Full length article

Bioleaching/enzyme-based recycling of aluminium and polyethylene from beverage cartons packaging waste

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ABSTRACT

Multilayer materials are frequently used in food and beverage packaging, delivering favourable properties for storage and protection. However, their complex construction, consisting of several layers of plastics, aluminium and paperboard (i.e., beverage cartons) makes them difficult for complete material recycling. Currently, the common treatment process used is hydropulping, resulting in partial recovery of the paperboard only. The so-called beverage carton reject fraction, consisting of plastics, aluminium, and some fibres, is incinerated for energy recovery, leading to the loss of secondary resources. Here a novel recycling procedure based on enzymatic hydrolysis of cellulose, followed by bioleaching of aluminium finally allowing recovery of pure polyethylene is presented. Application of a cellulase cocktail resulted in the release of 15 mM glucose within 24 h, followed by over 95% aluminium bio-extraction within 3–7 days using bacterially produced sulfuric acid. Dissolved aluminium could afterwards be completely recovered by selective precipitation at pH 6.4, resulting in the formation of aluminium hydroxide. Pure polyethylene at the end of the process was used in re-processing and film production, showing comparable results to commercially available materials. Additionally, scaling up in a 1 L stirred tank reactor proofed the feasibility of the process in reject recycling. With this innovative, environmentally friendly recycling method, maximum material recovery could be achieved, leaving a minimum of impurities for incineration (<5%) behind.

1. Introduction

Globally, plastic production accounted for around 367 million tons in 2020 of which around 55 million tons were produced in the European Union (EU) (Association of Plastic Manufactureres, 2021). The biggest share, of approximately 40%, is used in the food packaging industry, to produce single- and multilayer packaging materials (Association of Plastic Manufactureres, 2021; Kaiser et al., 2017). High-density (HD-PE) and low-density polyethylene (LD-PE), polypropylene (PP), polystyrene (PS), poly(ethylene terephthalate) (PET) and polyvinylchloride (PVC) are the most frequently used polymers in this area (Kaiser et al., 2017). The combination of different plastics allows the design of functional food packaging for optimal handling and protection of the packed goods

by providing structural integrity, abrasion resistance, scalability, as well as oxygen and other gas barriers properties (Morrison, 2017). Among these, composite materials consisting of different layers of plastics, paperboard and aluminium, such as liquid packaging boards (LPBs), represent the most complex structures (Gesellschaft für Verpackungsmarktforschung GmbH, 2016). In fact, these materials consist of up to 4 layers of PE, with additional layers of paperboard and aluminium in between (Terapak.com, 2022). A study comparing the recycling rates for LPBs in Germany, Spain, Sweden and the UK using data from the ACE (Alliance for Beverage Cartons and the Environment) Zero Waste Europe and Eunomia (Eunomia Research & Consulting Ltd, Bristol, UK), revealed that only 51% of beverage cartons placed on the market were recycled in 2019 and 2020 (Lahme et al., 2020). Furthermore, the

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existing data show that only 30–36% of paper board where recycled, while polymer and aluminium were not considered for recycling at all (Lahme et al., 2020). So far, existing live cycle assessments (LCA´s) of beverage cartons support that they provide a relatively sustainable packaging solution, but the recycling rates used in the calculations strongly differ when using the new methodology implemented by the European Commission in 2019 (European Commission, 2018a; Lahme et al., 2020). This leads to the assumption that existing LCAs of LPBs may reflect and over-estimation of recycling benefits. According to the European plastic strategy, which states that all used packaging should be reusable or recyclable by 2030, new methods for the recycling of multilayer materials need to be developed (European Commission, 2018b). The original TetraPak® patent describes a method for the easy delamination of multilayer films by the introduction of a water-soluble, thermoplastic resin film (Bergerioux, 1996). Nowadays, the separation of paperboard and PE-aluminium layers is called hydropulping, and has been successfully implemented commercially (Zawadiak, 2017). Despite the fact that paperboard can almost be completely recovered by these methods, the separation of PE and aluminium still is not possible. Consequently, the so-called reject material is incinerated for energy recovery which does not contribute to full closure of the carbon cycle (Pellis et al., 2016). More innovative technologies, dealing with an easy delamination of plastic-aluminium multilayer materials based on the introduction of an reversible crosslinking agent (Kaiser, 2020), chemical dissolution of aluminium followed by gravity separation of different plastics (Lee et al., 2006) or solvent-targeted recovery and precipitation based on the selective dissolution and precipitation of different plastics like PE, PVC and PET (Walker et al., 2020) have been investigated over the last years. In other studies, researcher tried to modify plastics used in multilayer packaging in a way to enhance mechanical recycling (Trossaert et al., 2022), investigate new recycling strategies like molten metal pyrolysis (Riedewald et al., 2022), or produce new materials out of multilayer waste such as cellulose-aluminium-plastic composites (Bonadies et al., 2022). In this study, we combined different innovative, bio-based solutions for a closed recycling loop of aluminium and PE based on a biotechnological approach. To remove cellulosic impurities which would extremely detrimental in regranulation of PE, an enzymatic approach using cellulases (enzymes capable of hydrolysing glycosidic bonds in cellulose) was investigated, resulting in the generation of glucose (Jeoh et al., 2017). Next, the application of extreme acid tolerant, iron- and sulfur-oxidizing bacteria was investigated to selectively dissolve the aluminium layer (bioleaching). These bacteria are able to dissolve metals directly *via* redoxolysis or indirectly *via* acidolysis and complexolysis (Sethurajan et al., 2018; Srichandan et al., 2019). Both bioleaching for metal recovery and cellulase enzyme treatment for second-generation glucose recovery from lignocellulosics has been implemented in multi-thousand tons scale for example in heap- and stirred-tank bioleaching of nickel and copper in Scandinavia and Chile (Gentina and Acevedo, 2016; Morin and D'Hugues, 2007; Saari and Riekkola-Vanhanen, 2012) or in the production of cellulosic ethanol (European Technology and Innovation Platform, 2022). The dissolved aluminium can afterwards be selectively precipitated and re-introduced in the production cycle (Kremser et al., 2021a). Finally, the remaining LPB reject fraction was purified by density separation, resulting in almost pure PE. Additionally, the re-processability of the purified PE was evaluated by rotational rheometry and Fourier-Transformed Infrared Spectroscopy (FT-IR) and compared with commercially available PE.

