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# Valorisation of waste pulp from materials recovery facility rejects for composite applications

Natalia Herrera [a], Diego Freire Ordóñez [a], Andre N. Gaduan [a], Kanjanawadee Singkronart [a], Daniel Hayes [b], Dhivya Puri [c], Koon-Yang Lee [a,d,*]

[a] Department of Aeronautics, Imperial College London, South Kensington Campus, London SW7 2AZ, United Kingdom
[b] Celignis Biomass Laboratory, Plassey Technology Park, Limerick V94 7Y42, Ireland
[c] Fiberight Ltd., Westfield Industrial Estate, Waunarlwydd, Swansea, SA5 4SF Wales, United Kingdom
[d] Institute for Molecular Science and Engineering (IMSE), Imperial College London, London SW7 2AZ, United Kingdom

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ABSTRACT

Waste pulp was recovered from MRF rejects, purified, processed into paper sheets and laminated with polylactide (PLLA) films. The purification process employed produced waste pulp of different qualities. The mechanical properties of the resulting waste pulp fibre-reinforced laminated PLLA composites was indifferent to the type of waste pulp treatment used. All composites showed significant improvements over neat PLLA, highlighting the viability of using waste pulp as a potentially cheap and sustainable reinforcement for polymers. Lifecycle assessment further showed that the composites possessed lower net global warming potential, as well as the end-point impact categories of human health and ecosystem quality compared to neat PLLA. This is due to the higher mechanical performance of the composites, which leads to higher weight saving of the functional unit. Our work paves the way for the use of pulp rejects from the recycling process for higher value applications, diverting them from landfill or incineration.

## 1. Introduction

The UK 25 Year Environmental Plan calls for the transformation of our society to be resource-efficient and have low fossil-carbon intensity, with a particular focus on turning waste into a primary feedstock resource [1]. To achieve this, the UK government put in place the ambitious Resource and Waste Strategy [2] that included the introduction of the £200 per tonne plastic packaging tax on manufacturers using $< 30$ % recycled plastic in packaging, as well as the mandate to achieve a household waste recycling rate of 50 % by 2020 and 65 % by 2030. These are ambitions targets as the “take-make-dispose” linear resource consumption model is still at the heart of our economy and society [3]. Manufacturers convert raw materials into products that can be sold to consumers, who then discard any waste generated. Data collected by the UK Department for Environment, Food and Rural Affairs showed that the UK households generate approximately 27 million tonnes of waste every year [4]. Despite various governmental campaigns and many new local authority dry recyclables collection schemes, the UK recycling rate of household waste has stagnated at 45 % since 2010.

Even though more resource recovery infrastructure can be built to increase recycling rates [5], there is some evidence suggesting that infrastructure expansion alone may not be sufficient [6–8]. This is due to the fact that the household is at the start of the waste re-processing chain, where waste materials are first segregated into dry recyclables (*i.e.*, clean materials with a recycling label) and non-recyclables (e.g., food or contaminated materials). Inevitably, this activity has flaws such as a high potential for contamination (incorrect materials) to enter the dry recyclables stream [9–11] and the heavy reliance on participation from the public, which could be low [12–14]. In any case, dry recyclables are then sent to a materials recovery facility (MRF), whereby the commingled collection of materials is separated into homogenous fractions of rigid plastics, mixed paper and cardboards, metals and glass. Each material fraction is baled and sent for onward recycling. However, up to 30 % of the dry recyclables entering a MRF are rejected due to contamination [15] and this accounts for $\sim$ 1 million tonnes of waste that are sent to landfill or incinerated in England every year [16].

It can be anticipated that the valorisation of MRF rejects could aid the UK to achieve its recycling targets. A typical MRF reject stream in the

* Corresponding author at: Department of Aeronautics, Imperial College London, South Kensington Campus, London SW7 2AZ, United Kingdom.
*E-mail address:* koonyang.lee@imperial.ac.uk (K.-Y. Lee).

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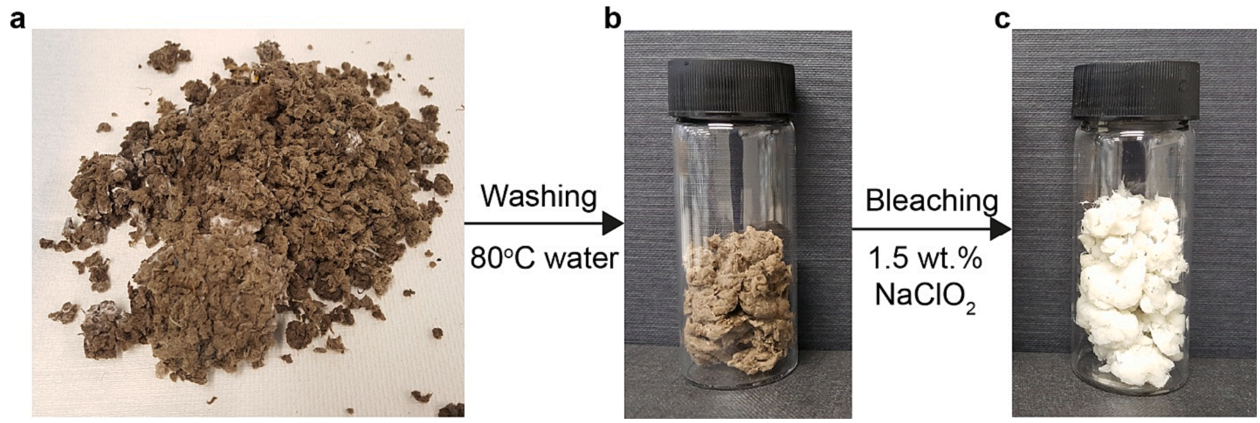


**Fig. 1.** Visual appearance of (a) as-received waste pulp, (b) waste pulp after hot water washing and (c) waste pulp after bleaching with $NaClO_2$.

UK consists of approximately 20 % mixed paper and cardboards, 50 % flexible plastics, 8 % metals, 14 % glass and the remainders are unidentifiable (personal communication). As the density between the different constituents is sufficiently large, a float-sink separation system similar to that used in polymer recovery facility (PRFs) can be employed to separate the different components [17]. Flexible plastics (mainly low-density polyethylene), metals and glass can be baled and sent for subsequent recycling. Due to the use of a water-based separation process, the mixed paper and cardboards fraction of the MRF rejects form a waste pulp that has a low or no market value in the recycled paper industry.

Here, we report the valorisation of low value waste pulp recovered from a water-based secondary sorting of MRF rejects as cheap reinforcing fibre feedstock for the production of high value sustainable laminated polylactide composites. This will create a stronger demand for waste pulp, diverting it away from landfill or incineration. As one of the main challenges in the utilisation of any waste material is contaminations, a process to purify the waste pulp is described in this work. The purified waste pulp was then formed into reinforcing paper sheets and laminated with polylactide to produce the waste pulp-reinforced polylactide composites. The chemical composition and thermal behaviour of the waste pulp, the (thermo-)mechanical properties of the waste pulp-derived paper sheets and their laminated polylactide composites were characterised and reported in this work.

## 2. Experimental

### 2.1. Materials

Waste pulp (solid content ~ 30 wt-%, see Fig. 1a) was kindly provided by Fiberight Ltd. (Waunarlwydd, Swansea, UK). It was recovered from MRF rejects using the proprietary HYDRACYCLE™ process [17]. Sodium chlorite ($NaClO_2$) (Merck, purity ≥ 80 %) was purchased from Sigma-Aldrich (Gillingham, Dorset, UK). Glacial acetic acid (AnalaR® NORMAPUR®, purity ≥ 99.8 %) was purchased from VWR International (Lutterworth, UK). Both chemicals were used as received to purify the waste pulp. Poly(L-lactic acid) (PLLA) (L9000, MW ≥ 150 kDa, D-content ~ 1.5 %) was purchased from Biomer GmbH (Schwalbach am Taunus, DE) and used as the polymer matrix for the laminated composites.

### 2.2. Purification of waste pulp recovered from MRF rejects

The as-received waste pulp had a distinct odour and contained impurities such as sand particles and small plastic fragments. Therefore, a method developed by Hietala et al. [18] was adopted. The waste pulp was first cleaned by dispersing in 80 °C water under magnetic stirring for 30 min at a consistency of 2 wt-%, followed by multiple cold water rinsing steps. During these steps, any observed impurities in the waste pulp were manually removed. The odour of the waste pulp was significantly reduced after this cleaning step but not completely eliminated. In addition to this, it still possessed the same brown colour of the as-received waste pulp (Fig. 1b). Thus, a sodium chlorite based bleaching step adopted from the work of Mtibe et al. [19] was conducted. Here, the waste pulp was dispersed in 1.5 wt-% $NaClO_2$ solution acidified with acetic acid to a pH of 4.2 at 80 °C for 1 h, followed by repeated washing with distilled water until a neutral pH was attained. This process removed the odour completely and increased the whiteness of the material (Fig. 1c). All pulp materials were stored in a 4 °C fridge prior to subsequent use.

