustrate the enzymatic and biological processes in four methods: compost, anaerobic digestion, enzymatic, and fermentation, where fermentation commonly occurs after an enzymatic depolymerisation step. Recycling processes are usually a combination of two or more different processes and often involve different methods. For example, chemical recycling often involves a mechanical aspect of shredding the material. Biological processes might require a blend of mechanical or green chemistry methods for pretreatment. For the purpose of this paper, we highlight the core process employed to enable circularity of textiles.

3.1. Mechanical processes for textile recycling

Mechanical recycling is the most established recycling process for textiles. Mechanical processes are scalable and associated costs are lower than chemical or bio-based processes however they result in low-cost materials with decreased quality, which reduces opportunities for high-value textile applications (Roos et al., 2019).

3.1.1. Fibre recycling

Fibre recycling usually refers to a mechanical recycling process in which the fabric is taken apart and the fibre is preserved (Rittfors, 2020). These processes have been adopted for all textile materials. The most

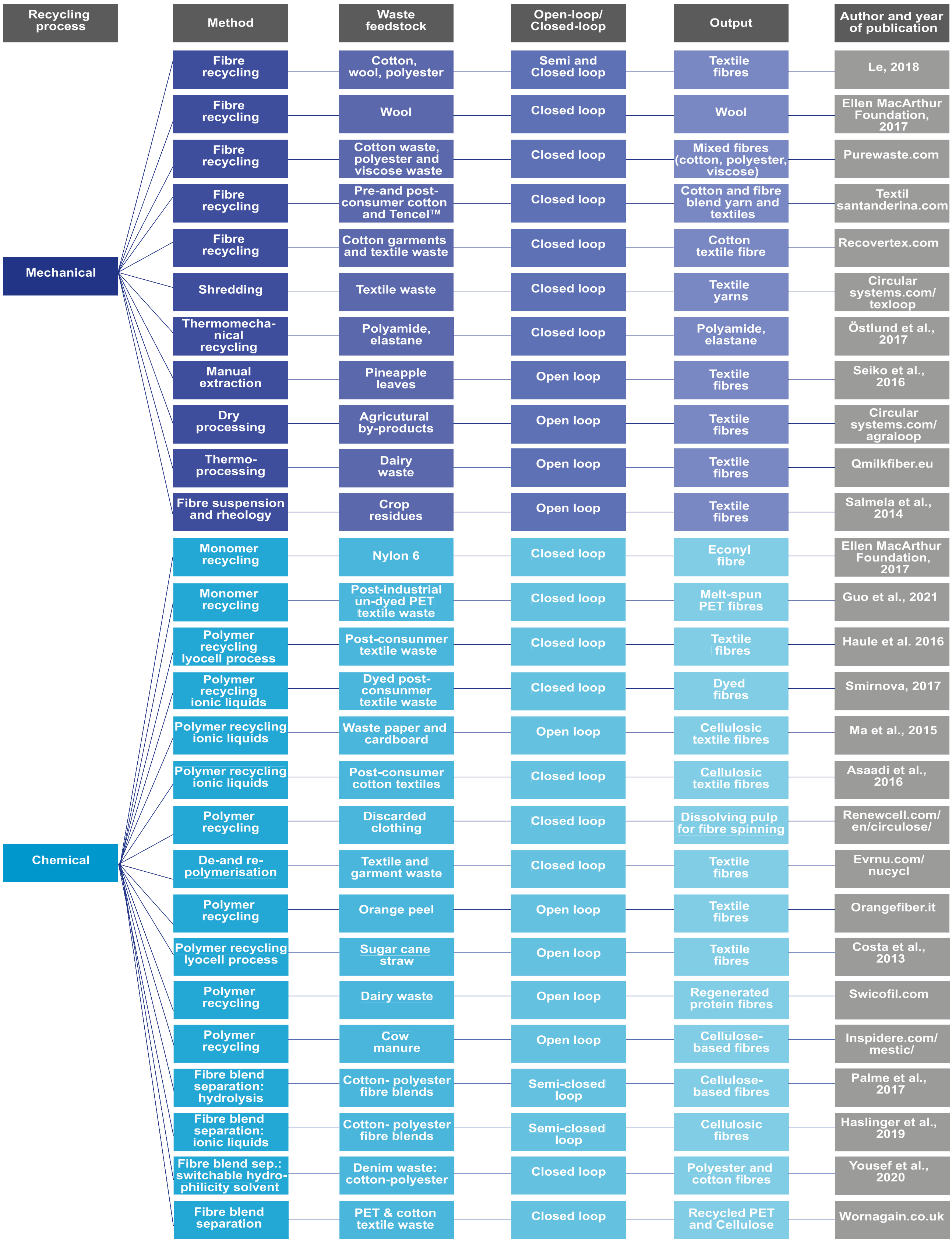

Fig. 2. Map of closed-loop mechanical and chemical recycling processes detailing different methods and fibre types, and open-loop processes that use agricultural by-products as the feedstock (Textil Santanderina; Worn Again; Recover Textile Systems, 2021; Swicofil).

