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Investigating waste mineral-filled cellulose sourcing in circular economy for regeneration into composite: Matching existing market volumes of oil-based plastics for packaging

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ABSTRACT

Global consumption of plastics has increased continuously in recent decades, leaving today's society with one of the most pressing environmental problems, plastic pollution. Current research has been focused on the development of bio-sustainable products with the aim of replacing the use of petroleum-based polymers with sustainable, renewable, and environmentally friendly materials. In this context, bioplastics have emerged, and where possible supporting biodegradability. The most abundant polymer occurring naturally is cellulose and remains one of the most promising renewable materials to replace plastic. This work forms part of a larger research activity studying the novel production of regenerated cellulose using ionic liquid dissolution, with the aim of drawing on filler-containing paper and board waste as a raw material for potential plastic replacement in circular economy. Analysis applied to a literature search is reported comparing the current consumption of plastics in packaging, the generation of packaging waste, the production and consumption of paper and cardboard and finally the recycling rate of these materials in Europe with special focus on material that either fails to enter, or is rejected during, the classical recycling process. Based on these data, commercialisation of cellulose regeneration made solely from the volume of paper and board waste that has failed to enter standard recycling, excluding single use products, e.g., sanitary, would be able to cover the current demand for plastic films used in packaging, and that no additional biomass in principle is needed. This finding not only supports the effort being made to scale-up the cellulose regeneration process commercially but relieves the pressure on agricultural land currently foreseen to be otherwise needed for extensive biomass production, rather allowing it to serve its main purpose in food production, so contributing to the circular economy quest for sustainability obviating environmental impact.

1. Introduction

Global consumption of plastics continues to increase, creating one of the most pressing environmental problems arising from today's society, plastic pollution (Lazarevic et al., 2010; Okoffo, et al., 2021; Wang, et al., 2021; Babaei, et al., 2023; Zhang, et al., 2023). Much current research has, therefore, been focused on the development of bio-sustainable materials with the aim of replacing petroleum-based polymer products with renewables. In this context, bioplastics have emerged as a potential alternative (Vinod, et al., 2020; Haque and Naebe, 2022; Stark and Matuana, 2021; Yu, et al., 2020).

Cellulose as the most abundant polymer occurring naturally remains the most promising amongst biopolymers (Shaghaleh, et al., 2018; Aimonen, et al., 2022; Yang, et al., 2022). However, a major difficulty it poses in respect to traditional high temperature polymer melt processing, is related to its inherently high crystallinity, resulting in temperatures much higher than its oxidation (combustion) point in air to reach melting, cellulose melting point being 467 °C (Dauenhauer, et al., 2016), thus precluding its use in traditional polymer processing lines. Therefore, to recover cellulose from biomass or waste practically requires

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forming a solution of cellulose in a suitable solvent medium, and then regenerating it into the extruded form desired, which, until recently has been challenging, frequently involving environmentally unfriendly chemistry (Medronho and Lindman, 2015; Imani, et al., 2022).

To date, most regenerated cellulose materials are produced by the viscose process, the oldest commercial production route (Kauffman, 1993). The main problems with this process, besides the use of highly toxic and ecologically harmful chemicals, are related to high energy and water consumption. In this context, many different sustainable solvents for cellulose have been investigated in recent years. Amongst these, ionic liquid (IL) has shown itself to be one of the most promising (Medronho and Lindman, 2015; Vocht, et al., 2021; Zhang, et al., 2017; Gupta and Jiang, 2015; Jedvert and Heinze, 2017; Pinkert, et al., 2009; McGill, et al., 2021; Shi and Wang, 2016; Xia, et al., 2020; Swatloski, et al., 2002). An essential consideration for the development of regenerated cellulose from solution in this way is the sourcing of the cellulose biomass. In this respect, waste materials, arising either within or falling outside standard recycling, are identified as highly interesting, giving the opportunity to establish circular economy (Al Rashid and Koç, 2023; Haque, et al., 2023).

Waste paper and board generally contain varying levels of mineral filler (Kostić, et al., 2022). Such filler is generally considered to be a linear economy product, i.e., virgin filler, extracted directly from natural deposits, is refined and ground to the desired particle size, or indirectly synthesised filler via extensive prior treatment to form a synthetic, usually precipitated, pigment product, and then applied across multiple industries. Typical application areas for filler include pulp and paper, paints and adhesives, plastics, and construction materials (Gane, et al., 2020; Barać, et al., 2022). Calcium carbonate in both its natural ground form (GCC) or as precipitated calcium carbonate (PCC) has become the main white filler component in many of these applications mentioned. PCC in these applications is derived from calcining the natural carbonate at high temperature, releasing CO_2 chemically and from the energy source used for calcining, to form calcium oxide, which subsequently undergoes slaking in water and exposure to CO_2. Paper and board-making predominantly uses calcium carbonate as filler, and, in high quality grades, additionally as concentrated coating suspension pigment. However, due to the difficulties in handling such mineral content in recycling wastepaper and board, and lack of viability, particularly due to its downgrading by printing ink etc., usually either unfilled waste paper, such as newsprint or demineralised paper after deinking, has been used to recover cellulose and prepare new paper and board materials.