### 2.3. Preparation of waste pulp paper sheets

In this work, paper sheets were made from hot water washed waste pulp, bleached waste pulp, as well as refined waste pulp. The refinement of waste pulp was conducted by passing bleached waste pulp through a 1.5 kW re-circulating colloid mill (JM-60, Shanghai Tiangang Machine Manufacture Co. Ltd., Shanghai, China) consisting of a toothed inner rotor and a toothed stator at a consistency of 0.7 wt-%. The minimum gap between the rotor and the stator was set to be 7 μm. A refining time of 7.5 min was chosen as preliminary study showed that the mechanical properties of the waste pulp paper sheets plateaued beyond this refining time. To produce the paper sheet, a waste pulp suspension with a consistency of 0.1 wt-% was first prepared and vacuum filtered onto a 125 mm diameter filter paper (Qualitative filter paper 413, VWR International Ltd., Lutterworth, UK) in a Büchner funnel. The filter cake was carefully removed from the used filter paper and press-dried into a flat, uniform sheet of waste pulp paper at 120 °C for 30 min under a weight of 1 t (Model 4122CE, Carver Inc., Wabach, IN, USA) between two sheets of fresh filter paper placed between two sheets of blotting paper (Grade 3MMCHR, GE Healthcare, Buckinghamshire, UK). The grammage of the waste pulp paper sheets was 120 ± 6 g $m^{-2}$. All waste pulp paper sheets were stored in a sealed environment containing silica gel pouches to avoid moisture absorption prior to subsequent use.

### 2.4. Production of waste pulp fibre-reinforced laminated PLLA composites

Prior to producing the laminated composites, thin PLLA film with a thickness of 180 ± 20 μm was prepared by compression moulding PLLA pellets at 180 °C for 2 min under a weight of 2 t (Model 4122CE, Carver Inc., Wabach, IN, USA). The prefabricated PLLA film and the waste pulp paper sheets were then cut into strips with dimensions of 15 mm × 70 mm using a Zwick/Roell manual cutting press (ZCP 020, Zwick Testing Machines Ltd., UK). A lay-up consisting of seven strips of PLLA films and six strips of waste pulp paper, positioned in an alternating sequence, was placed in a 15 mm × 70 mm metallic mould. The mould was then preheated to 185 °C for 4 min, followed by compression moulding at the same temperature under a weight of 0.5 t for 1 min (Model 4122CE, Carver Inc., Wabach, IN, USA). The resulting waste pulp fibre-reinforced laminated PLLA composite possessed a thickness of 1.5 mm and a waste pulp fibre loading ($w_F$) of 35 wt-%. PLLA composites reinforced with hot water washed waste pulp fibres, bleached waste pulp fibres and refined waste pulp fibres are herein termed PLLA/WF, PLLA/BF and PLLA/RF, respectively. As a benchmark for comparison, neat PLLA was produced by injection moulding (Haake Minijet II, Thermo Fisher Scientific, Hampshire, UK). The injection moulded rectangular PLLA specimens possessed dimensions of 80 mm × 12 mm × 3.2 mm and the injection moulded dog bone shaped PLLA specimens possessed an overall length of 60 mm, a gauge length of 10 mm, thickness of 3 mm and the narrowest part of the specimens was also 3 mm. The barrel and the mould temperatures of the injection moulder were set at 190 °C and 30 °C, respectively. The (post-)injection pressure and time were set to be 600 bar and 30 s, respectively.

### 2.5. Materials characterisation

#### 2.5.1. Chemical composition of waste pulp fibres

Prior to compositional analysis of the waste pulp fibres, an extraction process was carried out according to the National Renewable Energy Laboratory (NREL) standard operating procedure [20]. Hydrolysis of the dry extractives-free samples was then performed to determine the structural carbohydrates and lignin in the biomass. The procedure was divided in two main steps: a two-stage acid hydrolysis of the samples and the gravimetric filtration of the hydrolysate in order to separate it from acid-insoluble residue (AIR) [21]. Klason lignin was calculated by determining the weight difference between the AIR and its ash content. Acid soluble lignin was measured by determining the absorbance of an aliquot of the hydrolysate at 205 nm using an Agilent 8452 UV–vis spectrophotometer and an absorptivity constant of 110 $M^{-1}$ $cm^{-1}$ (TAPPI UM250-1991). The lignocellulosic sugars resulting from hydrolysis were determined by ion-chromatographic techniques as described by Hayes [22].

#### 2.5.2. Thermal stability of waste pulp fibres

The thermal degradation behaviour of the waste pulp after washing and bleaching was investigated using thermal gravimetric analysis (Discovery TGA, TA Instruments, Elstree, UK). Approximate 4 mg of sample was heated from room temperature to 600 °C at a rate of 10 °C $min^{-1}$ in $N_2$ atmosphere.

#### 2.5.3. Morphology of waste pulp fibres

Scanning electron microscopy (SEM) was used to investigate the morphology of the waste pulp fibres. It was performed using a large chamber electron microscope (S-3700 N, Hitachi, Tokyo, Japan). The accelerating voltage used was 15 kV. Prior to SEM, the waste pulp fibres were deposited onto aluminium stubs and Au coated (40 mA, 25 s) using an auto sputter coater (Agar Scientific, Stansted, UK).

#### 2.5.4. Mechanical properties of waste pulp paper sheets

Tensile properties of the waste pulp paper sheets were quantified using miniaturised rectangular test specimens with dimensions of 5 mm × 40 mm, cut using a Zwick/Roell ZCP 020 manual cutting press (Zwick Testing Machines Ltd., UK). Prior to tensile testing, two dots were marked on the surface of the test specimen in the direction of applied load in order to track the strain (iMetrum Ltd., Bristol, UK) experienced by the test specimen. The test specimen was then mounted onto a micro-tensile tester (MT-200, Deben UK Ltd., Woolpit, UK) equipped with a 200 N load cell. After mounting, the exposed length of the rectangular specimen was 25 mm. Tensile test was conducted using a crosshead displacement speed of 0.5 mm $min^{-1}$, which corresponded to a strain rate of 0.04 % $s^{-1}$. Average results of five test specimens are reported for each type of waste pulp paper sheet.

Fracture toughness of the waste pulp paper sheets was determined from single-edge notched tension (SENT) test specimens of 15 mm in width ($w$) and 35 mm in length. Before the test, an edge crack with length $a = \sim$3–4.5 mm was introduced using a sharp scalpel. The SENT test specimen was then mounted onto a micro tensile tester (MT- 200, Deben UK Ltd., Woolpit, UK) equipped with a 200 N load cell. The distance between the grips was set to be 25 mm. A crosshead displacement speed of 0.5 mm $min^{-1}$ was used in this test. To accurately track the displacement of the grips, a non-contact extensometer (iMetrum Ltd., Bristol, UK) was used. A total of five specimens were tested for each type of waste pulp paper sheet. The initial critical stress intensity factor ($K_{IC}$) of the paper sheets was calculated from:

$$K_{IC}, \mathrm{MPa\ m}^{0.5} = \sigma_{max} \times \sqrt{a} \times \left(1.99 - 0.41\left(\frac{a}{w}\right) + 18.7\left(\frac{a}{w}\right)^2 - 38.48\left(\frac{a}{w}\right)^3 + 53.85\left(\frac{a}{w}\right)^4\right) \quad (1)$$

whereby $\sigma_{max}$ is the stress at which the crack propagated.

#### 2.5.5. Porosity of waste pulp paper sheets and their laminated PLLA composites

The porosity ($P$) of the paper sheets and the laminated composites was calculated using the following equation:

$$P, \% = \left(1 - \frac{\rho_E}{\rho_T}\right) \times 100 \quad (2)$$

whereby He pycnometry (Accupyc II 1340, Micrometrics Ltd., Dunstable, UK) was used to measure the true density ($\rho_T$) of the samples. The envelope density ($\rho_E$) of the samples was determined by taking the ratio between the mass and envelope volume.

#### 2.5.6. Mechanical properties of neat PLLA and waste pulp fibre-reinforced laminated PLLA composites

Tensile and flexural (three-point bending) properties of neat PLLA and its laminated composites were determined in accordance with ASTM D638 and ASTM D790, respectively. These tests were conducted using an Instron universal tester (Model 5969, Instron, High Wycombe, UK) equipped with a 50 kN load cell. Average results of five test specimens are reported for each type of sample. Prior to tensile testing, poly (methyl methacrylate) end tabs with dimensions of 15 mm × 15 mm × 3 mm were glued onto the test specimens using cold curing epoxy resin (Araldite 2011, Huntsman Advanced Materials, Cambridge, UK). A non-contact optical extensometer (iMetrum Ltd., Bristol, UK) was used to monitor the strain experienced by the test specimen loaded under uniaxial tension based on the two points marked within the gauge section. The crosshead displacement speed used in tensile testing was 1 mm $min^{-1}$ (strain rate = 0.08 % $s^{-1}$). Flexural test was conducted using a crosshead displacement speed of 2 mm $min^{-1}$. The support span length used for the waste pulp fibre-reinforced laminated PLLA composites was 40 mm whilst neat PLLA was tested using a support span length of 55 mm due to its larger thickness.