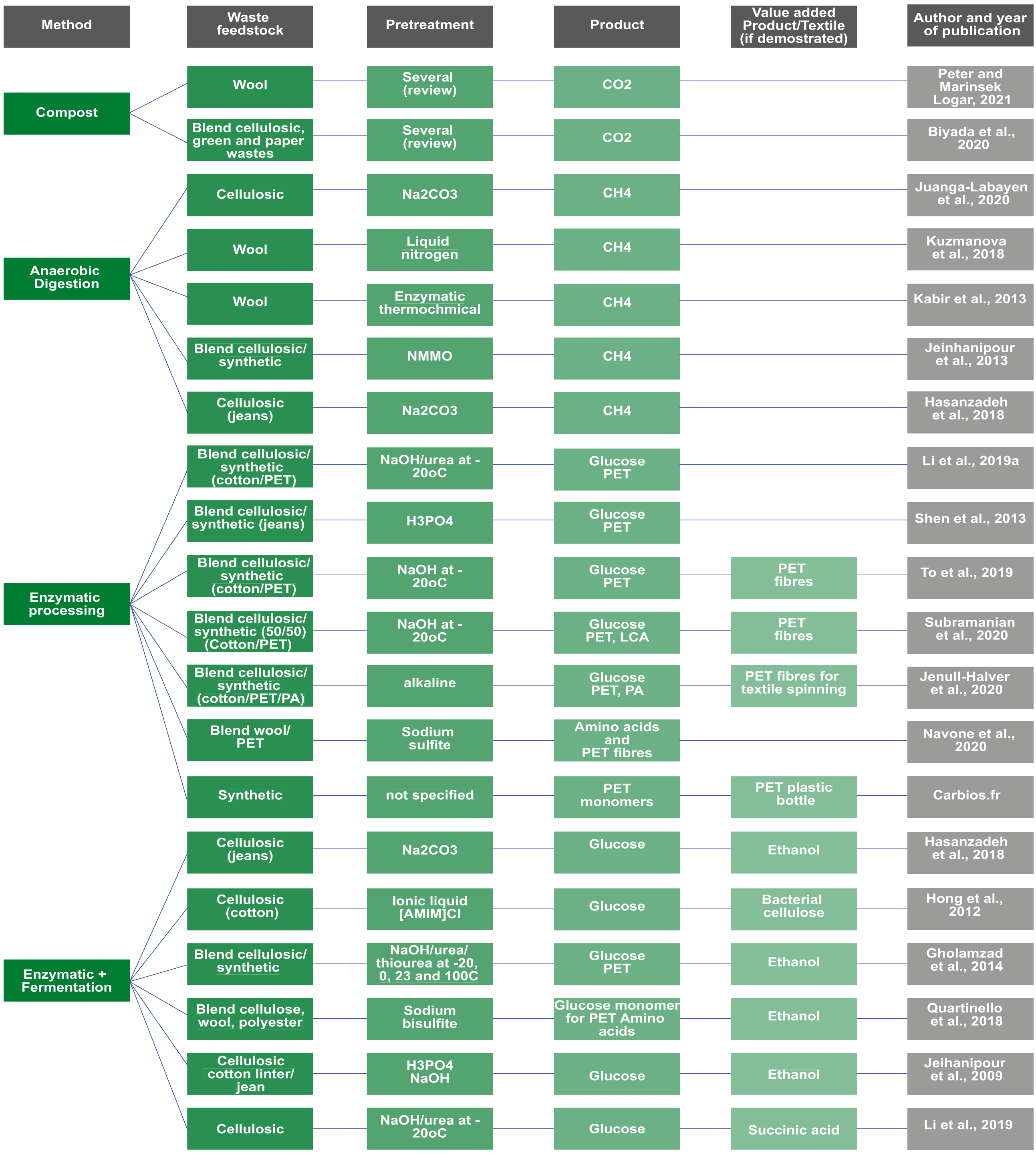

Fig. 3. Map of open-loop enzymatic and biological processes where textile is the feedstock (Kuzmanova et al., 2018; Kabir et al., 2013; Jeihanipour et al., 2013; NatureWorks, 2021).

common mechanical recycling method to process any type of textile fibre is through shredding or cutting. Before textiles can be shredded into smaller pieces, any hardware part such as zips and buttons need to be removed, which often requires human intervention (WRAP, 2014). The shredded textile is subsequently torn to produce a sliver that can be spun into new yarns (Roos et al., 2019). One of the limits of mechanical recycling is that fibres in the shredding process are shortened and therefore the production of new yarn often necessitates blending the recycled fibre with virgin fibres, in order to obtain the necessary strength and quality for apparel (Le, 2018). This mechanical recycling method is established at industrial scale for textile waste recycling of cotton fibres (Le, 2018). Cotton adopts a semi closed-loop recycling process, in which the recycled fibre is mixed with virgin cotton fibre and spun into new yarn. Wool adopts a closed-loop process without the need of mixing in virgin fibres when the textile is recycled for the first time (Ellen MacArthur Foundation, 2017). Fibre to fibre recycling for wool is also referred to as ‘regenerated wool’ by Italian wool producers operating in the area of Prato (Ellen MacArthur Foundation, 2017, p.98). If recycled for a second time, the fibre loses its quality and an open-loop process is adopted where the fibre is downcycled (Russell et al.,