The deinking process applied as standard technology today relies on flotation of hydrophobic ink components, as exemplified in the INGEDE standards (INGEDE, Oetztalerstrasse 5B, 81,373 Munich, Germany). This limits its suitability to the traditional common printing processes, including offset and solvent-based rotogravure, whilst precluding to a large extent water-based printing ink technologies. Mixed office and label waste, for example, is separated out of wastepaper collection due to the dominating digital printing technologies used for documentation, such as inkjet and electrophotography using water-based and toner inks, respectively. If office and label waste are included in the standard recycling process, colorant ink residues remain in the recycled fibre product, making it unsuitable for the majority of printing and writing, and packaging label grades (Ma, et al., 2016; Tsatsis, et al., 2017; Vukoje and Rožić, 2018). If one adds the quantity of suitable paper and packaging waste, which, for whatever reason, has evaded the recycling collection, typically due to poor recycling habits, ineffective collection by some local authorities and, on occasion, less than diligent subcontracted private companies, together with uneconomic waste material separation etc., then, rather than being re-used, environmentally valuable cellulose and mineral filler is ending up in incinerators, or, worse still, landfill (Platnieks, et al., 2020).

Recent work (Kostić, et al., 2022) has illustrated how calcium carbonate-cellulose composite filament can be produced via a dry-jet wet spinning process using 1,5-diazabicyclo[4.3.0] non-5-enium acetate ([DBNH][OAc]), constructed from DBN and glacial acetic acid, as ionic liquid for cellulose dissolution (Parviainen, et al., 2015), adding ground calcium carbonate (GCC) as filler. Additionally, in the same research, digitally printed office waste paper containing already present filler content (PCC) was used for cellulose filament making under the same conditions without any pretreatment. It was found that the process is robust and simple in concept. Moreover, the results showed that the presence of calcium carbonate filler in the composite matrix containing printed toner enhanced the tensile strength, elasticity, and Young's modulus under initial strain, whilst increasing thermal stability in comparison to filament made from virgin cellulose alone, demonstrating that digitally printed office paper, can be effectively used in cellulose composite filament manufacturing without deinking.

This paper is part of the current work discussed above, studying a novel cellulose-based film production from mineral filler-containing paper and board waste, forming a solution using IL, retaining the particulate nature of the filler, regenerating the cellulose polymer structure from solution, and evaluating the resulting film properties as a suitable product for replacing plastic whilst simultaneously supporting circular economy. The specific question being addressed here is whether the volume of available waste cellulose material resource within the paper and packaging usage chain is sufficient without having to revert to virgin cellulose from dedicated forest and agricultural biomass production. Focus is also given to a first analysis of the commercial viability of composites made from calcium carbonate and regenerated cellulose (Imani, et al., 2022; Perišić, et al., 2022). On the one hand, the current demand for plastic packaging film in Europe and the availability of cellulose waste with filler content have been analysed to determine whether it is possible to cover the current demand. On the other hand, the price of the respective resources conventionally used to produce packaging has been considered, namely, the price of fossil oil to form a typical plastic film, such as low-density polyethylene (LDPE), versus the price of virgin cellulose in the case where the novel film replacement should be made initially solely from dissolution of virgin fibre. Potential sources of cellulose waste that could be rapidly integrated have also been studied, i.e., those which currently pay fees (depending on the country) to deposit such waste in landfill.

Finally, being also a part of the validation of the business model, an example of such a biomaterial cellulose-based film has been produced on a laboratory scale in order to confirm the process feasibility and to validate the convenience of the product properties for the customer. The material formulation content of the example is used to provide resource material costings.

2. Methods and material parameter tools for analysis

One of the most viable methods for creating environmentally friendly composites is the use of natural fibres as reinforcement, and cellulose appears frequently to be an excellent candidate. However, to achieve consistency and strength, virgin fibres and fillers generally have to be used, since recycled fibre alone can be too weak and unsuitable for combining with filler, the particles of which, in the case of fibre-based composites, act to interfere physically with the fibre-fibre bonding, so weakening the composite even further. Thus, in such traditional composites it is rarely possible to source cellulose from the recycling chain, rendering the environmental use of cellulose frequently uneconomic (Medronho and Lindman, 2015). Therefore, the concept of regenerating cellulose from waste, i.e., renewing the crystal structure from the chemical cellulose units is a technical breakthrough enabling full use of cellulose in whatever physical condition. As background to this viability analysis, the production of cellulose-$CaCO_3$ composite filaments and films from cellulose pulp and office paper waste, itself containing 27 w/w% precipitated $CaCO_3$ filler (PCC) without any pretreatment, was undertaken using [DHBN][OAc] as the ionic liquid solvent to form a dope, from which the filament or film is generated. This process is summarised here, so that subsequently the material parameters, such as sourcing and processing, can be referred to as they are progressively built into the subsequent market and viability analysis - the main subject of this paper.

2.1. Film composite materials

The materials used in the starting experimental series, as published in Kostić, et al., 2022, were:

virgin cellulose pulp (pre-hydrolysed Kraft (PHK)) birch pulp, delivered in sheet form (494 cm^3 g^{-1}) from Stora Enso Enocell Specialty Cellulose, Finland

ground calcium carbonate (GCC)

copy paper; electrophotographically printed (toner) and unprinted, containing in-situ *precipitated calcium carbonate* (PCC) at 27 w/w% filler loading

DBN (1,5-Diazabicyclo[4.3.0] non-5-ene) 99 %, Fluorochem, UK

acetic acid (glacial) 100 %, Merck, Germany

2.1.1. Cellulose sources

The two sources of cellulose, namely, ground (refined) virgin pulp, into which GCC was introduced during cellulose dissolution, and ground copy paper, which already contained PCC in it. This latter cellulose source (copy paper) was obtained from A4 sheets of 80 g m^{-2} basis weight, consisting of fully bleached chemical pulp containing 27 w/w% of PCC measured by ash. Copy paper was used either unprinted or printed, i.e., office waste paper, containing residues of ink toner, binders and surfactants.