#### 2.5.7. Differential scanning calorimetry (DSC) of neat PLLA and waste pulp fibre-reinforced laminated PLLA composites

The crystallisation and melt behaviour of neat PLLA and its laminated composites was quantified using DSC (Discovery DSC, TA Instruments, Elstree, UK). A sample mass of approximately 10 mg was used. The sample was heated from room temperature to 200 °C using a rate of 10 °C $min^{-1}$ in $N_2$ and the degree of crystallinity of the injection moulded sample ($\chi_{c,moulded}$) was calculated using the following:

$$\chi_{c,moulded}, \% = \frac{\Delta H_m - \Delta H_c}{(1 - w_f) \times \Delta H_m^o} \times 100 \quad (3)$$

where $\Delta H_m$ and $\Delta H_c$ denote the melting and crystallisation enthalpy, respectively. The symbol $\Delta H_m^o$ is the melting enthalpy of 100 %

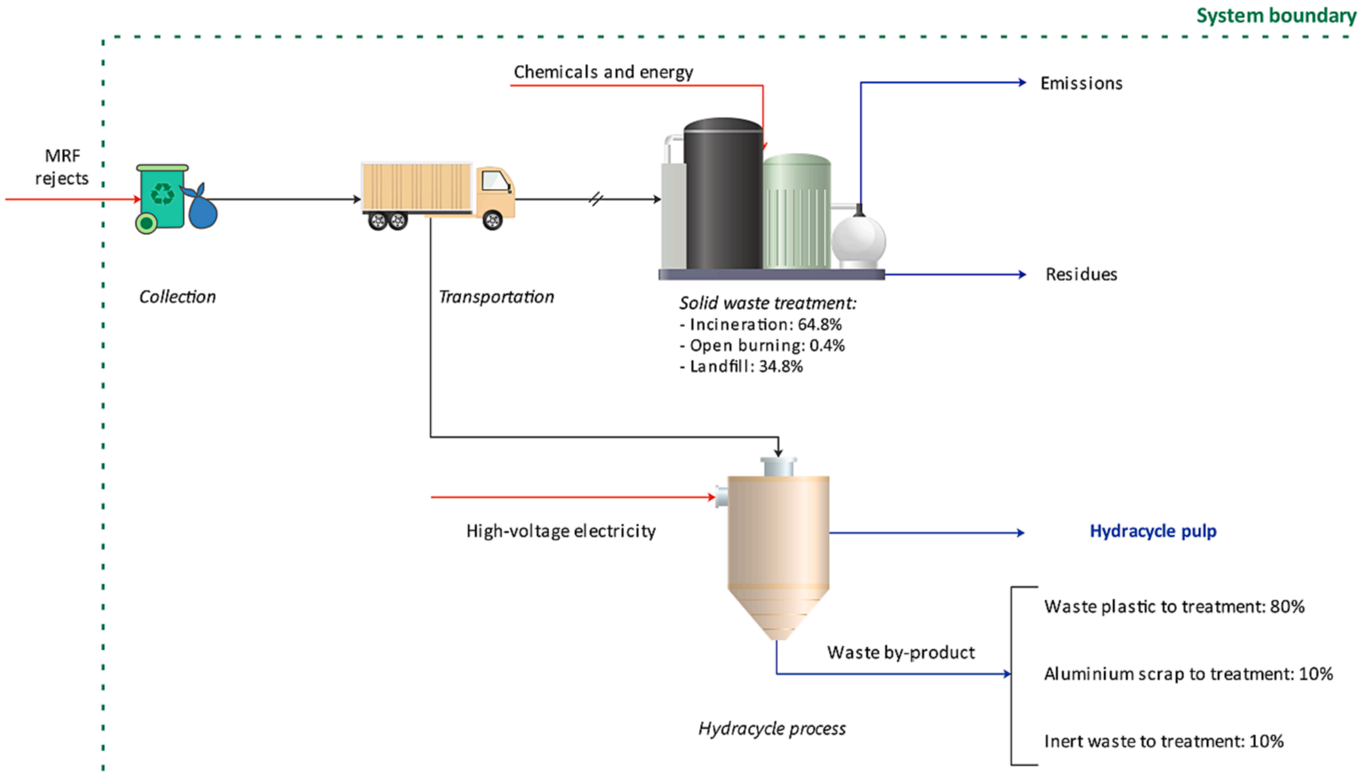


**Fig. 2.** System boundary of the HYDRACYCLE™ process.

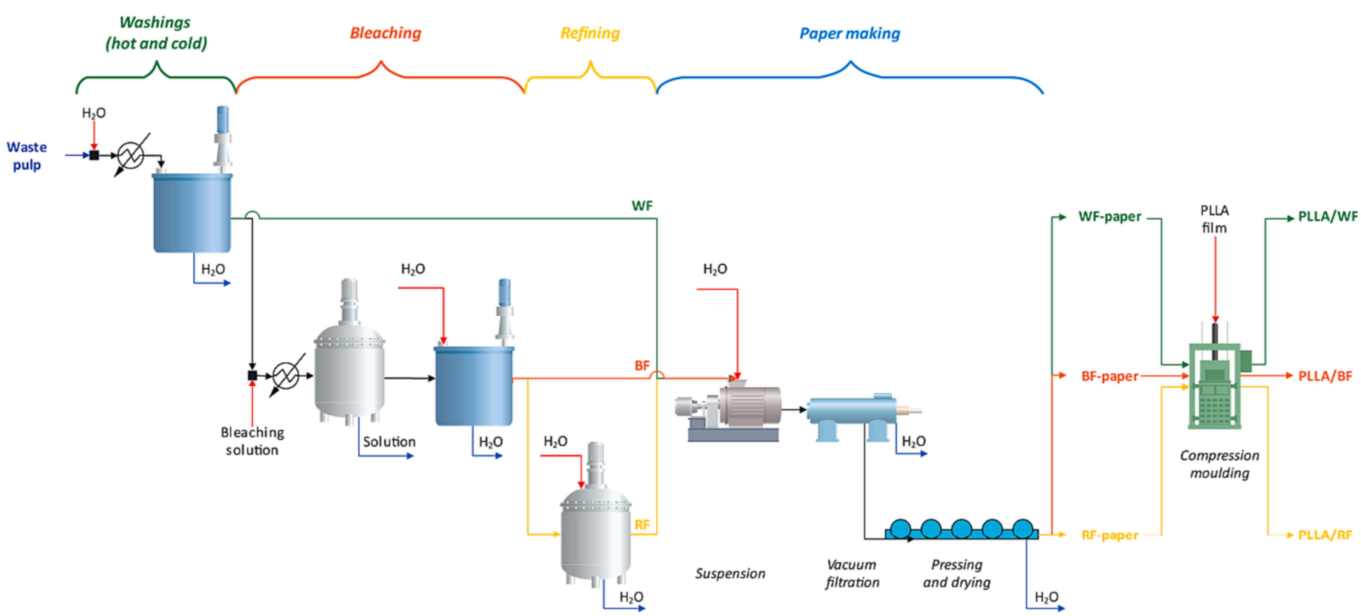


**Fig. 3.** Process flowsheet of manufacturing laminated PLLA composites reinforced with waste pulp from MRF rejects.

crystalline PLLA, taken as 90.9 J $g^{-1}$ based on value reported in the literature [23]. The overall crystallinity ($\chi_c$) of PLLA was calculated from

$$\chi_c, \% = \frac{\Delta H_m}{(1 - w_f) \times \Delta H_m^o} \times 100 \quad (4)$$

### 2.5.8. Lifecycle assessment (LCA)

To assess the environmental impacts stemming from the manufacturing of laminated PLLA composites reinforced with waste pulp fibres, we simulated the production process associated with the foreground system, which interacts with the technological and ecological spheres by exchanging matter and energy using a process simulation software (Aspen Plus V11, Aspen Technology Inc.). Once the mass and energy requirements were estimated, LCA was applied according to the guidelines given by Brujin et al. [24]. The LCA models were implemented in a lifecycle engineering software (SimaPro v9.0, PRé Sustainability, Amersfoort, NL), interfacing with ecoinvent v3.8. The modelling approach and assumptions are further explained below.