2. Materials and methods

2.1. Reject material

Around 25 kg multilayer reject material from liquid beverage packaging cartons was provided by the German paper and recycling company Niederauer Mühle (Kreuzau, Germany) after the process of hydropulping. It consisted of up to A4 sized sheets of multilayer materials containing different polymers, aluminium and cellulosic remaining. Prior to characterization, the sample was dried at 60 °C for up to 48 h and shredded in a kitchen blender to a size smaller than 11 mm, followed by homogenization via sufficient mixing. Bigger plastic parts such as caps were manually removed before shredding.

2.2. Sample characterization

Fourier transformed infrared spectroscopy (FT-IR) was used for identification of different polymers and cellulose in the LPB reject material, as well as polymer characterization after bioleaching and polyethylene recovery. Therefore, polymer samples were measured between 4000 and 650 cm^{-1} on a Perkin Elmer Spectrum 100 FT-IR Spectrometer (PerkinElmer, Traiskirchen, Austria). All samples were acquired using 64 scans and a resolution of 2 cm^{-1}.

Surface characterization before and after each treatment step was performed with a Hitachi TM3030 scanning electron microscope (SEM) coupled with energy-dispersive X-ray spectroscopy (EDS) to show changes of the material composition during the different process steps (i. e. polymer, cellulose and aluminium).

2.3. Cellulose hydrolysis

The commercially available enzyme mixture Cellic® CTec3 (Novozymes, Baksvaerd, Denmark) consisting of different cellulases and hemicellulases was used in the hydrolysis experiments for cellulose removal. An enzymatic activity of 196 FPU mL^{-1} on filter paper and a protein content of around 183 mg mL^{-1} were previously determined and confirmed by other studies (Novy et al., 2019; van der Zwan et al., 2019). Different enzyme concentrations (i.e. 2.5, 5.0 and 7.5% (v/v)) were tested on their effectiveness to reduce the cellulose content of the LPB reject material. Therefore, 1 g of shredded material was added to a 250 mL Erlenmeyer flask, followed by the corresponding amounts of a 0.5 M citric acid buffer (pH=4.8) and enzyme to give a total reaction volume of 100 mL. All experiments were carried out at 50 °C and 150 rpm in a rotary shaker in biological triplicates and samples for glucose determination were taken after 3, 6 and 24 h. After hydrolysis, samples were sieved through a 100 μm sieve plate and dried in a 60 °C oven until complete dryness. By weighting the samples before and after cellulase treatment, the weight loss corresponding to the cellulose degradation was recorded.

2.4. Glucose determination

To measure the released glucose concentration over cellulose hydrolysis, samples were precipitated via Carrez precipitation to remove impurities. Therefore, 960 μL of diluted sample was mixed with 20 μL of a 0.25 M K$_4$[Fe(Cn)$_6$] x 3H$_2$O solution and vortexed, followed by the addition of 20 μL of a 1 M ZnSO$_4$ x 7H$_2$O solution and mixing. Afterwards, samples were centrifuged for 30 min at 12,700 rpm. The supernatant was filtered into HPLC vials through a 0.2 μm polyamide filter and stored at 4 °C. HPLC analysis was conducted using a 1260 HPLC system (Agilent Technologies, Santa Clara, USA) equipped with an ION-300 column (New Haven, USA). 0.01 N H$_2$SO$_4$ was used as mobile phase at 45 °C with an isocratic flow rate of 0.325 ml min^{-1}. A glucose calibration in the range from 1 to 100 mM was prepared and used for calculation of the released sugar concentration.

2.5. Bioleaching experiments

Indirect, non-contact experiments were carried out using a biogenically produced sulfuric acid which was provided from an ongoing in-lab study (see Supplementary material) (Kremser et al., 2022). To test the effect of LPB reject concentration on the aluminium leaching efficiency, three different concentrations (i.e., 1.0, 2.5 and 5% (w/v)) were used in the indirect leaching experiments. Therefore, the corresponding

amount of shredded material was added to a 250 mL Erlenmeyer flask and 100 mL of biogenic sulfuric acid was added. All experiments were carried out in biological triplicates and one blank for the highest LPB reject concentration, consisting of 100 mL of a 1.4 M commercial sulfuric acid was run in parallel. The flasks were incubated at room temperature and 150 rpm in an orbital shaker and samples for aluminium determination were taken after 1, 3 and 7 days. After aluminium leaching, samples were as well sieved through a 100 μm sieve plate, followed by a washing step with double distilled water (ddH$_2$O) to remove the remaining acid. The washing was conducted by putting the reject samples back into their flasks, adding 200 mL ddH$_2$O and shaking the samples at 150 rpm for 30 min. This procedure was repeated 3 times in total, ending up with drying the samples at 60 °C in the oven. The weight difference before and after aluminium leaching was used to calculate the aluminium content and compared to the spectrophotometrically aluminium determination.

Direct bioleaching experiments were performed using the sulfur- and iron-oxidizing bacterium *Acidithiobacillus ferrooxidans* (DSM 583, newly classified as *Acidithiobacillus ferridurans* (Moya-Beltrán et al., 2021)). Three different ferrous iron concentrations of 1.0, 2.5 and 5.0% (w/v) were used in the experiments by adding the corresponding amounts of FeSO$_4$·7H$_2$O to 1 L of DSMZ culture medium 70 adjusted to pH=1.4 with 18 M sulfuric acid. Prior to bioleaching experiments, 50 mL of culture was added to 450 mL of each ferrous iron medium and pre-cultivated for one week at 30 °C and 150 rpm in order to reach a full conversion of ferrous iron to ferric iron. After one week, 100 mL of culture put into 250 mL Erlenmeyer flasks in biological triplicates and two different LPB reject concentrations (i.e. 1.0 and 2.5% (w/v)) were added. Furthermore, 1.0% (w/v) of elemental sulfur was added to each flask to favour the reduction in pH by sulfur metabolization and the corresponding sulfuric acid production. Bioleaching experiments were carried out at 30 °C and 150 rpm for up to 14 days and samples for aluminium determination were taken after 1, 3, 7, 9 and 14 days of incubation. An abiotic blank experiment for each LPB reject concentration was run in parallel containing only culture medium and shredded material. Washing and drying of the samples after bioleaching was conducted as in the indirect, non-contact experiments.