2.1.2. Filler

GCC as virgin filler was provided by OMYA Hustadmarmor AS (Elnesvågen, Norway). This filler was produced chemical-free by wet grinding and subsequent dewatering. It was used by addition into the cellulose regeneration process in each of two forms, (i) chemical-free, and (ii) treated by vigorous premixing in ionic liquid (IL). Each filler was introduced into the process by preconditioning at 80 °C for 10 min prior to mixing with liquefied IL at the same temperature under high shear.

2.1.3. Ionic liquid

The ionic liquid 1,5-diazabicyclo[4.3.0] non-5-ene1-ium acetate ([DBNH][OAc]) was synthesised in the laboratory by slow addition of equimolar amounts of glacial acetic acid to DBN. The mixture was stirred for 1 h at 80 °C.

2.1.4. Dope formulation

Dopes containing 13 w/w% cellulose, either as virgin cellulose pulp together with 0.26 w/w% of GCC (2 parts by weight filler on 100 parts by weight cellulose), printed or unprinted copy paper or mixtures thereof, were formed by dissolution in [DBNH][OAc] according to the following procedure:

1. pulp and/or paper sheets were ground and sieved through 100 μm mesh,
2. [DBNH][OAc] was liquefied in a vessel held in a water bath for 1 h at 80 °C, followed by filtering through a metal filter (5 − 6 μm absolute fineness) in an oven,
3. the designated amount of cellulose source powder was added to the melted (at 80 °C) [DBNH][OAc] and stirred at 80 °C under 30 min^{-1} (rpm) for 30 min using an in-house designed water-heated mixer.

Five different dopes were prepared to analyse their properties, including before and after GCC filler particles were added to the virgin cellulose dope, and in the case of copy paper when PCC was already a constituent.

The five different dopes were each formed in 15 mm diameter cylindrical moulds (dimensions corresponding to the filament spinning cylinder), wrapped, and sealed against moisture, and stored in a refrigerator (~8 °C).

2.1.5. Extrusion/spinning

Extrusion of dope in the spinning process was performed under controlled temperature to maintain the liquefaction of the IL (80 °C). Immediately upon extrusion through the chosen nozzle, the filament or film was subjected to cooling under constant stress passing first through an air gap from the spinneret towards a cool water bath used to “set” the dope.

The extrudate was drawn at a defined draw ratio (linear extension) to orientate the crystallising cellulose in the machine direction during cooling from 80 °C to 23 °C, using a controlled bobbin winding rate.

3. Market analysis

3.1. Global plastic production and single-use plastics consumption

Since the last century, global plastic production and consumption has increased relentlessly, realising the value of the material properties. In 1950 the worldwide plastic production was 1.5 million metric tonnes (Mt), and in 2020 it reached the enormous figure of 367 Mt, of which 55 Mt alone were produced in Europe (Statista, 2020).

Bibliographic data show that consumption of plastic for packaging in the EU has remained stable over time, 20 Mt. The consumption of plastic packaging remains stable over time while the consumption of paper and board packaging is growing slightly. It is understood that consumption is not growing in Europe but in other countries, mainly in China and the United States (Plastics Europe, 2021). This trend is also reflected in respect to production, Fig. 1**,** where it can be observed that plastic production in Europe has also remained fairly stable over time at around 60 Mt. This is because most of the plastics consumed in the world are produced in China, which in 2020 accounted for 32 % of the world's plastics production. NAFTA (North American Free Trade Agreement) is the second largest producer of plastics accounting for 19 %, followed by the rest of Asia (17 %) and Europe accounting for only 15 % of global plastics production in 2020 (Plastics Europe, 2021).

The growing demand for single-use plastics has contributed to the high global production rate of plastics (Plastics Europe, 2021). Single-use plastics are the most consumed polymer material today and is almost exclusively made from petrochemicals (Natural Resources Defense Council, Inc., 2020). These single-use plastic materials are typically used for packaging and are intended for one-time use only. The fact that single-use plastics are difficult and expensive to recycle results in only 14 % of packaging waste being recycled. After their single use they are, therefore, typically disposed of in landfills (Chen, et al., 2021), or to a lesser extent incinerated (energy recovery). This results in more than

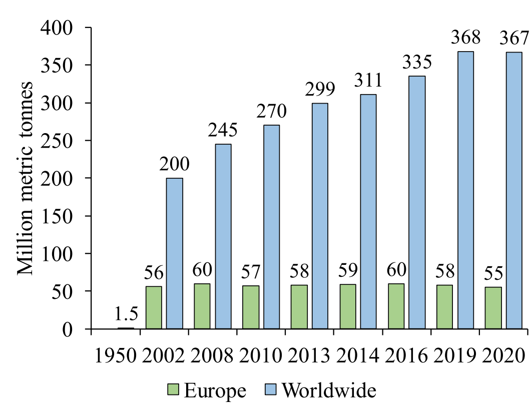

Fig. 1. Global plastic production, summarised with copyright permission from (Statista, 2020).

80 % of plastic packaging waste being disposed of (Chen, et al., 2021).

The case above of single-use plastics is considered the most environmentally critical, however the problem is also generally critical across all areas of plastic consumption. In Europe, 29 Mt of post-consumer plastic waste were collected in 2020, of which 34 % was recycled, 42 % went to energy recovery and 24 % ended up in landfill (Plastics Europe, 2021).