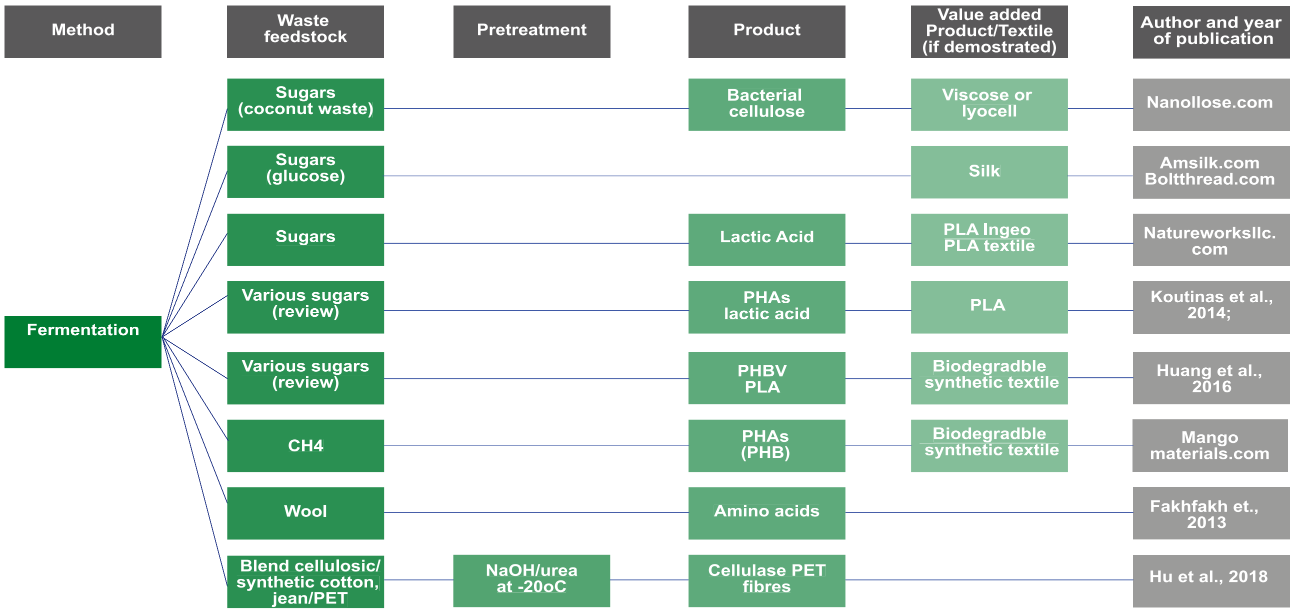

Fig. 4. Map of open-loop biological processes where textile is the final product from fermentation or when fermentation produces amino-acids and enzymes.

2016). Polyester (Drochytka et al., 2017) and acrylic (El Wazna et al., 2017) synthetic wastes have been recycled in an open-loop process where the fibre is downcycled into a non-woven fabric or insulation materials.

3.1.2. *Thermomechanical recycling*

This process is also referred to as mechanical polymer recycling (Ellen MacArthur Foundation, 2017) or thermal recycling (Niinimäki et al., 2020). Thermomechanical recycling melts synthetic fibres before these can be re-spun into new fibres or reshaped into other forms. Thermomechanical recycling is used for pure synthetic fibres such as nylon 6, thermoplastic polyurethane, elastane and polyamide 6 (Kunchimon et al., 2019), however properties of re-spun elastane fibres demonstrate breakage, unless it is transformed into solid shapes such as bars (Östlund et al., 2017). Despite this process being technologically feasible and a promising solution for polyester recycling, it is not yet scaled at industry level due to high costs involved in removing contaminants (Ellen MacArthur Foundation, 2017), except for limited commercial examples such as Japanese company Teijin fibres (Hamouda and Lu, 2014).

3.1.3. *Commercial scale transformation of agricultural by-products into textiles*

Larger scale open-loop processes transforming agricultural by-products into textile fibres have been developed. Recent circular economy approaches propose to make 'the most of food' and to transform food by-products into value-added materials such as textile fibres (Ellen MacArthur Foundation, 2019). These include open-loop mechanical recycling methods for the extraction of fibres from agricultural by-products and waste for textile production. Whilst the process requires manual and time-intensive methods to separate the fibres from the by-product by hand, it is common in the Philippines to use pineapple leaf fibres to produce Pina cloth garments (Seiko et al., 2016), which has been commercialised in Europe under the product name Piñatex® (Ananas Anam, n.d.). Agraloop BioFibre™ by Circular Systems™ uses a mixed mechanical and wet spinning approach to produce a biofibre from a range of agricultural by-products such as the fibre-rich stems or straw of oil-seed hemp, oil-seed flax, rice straw, pineapple leaves, banana tree trunks and sugar cane bagasse (Circular Systems, n.d.). The biorefinery concept produces a range of co-products such as packaging, fertiliser and bio-energy (Circular Systems, n.d.).

Recycling of agricultural by-products and waste into fibres for textiles is also enabled by the development of new technologies. Spinnova in Finland have patented a mechanical process that is inspired by a spider web to use fibre suspension and rheology for the production of cellulose that is converted into textile fibres 'using technologies currently available in [the] pulp and paper industry' (Salmela et al., 2014). Whilst originally this process used virgin feedstock such as wood pulp, it has been adapted to produce prototype textiles from agricultural residues such as straw (Remington, 2019). CelluComp in Scotland have extracted cellulose nanofibres from potato and sugar beet using a micro fibrillated cellulose process which could, in principle, be used for textiles (Holland et al., 2019). Another mechanical process uses heat to form man-made QMilk fibres from casein using waste from dairy farms (QMilkfibre, n.d.).