2.6. Aluminium determination and precipitation

Aluminium concentrations during bioleaching experiments were measured using the Spectroquant® aluminium test (Merck KGaA, Darmstadt, Germany), in which aluminium ions react with chromazurol S to form a blue-violet complex. To use the method in 96-well plates, the protocol was adjusted in a previous study, accordingly (Kremser et al., 2021a).

Dissolved aluminium was recovered by selective precipitation at pH=6.5 (Wei et al., 2005). Therefore, the pH-value of the lixiviant was dropwise adjusted with 1 M NaOH and the precipitated Al(OH)$_3$ was collected by filtration through a Durapore® PVDF filter membrane with a pore size of 0.45 μm. The effectiveness of this method was assessed by measuring the aluminium concentration before and after precipitation.

2.7. Polyethylene recovery and characterization

The plastic material recovered after the cellulose and bioleaching treatments was subjected to a density separation in order to remove eventual polyester and polyamide impurities. 5 g of dried plastic LPB reject material was weighted in a 1.5 L beaker and 1 L ddH2O was added. The mixture was stirred for 15 min at 400 rpm and 25 °C and was then let settle for other 15 min. The "floating fraction" of the material, consisting mainly of PE, was recovered using a sieve while the "sinking fraction" was recovered via filtration. The two fractions were then freeze dried before further analysis.

Commercially available LDPE ALCUDIA® PE033 with a melt flow rate of 0.3 g/10 min and a density of 921 kg/m^3 was purchases from REPSOL (Madrid, Spain) and used for comparative experiments with the recycled PE fraction.

The homogenization of the samples was carried out in melt and therefore, a laboratory scale was used a co-rotating conical twin-screw extruder HAAKE™ MiniLab II Microcomponent. This extruder allowed the homogenization of polymer, obtaining 3–5 g samples for subsequent characterization. The HAAKE extruder allows working in a discontinuous way, monitoring and adjusting parameters such as temperature, motor torque, and viscosity during the process. In addition, it can be used in recirculation mode with the extraction of small aliquots if necessary. The homogenization of polymers by extrusion was carried out at 160 °C, 60 rpm for 5 min.

Films with a thickness of 0.1 mm and a size of 100 × 100 mm of recycled commercial PE and recovered PE with and without prior extrusion were produced using a FONTIJNE GROTNES BV LPC 600 hydraulic press at 160 °C. The films were generated in two cycles within a total time of 5 min. Cycle one was operated at 160 °C, 60 KN for 3 min, followed by cycle two at 160 °C, 200 KN for 2 min.

Rheological analysis measurements were performed using a Rotational Rheometer TA Instruments AR-G2 under nitrogen atmosphere. Measurements were done on a 25 mm plate at an angle of 0° and 180 °C in share rate swept mode. 5–10 g of the sample were placed between the plates and heated up to the testing temperature for 5 min. Then, the gap was closed and the excess of the sample from the plates removed. To start the test, the upper plate is rotated to apply the required shear rate from 0,001 to up to 500 s^{-1}.

Fourier transformed infrared spectroscopy (FT-IR) was used to determine the nature of the produced films from recycled commercial PE and the PE reject fraction from LPB's. Therefore, polymer samples were measured between 4000 and 650 cm^{-1} on a Perkin Elmer Spectrum 100 FT-IR Spectrometer (PerkinElmer, Traiskirchen, Austria). All samples were acquired using 64 scans and a resolution of 2 cm^{-1}.

2.8. Integrated experiments

After experiments in 100 mL small scale, a larger scale experiment was performed in 1 L. Therefore, 50 g L^{-1} LPB reject sample was added to a 2 L wide neck bottle followed by 925 mL 0.5 M citric acid buffer at pH=4.8 and 75 mL (7.5% v/v) Cellic® CTec3. The bottle was afterwards incubated at 50 °C and 170 rpm for 4 days to ensure a sufficient cellulose hydrolysis. After 4 days, the hydrolysed LPB reject was filtered through a 100 μm sieve and washed by rinsing it with ddH$_2$O. Directly after washing and filtration, the sample was again added into the 2 L bottle and 1 L of 1.4 M biogenic acid was added. The aluminium bioleaching experiment was performed at 30 °C and 170 rpm for up to one week. After bioleaching, the LPB reject was washed three times with ddH$_2$O and dried at 60 °C until complete dryness. Prior to selective precipitation, the lixiviant was filtrated through a Durapore® PVDF filter membrane with a pore size of 0.45 μm. Afterwards the pH was dropwise adjusted to pH=6.5 with 10 M NaOH and the precipitated aluminium hydroxide was collected by filtration, followed by drying at 60 °C until complete dryness.

3. Results and discussion

3.1. Reject characterization

LPB reject was characterized using Scanning Electron Microscopy (SEM) coupled with Elemental Analysis (EDX) and Fourier Transformed Infrared Spectroscopy (FT-IR). Depending on the manufacturer and the use of the multilayer packaging, different polymers such as polyethylene (PE), polypropylene (PP), nylon and others are used. A total number of 26 polymer samples, differing in colour and material, were collected from the shredded LPB reject and used for characterization. FT-IR measurements (Fig. 1) revealed polyethylene to be the most prominent material in the sample, showing characteristic bands in the regions
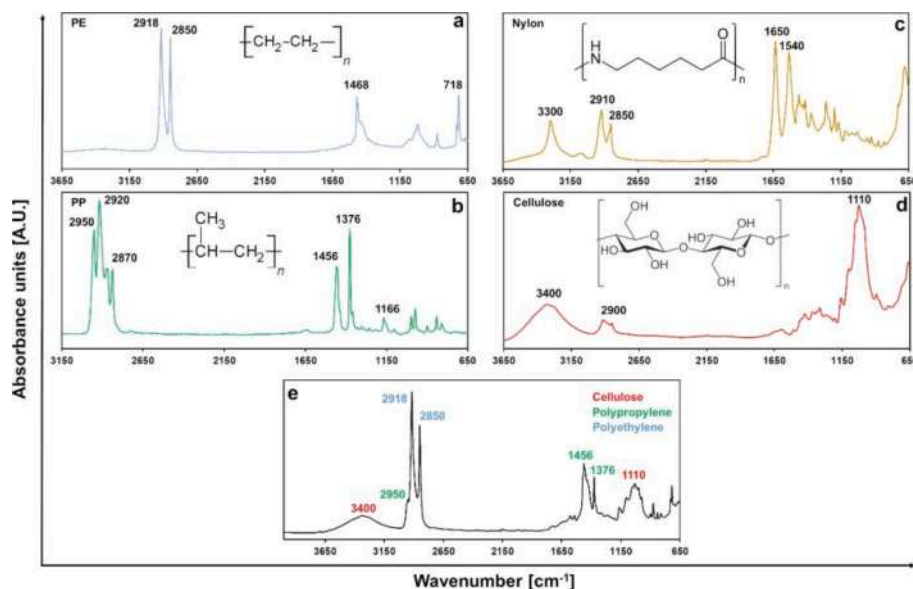
Fig. 1. FT-IR spectra of different components identified in beverage carton reject samples. a, polyethylene with the chemical structure and characteristic peaks labelled. **b,** polypropylene with the chemical structure and characteristic peaks labelled. **c,** with the chemical structure and characteristic peaks labelled. **d,** with the chemical structure and characteristic peaks labelled. **e,** multilayer material.