3.2. Waste generation and packaging waste

The consumption of packaging in particular has been steadily increasing over time and, in step, so has the concern of companies for the whole life cycle of their products (Klaiman, et al., 2016). In response to this concern, it is the task of the analysis undertaken here to know the amount of packaging waste, contrasting between cellulose-containing and plastic, that is generated, and, thus, demonstrate through market data that the accepted urgent need to improve waste management today could provide a source for cellulose raw materials and fillers as an alternative to plastic. Parallel to the plastic waste issue, therefore, it is crucial to understand how the waste cycle in general works in Europe. To this end, bibliographic research has been carried out.

In addition to the composition of packaging waste, it is the responsibility of this project to know the amount of packaging waste that is generated and thus demonstrate through market data the urgent need to improve waste management today.

In order to collect data on this overall activity in the paper market, several databases have been consulted mainly CEPI, Eurostat and Statista.

Fig. 2 shows the evolution of general packaging waste generated in Europe. Over time, the generation of packaging waste has been increasing steadily (Eurostat, 2019). In one decade, more than 10 Mt of additional packaging waste have been generated, raising the amount to 78 Mt of packaging waste in Europe alone. This immense amount of waste comes largely from packaging materials that are thrown away after use.

Traditionally, and during the last decade, Europe exported most of its plastic, and paper and board waste to other countries (mainly to China), where it was managed and recycled by its recycling industry (Wen, et al., 2021). China has been seen to be the largest importer of waste of all kinds, however, in recent years negative impacts on the environment have been observed due to imported recyclable waste. For this reason, the Chinese government have increasingly tightened restrictions on waste imports. In 2017 it banned the import of 24 types of solid waste, including plastic and paper waste. Prior to 2017 the recycling rate of packaging waste was increasing in line with the generation of waste, however, due to this new law in China, from 2017 onwards a decrease in recycling rate can be clearly observed in respect to the European Union (EU), Fig. 3.

While more than 1.6 Mt of plastic waste were exported from Europe in 2015, only 0.41 Mt were exported in 2017 (after the ban in China) and

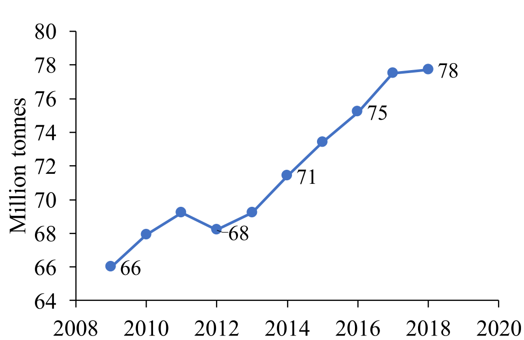

Fig. 2. Packaging waste generated in EU, summarised with EU copyright freedom from (Eurostat, 2019).

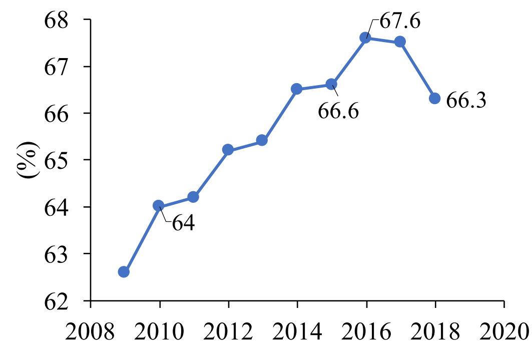

Fig. 3. Recycling rate of packaging waste in Europe, summarised with EU copyright freedom from (Eurostat, 2019).

less than 14 kt in 2019 (Statista, 2020). Paper waste exports to China decreased from 5 Mt to less than 0.7 Mt between 2016 and 2019 (Eurostat, 2019).

In parallel to the discussion considering plastic entering waste streams there is increasing activity invested in plastic recycling and upcycling to generate a circular economy based on plastic reuse. Methods employed include pyrolysis, gasification, photoreforming, and mechanical reprocessing. Careful focus is required, however, to realise the process and the final potential, particularly on the plastic waste composition, the resulting conversion products arising from processing, the relevant polymer reaction mechanisms, in turn subject to the need for catalyst selection governing conversion efficiencies, and the opportunities present in each case for subsequent polymer modification (Zhao, et al., 2022). The demand for single component, or fixed component ratio polymer content within cycles is derived from the nature of polymer matrix formation, which, based on thermodynamic grounds, fails to promote internal bonding when the matrix alignment is disturbed, or the bonding moieties present are incompatible. The difficulties surrounding ensuring separation of different polymer chemistries remains the dominant major obstacle within the consumer society acting as the recycling source, together with the economic challenges of meeting the costs of the methods described for upcycling (Shi, et al., 2024; Wamba, et al., 2023). This specificity for plastic polymer differs from the basic universal building block continuity of pure crystalline cellulose, comprising multiple subunits of glucose, in which the chemistry remains constant. Ancillary materials, such as lignin and non-crystalline celluloses, hemicelluloses, become separated into either the desired soluble cellulose species in ionic liquid, which then contribute to the final regenerated cellulose target product derived from recrystallisation, or insoluble components, which, in principle, can either be removed or incorporated as effective additional filler, thus not disturbing the intermolecular hydrogen bonding of the intrinsic cellulose structure. Nonetheless, provided single polymer comprising plastic waste can be effectively sourced, in reality most effectively achievable within speciality applications rather than widespread packaging, keeping plastic within a circular economy mitigates to some large extent its loss into long lasting pollutant.