**Table 1**
Chemical composition and onset thermal degradation temperature ($T_{d,onset}$) of washed waste pulp and bleached waste pulp.

| Waste pulp | Cellulose (%) | Hemicellulose (%) | | | | Lignin (%) | | Ash (%) | Extr.[g] (%) | $T_{d,onset}$(°C) |
|---|---|---|---|---|---|---|---|---|---|---|
| | | Xy[a] | Mann[b] | Arab[c] | Galac[d] | KL[e] | ASL[f] | | | |
| Washed | 55.6 | 3.9 | 3.2 | 0.2 | 0.4 | 12.7 | 1.2 | 9.4 | 5.7 | 197 |
| Bleached | 76.6 | 5.9 | 5.0 | 0.3 | 0.5 | 5.8 | 1.8 | 1.4 | 1.7 | 237 |

$T_{d,onset}$ is defined as the temperature at which a deflection in the TGA curve was first observed from the established baseline prior to the thermal event.
[a] Xylan.
[b] Mannan.
[c] Arabinan.
[d] Galactan.
[e] Klason lignin.
[f] Acid soluble lignin.
[g] Extractives.

*2.5.8.1. Goal and scope definition.* We considered a cradle-to-gate approach, which includes all life cycle emissions from waste collection (Fig. 2) to the production of the laminated PLLA composites (Fig. 3). Our LCA is based on the UK electricity grid. The functional unit in our LCA model is a neat PLLA or reinforced PLLA composite panel. A performance indicator based on the specific flexural modulus of the materials was used to calculate the mass of the functional unit ($m_{f.u.}$) required to achieve the same level of flexural performance as neat PLLA. The term $m_{f.u.}$ is determined from:

$$m_{f.u.} = m_{PLLA} \times \left(\frac{E_{f,PLLA}}{E_{f,\ f.u.}}\right)^{1/3} \times \left(\frac{\rho_{f.u.}}{\rho_{PLLA}}\right) \tag{5}$$

where $m_{PLLA}$, $E_{f,PLLA}$, $E_{f,\ f.u.}$ , $\rho_{PLLA}$ and $\rho_{f.u.}$ are the reference mass of PLLA (1 kg in our work), flexural modulus of PLLA, flexural modulus of the functional unit, density of PLLA and the functional unit, respectively. The derivation of equation (5) can be found in the work of Gaduan et al. [25].

*2.5.8.2. Lifecycle inventory (LCI).* The LCI combines the data from the foreground system, *i.e.*, mass and energy flows from the Aspen simulation, and from the background system, *i.e.*, surrounding processes that provide inputs to the main process. The foreground system information was obtained from the model simulation in Aspen Plus. On the other hand, the background system data were obtained from the ecoinvent v3.8 database. Allocation at the point of substitution (APOS) was applied because it includes both the impacts of the production and treatment processes [26]. The LCI for PLLA production was collected directly from the ecoinvent v3.8 database and an electricity consumption of 17 MJ $kg^{-1}$ for compression moulding [27] was assumed. For the water output of each sub-process, it was assumed that 10 % of water was sent to wastewater treatment and 90 % was recirculated to the process. Utility consumption for the refining and papermaking process was estimated from literature information [28,29].

*2.5.8.3. Environmental impact assessment (EIA).* Following the hierarchist cultural perspective [30], the ReCiPe 2016 framework [31] was applied as implemented in SimaPro. Our study primarily centred on the assessment of global warming potential (GWP) and the three endpoint indicators, *i.e.*, human health (HH), ecosystem quality (EQ), and resource scarcity (RS). These endpoint indicators were derived from the aggregation of various midpoint indicators, as detailed below:

- HH (DALY): Global warming (HH), stratospheric ozone depletion, ionising radiation, ozone formation (HH), fine particulate matter formation, human carcinogenic toxicity, human non-carcinogenic toxicity and water consumption (HH).
- EQ (species.yr): Global warming (terrestrial ecosystems), Global warming (freshwater ecosystems), ozone formation (terrestrial ecosystems), terrestrial acidification, freshwater eutrophication, marine eutrophication, terrestrial ecotoxicity, freshwater ecotoxicity, marine ecotoxicity, land use, water consumption (terrestrial ecosystems) and water consumption (aquatic ecosystems).
- RS ($USD_{2013}$): Mineral resource scarcity and fossil resource scarcity.

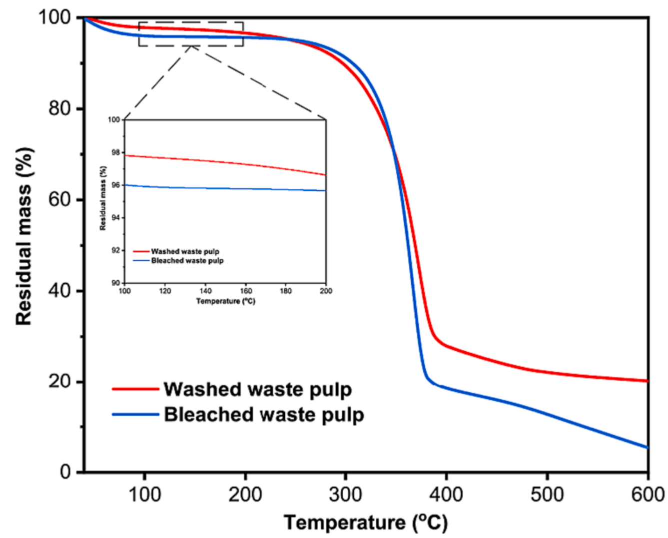


**Fig. 4.** Thermal degradation behaviour of the waste pulp in $N_2$.

## 3. Results and discussion

### 3.1. Chemical composition and thermal degradation of waste pulp

Table 1 summarises the chemical composition of the waste pulp recovered from MRF rejects after washing with hot water only and after acidified $NaClO_2$ bleaching step (see Fig. 1 for the visual appearance of the materials). The high lignin content of the washed waste pulp suggests that the mixed paper and cardboards ended up in a MRF reject stream was mainly mechanical pulp as chemical pulp typically contains low residual lignin [32]. The presence of mechanical pulp in MRF rejects is not surprising as the primary application of mechanical pulp is for products with relatively short life span, including newspaper and cardboard packaging [33], which often ends up in dry recyclables. It can also be seen from Table 1 that the acidified $NaClO_2$ treatment of waste pulp reduced the lignin content and increased the relative content of both cellulose and hemicellulose. This is because acidified $NaClO_2$ removes lignin selectively from lignocellulosic pulp [34]. It should be noted that the delignification is incomplete due to the formation of lignin-carbohydrate complex [35].

The thermal degradation behaviour of the waste pulp washed with hot water only and bleached with acidified $NaClO_2$ in $N_2$ is presented in Fig. 4. The initial weight loss between 50 and 100 °C corresponds to the