of 2918 cm^{-1} and 2850 cm^{-1} (CH$_2$ symmetrical stretching), 1468 cm^{-1} (bending deformation) and 718 cm^{-1} (rocking deformation) (Gulmine et al., 2002). In the PE spectrum, additional peaks at around 1425 cm^{-1} and 870 cm^{-1} were detected, which can be assigned to CaCO$_3$, a frequently used inorganic filler which modifies their mechanical and optical properties (Jimoh et al., 2018; Wihlborg and Rubinovitz, 2016). Furthermore, a peak at around 1110 cm^{-1} in the PE spectrum indicates remaining cellulose fibres on the polymer surface. Additionally, polymers such as polypropylene (CH$_3$ asymmetric stretching at 2950 cm^{-1}, CH$_3$ symmetrical stretching at 1456 cm^{-1} and CH$_3$ symmetrical bending at 1376 cm^{-1}) (Long, 2004) and nylon (NH stretching at 3400 cm^{-1}, Amide I and II stretching and bending at 2910 and 2850 cm^{-1}, C=O stretching at 1650 cm^{-1}, and NH bending at 1540 cm^{-1}) (Long, 2004) were detected. Furthermore, traces of cellulose fibers were found on the surface of some of the measured polymer reject samples, showing characteristic bands at 3400 cm^{-1} (OH-stretching), 2900 cm^{-1} (CH-stretching) and 1110 cm^{-1} (C-O-C-stretching) (Zhuang et al., 2020).

The complex mixture of different materials in the LPB reject was further visualised using SEM/EDX measurements. In Fig. 2a and b, a multilayer material consisting of plastic, aluminium and cellulose out of the LPB reject was analysed, revealing additionally, that cellulose removal by hydropulping was not completely sufficient and on the other hand it shows aluminium to be incorporated between different polymer layers.

3.2. Cellulose removal

Hydropulping is the state-of-the-art process to separate paperboard from LPB multilayers, reducing the cellulose content to less than 5% (Georgiopoulou et al., 2021; Zawadiak, 2017). Nevertheless, up to 11% of cellulosic fibre material was found in the present LPB reject sample (Fig. 2a–c), potentially reducing the purity of PE and aluminium during recycling. Especially in reprocessing of PE, degradation of cellulosic residuals causes a change of colour and reduces its mechanical properties at temperatures above 200 °C (Brebu and Vasile, 2010; Yang et al., 2007), which will negatively affect the quality of recycled polymers. By applying different concentrations of cellulases (i.e., 2.5, 5.0 and 7.5%), up to 15 mM of glucose could be released at the highest cellulase concentration after 24 h (Fig. 3e). With increasing enzyme concentration,

characteristic peaks for cellulose at 3400 cm^{-1} and 1110 cm^{-1} decreased in intensity, whereas peaks related to PE at 2918 cm^{-1} and 2850 cm^{-1}, 1468 cm^{-1} and 718 cm^{-1} significantly increased (Fig. 3a). The results indicated a more efficient cellulose removal at higher enzyme concentrations, comparable to the acid hydrolysis. SEM analysis after the cellulase treatment revealed that the fibrous material was almost completely removed, leaving only aluminium and PE in the LPB reject (Fig. 2d–f). Nevertheless, it was still possible to identify some isolated remaining fibres after the cellulase treatment. FT-IR analysis of these fibres indicated that the lignin content of the treated samples increased by showing more intense bands at 1600–1650 cm^{-1} (C=O stretching), 1513 cm^{-1} (C=C-C aromatic ring stretching), 1460 cm^{-1} (C-H deformation) and 1268 cm^{-1} (C-O stretching of C$_5$ substituted aromatic units) after cellulose removal (Rashid et al., 2016; Zhuang et al., 2020) (Fig. 3c and d). Hydrolysing cellulose and hemicellulose, results in removal of glucose and xylose, consequently increasing the intensity of entrapped lignin (Singh et al., 2016). Almost complete hydrolysis of cellulose is in agreement with previous studies on enzymatic recovery of valuable glucose from deinking rejects for production of bioethanol and other platform molecules (Annamalai et al., 2020; Haske-Cornelius et al., 2021).