When considering cellulose materials, two readily identifiable positives repeatedly emerge: the sustainability of its biomaterial sourcing and the product biodegradability, intrinsic to the nature of natural cellulosic material. In the realm of the plastic economy, sourcing from biomaterials and biodegradability have also become a major focus for development, often associated within the confinement of speciality applications, where the economy can support the novelty and cost structure of deriving these polymers from bioresources (Mousavi, et al., 2024). The sourcing of biomaterial to synthesise biopolymers is also partly driven by the environmental desirability to upcycle the vast quantities of readily available arable agricultural waste, and, thus, brings the topic of regenerated cellulosic materials, retaining the natural chemical structure, into companionship with synthetic polymerisation derived from biopolymeric component material. The review provided by Ghosal et al. (Ghosal and Ghosh, 2023) critically discusses recent trends in this field of biopolymers, recognising the global environmental threat of plastics in general and presenting the drivers for the contrasting recycling routes of lignocellulosic biomass and synthetic polymer wastes. Once again, the opportunities for speciality applications including biodegradability and natural material tolerance by living tissue are identified, embracing drug delivery, tissue engineering, and antimicrobial applications.

As European waste exports fall sharply, while in the short term an "Exporter substitute" is sought that can recycle waste generated in Europe, waste is accumulating in landfills. As a result, not only is the value of waste in Europe decreasing, but also the fees for depositing waste in European landfills are expected to increase. Non-recycled waste paper accumulating in Europe loses its value, while at the same time the cost of the reduced amount of recycled pulp supply in the EU increases. This raises the question whether it is possible to use existing paper and board waste generated in the EU, which now lies outside European recycling capacity, either due to reduced recycling activity in China or because the waste is not considered suitable for recycling or simply bypasses recycling, as a source of cellulose raw material to replace the current use of plastic in the production of packaging films. The analysis thus focuses on what the current consumption of packaging films in Europe is, and to estimate whether the amount of paper and board waste generated in the EU that could be used as raw material for these products can cover the demand.

Next, a literature search was carried out on the European market of the paper and board industry, to be able to estimate the amount of cellulose waste that is available outside the recycling circuit.

3.3. Waste cellulose sources, availability in Europe

As mentioned before, cellulose is one of the most used materials as biopolymer. Here we focus on the potential for producing a biomaterials-based film from waste cellulose sources. For this reason, in this section we identify the trends in the European paper market to understand the amount of paper and board that is consumed and actually recycled in order to estimate later the amount that is not recycled and so predict future scenarios for accessing this non-recycled raw material source based on trends identified by the paper industry.

Fig. 4 is a compilation drawn from the various Confederation of European Pulp and Paper Industries (CEPI) reported production and consumption data for the last few years. In contrast to the consumption of plastics, the consumption of paper and board in the European market has been declining gently over the years from 82 Mt in 2000 to 71 Mt in 2020 (CEPI, 2020). In the last 20 years technology has been advancing to evolve to an industry focused on digitisation.

In addition to the production and consumption of paper and cardboard, CEPI's annual report for 2020 shows the recycling rate in recent years within the European Union. Fig. 5 shows that the European industry has been increasing the recycling rate of paper and board in recent years. The recycling rate of paper and cardboard was 74 %. This leaves a big question that is important to ask, what happens with the remaining 26 %?

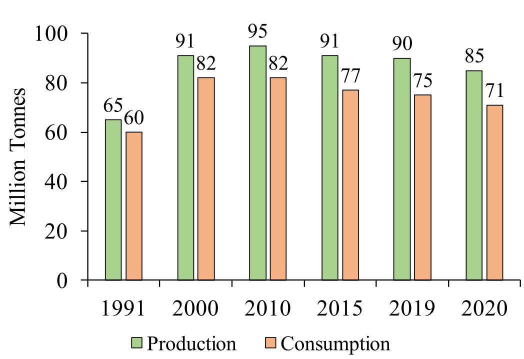

Fig. 4. Paper and Board in Europe, summarised with copyright permission from (CEPI, 2020).

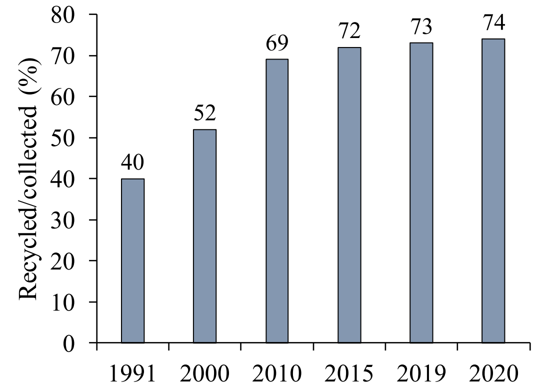

Fig. 5. Recycling rate in EU, summarised with copyright permission from (CEPI, 2020).

The report collects annual data from the European paper industry on the collection and recycling (utilisation) of paper for recycling. The illustration in Fig. 6 shows the evolution of these data over the last few years and shows that since 2010 the amount of paper for recycling collected has remained stable at 56 Mt, but that in the last year fewer tonnes were collected and recycled than in previous years.

Fig. 6, thus, confirms that in 2020 there were 6 Mt that were collected but not recycled, thus 6 MT of losses in paper for recycling. In this context, it is important to ask, where do the tonnes of paper for recycling that are collected but not recycled go?