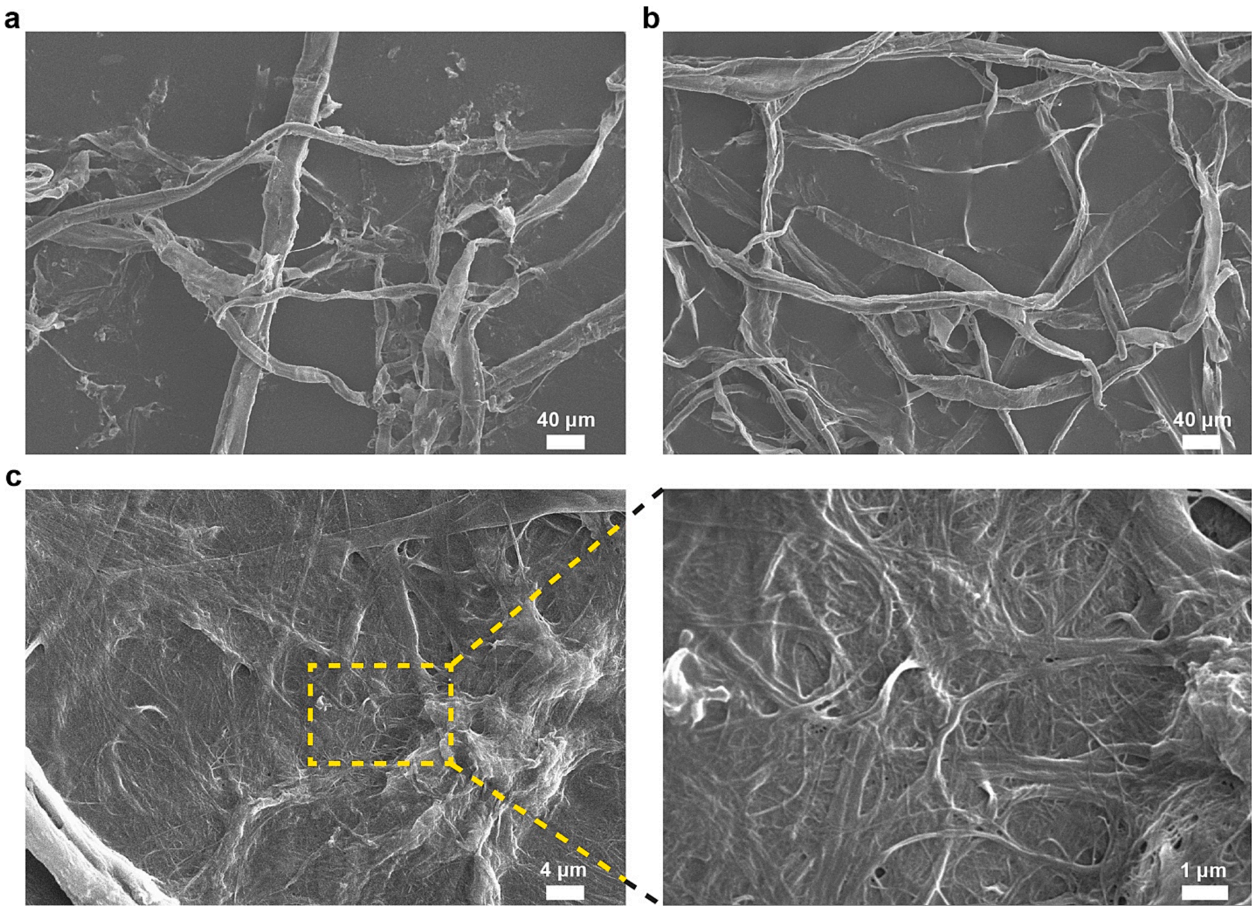


**Fig. 5.** SEM images of (a) WF pulp, (b) BF pulp and (c) BRF pulp.

evaporation of moisture from the pulp. A slow weight loss between 100 and 200 °C was observed for the waste pulp washed with hot water only (see inset in Fig. 4). Over the same temperature range, the bleached waste pulp was thermally stable. This is hypothesised to be due to the presence of non-structural biomass components as evident from the higher extractive fraction in the hot water washed waste pulp. These non-structural biomass includes low molecular weight fats, waxes and simple sugars [36], which simple hot water washing is insufficient to dissolve but the more aggressive $NaClO_2$ treatment could. The origin of these non-structural biomass components may stem from the virgin mechanical pulp itself or contact-transferred (*i.e.*, contamination) to the mixed paper and cardboards during segregation into dry recyclables.

Both types of pulp then underwent single step degradation between 300 and 400 °C, which corresponds to the decomposition of the cyclic structure of cellulose [37,38], as well as the cleavage of functional groups [39] and aryl-ether linkages [40] in lignin. The latter results in the formation of free radicals that leads to self-condensation and the formation of products with increased thermal stability. Due to this, the hot water washed waste pulp that had a higher lignin content also possessed a higher char content compared to bleached waste pulp. As lignin starts to decompose at a lower but over a broader range of temperatures (200–500 °C) than cellulose (300–400 °C) [39,41–44], waste pulp washed with hot water only also possessed a lower onset thermal degradation temperature than bleached waste pulp (see Table 1).

### 3.2. Morphology of waste pulp fibres

The SEM images showing the morphology of the different waste pulp fibres after washing with hot water only, bleached with acidified $NaClO_2$, as well as refined with a re-circulating colloid mill are shown in Fig. 5. Both the hot water washed waste pulp fibres (Fig. 5a) and bleached waste pulp fibres (Fig. 5b) possessed a clean and smooth surface morphology and a similar width of *ca.* 25 μm. By passing the bleached waste pulp through a re-circulating colloid mill, signification fibrillation occurred. This produced refined waste pulp fibres that possessed a mixture of fibrils with a width of ~ 1–2 μm, as well as cellulose nanofibrils (CNFs) with a width of ~ 100 nm. The presence of CNFs in refined waste pulp is surprising as CNFs is typically produced using high-pressure homogenisers [45,46] or energy intensive stone grinders [47]. To the best of our knowledge, no studies have reported CNF production using a colloid mill. Conventional laboratory mill used in pulp and paper research, such as a PFI mill, generates only limited shear force and can only reduce the size of pulp fibres to ~ 1 μm [48]. This is because a PFI mill consists of an inner toothed beater and a smooth outer housing. The re-circulating colloid mill used in this work, which was designed to produce food paste, possesses two toothed surfaces akin to a stone grinder typically used to produce CNFs. This two-

**Table 2**
Porosity ($P$), tensile modulus ($E$), tensile strength ($\sigma$) and the initial critical stress intensity factor ($K_{IC}$) of the paper sheets produced from waste lignocellulosic pulp.

| Waste pulp | $\rho_E$(g cm$^{-3}$) | $\rho_T$(g cm$^{-3}$) | $P$(%) | $E$(GPa) | $\sigma$(MPa) | $K_{IC}$(MPa m$^{0.5}$) |
|---|---|---|---|---|---|---|
| Washed | 0.53 ± 0.07 | 1.58 ± 0.03 | 66 ± 5 | 2.9 ± 0.4 | 17 ± 3 | 0.4 ± 0.1 |
| Bleached | 0.71 ± 0.09 | 1.60 ± 0.02 | 55 ± 8 | 3.2 ± 0.2 | 16 ± 1 | 1.8 ± 0.3 |
| Refined | 0.95 ± 0.05 | 1.55 ± 0.01 | 39 ± 3 | 8.6 ± 0.5 | 63 ± 6 | 4.3 ± 0.5 |

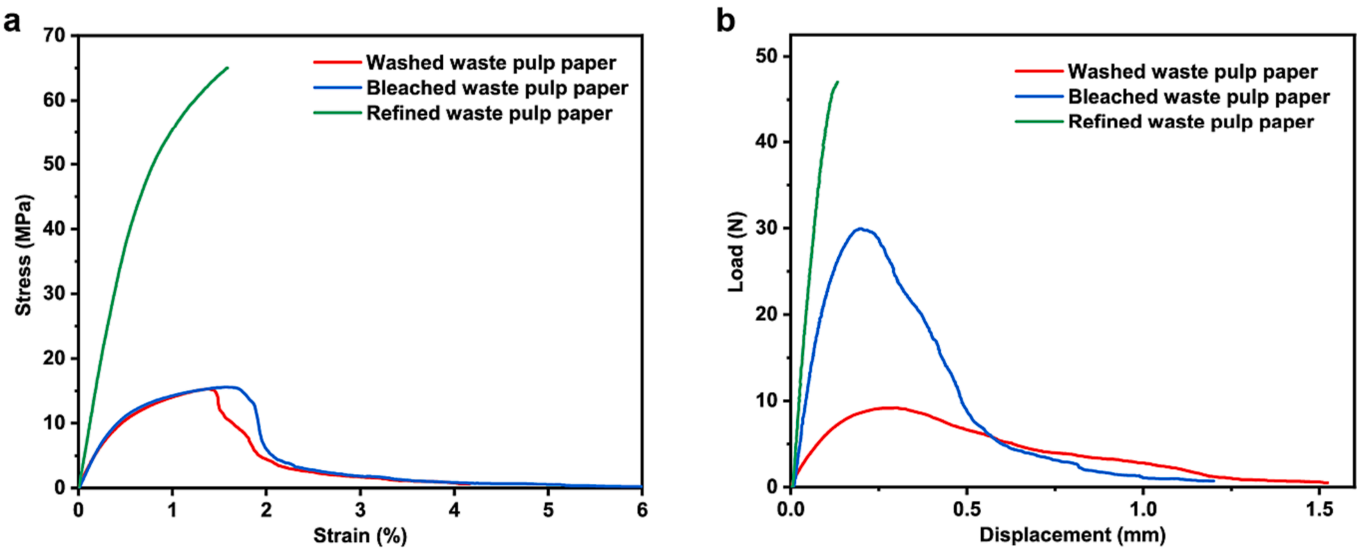


**Fig. 6.** Representative mechanical response of the paper sheets under (a) uniaxial tension and (b) single edge notched tension.

**Table 3**
Mechanical properties of paper based laminated PLLA composites. $E_T$, $\sigma_T$, $E_F$ and $\sigma_F$ denote tensile modulus, tensile strength, flexural modulus and flexural strength, respectively.

| Materials | $E_T$(GPa) | $\sigma_T$(MPa) | $E_F$(GPa) | $\sigma_F$(MPa) | $P$(%) | $P_{theoretical}$(%) |
|---|---|---|---|---|---|---|
| Neat PLLA | 3.5 ± 0.1 | 61 ± 1 | 3.3 ± 0.1 | 102 ± 5 | 0 | |
| PLLA / WF | 5.9 ± 0.4 | 43 ± 3 | 6.3 ± 0.4 | 106 ± 6 | 6 ± 1 | 36 ± 1 |
| PLLA / BF | 6.5 ± 0.4 | 61 ± 6 | 6.6 ± 0.5 | 115 ± 9 | 5 ± 1 | 27 ± 1 |
| PLLA / RF | 6.6 ± 0.6 | 66 ± 6 | 6.2 ± 0.7 | 115 ± 5 | 9 ± 2 | 17 ± 1 |

toothed surface configuration increased the efficiency of fibrillation, leading to the production of a coarse mixture of larger fibrils and CNFs.