3.3. Selective aluminium bioleaching and recovery

In order to selectively separate the aluminium from the LPB reject, different methods including acid and alkaline dissolution (Lee et al., 2006) or the introduction of reversible crosslinking agents (Cinelli et al., 2016; Kaiser, 2019) have been investigated and patented in recent years. The application of bioleaching presents a completely new approach for the selective dissolution of the aluminium layer from the LPB reject material as it has already been successfully used in the recovery of metals from other waste streams (Brar et al., 2021; Gomes et al., 2018; Kremser et al., 2021b; Pathak et al., 2021; Yesil et al., 2021). To evaluate the potential of different bioleaching applications, two methods including indirect (non-contact) and direct bioleaching of the LPB reject were tested. For the indirect, non-contact bioleaching, a mixed culture of the two sulfur-oxidizing bacteria *A. thiooxidans* and *A. caldus* was cultivated in a minimal medium containing elemental sulfur and the resulting cell-free supernatant (1.4 M biogenic acid) was incubated with different concentrations of LPB reject for up to 9 days. Independently
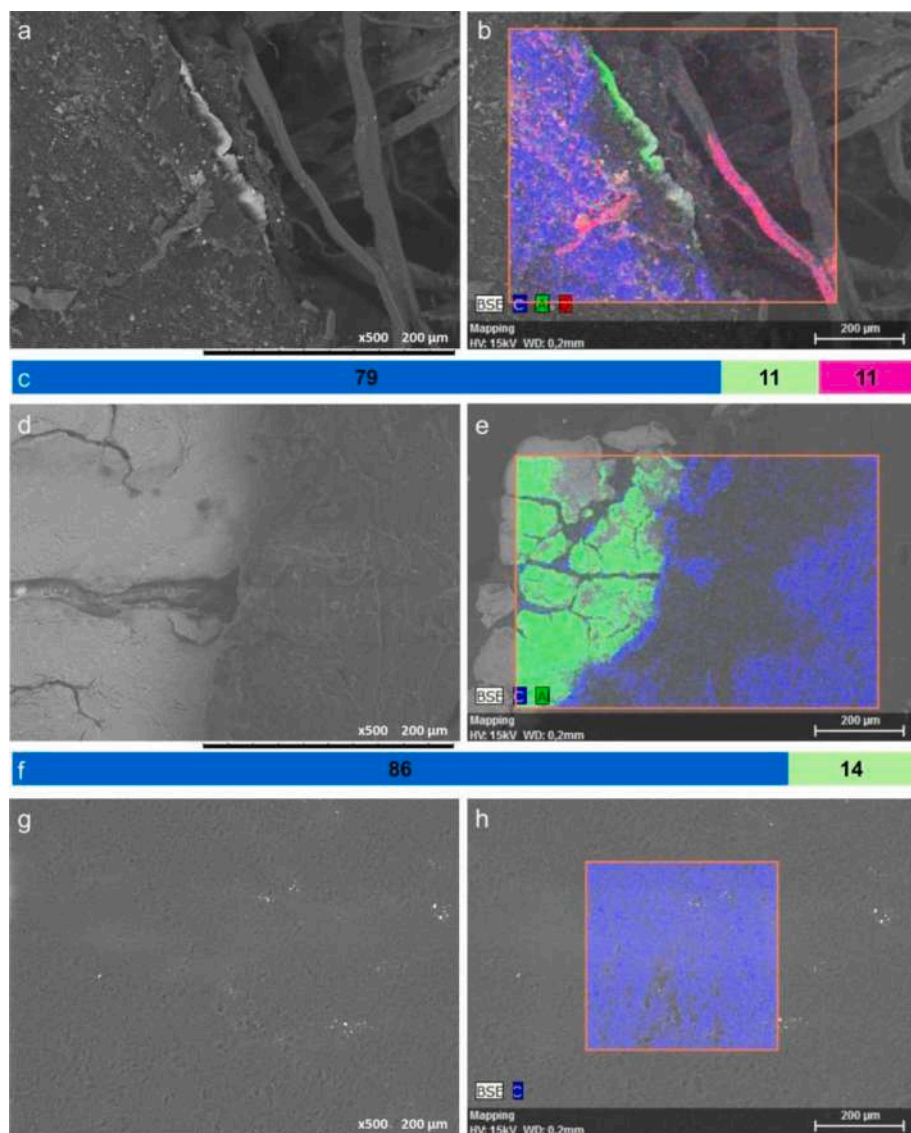
Fig. 2. SEM and EDX analysis of the original and treated beverage carton reject samples. a, SEM image of the original LPB reject material showing the different components: plastic, aluminium and cellulose fibres. **b**, elemental analysis revealing the different materials: plastic (blue), cellulose fibres (pink) and aluminium (green). **c**, bar chart showing the composition of the original material before further treatment (79% plastic, 11% cellulose and 11% aluminium.). **d** and **e**, SEM and EDX analysis of the LPB reject after cellulase treatment with no cellulose present. **f**, composition of the LPB reject after cellulose hydrolysis (86% plastic and 14% aluminium). **g** and **h**, Pure plastic after bioleaching removal of the aluminium layer.

from the concentration, around 80% of the aluminium could be removed by the biogenic acid within the first 3 days, ending up with complete dissolution after 7 days of incubation (Fig. 4a and d). The control experiment using the same molarity of commercial sulfuric acid resulted in comparable leaching efficiencies, indicating that the biogenic produced acid was as effective as the commercially available one.

For the direct bioleaching approach, the iron- and sulfur-oxidizing bacteria *Acidithiobacillus ferrooxidans* was tested with different ferrous iron concentrations (i.e., 1.0, 2.5 and 5.0% (w/v)) on similar LPB reject concentrations (i.e., 1.0 and 2.5% (w/v)) as in the indirect, non-contact experiments. Ferric iron (Fe^{3+}) was previously reported to be a major contributor as a strong oxidant in the bioleaching of metals from solid materials (Hubau et al., 2018; Jafari et al., 2019; Kaksonen et al., 2020; Kremser et al., 2021b) which can be continuously regenerated by *A. ferrooxidans*. Nevertheless, in the bioleaching of aluminium from multilayer LPB reject material, the concentration of ferrous iron showed only minor effects. Compared to the indirect bioleaching, lower leaching efficiencies were achieved within the first week reaching only 75% after 7 days, independent from the ferrous iron concentration (Fig. 4b). Even after 14 days of incubation, only a maximum of around 88% aluminium leaching could be reached. More severe impacts on the leaching efficiency could be observed regarding the LPB reject concentration. A higher concentration resulted in a decrease in the leaching efficiency