One of the key points to obtaining answers to the question of cellulose waste availability is the correct interpretation of the data found regarding the EU paper and board market. It has been assumed that the total available quantity of not recycled paper and board comes from the sum of:

$$\begin{gathered}\textit{waste cellulosic between production and consumption}\\ +\\ \textit{waste cellulosic between collection and recycling}\\ +\\ \textit{single use products, e.g., sanitary}\end{gathered} \tag{1}$$

In order to estimate the amount of paper and board that is actually recycled and that which is not, some data have been taken from the available data sources and other data have been estimated by combining complementary information. The following data have been taken from the CEPI database. Annual CEPI reports on the market data of the paper industry in the EU countries are published on their website (https://

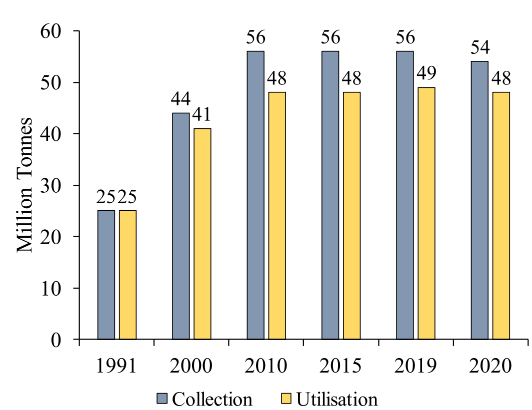

Fig. 6. Paper for recycling in EU, summarised with copyright permission from (CEPI, 2020).

www.cepi.org/):

production and consumption of paper and board (in tonnes), and recycling rate (%).

From the definition of recycling rate, Eq. (2), provided by CEPI, the tonnes of recycled paper and cardboard have been determined.

$$\textit{recycling rate} = \frac{\textit{recycled paper and board}}{\textit{consumption of paper and board}} \tag{2}$$

Data can be found on production, consumption, exports, imports, recycling rate of paper and board, as shown in Fig. 7.

The important information provided by Fig. 7 is the amount of waste from cellulosic sources that is not currently recycled in Europe, as expressed in Eq. (1). It is important to understand that certain single use streams are *per force* currently not included in the recycling loop. These are typically excluded either for reasons of hygiene (such as hygienic and sanitary products, including tissue paper), unrecoverable (such as wallpaper, honeycomb structures in furniture, plasterboard), or destroyed in use (such as cigarette paper, technical filters, teabags) (CEPI, 2022).

According to the data, the EU consumed 78 Mt of cellulosic sources and recycled 55 Mt of them in 2022. The recycling rate of paper was 70.5 %. This implies that 23 Mt of cellulosic waste, which may comprise single-use products, remained unrecycled in Europe (defined as EU member States (Eu-27) + Switzerland (CH) + Norway (NO) + United Kingdom (UK)). These trends indicate that the paper value chain has improved its recycling performance and reduced its environmental impact in 2022. However, there is still a gap between production and consumption, and between consumption and recycling, which means that some paper and board products are either exported or disposed of in other ways.

It should be noted that in reality there may be differences in the availability of the identified waste paper and board that is not recycled, as it is derived from the difference between the statistical values,

$$\textit{available cellulosic outside recycling} = \textit{production} - \textit{consumption} - \textit{recycled} \tag{3}$$

Moreover, the crucial question that should be answered from an environmental perspective is why are there these losses? Some of the answers could be paper mill losses, manufacturing losses, end use losses etc. For example, material that is collected for recycling is subsequently sorted into acceptable product and reject. The reject takes on many forms, including

(i) the number of times it has already been recycled, due to the weakening effect on cellulose fibre transitioning many times through paper/ board making and recycling
(ii) the technology content:
  a. combined packaging, e.g., lamination of plastic and metallic film with board (milk and juice cartons etc.), printed electronics labelling
  b. digitally printed material, e.g., inkjet and electrophotography (mixed office waste), water-based flexography, speciality inks (labelling, conductive, and metallic) which cannot enter the traditional flotation-based deinking process
  c. contaminated packaging, e.g., food, oil, and chemical products
(iii) selectivity of the recycling end use business, e.g., colour, dye content

In addition, there is the inefficiency of collection on behalf of local authorities or outsourced collectives. It can be the case that carefully separated household waste becomes remixed and so uneconomic to process further. Industry and business left unmonitored can follow the more economic path of incineration or even shipping waste to lower cost regional landfill.

All of the above-mentioned sources and reasons for cellulosic bypassing recycling are either technological (best case) or economically unviable (worse case), or mismanagement, including personal habits (worst case). In each of these cases, there is either economic cost or environmental cost, or both. With incentive for using this not recycled waste to form an alternative to plastic at many levels of application, depending on raw material quality (mainly contamination level), there will be associated immediate economic benefit – a major driving force – and a longer-term environmental benefit. It is not often that economic and environmental benefits go hand in hand!

4. Price of raw material

To analyse the viability of forming filaments and films from regenerated cellulose via the dissolution using ionic liquid (IL), it is necessary to create a business model structure that shows proven evidence that such a biomaterials-based film production is a suitable product to produce from which companies will make a profit.

Currently, the regeneration concept is a novel process, and many aspects need further optimisation. For example, the IL used to dissolve the source cellulose, from which the regenerated material in the form of, for example, films and filaments, is produced, must be recovered for reasons of material purity free from residual processing chemical as well as from economic necessity. IL recovery, in the case studied here, can be achieved by extraction of the residue after distillation of the in-line coagulation water bath. In addition, selection of the cellulosic waste source itself needs to be evaluated and optimal resource streams established. These factors will affect future research on the actual cost of regenerated cellulose film production and the benefits compared to conventional fossil oil polymer-based film production. Therefore, a rough approximation has been made to determine a first order estimate