3.1.4. *Sustainability of mechanical textile recycling process*

There is very little information on the environmental impact of recycling processes. Much of it is either too specific to generalise, or not sufficiently transparent for broad conclusions to be made. A large number of environmental impacts need to be considered (e.g. land use, global warming potential, energy use, chemical use and associated pollution, water use etc.) and it is difficult to summarise. However, energy consumption can be used as a rough proxy for environmental impact for preliminary assessments. Production processes for virgin cotton fibre tend to consume around 50 MJ/kg of fibre (Rana et al., 2015). Processes common to all recycling schemes (sorting, transport etc.) consume between 2 and 5 MJ/kg; if a washing and drying process is required, a further 5 MJ/kg is added. The mechanical recycling process itself is relatively low energy, between 0.2 and 0.4 MJ/kg (for substitution of virgin yarn) and 0.9 MJ/kg for shredding and carding for flax replacement (Schmidt et al., 2016). Thus, in most cases, mechanical recycling will use between 5% and 20% of the energy used in virgin fibre production.

3.2. *Chemical processes for textile recycling*

Chemical recycling provides substantial opportunities to maintain materials in a closed loop. However, with the rate of closed-loop processes in chemical textile recycling still only at 1%, these technologies are yet to scale to industrial capacity (Ellen MacArthur Foundation, 2017). There are two types of chemical processes for textile recycling:

monomer and polymer recycling. While current chemical methods for polymer recycling often degrade the polymer chain, leading to a loss of quality in the recycled fibre, monomer recycling transforms end-of-use materials into virgin-quality fibres (Guo et al., 2021). Chemical recycling can, in principle, be applied to most textile fibres, however, monomer recycling is currently only being used for synthetic fibres.

3.2.1. Monomer recycling

In monomer recycling, the polymer chain is disassembled to obtain intact monomers. The monomers can then be transformed into new virgin polymers via polymerisation. This process has been adopted to transform different types of textile waste into fibres. An example of this is ECONYL fibre, which is made from Nylon 6 in discarded carpet and fishing nets (Ellen MacArthur Foundation, 2017). Recycling of PET plastic into textile fibres has been established using different chemical methods and a wide range of literature is available on this open-loop process. The dyes of polyester fabrics can hinder closed loop recycling, and methods such as sodium formaldehyde sulfoxylate have been proposed (Fei et al., 2020). Most recently, recycling of post-industrial un-dyed PET textile waste that is melt-spun into fibres has been established using Mg–Al double oxides pellets as a sustainable catalyst (Guo et al., 2021). Monomer recycling for cellulose and wool based textile materials has been explored to produce new products but has so far not resulted in textiles. The European RESYNTEX project transforms protein-based waste such as wool into amino acids and peptides via chemical degradation that are used to produce adhesives (Delahay, 2019).

3.2.2. Polymer recycling

Chemical polymer recycling has been investigated for cellulose-based, synthetic and blends of fibres (Roos et al., 2019). In polymer recycling, the fibre is taken apart using mechanical processes such as shredding, followed by chemical dissolution with specific often dangerous solvents. The polymer remains preserved and the fibres are regenerated and spun (Sherwood, 2020). Hardware parts and dyes need to be removed for these processes and the latter involve toxic chemicals such as bleach. Recently, the reintroduction of dyed post-consumer textile waste into a new lifecycle has been achieved using ionic liquid solvents and this would reduce its environmental impact significantly (Smirnova, 2017). Historically, chemical polymer recycling of post-consumer cotton or cellulosic by-products has evolved from the processes developed for the production of man-made fibres from virgin wood pulp, the viscose and lyocell processes. In particular, N-methylmorpholine-N-oxide (NMMO) is used industrially by Lenzing to produce Tencel™ fibres in the lyocell process (Lyocell.info, n.d.). The use of NMMO is seen as more sustainable than carbon disulfide used for viscose, because it is carried out in mild conditions and the solvent can be recycled (Jiang et al., 2020). The lyocell process has been adapted for the development of fibres from pre-consumer and post-consumer waste added to wood pulp (Tencel, 2021) and cotton waste garments with and without wood pulp (Haule et al., 2016). Instead of NMMO, ionic liquids (ILs) can be used to solubilise cellulose. ILs are salts in a liquid state usually with a melting point lower than 100 °C (Ma et al., 2015). ILs have been applied to dissolve post-consumer cotton waste textiles (Asaadi et al., 2016), paper and cardboard waste in order to regenerate cellulose-based textile fibres using the Ioncell-F technology (Ma et al., 2015). Compared to NMMO from the Lyocell process, ionic liquids are greener molecules that can dissolve cellulose at moderate temperatures with no cellulose or solvent degradation. Research is also invested in solvent recovery by 'successive thermal treatments under reduced pressure' (Elsayed et al., 2020). Companies including Circulose® (Re:newcell, 2021) and Nucycle™ (Evrnu, 2020) are currently bringing chemical polymer recycling of cellulose-based textiles to a commercial scale. Polymer recycling has also been applied to the recycling of cellulosic by-products. For example, Orange Fiber in Italy uses orange peel, a by-product of the juice industry, as a cellulose source to make textile fibres for yarns and fabrics (Orange Fiber, n.d.). The NMMO process has also been demonstrated with sugar cane straw, a by-product from the sugar cane industry (Costa et al., 2013). Other by-products resulting from animal farming include cow manure for cellulose-based viscose (Essaïdi, 2015) and dairy waste in the form of regenerated protein fibres named Swicofil, using acrylonitrile instead of formaldehyde (Cordis, 2017).