(maximum 20% after 14 days with 25 g L^{-1}) which was not observed in the indirect mechanism. Therefore, a concentration of 5.0% was not applied in the direct bioleaching. A positive impact of the bacteria in direct bioleaching could be observed by comparing the results to the chemical blank experiments. After 7 and 14 days, the bioleaching efficiency was around 20 and 10% higher, respectively. Unfortunately, the increasing ferric iron concentrations resulted in orange to red colouring of the biologically treated LPB reject samples, which is assumed to have negative impacts on the sample purity at the end of the process (Fig. 4f). FT-IR investigations of the coloured samples revealed oxidative changes in the polymer structures (Fig. 4g). With increasing ferrous iron concentrations, bands at 1045 cm^{-1} (CH$_2$-O stretching vibration), 1100 cm^{-1} (C-O-C stretching vibration), 1170 cm^{-1} (C-O stretching vibration) (Hamzah et al., 2018) and around 3400 cm^{-1} (hydroxyl group) (Qin et al., 2003) characteristic for the oxidative degradation of PE show increasing intensities. The oxidative degradation may result from a persulfate-based advanced oxidation. Persulfate is one of the intermediate sulfur species in *Acidithiobacillus* species during sulfur metabolization (Rohwerder and Sand, 2003) which can be activated by ferric iron at low pH-values, resulting for example in the degradation of different organic materials (Kaur et al., 2019; Wang and Wang, 2017), comparable to a Fenton-like oxidation.
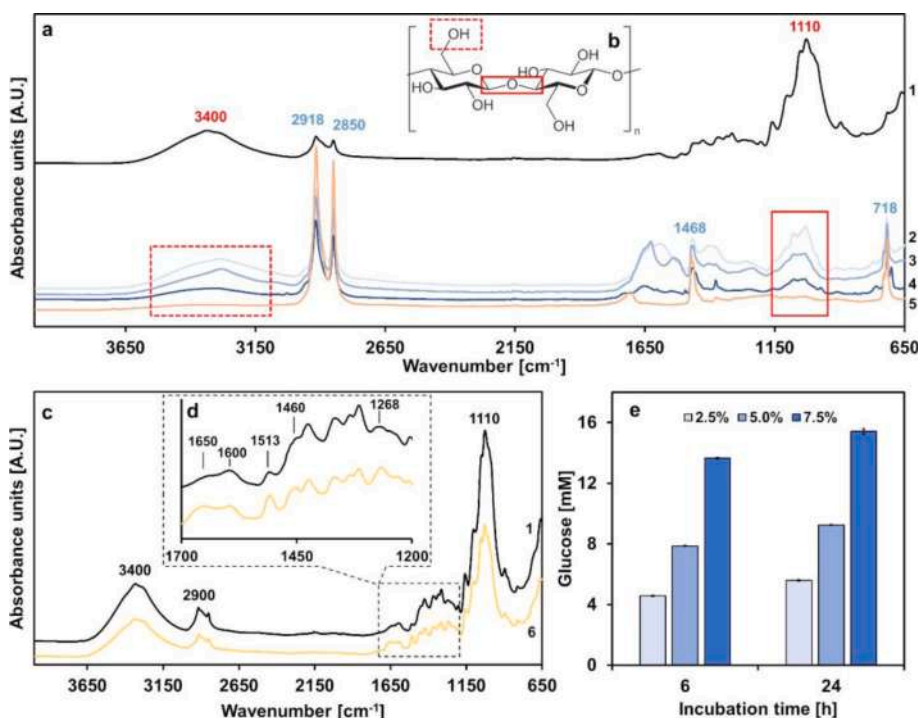
Fig. 3. Characterization of the LPB reject material before and after enzymatic cellulose hydrolysis. a, FT-IR spectra of cellulosic material in the LPB reject before (1) and after treatment with increased cellulase concentration (2 = 2.5%, 3 = 5.0% and 4 = 7.5% (v(/v)) or acid hydrolysis (5). b, Structure of cellulose with the characteristic regions in red squares, in which a decrease in intensity was observed during the enzymatic and chemical hydrolysis. c and d, FT-IR spectra of the remaining fibres after cellulase treatment (6) compared to the original cellulosic material (1). e, Glucose concentration released during the hydrolysis of the reject material with increasing enzyme concentrations of 2.5–7.5% (v/v) after 6 and 24 h.

3.4. Integrated treatment

After successful application in small scale, an experiment with 50 g L^{-1} LPB reject was performed in a 2 L wide neck bottle (Fig. 5). Cellulose hydrolysis resulted in around 18.3 mM of released glucose which was higher compared to the individual experiments above. This was also reflected by a reduced amount of remaining cellulosic fibres after washing. Directly after washing in the bottle, 1 L of biogenic acid was added, resulting in complete aluminium dissolution after 7–8 days which was in line with leaching efficiencies achieved in the batch experiments. Finally, the aluminium in the leaching lixiviant was selectively precipitated with 10 M NaOH to form aluminium hydroxide (Al (OH)$_3$) and dried in the oven. The precipitation efficiency, calculated by the amount of dissolved aluminium before and after the precipitation, resulted in almost 100%, as no aluminium could be detected in the solution after precipitation.

3.5. PE purification, recovery, and characterization

The original LPB reject material reflects a complex mixture of different materials including plastics (PE as major component, PP and PA (polyamide) in very small amounts, <5% w/w), aluminium and cellulose. Depending on how effective the hydropulping process is in removing the paperboard as well as the use of different plastics on the inner and outer layer of a multilayer packaging, the purity of the recovered materials is affected. In Fig. 1e the complex mixture in the original LPB reject can be observed. Within the spectra, characteristic bands for three different materials, comprising cellulose, polyethylene and polypropylene can be detected. For PE purification and recovery, LPB reject after cellulase treatment and aluminium bioleaching from the integrated treatment was used (Fig. 4c). After density separation, two fractions could be separated consisting of PE (Fig. 5) and another fraction of mixed plastic impurities. The PE fraction was afterwards used for characterization and compared to commercially available PE. In rheology tests, the viscosity of the recycled PE fraction was measured and compared to commercially available PE. Processing steps such as extrusion and film production were evaluated regarding their influence on homogeneity of the recycled PE. In Fig. 6a, a clear effect of the

extrusion in general and in the applied residence time for extrusion was presented. Viscosity of the recovered PE without extrusion was the lowest, and a high inhomogeneity of the material was reflected by a high standard deviation. Extrusion for 2 or 5 min (Fig. 6c and d) prior to rheology tests, significantly increased the viscosity of the material by increasing the homogeneity of the PE fraction. Film pressing of extruded commercial PE and recovered PE from LPB reject before rheology measurements resulted in highly homogeneous materials, comparable to each other (Fig. 6b). The results indicate that the recovered PE from multilayer packaging and commercial PE have similar properties in terms of processability and can be considered suitable for traditional plastic processing.