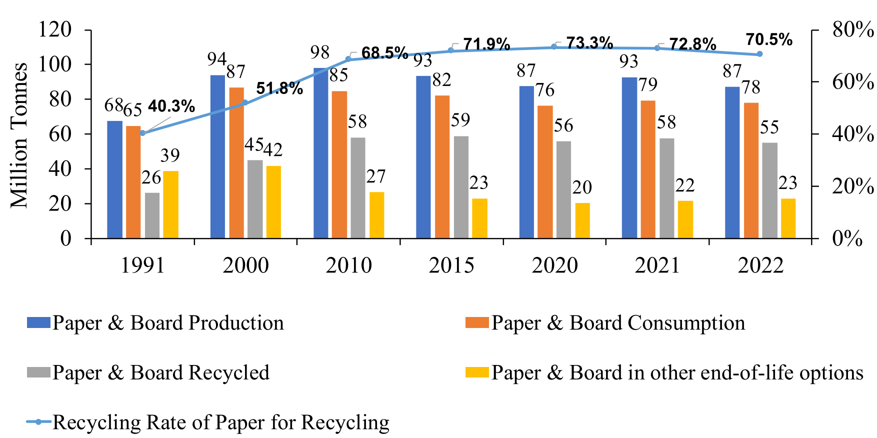

Fig. 7. Recycling of Paper & Board in Europe (Eu-27 + CH, NO & UK), summarised with copyright permission from (CEPI, 2022).

of the economic implications of this biomaterials-based production. The variable that has been selected to define the regenerated cellulose film business model is the price of the raw materials used.

In order to compare traditional plastic film production from fossil oil versus regenerated cellulose film from both virgin and from waste sources, the respective raw material price data were sought. The virgin sources for film production that have been considered are:

- fossil hydrocarbon for plastic film production, e.g., low density polyethylene (LDPE)
- virgin cellulose pulp
- waste sourced cellulosic
- filler particles (calcium carbonate)

as summarised in Table 1.

The residual cellulosic sources considered for this analysis are waste paper and board, and other biomass residues from waste between production and consumption and waste between collection and recycling, i.e., according to Eq. (1). This naturally ignores other the accessible sources outside the paper and board consideration, such as annual agricultural waste from material such as crop husks and stems. The key point to understand the potential benefits that this project can provide to the paper and board industries is based on the idea that conventional film production has a raw material cost that directly affects the price of the film, while regenerated cellulose film production based on waste paper and board outside the recycling chain does not have a raw material cost because the raw material is a waste that ends up in landfill. To a first approximation, the value of waste paper and board falling outside the recycling circuit is considered here as being necessarily equal to the current landfill costs. This assumption is based on the fact that the waste must be transported to the point of use as a raw material, and so should have at least an economic value equal to relieving the industry of landfill tax, whilst still supporting transport costs.

It should also be noted that in considering filler (Table 1), the analysis here ignores the value obtained from existing filler in the waste paper and board. Filler already in the waste cellulosic source has been shown to add stiffness and impact resistance to the film produced (Kostić, et al., 2022; Imani, et al., 2022). Furthermore, such filler is normally an economic burden following deinking in recycling, leading to massive high-density landfill. Although the predominant filler is calcium carbonate, and, as such, a material that is continually being renewed on a planetary eon scale, the concept of a single use filler remains anathema to environmental conservation. This process of retaining it in the filaments and films produced, thus enables otherwise waste mineral to be reused, essentially infinitely.

It is foreseen that companies which could potentially produce this novel cellulose-based film could offset taxes to the companies that currently have to deposit their waste paper to landfill, and so both will have benefits, i.e., companies that have to pay for the disposal of their waste will have to pay less and companies interested in producing this biomaterial film will have an income from their raw material acquisition. This is where one of the most important economic benefits that makes cellulose film production attractive for all companies active in the cellulose product market lies, as they can revalue their waste by supplying to the production of a film, which is biodegradable, and, in principle, essentially infinitely recyclable through the regeneration process (the cellulose molecule remains intact throughout unlike the weakening of fibre during standard recycling). In turn, such a film could replace plastic made from fossil oil that remains non-biodegradable and polluting on a planetary level.

Table 1
The virgin and material sources considered readily available currently comparing plastic film with regenerated cellulose from residual sources for regenerated cellulose film production.

Raw Material	Price Indicator	€/tonne
LDPE	Fossil oil price	1 200
Ionic liquid (largely recoverable and recyclable)	Current market price	2 000
Virgin pulp	Pulp price	800
Calcium carbonate	Pigment price	200
Waste paper and board (outside the recycling chain)	*Landfill cost for waste disposal (Finland)	73

*Landfill cost is the landfill tax (used as an estimate of the value of paper and board waste lost outside recycling).

5. Results

Various possible economic paths have emerged based on formulating the following questions:

(i) How big is the packaging film production demand?
(ii) Will it be possible to match the current plastic packaging film production demand from waste paper and board lying outside the recycling chain?
(iii) Is it necessary in the short term to find other cellulose biomass sources (virgin or recycled)?
(iv) Is the cost structure viable in comparison to the current plastic film costs?

Firstly, the answers to questions (i)-(iii) lie in the single analysis of comparing the current demand for plastic film and the available amount of waste paper and board that is not recycled today. If the available non-recycled waste paper and board can meet the demand, no additional biomass would be needed in the immediate future, but if it cannot meet the current demand, other sources of cellulose waste will have to be found to meet the current demand.

Turning to the question of whether demand can be met, the assumption made is as follows:

- *If the amount of non-recycled paper and board available is greater than or equal to the consumption of plastic packaging, the demand could be met.*
- *Vice versa, if the amount of non-recycled paper and board available is less than the consumption of plastic packaging, demand cannot be met.*

Recalling the data from Section 3, European market analysis, the bibliographic research undertaken here shows, firstly, that if we follow the first trial formulation published by (Kostić, et al., 2022), i.e., the production via IL dissolved cellulose dope, we may conclude the serendipitous finding, valid for Europe, that

$$\begin{gathered} \textit{volume of packaging plastic consumed} \\ \approx \textit{volume of paper and board waste lying outside the recycling chain,} \\ \textit{including single use} \\ \approx 23\,\mathrm{Mt} \end{gathered} \tag{4}$$

Although it is difficult to estimate the amount of single use grades included in this figure, to a first approximation a considered available amount remaining, excluding single use, could reasonably be considered to be of the order of ~20 Mt. To a first approximation, therefore, the amount of cellulose waste available in Europe lying outside the recycling chain could indeed alone cover the current demand for packaging film, or at least come close to it.