3.2.3. The separation of fibre blends

A main barrier to recycling post-consumer textiles is the fact that they usually consist of fibre blends Niinimäki et al., 2020. Chemical recycling methods can only be applied to one type of feedstock. Therefore, recent technologies are being upscaled that enable the chemical separation of these materials for polymer and monomer recycling. Södra OnceMore™ (n.d.) can dissolve polyester from blends of white poly-cotton to recover cellulose that can be combined with wood pulp for the making of new cellulose-based textiles (Palme et al., 2017). Palme et al. (2017) developed a chemical process that uses sodium hydroxide to depolymerise the PET component of polyester, so that the intact cotton component can be recovered for fibre production. Ionic liquids have also been applied to dissolve the cellulose component of cotton polyester blended fibres in order to spin cellulosic fibres (Haslinger et al., 2019). A switchable hydrophilicity solvent was used to dissolve polyester from cotton fibres in denim waste, in which both fibres types can be reused (Yousef et al., 2020). The separation of fibre blends recovered 98% of the polyester fraction while the cellulose was fermented into ethanol following enzymatic hydrolysis (Gholamzad et al., 2014). Other processes that achieve a separation of fibre blends are Worn Again's technology and the partnership between the Hong Kong Research Institute for Textiles and Apparel and H&M Foundation (Ellen MacArthur Foundation, 2017), however these have yet to reach a commercial scale.

3.2.4. Sustainability of chemical textile recycling processes

Chemical recycling processes can achieve the development of high-value fibres in textile recycling, however the toxicity of the solvents used for dissolution is a big issue. The environmental impacts of chemical recycling processes also lie in the energy required and water use. Solvent reuse is established for scaled-up processes such as NMMO but is not yet fully efficient for ionic liquids. While processes using ionic liquids can function at lower temperatures than NMMO, these are associated with a higher cost. The reliance of the textile and apparel industry on virgin synthetic or cotton feedstock for fibre production is exacerbated by the higher costs of fibres resulting from chemical recycling. There is now a need for governmental policy to support the scale-up and uptake of these technologies. Current investments are being put in place to increase novel polymer recycling technologies from a pilot to a commercial scale by 2030 (Östlund et al., 2015), which would support reducing costs as well as improving the process efficiency and environmental impacts.

As with mechanical recycling, data is sparse, but one source suggests that chemical recycling of polyester consumes around 12 MJ/kg; considerably more than mechanical recycling, but still less than virgin polyester production (>100 MJ/kg) (Schmidt et al., 2016).

3.3. Bio-based processes adaptable to textile recycling

Bio-based processes will be discussed under three main headings: biological, i.e. end-of-life biological decomposition where microorganisms are deconstructing textile materials to simple molecules (compost and anaerobic digestion), and biochemical, i.e. enzymatic depolymerisation which uses enzymes to deconstruct textile polymers into monomers and fermentation which, in this article, is the use of microorganisms to transform feedstock into desirable products.

3.3.1. Biological decomposition

Composting is a biological process where aerobic microorganisms transform organic materials into carbon dioxide (CO2), ammonia, water and heat in the presence of oxygen, water and nitrogen. Composting is relatively cheap and has been applied to organic solid waste (green waste) and human and animal wastes but not much to textiles yet, although a study of the microbial composition of a textile-based compost will help develop this further (Biyada et al., 2020). Composting of waste textiles is limited to natural and semi-synthetic fibres and is seen as better than incineration because it can produce organic fertilisers that can improve the level of soil organic matter, long-term soil fertility, and productivity (Ellen McArthur Foundation, 2017). Wool and wool sludge can be composted to produce fertilizer and raw wool has also been used by itself to enhance plant growth (see review: Petek and Marinšek Logar, 2021). Composting can be an excellent tool for biotechnologists to mine for novel enzymes that can deconstruct plant cellulosic complexes (Alessi et al., 2018). A similar approach could be envisaged for textiles.

Anaerobic digestion (AD) occurs in the absence of oxygen and in the presence of methanogenic bacteria and leads to the production of small-chain acids and ultimately a gaseous mixture of methane (biogas) and CO2 from macromolecules such as lipids, proteins and carbohydrates (Oreggioni et al., 2017). It is usually used to transform food and agricultural waste into methane when mixed with an inoculum from a water treatment plant or manure. It has also been shown to be useful for the transformation of production effluents or by-products in energy that can then be used to fuel the process the by-products originate from. Anaerobic digestion has been reported using cellulose rich feedstock such as wheat straw (Reilly et al., 2015), and textile waste (Juanga-Labayen et al., 2021, and examples in Fig. 3).

The products from these processes cannot be reused to make materials so they could not constitute a step in closed-loop recycling. However, they constitute a relatively feasible alternative to incineration or landfill as long as the amount of GHG emitted is well managed which will need to be assessed when scaling up. Given that circularity of textiles encompasses closed-loop processes in order to retain the value of materials for longer, biological decomposition such as composting should be the final step in returning nutrients back to the soil.