FT-IR analysis of the films produced from the commercial recycled PE and the recycled PE from LPB reject (Fig. 6e and f) revealed similar chemical properties with only minor impurities still present in the PE recovered from the multilayer materials processing. Peaks characteristic for PE could be assigned for both produced films at 2918 cm^{-1} and 2850 cm^{-1} (CH$_2$ symmetrical stretching), 1468 cm^{-1} (bending deformation) and 718 cm^{-1} (rocking deformation) (Fig. 6g). Additionally, impurities in the form of orange particles could be observed in the PE film from LPB reject (Fig. 6f and g). FT-IR measurements indicate the presence of what most probably are polyurethane-based impurities, with peaks observed in the 1635 - 1725 cm^{-1} (C = O carbonyl stretching) and 1095 cm^{-1} (C-O-C stretching vibration) (Lee and Lin, 2006) range. Nevertheless, similar spectra of commercial PE film and film made of the PE from LPB reject underline the findings from rheology tests, confirming that the recovered material is suitable for traditional plastic processing.

4. Conclusion

Two-step enzymatic-bioleaching recycling was shown to be an effective and environmentally friendly method for recycling of complex, metal-containing multilayer waste materials due to the specificity of enzymes and of bioleaching. It can be seen as one of several new and innovative technologies, helping to increase the recycling rates for valuable raw materials. Applying enzymes highly specific for cellulose hydrolysis can help to increase the purity of the final polymer products by removing fibrous remains.. The mild treatment technique of using
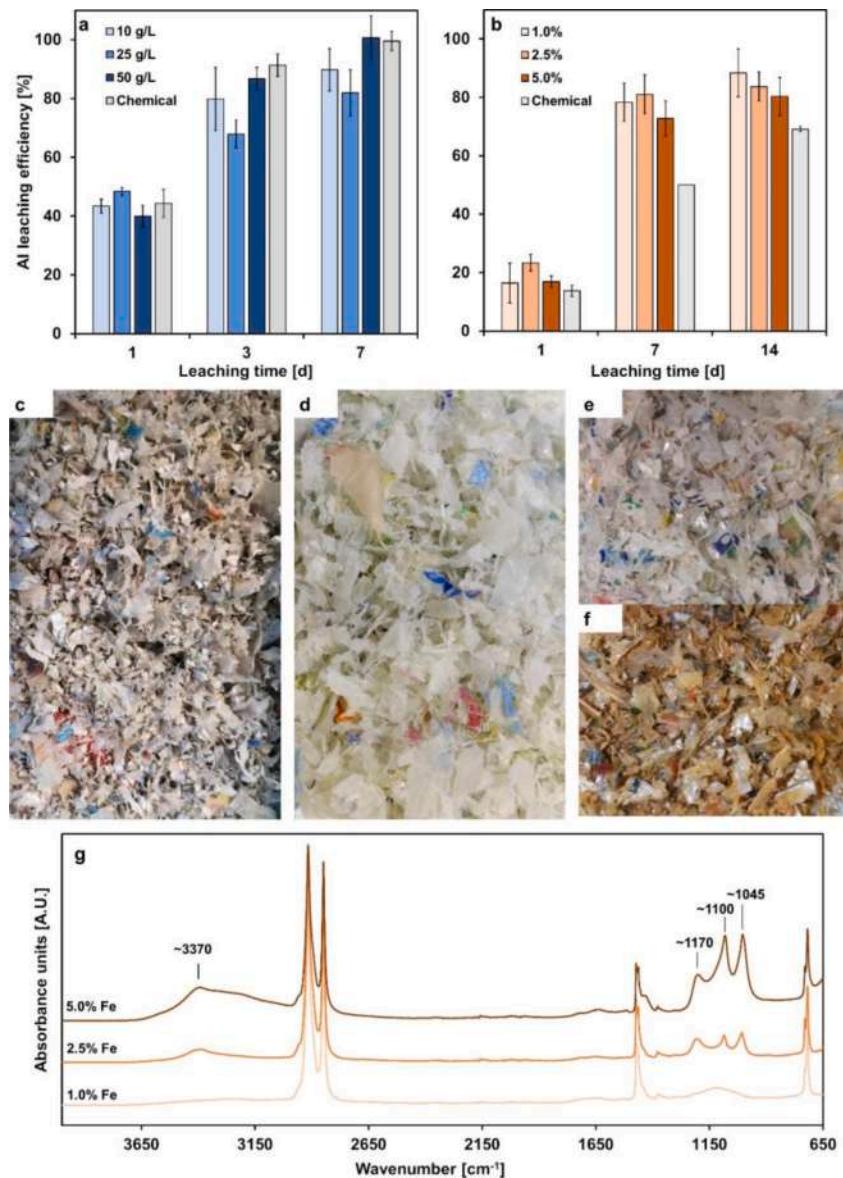
Fig. 4. Indirect and direct bioleaching of LPB reject material. a, aluminium leaching efficiencies of the indirect, non-contact bioleaching after 1, 3 and 7 days with increasing LPB reject concentrations from 10–50 g L^{-1}. **b,** aluminium leaching efficiencies of the direct bioleaching after 1, 7 and 14 days with 10 g L^{-1} LPB reject and 1.0, 2.5 and 5.0% (w/v) ferrous iron. **c,** reject material after cellulose hydrolysis consisting of different plastics and aluminium. **d,** LPB reject after indirect bioleaching showing complete removal of aluminium. **e,** LPB reject after bioleaching of 10 g L^{-1} with the lowest ferrous iron concentration of 1.0% (w/v). **f,** LPB reject after bioleaching of 25 g L^{-1} with the highest ferrous iron concentration of 5.0% (w/v) showing the remaining aluminium and colouring of the material. **g,** FT-IR spectra of PE after direct bioleaching with increasing ferrous iron concentrations. Characteristic peaks indicating oxidation of the polymer are labelled.
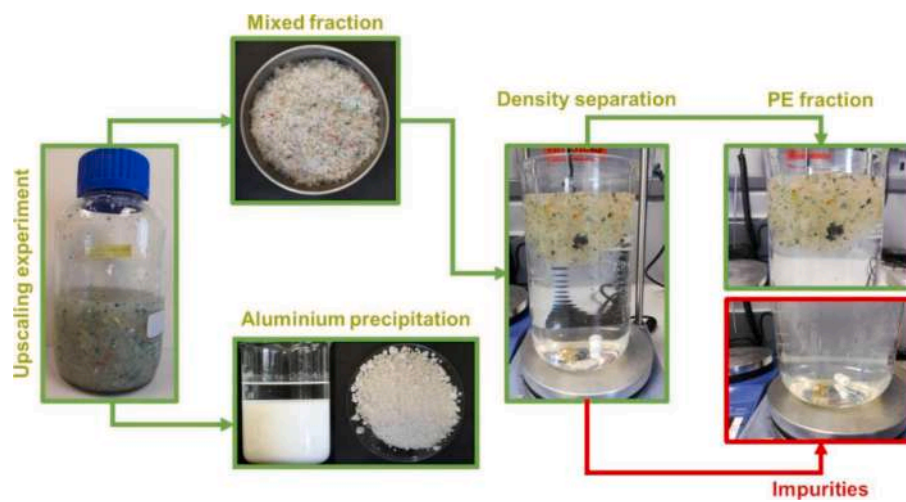