Secondly, the viability question posed in (iv) can be answered from the summary in Table 1, i.e., assuming processing costs are similar between oil extraction, refining, polymer synthesis and film production versus those for regenerated cellulose film production we have two scenarios,

1. equating raw material costs for fossil oil and virgin materials for regenerated cellulose, considering a filler loading of, say, 10 w/w% calcium carbonate added to 90 w/w% cellulose with an assumed

minimum recovery level of ~80 % for IL

$$\begin{aligned} &\textit{fossil oil price} = 1\ 200€/\text{tonne} \\ &\approx (\textit{virgin pulp price} \times 0.9) + (\textit{virgin filler price} \times 0.1) + (\textit{ionic liquid price} \times 0.2) \\ &\approx (800 \times 0.9) + (200 \times 0.1) + (2\ 000 \times 0.2) = 720 + 20 + 400 = 1\ 120€/\text{tonne} \end{aligned} \tag{5}$$

2. equating raw materials costs for fossil oil and waste cellulosic materials sourced outside the recycling chain, also considering a naturally occurring average filler load of 10 w/w% in the waste, thus obviating the need for virgin filler, and the same minimum 80 % recovery for IL

$$\begin{aligned} &\textit{fossil oil price} \\ &>> \textit{waste paper and board outside the recycling chain}\ (\textit{landfill tax}) \\ &+(\textit{ionic liquid price}\ \times 0.2) \\ &\Rightarrow 1\ 200\ >> 73 + 400 = 473\ €/\text{tonne} \end{aligned} \tag{6}$$

from which, we see that if one uses virgin cellulose and filler materials (case 1.) the costs between plastic film and regenerated cellulose film are similar, whereas using waste cellulose (already containing filler) sourced outside the recycling chain (case 2.) has the potential for large economic savings of ~700 €/tonne.

6. Conclusions

The main conclusions drawn from the study are that

- the sourcing of cellulosic material from waste lying outside the recycling chain to feed the process of regenerated filler-containing cellulose filament and film production according to, respectively, Kostić et al., 2022 and Imani et al., 2022, is quantitatively robust in terms of the analysed volume being available to meet the current demand for plastic film
- using virgin cellulose and filler materials the costs between plastic film and regenerated cellulose film are similar, whereas
- using waste cellulose (already containing filler) sourced outside the recycling chain has the potential for large economic savings of ~700 €/tonne of film.

Thus, the proposed solution to plastic packaging film replacement using biodegradable regenerated cellulose-filler composite, as reported by Kostić et al., 2022, not only contributes to circular economy at equal cost-quality value but could provide potential for economic gain for lower grade films derived from cellulosic waste lying outside the recycling chain today. Furthermore, this latter could be achieved solely from the cellulosic packaging waste volume analysed as lying outside the recycling chain without requiring further dedicated biomass, therefore contributing also to environmental conservation.

The packaging, agriculture and construction sectors are the main consumers of plastic films. The production of regenerated cellulose film could offer these sectors a renewable product that provides them with a cost-effective solution to replace their fossil oil-based products.

The contributions provided by this paper motivate the following issues to be investigated further.

- Collection of more accurate data on the amount of various segments in Europe contributing to the cellulosic waste that lies outside traditional recycling, such as office paper waste. This will require a comprehensive market analysis to which data from a spectrum of European inputs producing and handling office paper waste will require to be reported. This has proven to be one of the difficulties encountered in collecting market data, as the market data found refer generally only to the traditionally accepted paper and board groups of printing and writing, and packaging, for example, but not to detailed subgroups based on recyclability.
- Future possible sources of biomass remain to be investigated. It can be envisaged that agricultural waste biomass, including, for example, annual crop waste and stems could fit well into the novel regenerated cellulose process, especially now that the economic value, and mechanical and optical properties of cellulose-filler composites have been demonstrated in this context.

It is recognised that further research is needed on the production process of regenerated cellulose film. Questions such as, what will be the scope of production, the selling price of the film in terms of grade structure, and what could be the market share, need to be answered.

6.1. Future perspective

In addition, companies producing filler particles will also be interested, since valorised composites made including waste mineral would provide a solution to the waste management challenge from their customers as to the cost and environmental load of the current linear economy of mineral filler usage today.

CRediT authorship contribution statement

Monireh Imani: Investigation, Data curation, Writing – original draft, Visualization. **Isabel María Vidal Carreras:** Methodology, Formal analysis, Investigation, Data curation, Writing – original draft, Visualization. **Katarina Dimić-Mišić:** Investigation, Visualization. **Mirjana Kostić:** Methodology, Validation, Investigation, Writing – review & editing. **Ernest Barceló:** Conceptualization, Validation, Supervision, Funding acquisition. **María Alicia Cardete García:** Validation, Resources, Writing – review & editing, Supervision, Project administration, Funding acquisition. **Patrick Gane:** Conceptualization, Methodology, Validation, Formal analysis, Resources, Writing – original draft, Writing – review & editing, Visualization, Supervision, Project administration, Funding acquisition.

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.

Data availability

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

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