### 3.3. Mechanical properties of waste pulp paper sheets

The mechanical properties of the waste pulp paper sheets are tabulated in Table 2. The representative uniaxial tensile stress–strain curves and SENT load–displacement curves are presented in Fig. 6a and Fig. 6b, respectively. It can be seen from both figures that after the initial linear elastic response followed by yielding, paper sheets derived from hot water washed waste pulp and bleached waste pulp fractured in a progressive manner, characterised by a gradual decrease in mechanical load. Such mechanical response can be attributed to the poor degree of fibre–fibre bonding of recycled pulp [49], which led to slippage, reorientation and pull out of the waste pulp fibres during deformation. Paper sheet derived from refined waste pulp, on the other hand, fractured in a catastrophic manner, characterised by a sharp drop in load to 0 N. This can be ascribed to the increased degree of fibre–fibre bonding due to the presence of CNFs in the coarse mixture of the fibrils. The presence of CNFs also lowered the porosity of the resulting refined waste pulp paper sheet due to "pore-filling" effect [50]. As a result, paper sheet produced from refined waste pulp possessed a higher tensile modulus, tensile strength and $K_{IC}$ over paper sheets derived from hot water washed waste pulp and bleached waste pulp (see Table 2). This increase in tensile and fracture performance of refined waste pulp paper sheet however, is at the expense of plasticity as the mobility of the fibres in a tightly bonded fibrous network showed little deformation under creep [51].

### 3.4. Mechanical properties of waste pulp fibre-reinforced laminated PLLA composites

Table 3 summarises the tensile and flexural properties of waste pulp fibre-reinforced laminated PLLA composites. It was found that waste pulp fibres, independent of purification and refinement, acted as effective stiffening agent for PLLA (see Table 3). Both the tensile and flexural modulus of the waste pulp fibre-reinforced laminated PLLA composites increased by ∼ 90 % when compared to neat PLLA. In a laminated construct, the elastic modulus of the composite is the volume weighted average of the elastic modulus of the reinforcement and the polymer matrix [52]. Since the paper sheet derived from refined waste pulp possessed a higher tensile modulus compared to neat PLLA, the modulus of the resulting PLLA/RF composite increased as expected. Considering that the paper sheets derived from hot water washed waste pulp and bleached water pulp had a lower tensile modulus, it is surprising to see that both PLLA/WF and PLLA/BF possessed a higher tensile modulus than neat PLLA (see Table 3).

To investigate this further, we estimated the theoretical porosity of the laminated PLLA composites ($P_{theoretical}$), assuming that the reinforcing paper sheets are uniform, non-porous and incompressible rectangular slabs that are impermeable to molten PLLA using the following equation [53]:

$$P_{theoretical}, \% = 1 - \frac{\rho_E}{\rho_T}\left(\frac{[1 - w_f]\rho_f + w_f\rho_m}{[1 - w_f]\rho_E + w_f\,\rho_m}\right) \times 100 \tag{6}$$

where $\rho_m$ is the density of PLLA, taken as $1.26 \pm 0.01$ g cm$^{-3}$ based on previous measurements [54]. These results are tabulated in Table 3. It was found that the porosity of laminated PLLA/WF, PLLA/BF and PLLA/RF was consistently lower than $P_{theoretical}$. These findings confirmed that molten PLLA successfully infiltrated into the reinforcing paper sheet, bridging the small structural gaps. The pulp fibre network is now bonded together by the PLLA matrix over a substantial part of their length. As a result, the tensile modulus of PLLA/WF, PLLA/BF and PLLA/BRF composites is dominated by the individual waste pulp fibre instead of the tensile modulus of the paper sheet.

The same principle is also true for the strength properties of the laminated PLLA composites. The tensile strength of hot water washed pulp paper and bleached pulp paper was found to be only 17 MPa (Table 2) but the resulting laminated PLLA/WF and PLLA/BF composites achieved a tensile strength of 43 MPa and 61 MPa, respectively. The lower tensile and flexural strength of laminated PLLA/WF composite

**Table 4**
Glass transition temperature ($T_g$), crystallisation temperature ($T_c$) and melting temperature ($T_m$) of PLLA and its laminated composites. The symbols $\chi_{c,moulded}$ and $\chi_c$ are the degree of crystallinity of the PLLA matrix in the moulded samples and the overall crystallinity of PLLA, respectively.

| Samples | $T_g$(°C) | $T_c$(°C) | $T_m$(°C) | $\chi_{c,moulded}$(%) | $\chi_c$(%) |
|---|---|---|---|---|---|
| Neat PLLA | 62 ± 1 | 105 ± 1 | 171 ± 1 | 19 ± 2 | 48 ± 3 |
| PLLA / WF | 65 ± 1 | 105 ± 1 | 170 ± 1 | 40 ± 1 | 52 ± 1 |
| PLLA / BF | 65 ± 1 | 110 ± 2 | 170 ± 1 | 43 ± 3 | 53 ± 1 |
| PLLA / RF | 65 ± 1 | 106 ± 2 | 170 ± 1 | 40 ± 2 | 53 ± 1 |

compared to laminated PLLA/BF and PLLA/RF is postulated to be due to the thermal degradation of WF pulp induced by composite moulding due to its lower $T_{d,onset}$. Nevertheless, tensile and flexural strength of the laminated PLLA composite increased by as much as 8 % and 13 %, respectively, over neat PLLA as seen in PLLA/RF. These results showed that waste pulp can indeed serve as a reinforcement for polymers.

### 3.5. Crystallisation and melt behaviour of waste pulp fibre-reinforced laminated PLLA composites

The characteristic glass transition ($T_g$), crystallisation ($T_c$) and melting ($T_m$) temperatures of neat PLLA and its laminated PLLA composites are summarised in Table 4. We also present $\chi_{c,\text{ moulded}}$ and $\chi_c$ as calculated from equations (3) and (4) in the same table. It can be seen that the addition of waste pulp did not have a profound effect on the $T_g$ and $T_m$ of PLLA, signifying that the chain mobility of PLLA was not affected by the presence of waste pulp. All waste pulp fibre-reinforced laminated PLLA composites possessed a higher $\chi_{c,\text{ moulded}}$ (~40 %) compared to neat PLLA (20 %). However, no difference in $\chi_c$ was observed, implying that the presence of waste pulp influenced only the crystallisation kinetics of the PLLA matrix. PLLA is known to possess a slow crystallisation kinetics [55]. However, *trans*-crystallisation of PLLA can occur from the surface of lignocellulosic fibres [56]. This accelerated the crystallisation of the PLLA matrix in the laminated PLLA composites and ultimately increased $\chi_{c,\text{ moulded}}$ but not $\chi_c$. Similar results have also been observed, whereby highly crystalline bacterial cellulose lowered $T_c$ but not $\chi_c$ [57–59].

### 3.6. LCA of waste pulp fibre-reinforced laminated PLLA composites

As there is also an environmental burden associated with the recovery and re-processing of waste pulp fibres from MRF rejects for use in composite applications, LCA was used to ascertain whether the valorisation of waste pulp as reinforcement for PLLA is still a sustainable option. We begin by analysing the environmental footprint of obtaining waste pulp fibres from the HYDRACYCLE™ process (see Fig. 7a). In the "do nothing" scenario (*i.e.,* in the absence of the HYDRACYCLE™ process), the MRF rejects are sent to landfilled or incinerated as per the usual practice. This will lead to a scenario with GWP, HH, EQ and RS of 3.2 kg $CO_2$e, 1.1 × $10^{-5}$ DALY, 1.4 × $10^{-8}$ species.yr and 0.02 $USD_{2013}$ per kg of MRF rejects, respectively. With the implementation of the HYDRACYCLE™ process, the MRF rejects are recovered and sorted. As the waste pulp is recovered for subsequent used as reinforcement for PLLA, it receives environmental credits due to the avoidance of solid waste treatment (landfill / incineration). Most of the environmental impacts associated with the production of waste pulp come from the treatment of the by-products after the HYDRACYCLE™ process, *i.e.,* treatment of the plastic waste and aluminium scrap, which together account for 92 %, 93 %, 93 % and 75 % of the total positive impacts on GWP, HH, EQ and RS, respectively. The net GWP, HH and EQ impacts are −2.3 kg CO2e, −8.8 × $10^{-6}$ DALY, −9.3 × $10^{-9}$ species.yr and 0.02 $USD_{2013}$ per kg of waste pulp produced, respectively (hollow diamond icon in Fig. 7a). The net positive environmental impact on RS is mainly due to the consumption of mineral and fossil resources for the treatment of aluminium scrap, followed by the treatment of plastic waste and the transportation of waste to the processing plant.