3.3.2. Enzymatic depolymerisation

Most enzymatic processes which use cellulosic biomass as a feedstock for fermentation require a chemical pretreatment before the enzymatic step that converts the cellulose into sugars. A wide range of pretreatments for lignocellulosic biomass have already been developed and reviewed (Beig et al., 2020). Processes include size reduction, the use of acid or alkaline solution, ammonia, pressure and/or high temperature, microwave (Yu et al., 2020), and ionic liquid (George et al., 2015). A detailed review has been commissioned by the Biomass Biorefinery Network (2021). In the case of cellulosic textiles, pretreatments are less developed but interest in the area is expanding, and mainly consist of size reduction and high concentration of caustic alkaline solutions (sodium hydroxide 7–20%) in a process similar to mercerisation (Shuhua et al., 2020). In contrast to usual cellulosic feedstocks, the use of alkaline solution to pretreat cellulose from textile waste is usually more efficient at low temperatures such as −20 °C and in the presence of urea or thiourea (Figs. 3 and 4). Gholamzad et al. (2014) compared a range of temperature and Hu et al. (2018) used alkaline pretreatment at low temperature with blend feedstocks to produce cellulases. Li et al. (2019a) recovered glucose from textile waste (mix of cotton and polyester) after a pre-treatment in alkaline conditions at −20 °C. The energy demand of the cooling step was reported to be the most prominent detrimental step in the life cycle analysis produced (Subramanian et al., 2020). Pretreatments with phosphoric acid and sodium carbonate which may offer less impact on the environment have been previously studied (Hasanzadeh et al., 2018), resulting in a reduction of the cotton crystallinity (Shen et al., 2013). Organic solvent such as ionic liquid or N-methylmorpholine-N-oxide (NMMO) are novel and greener molecules used to dissolve cellulose. The ionic liquid (IL) 1-allyl- 3-methylimidazolium chloride ([AMIM]Cl) was efficient in pretreating cotton waste for enzymatic hydrolysis but the authors concluded that the cost of the ionic liquid would hinder commercialisation (Hong et al., 2012). Viable use of ionic liquid has been reported with biomass and the same strategy may be applied to textile feedstocks (Brandt-Talbot et al., 2017). Further research is now needed to lower the cost and environmental impact of textile waste pretreatments to enable the development of a commercially viable biochemical recycling route of waste textile.

Cellulolytic enzyme cocktails (cellulases) have been used to depolymerise cellulose polymers into their constituent monomer glucose. The efficiency of the hydrolysis step also called saccharification is dependent on the efficiency of the cellulases which account for the second most expensive step in the process after pretreatment (IEA Bioenergy, 2020) Cellulase research is extensive and cellulose cocktails are available commercially from companies such as Novozymes (Denmark), DuPont (USA), and Roche (Switzerland). In the context of textiles, the use of cellulase cocktails with pretreated textile blends to produce soluble sugars and PET fibres has been reported (Jenull-Halver et al., 2020 and examples in Fig. 3). The advantages of an enzymatic hydrolysis are that the cellulose fraction becomes soluble and can be separated from other fractions in the textile blend without the need for manual sorting. Interestingly, Vecchiato et al. (2018) recovered the flame-retardant pigment from dyed viscose. In a similar concept, a blend of wool-cotton-polyester textile waste could be recycled by sequential and selective use of enzymes to depolymerase the wool and cotton before recovering the polyester synthetic fibre fraction and fermenting the glucose to ethanol (Quartinello et al., 2018).

Wool is constituted by a fibrous polymer of amino acids called keratin and is characterised by its resistance to digestion. This recalcitrance is due to the large number of cysteinyl residues, crossed-linked with disulphide bridges that confer additional strength and rigidity to the polymer. These disulphide links require the use of reducing agents for chemical dissociation. The enzymes responsible for wool depolymerisation are proteases called keratinases that can convert wool polymers in its constituent amino acids. Research on keratinases focussed on recycling the keratin rich waste from the sheep and poultry industry and their aim is to hydrolyse the keratin for the production of protein rich fish meal. Raw wool from sheep is a good environment to find or grow microbes that can metabolise keratin (Petek and Marinšek Logar, 2021). Fang et al. (2013) identified and purified 3 proteins K1, K2 and K3 from a poultry farm. K1 and K2 were able to modify the cuticle layer of wool, which would have commercial application for wool textile processing while K1, K2 and K3 were able to fully hydrolyse wool. Hydrolysis of keratin is usually carried out as microbial hydrolysis where microorganisms grow in media containing keratin and secrete keratinases in the liquid medium. The use of enzymes for recycling wool textile waste as part of textile waste blend has been reported by Navone et al. (2020) where they use an enzyme available commercially to specifically solubilise the wool fraction of the textile blend.

In the case of synthetic materials, nature has no experience in constructing or deconstructing man-made polyester type material and it is essential to use state of the art biotechnology tools to achieve significant progress. There has also been much interest in mining for enzymes that could depolymerise PET used for plastic bottles because the process can be seen as easier than mechanical sorting of the different class of plastic (Koshti et al., 2018). In order to engineer such a new enzyme, several already known enzymes from the esterase, cutinase and lipase families have been studied in microorganisms that were cultured on PET as a source of carbon. In 2016, a novel enzyme called Polyethylene Terephthalate hydrolase or PETase was characterised by Yoshida et al. (2016). The enzyme resembled a putative lipase and could specifically depolymerase PET, but the use of genetic engineering enabled it to improve its productivity (Austin et al., 2018). Since 2016, other PETases have been discovered and improved. Recently, Tournier et al. (2020) demonstrated that their improved PETase could hydrolyse post-consumer PET flakes and that the monomers could be used to produce *de novo* PET plastic of commercial quality, achieving closed loop recycling for plastic bottles. This was further applied recently by the French company Carbios (Carbios, n.d) that produced clear bottles containing 100% recycled Purified Terephthalic Acid (rPTA) from synthetic textile waste. The same concept has been pursued to engineer enzymes that could depolymerise nylon (Biundo et al., 2019).

3.3.3. Fermentation

Fermentation is a metabolic process that produces chemical changes in organic substrates through the action of enzymes. Practically, fermentation is the intentional use of microorganisms such as bacteria and fungi as well as eukaryotic cells, to make products useful to humans.