Fig. 5. Upscale experiment of bioleaching-based reject recycling. After cellulose removal and aluminium bioleaching, a mixed plastic fraction and aluminium hydroxide were obtained. Polyethylene from the mixed fraction was purified using density separation. At the end a pure PE fraction was obtained and a minimum number of impurities (<2% w/w) could be separated.
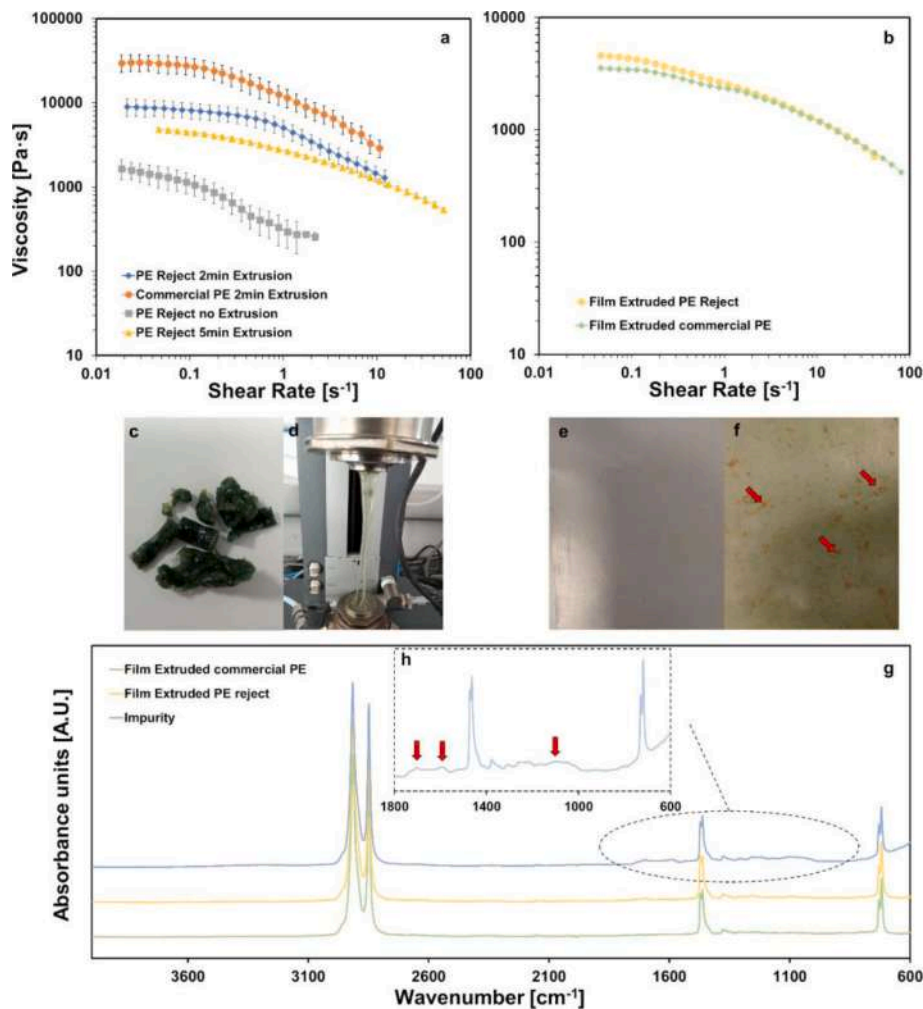
Fig. 6. Characterization of the purified PE from LPB reject. a, rheology tests of commercial PE and recovered PE with and without prior extrusion and changing extrusion time. **b**, rheology tests of produced PE films after extrusion. **c**, extruded PE from LPB reject. **d**, extruded PE from LPB reject after rheology tests. **e**, film produced out of commercial extruded PE. **f**, film produced out of extruded PE from LPB reject with red arrows indicating the impurities in the material. **g**, FT-IT spectra of the films produced from commercial extruded PE, extruded PE from LPB reject and the visible impurities. **h,** detailed FT-IR spectra of the impurities in the PE film from LPB reject with arrows indicating peak assignments for polyurethane.

biologically produced sulfuric acid which can be generated out of waste elemental sulfur, led to complete aluminium dissolution in a period of only one week, by causing no harm to the polymeric structures. At the end, pure polyethylene and aluminium hydroxide are the final products, which can again be integrated in a production cycle, minimizing the need of incineration. The achieved results therefore contribute to the ambitious goal of the European Union, stating that all packaging waste should be reusable or recyclable by the year 2030 (European Commission, 2018b).

Author's contributions

K.K. and P.G. carried out the bioleaching/enzymatic depolymerization experiments and analysed the data. K.K. and A.P. wrote the manuscript. G.M.G. and A.P. corrected the manuscript. G.M.G. and A.P. acquired the funding and designed and supervised the work. A.B.B., D.E. R. and B.M.M. carried out the mechanical testing of the recovered PE material.The authors declare no competing interests

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.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.resconrec.2022.106444.

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