Fig. 7b shows the GWP and the various endpoint-level environmental impact of producing neat PLLA or the different waste pulp fibre-reinforced laminated PLLA functional units based on the flow sheet described in Fig. 3. The laminated composites PLLA/WF, PLLA/BF and PLLA/RF possessed a lower net GWP, HH and EQ than our benchmark PLLA functional unit. This can be attributed to the higher flexural modulus of the various waste pulp fibre-reinforced laminated PLLA composites (see Table 3), which leads to a higher weight saving. Amongst the waste pulp fibre-reinforced laminated PLLA composites, PLLA/WF possessed the lowest net GWP, HH and EQ of 1.9 kg CO2e, 1.4 × $10^{-7}$ DALY, 1.5 × $10^{-8}$ species.yr and 0.41 $USD_{2013}$ per functional unit,

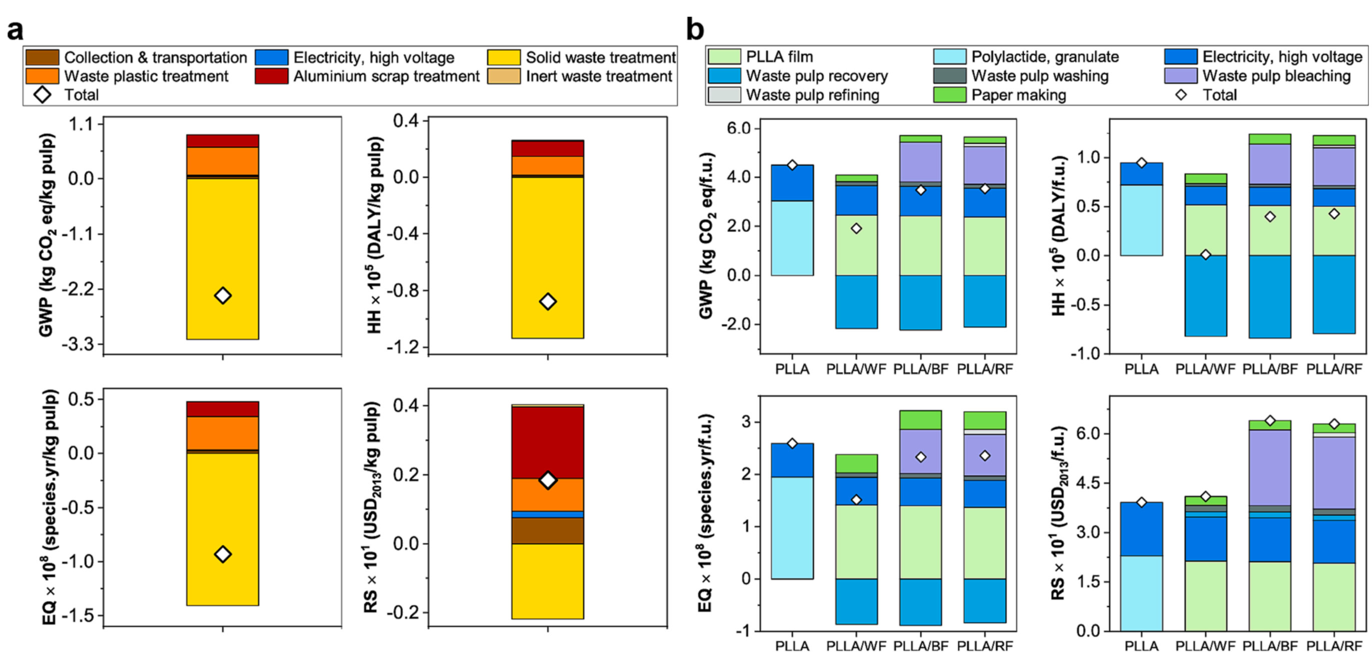


**Fig. 7.** Environmental footprint of (a) recovering waste pulp from the HYDRACYCLE™ process and (b) producing laminated PLLA composites reinforced with waste pulp fibres.

respectively (hollow diamond icon in Fig. 7b). This is because the bleaching process, which is energy intensive as it is conducted at elevated temperature, adds a significant environmental burden (~12 MJ per kg of waste pulp treated, see supplementary information) to the production of PLLA/BF and PLLA/RF. The bleaching process also led to a higher RS for both PLLA/BF and PLLA/RF compared to PLLA/WF.

## 4. Conclusions

We have successfully demonstrated in this work that low value waste pulp recovered from MRF rejects can be recovered and valorised as reinforcing fibres for polymers. High performance laminated PLLA composites reinforced with a $w_f$ of 35 wt-% can be fabricated by laminating waste pulp paper sheets with PLLA films. The laminated PLLA composites exhibited a tensile modulus and strength of up to 6.6 GPa and 66 MPa, respectively. Three-point bending test also showed that the flexural modulus and strength of laminated PLLA composites can achieve up to 6.6 GPa and 115 MPa, respectively. It was found that the PLLA infiltrated the waste pulp paper sheet. As a result, the tensile properties of the laminated PLLA composites are dominated by the individual waste pulp fibre instead of the mechanical properties of the paper sheet. The simplicity of manufacturing waste pulp fibre-reinforced PLLA composites, *i.e.*, papermaking, film casting and compression moulding, coupled with the use of waste pulp as cheap and sustainable reinforcement feedstock, is expected to create a stronger demand for MRF rejects, diverting them away from landfill or incineration and increase the national recycling rate of the UK.

## CRediT authorship contribution statement

**Natalia Herrera:** Writing – review & editing, Writing – original draft, Validation, Resources, Methodology, Investigation, Formal analysis. **Diego Freire Ordóñez:** Writing – review & editing, Writing – original draft, Validation, Software, Methodology, Investigation. **Andre N. Gaduan:** Writing – review & editing, Writing – original draft, Methodology. **Kanjanawadee Singkronart:** Writing – review & editing, Writing – original draft, Methodology. **Daniel Hayes:** Writing – review & editing, Investigation. **Dhivya Puri:** Writing – review & editing, Funding acquisition. **Koon-Yang Lee:** Writing – review & editing, Writing – original draft, Supervision, Funding acquisition, Conceptualization.

## Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Koon-Yang Lee reports financial support was provided by European Commission. If there are other authors, they 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

Data will be made available on request.

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## Appendix A. Supplementary material

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## References

[1] Defra. 25 Year Environmental Plan. https://www.gov.uk/government/publications/25-year-environment-plan; 2018.
[2] Defra. Resources and waste strategy for England. https://www.gov.uk/government/publications/resources-and-waste-strategy-for-england; 2018.
[3] Towards the circular economy. https://ellenmacarthurfoundation.org/towards-the-circular-economy-vol-1-an-economic-and-business-rationale-for-anEllen MacArthur Foundation; 2013.
[4] Defra. UK statistics on waste. https://www.gov.uk/government/statistics/uk-waste-data; 2022.
[5] Purnell P. On a voyage of recovery: a review of the UK's resource recovery from waste infrastructure. Sustain Resilient Infrastruct 2019;4(1):1–20.
[6] Moura JMBM, Gohr Pinheiro I, Carmo JL. Gravimetric composition of the rejects coming from the segregation process of the municipal recyclable wastes. Waste Manag 2018;74:98–109.
[7] Catagna A, Casagrande M, Zeni A, Girelli E, Rada E, Ragazzi M, et al. 3R'S from citizens point of view and their proposal from a case-study. UPB Sci Bull, Series D: Mech Eng 2013;75:253–64.
[8] Lima FPA, de Oliveira FG. Recycling and social technologies for sustainability: The Brazilian experience of wastepickers' inclusion in selective collection programs. Work 2017;57:363–77.
[9] Welle F. Twenty years of PET bottle to bottle recycling—An overview. Resour Conserv Recycl 2011;55(11):865–75.
[10] Awaja F, Pavel D. Recycling of PET. Eur Polym J 2005;41(7):1453–77.
[11] Pivnenko K, Eriksson E, Astrup TF. Waste paper for recycling: Overview and identification of potentially critical substances. Waste Manag 2015;45:134–42.
[12] Oke A, McDonald S, Korobilis-Magas E, Osobajo OA, Awuzie BO. Reframing Recycling Behaviour through Consumers' Perceptions: An Exploratory Investigation. Sustainability 2021.
[13] Norton V, Waters C, Oloyede OO, Lignou S. Exploring Consumers' Understanding and Perception of Sustainable Food Packaging in the UK. Foods 2022.
[14] Timlett RE, Williams ID. Public participation and recycling performance in England: A comparison of tools for behaviour change. Resour Conserv Recycl 2008;52(4):622–34.
[15] Hahladakis JN, Purnell P, Iacovidou E, Velis CA, Atseyinku M. Post-consumer plastic packaging waste in England: Assessing the yield of multiple collection-recycling schemes. Waste Manag 2018;75:149–59.
[16] Defra. Rejected recycling. https://www.gov.uk/government/publications/rejected-recycling; 2019.
[17] Banks C, Thompson NM, Puri DJ, Speller P, Stuart-Paul C. Method for waste processing. US Patent 20180361394: Fiberight Ltd.; 2016.
[18] Hietala M, Varrio K, Berglund L, Soini J, Oksman K. Potential of municipal solid waste paper as raw material for production of cellulose nanofibres. Waste Manag 2018;80:319–26.
[19] Mtibe A, Linganiso LZ, Mathew AP, Oksman K, John MJ, Anandjiwala RD. A comparative study on properties of micro and nanopapers produced from cellulose and cellulose nanofibres. Carbohydr Polym 2015;118:1–8.
[20] Sluiter A, Ruiz R, Scarlata C, Sluiter J, Templeton D. Determination of Extractives in Biomass; Laboratory Analytical Procedure (LAP). National Renewable Energy Laboratory; 2008: 1–9.
[21] Sluiter A, Hames B, Ruiz R, Scarlata C, Sluiter J, Templeton D, et al. Determination of structural carbohydrates and lignin in biomass, in: Laboratory Analytical Procedure (LAP). National Renewable Energy Laboratory; 2008.
[22] Hayes DJM. Development of near infrared spectroscopy models for the quantitative prediction of the lignocellulosic components of wet Miscanthus samples. Bioresour Technol 2012;119:393–405.
[23] Pyda M, Bopp RC, Wunderlich B. Heat capacity of poly(lactic acid). J Chem Thermodyn 2004;36(9):731–42.
[24] Bruijn H, Duin R, Huijbregts MAJ, Guinee JB, Gorree M, Heijungs R, et al. Handbook on Life Cycle Assessment: Operational Guide to the ISO Standards. The Netherlands: Springer, Dordrecht; 2002.
[25] Gaduan AN, Singkronart K, Bell C, Tierney E, Burgstaller C, Lee K-Y. Mechanical Upcycling Immiscible Polyethylene Terephthalate-Polypropylene Blends with Carbon Fiber Reinforcement. ACS Appl Polym Mater 2022;4(5):3294–303.
[26] Wernet G, Bauer C, Steubing B, Reinhard J, Moreno-Ruiz E, Weidema B. The ecoinvent database version 3 (part I): overview and methodology. Int J Life Cycle Assess 2016;21(9):1218–30.
[27] Tchana TG. Life Cycle Evaluation of Manufacturing and Mechanical Properties for Novel Natural Fibre Composites. UK: University of Hertfordshire; 2021.
[28] Suhr M, Klein G, Kourti I, Gonzalo MR, Santonja GG, Roudier S, et al. Best Available Techniques (BAT) Reference Document for the Production of Pulp, Paper and Board. Best Available Techniques (BAT) Reference Document for the Production of Pulp, Paper and Board - Industrial Emissions Directive 2010/75/EU (Integrated Pollution Prevention and Control). Seville, Spain: European Commission Joint Research Centre Institute for Prospective Technological Studies; 2015.
[29] SaintyCo. https://wwwsaintycocom/pharma-equipment/semi-solids-production/cm-series-colloid-mill/. Last accessed 22nd October 2023.