3.3.3.1. Solid-state fermentation using textile wastes. Solid-state Fermentation (SSF) is a bioprocess carried out in absence or near absence of free water, but with the moisture necessary to allow growth and metabolic activity of microorganisms. These conditions resemble the natural environment in which they exist and from which microorganisms are isolated (Thomas et al., 2013). It has been used to produce enzymes industrially for biochemical processes especially cellulases (Review Maftukhah, 2019). Other enzymes involved in the depolymerisation of lignocellulosic substrates were also produced by using cellulosic-rich residues as substrate (Verma et al., 2021). Hu et al. (2018) used waste textiles as a substrate for SSF with *Trichoderma reesei* to produce cellulases that can be further used to hydrolyse pretreated waste textiles into monomer sugars efficiently allowing the recovery of the insoluble polyester fraction. The work was further advanced by optimising pretreatment and hydrolysis to enable the separation of the textile cellulose (depolymerised into sugar which are soluble) and the insoluble polyester.

3.3.3.2. Submerged fermentation using textile wastes. Submerged fermentation is more commonly used. In submerged fermentation, a liquid is the reaction medium and the compounds of interest are secreted in the liquid fraction. This process is used for the production of a range of fuel molecules and platform chemicals from hydrolysed cellulose rich feedstocks and include ethanol, lactic acid, citric acid, succinic acid, butanol as further described below (Sheldon, 2014). The use of wastes instead of virgin resources as feedstock for fermentation has been described by Koutinas et al. (2014). Submerged fermentation using textiles has been described in the literature. Wang et al. (2018) used submerged fungi fermentation to produce cellulases from textile wastes. Yeast fermentation was reported from cellulosic textile to produce bioethanol (Jeihanipour and Taherzadeh, 2009) as well as succinic acid (Li et al., 2019b). Gholamzad et al. (2014) carried out simultaneous saccharification and fermentation from mixed textile waste to produce ethanol and recover polyester. Sheep wool waste has been fermented using a new keratinolytic bacterium, *Bacillus pumilus A1,* to produce a hydrolysate rich in amino acid that could be used for the production of fishmeal (Fakhfakh et al., 2013).

To minimise costs and increase efficiency in biorefineries, scientists are trying to simplify processes by making the fermenting organism(s) produce all the necessary enzymes to transform the biomass into products (Yee et al., 2014). This decrease in the number of steps aims to increase the cost efficiency of the overall process. There are two options to enable this. The fermenting organism needs either to be able to express the cellulases and other enzymes, or more than one micro-organism (co-cultures) has to be fermented at the same time. This is described as consolidated bioprocessing. One main application is hydrolysis and co-fermentation of lignocellulosic biomass in a single step for biofuel production (Rastogi and Shrivastava, 2017). There is a need for the enzyme-based biorefinery sector to develop novel microorganisms with desired properties that are both robust and adapted to specific feedstock and products such as textiles. Advanced biotechnological approaches such as metagenomics, next-generation sequencing and metabolic engineering can help in developing novel strains that will be suitable for performing multiple functions in a single bioreactor.

3.3.3.3. Biopolymer bio-manufacturing. The polymers used to make textiles–cellulose for cotton, amino acids and polyester–can be produced directly by specific microorganisms (Fig. 4). We believe biopolymer biomanufacturing by microorganisms can be a step in textile recycling if the organism ferments feedstock that originates from textiles. In nature, cellulose is found in plants but it can also be biosynthesised by a range of microorganisms (Chawla et al., 2009). The most efficient producers of cellulose are *Acetobacter hansenii*, *A. pasteurianus* and mainly *Komagataeitobacter xylinus*, a gram-negative bacterium, that was discovered in vinegar by Brown (1886) and is an essential ingredient for the preparation of Kombucha tea and 'nata de coco'. Bacterial cellulose (BC) microfibrils have unique physical properties that have enabled their use at commercial scale for various applications. These have been extensively reviewed and include packaging (Azeredo et al., 2019), medical industry as wound dressing and the production of loudspeaker diaphragms (Hussain et al., 2019). There is extensive information on its culture and biochemical pathway and the need to improve its culture for large scale commercial production has also been widely reported (Lee et al., 2014). Recently, the genome of several bacterial cellulose producing strains has been sequenced (Prust et al., 2005) and genetically engineered lines were generated (Florea et al., 2016). A Symbiotic Culture of Bacteria and Yeast also called SCOBY is commercially used for the fermentation and production of kombucha tea. This concept similar to consolidated processing was used by Gilbert et al. (2021) to engineer co-cultures of a BC-producing bacterium with engineered lab strains of yeast in order to develop a synthetic SCOBY providing an example of engineered living materials. The interest in bacterial cellulose for the textile industry has two facets. Firstly, the static fermentation produces an intertwined mat of cellulose microfibers across the surface of the culture vessel. This material can be grown in the shape and size of the growth vessel, allowing simple shaping of non-woven textiles by designers (Rathinamoorthy and Kiruba, 2020). The first research on bacterial cellulose from the fashion domain was performed by the British fashion designer Suzanne Lee for her research project Biocouture. The inherent properties of the bacterial cellulose are however not ideal for it to be used directly as clothes, especially its hydrophilicity and paper-like feel. Beyond textiles, bacterial cellulose has also been processed for its potential use as a vegan leather alternative using coconut water (Malai Biomaterials Design, 2021) or Kombucha/tea (ScobyTec, n.d.). The second interest of bacterial cellulose is to use it as a source of cellulose for textile spinning in a similar principle as viscose made from tree cellulose as demonstrated by the company Nanollose™ (Nanollose, 2021). Finally, cotton-based waste textiles were pretreated with ionic liquids and hydrolysed using commercial cellulases before bacterial cellulose fermentation, but a closed-loop process for textile production was not explored (Hong et al., 2012).

Silk is a protein-based fibre historically used for textiles. Silk's unique physical and biological properties are produced by spiders and the larvae of the mulberry silkworm. Because of silk's outstanding mechanical properties and biocompatibility, and the difficulty in obtaining large amounts naturally with farming, research has been carried out to produce it using controlled processes. A chemical process was reported by Courtaulds Ltd but did not lead to commercial scale (Bamford, 1996). Engineered yeast (Bowen et al., 2018) and *E. coli* (Xia et al., 2010) have been reported to be able to produce silk proteins successfully; and the process has been developed industrially for textile applications in Germany (AMSilk, n.d.) and the U.S. (Bolt thread, n.d.). For example, Microsilk™ is produced using engineered yeast fermentation of sugar followed by yarn spinning to produce a novel protein-based textile (Boltthread.com).

Many bacteria are capable of accumulating polyesters of hydroxyalkanoates in the form of cytoplasmic granules when carbon sources are in excess and nutrients depleted. Polyhydroxyalkanoates (PHA) are a large family of polymers that can be blown and moulded, foamed and processed into yarns. PHAs have biocompatibility, biodegradability, and material properties like those of commodity plastics and are produced commercially by fermentation from sugars to make bioplastic (Koutinas et al., 2014) and textiles (Mango Materials, 2021) or mixed with the bio-based polylactic acid to make biodegradable bio-based textiles (Huang et al., 2017).

3.3.4. Sustainability of bio-based processes

The advantage of bio-based processes is that they have typically very low energy demand, use benign solvents and chemicals and are based on renewable rather than fossil carbon. It is important to note that while bio-based processes occur under mild conditions, this alone is no guarantee of overall process sustainability, which needs to be considered from both an economic and environmental point of view. Feedstocks usually require a pretreatment that involve the use of caustic chemicals, specific vessels and energy (described in 3.3.2). The use of commercial enzymes increases costs and environmental impact significantly. These elements have hindered the development of commercially viable biofuel production from lignocellulosic feedstock and the same can be expected when using textile material as feedstock. Another issue that affected biorefinery scale-up for the production of bioproducts is concentration. In comparison to other processes, feedstocks are usually not pure and enzymatic reactions can be efficient but lead to a concentration of sugars too low for further economically viable fermentation. The advantage of end-of-life textiles is that cotton is 100% cellulose instead of 35% in lignocellulosic feedstock such as straw. Finally, many processes described in this review include the use of Genetically Modified Organisms (GMO) which attracts public concerns that will need to be dealt with before full commercial development.

4. Summary and perspectives

In this paper, we review the current concepts and associated mechanical and chemical processes developed for textile recycling in the context of a circular economy. Mechanical recycling is well established but cannot take place infinitely as the process degrades the quality of the textiles. Whilst mechanical recycling is adopted in industry, chemical recycling can achieve a high-quality output but is not yet sufficiently scaled. In recycling models, the use of microorganisms is usually limited to biodegradation with or without co-generation of products such as methane.

We introduce two novel options: enzymatic and biological processes as an alternative to chemical recycling coupled with bio-manufacturing of biopolymers as a source of novel materials for textile applications. While these options are not yet used in a closed-loop recycling format, i.e. textile to textile, these processes have been demonstrated in open loop recycling i.e. use textiles to make feedstock for fermentation or the use of biopolymers to make textile fabrics. For example, textiles have been demonstrated as feedstock for the production of ethanol and succinic acid. PLA obtained from lactic acid have been used to make novel textile materials. The aim of this paper is to highlight the potential of research to close the loop where end-of-life textiles would be used to make novel virgin textile materials using enzymes and fermentation.

The advantage of using enzymes to deconstruct end-of- life textiles is that they can overcome the issues associated with the multi-material nature of textiles such as polycotton. Enzymes are specific to individual textile types and can solubilise specific polymers/fabric types enabling further processes with the non-hydrolysed fractions. To produce textiles from current end-of-life textiles commercially will still require further research on reusing dyes and finishes. Pretreatments will have to be improved as described in this paper so that the process remains sustainable and economically viable. Novel micro-organisms will have to be developed to increase substrate conversion rate and produce biopolymers that can be efficiently spun into novel textile fibres. The sustainability and potential for scalability will have to be considered early on in the development of these new materials. The use of energy and demand for water will also have to be monitored as enzymatic and biological processes have been found to require large amounts of water. It will also be important to design novel textiles from the fibres and ensure that the resulting garments can be reused or recycled within a circular economy context.

This paper aims to demonstrate the potential of a multi-disciplinary approach for recycling where textile materials would be regenerative i.e. textile fibres would have the potential to be recycled to novel virgin-quality material instead of down-cycled to lower quality products. This approach involves the use of enzymatic and biological processes not only to decompose the textile at the end of life but also to produce it. This is an opportunity to inspire future research into biochemical, enzymatic and biological processes focused on innovative textile processes. This paper also proposes a new model for textile recycling that modifies the current concept of separate biological and technical cycles in the circular economy.

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Miriam Ribul: Conceptualization, Methodology, Writing – original draft, Visualization. **Alexandra Lanot:** Conceptualization, Methodology, Writing – original draft, Visualization. **Chiara Tommencioni Pisapia:** Writing – original draft, Visualization. **Phil Purnell:** Writing – review & editing. **Simon J. McQueen-Mason:** Writing – review & editing. **Sharon Baurley:** Writing – review & editing.

Declaration of competing interest

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

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