[30] Tomatis M, Moreira MT, Xu H, Deng W, He J, Parvez AM. Removal of VOCs from waste gases using various thermal oxidizers: A comparative study based on life cycle assessment and cost analysis in China. J Clean Prod 2019;233:808–18.

[31] Huijbregts MAJ, Steinmann ZJN, Elshout PMF, Stam G, Verones F, Vieira M, et al. ReCiPe2016: a harmonised life cycle impact assessment method at midpoint and endpoint level. Int J Life Cycle Assess 2017;22(2):138–47.

[32] Holtzapple MT. LIGNIN. In: Caballero B, editor. Encyclopedia of Food Sciences and Nutrition. Second Edition. Oxford: Academic Press; 2003. p. 3535–42.

[33] El-Haggar SM. Chapter 5 - Sustainability of Municipal Solid Waste Management. In: El-Haggar SM, editor. Sustainable Industrial Design and Waste Management. Oxford: Academic Press; 2007. p. 149–96.

[34] Wang Y, Yang Y, Qu Y, Zhang J. Selective removal of lignin with sodium chlorite to improve the quality and antioxidant activity of xylo-oligosaccharides from lignocellulosic biomass. Bioresour Technol 2021;337:125506.

[35] Gupta R, Lee YY. Investigation of biomass degradation mechanism in pretreatment of switchgrass by aqueous ammonia and sodium hydroxide. Bioresour Technol 2010;101(21):8185–91.

[36] Pattiya A. 1 - Fast pyrolysis. In: Rosendahl L, editor. Direct Thermochemical Liquefaction for Energy Applications. Woodhead Publishing; 2018. p. 3–28.

[37] Seifert M, Hesse S, Kabrelian V, Klemm D. Controlling the water content of never dried and reswollen bacterial cellulose by the addition of water-soluble polymers to the culture medium. J Polym Sci A Polym Chem 2004;42(3):463–70.

[38] Cheng K-C, Catchmark JM, Demirci A. Enhanced production of bacterial cellulose by using a biofilm reactor and its material property analysis. J Biol Eng 2009;3(1):12.

[39] Brebu M, Vasile C. Thermal degradation of lignin - A review. Cellul Chem Technol 2010;44(9):353–63.

[40] Afifi AI, Hindermann JP, Chornet E, Overend RP. The cleavage of the arylOCH3 bond using anisole as a model compound. Fuel 1989;68(4):498–504.

[41] Yang H, Yan R, Chen H, Lee DH, Zheng C. Characteristics of hemicellulose, cellulose and lignin pyrolysis. Fuel 2007;86(12):1781–8.

[42] Waters CL, Janupala RR, Mallinson RG, Lobban LL. Staged thermal fractionation for segregation of lignin and cellulose pyrolysis products: An experimental study of residence time and temperature effects. J Anal Appl Pyrol 2017;126:380–9.

[43] Lv D, Xu M, Liu X, Zhan Z, Li Z, Yao H. Effect of cellulose, lignin, alkali and alkaline earth metallic species on biomass pyrolysis and gasification. Fuel Process Technol 2010;91(8):903–9.

[44] Várhegyi G, Antal MJ, Jakab E, Szabó P. Kinetic modeling of biomass pyrolysis. J Anal Appl Pyrol 1997;42(1):73–87.

[45] Turbak AF, Snyder FW, Sandberg KR. Microfibrillated cellulose, a new cellulose product: Properties, uses and commercial potential. J Appl Polym Sci: Appl Polym Symp 1983;37:459–94.

[46] Herrick FW, Casebier RL, Hamilton JK, Sandberg KR. In: Microfibrillated cellulose: morphology and accessibility. New York: Wiley; 1983. p. 797–813.

[47] Nakagaito AN, Yano H. The effect of morphological changes from pulp fiber towards nano-scale fibrillated cellulose on the mechanical properties of high-strength plant fiber based composites. Appl Phys A-Mater Sci Process 2004;78(4):547–52.

[48] Chakraborty A, Sain MM, Kortschot MT, Ghosh SB. Modeling energy consumption for the generation of microfibres from bleached kraft pulp fibres in a PFI mill. BioResources 2007;2(2):210–22.

[49] Jin H, Kose R, Akada N, Okayama T. Relationship between wettability of pulp fibers and tensile strength of paper during recycling. Sci Rep 2022;12(1):1560.

[50] Li A, Xu D, Luo L, Zhou Y, Yan W, Leng X, et al. Overview of nanocellulose as additives in paper processing and paper products. 2021;10(1):264-81.

[51] Santmarti A, Liu HW, Herrera N, Lee K-Y. Anomalous tensile response of bacterial cellulose nanopaper at intermediate strain rates. Sci Rep 2020;10(1):15260.

[52] Hull D, Clyne TW. An introduction to composite materials. 2 ed. Cambridge: Cambridge University Press; 1996.

[53] Hervy M, Blaker JJ, Braz AL, Lee K-Y. Mechanical response of multi-layer bacterial cellulose nanopaper reinforced polylactide laminated composites. Compos A Appl Sci Manuf 2018;107:155–63.

[54] Hervy M, Bock F, Lee K-Y. Thinner and better: (Ultra-)low grammage bacterial cellulose nanopaper-reinforced polylactide composite laminates. Compos Sci Technol 2018;167:126–33.

[55] Wang Y, Liu C, Shen C. Crystallization behavior of poly(lactic acid) and its blends. Polym Crystalliz 2021;4(3):e10171.

[56] Wang Y, Tong B, Hou S, Li M, Shen C. Transcrystallization behavior at the poly (lactic acid)/sisal fibre biocomposite interface. Compos A Appl Sci Manuf 2011;42(1):66–74.

[57] Lee K-Y, Bharadia P, Blaker JJ, Bismarck A. Short sisal fibre reinforced bacterial cellulose polylactide nanocomposites using hairy sisal fibres as reinforcement. Compos A Appl Sci Manuf 2012;43(11):2065–74.

[58] Lee K-Y, Blaker JJ, Bismarck A. Surface functionalisation of bacterial cellulose as the route to produce green polylactide nanocomposites with improved properties. Compos Sci Technol 2009;69(15):2724–33.

[59] Montrikittiphant T, Tang M, Lee K-Y, Williams CK, Bismarck A. Bacterial cellulose nanopaper as reinforcement for polylactide composites: renewable thermoplastic NanoPaPreg. Macromol Rapid Commun 2014;35(19):1640–5.