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A greener approach for physical separation of polycotton textile waste

Ying Siew Khoo^{a,b}, Yen Nan Liang^{a,b}, Xiao Hu^{a,b,*}, Jia Wei Chew^{b,c,d,**}

^a School of Materials Science and Engineering, Nanyang Technological University (NTU), 50 Nanyang Ave, Block N4.1, 639798, Singapore

^b RGE-NTU Sustainable Textile Research Centre, Nanyang Technological University (NTU), 639798, Singapore

^c School of Chemistry, Chemical Engineering and Biotechnology, Nanyang Technological University (NTU), 62 Nanyang Drive, 637459, Singapore

^d Chemical Engineering, Chalmers University of Technology, Gothenburg 412 96, Sweden

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ABSTRACT

Most textile waste ends up in the landfill. Clearly, enhancing textile recycling is urgently needed. For circularity, efficient means for separating the constituents of the textile blends is crucial. Targeting the popular polycotton textile blend, a method was developed to physically separate cotton and PET (polyethylene terephthalate) from waste textiles. First, the waste polycotton textile was milled into smaller fibers to mitigate entanglement. Then, leveraging the relatively higher hydrophilicity of cotton, three liquids (namely, water, mineral oil, and ethanol) were mixed to form a two-phase liquid-liquid system, which induced the preferential segregation of cotton to the water-rich bottom liquid phase. Factors studied included liquid composition and milling method. The best-performing liquid mixture allowed for 84.7 % of the cotton and 85.5 % of the PET from the waste polycotton (WPC) sample to be recovered respectively from the bottom liquid phase (BLP) and top liquid phase (TLP). The separation effectiveness can be further enhanced if fiber entanglement issue can be mitigated. The recovery of about 99.9 % PET and 86.7 % cotton from a prepared mixture of pure cotton and pure PET textile wastes is a case in point, demonstrating the efficacy of the method that is fast, at ambient condition, and does not require extensive chemicals. While the interwoven design of cotton and PET in textiles confers benefits like comfort and strength, the resulting entanglement is detrimental for circularity.

Synopsis: An eco-friendly yet rapid two-phase liquid-liquid system was developed for separating polycotton textile fibers, aimed at enhancing sustainability and circularity

1. Introduction

Propelled by population growth, fast-changing fashion trends and improvement of living standards especially in developing countries, the textile industry has emerged as a major polluter [1,2]. More textile wastes are expected to be generated, leading to higher demands for landfills. Of the 80–100 billion new clothing garments produced annually by the textile industry, only 1 % is recycled back into new garments [3]. Textile waste not only burdens the limited landfill spaces [4], the dyes leached out from the discarded textile waste can result in environmental risks, posing health hazards to the underground water sources and ecosystem [5]. Furthermore, 8–10 % of global greenhouse gas emission was contributed by the textile, apparel, and fashion (TAF) industries [6], which represent the second largest water consumers of approximately 215 trillion liters annually [7]. Therefore, urgent

attention must be paid to managing textile waste, including developing efficient collection systems and recycling technologies.

Reuse is the most circular option for clean textiles in good condition, but not applicable for the worn out, non-rewearable and damaged textiles [8], thus necessitating chemical, biological or mechanical methods [7,9]. One of the major challenges of textile recycling is that the fabrics are typically blends of different types of intertwined fibers, which requires separation into the constituent components before further treatment [8]. Among the blended fabrics, polyester-cotton is the most prominent and common fabric blends, which is also known as polycotton [10,11]. PET offers advantages of durability, strength and lower price, while cotton is beneficial for higher water absorbency and softness, making their combination a perfect blend for fabrics [12]. In view of circularity and sustainability, the recycling of blended fabric is of utmost importance and has been attracting increasing attention.

* Corresponding author at: School of Materials Science and Engineering, Nanyang Technological University (NTU), 50 Nanyang Ave, Block N4.1, 639798, Singapore.

** Corresponding author at: Chemical Engineering, Chalmers University of Technology, Gothenburg 412 96, Sweden

E-mail addresses: asxhu@ntu.edu.sg (X. Hu), jia.chew@chalmers.se (J.W. Chew).

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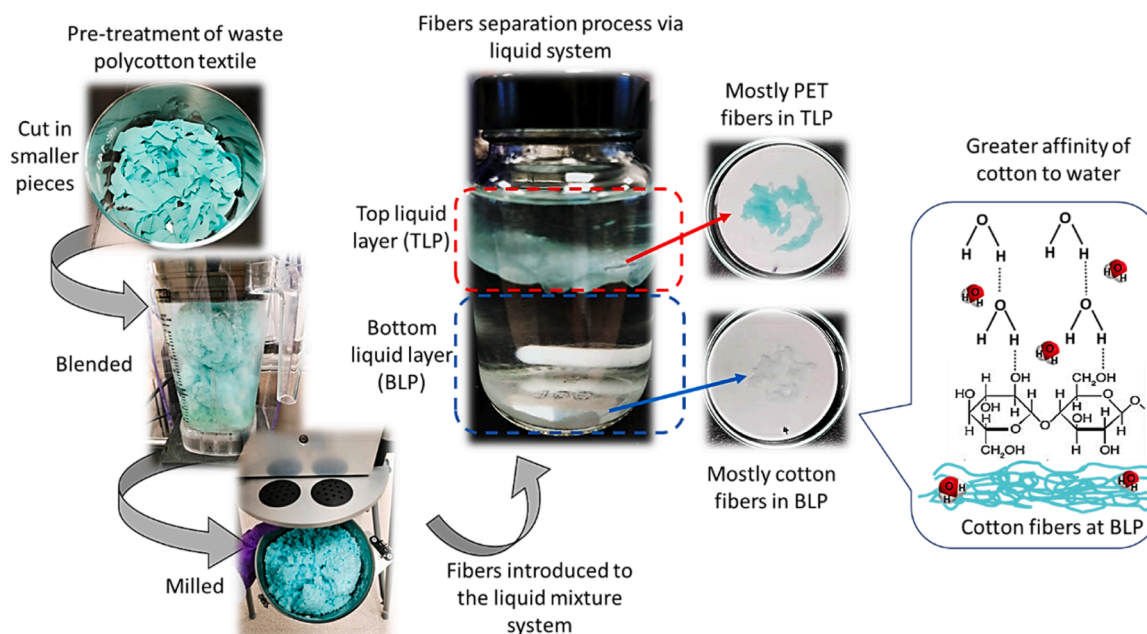


Fig. 1. Overview of the devised fiber separation process, which involves milling followed by separation via a two-phase liquid-liquid mixture composed of DI water, mineral oil and ethanol (or IPA).

Numerous chemical recovery studies, including cellulose dissolution, PET dissolution, acid hydrolysis of cellulose, enzymatic hydrolysis and alkaline hydrolysis of PET, have been reported for recycling polycotton fabrics by depolymerizing or dissolving one of the components [10, 12–16]. For instance, Xia et al. [10] successfully obtained regenerated cellulose (55–65 %) and high-purity recovered PET (92–96 %) from waste PET-cotton blended textiles (PET:cotton = 30:70) using an ionic liquid, namely, 1-allyl-3-methylimidazolium chloride (AmimCl), at 80 °C for 3 h under vigorous mechanical stirring. The dissolution of cellulose in ionic liquids was tied to the separation of hydroxyl groups of different chains upon interaction between cellulose and ionic liquid [17]. Moreover, dimethyl sulfoxide (DMSO) has been utilized as an organic solvent to dissolve the organic materials, including PET and organic residues, remaining from the textile dyes [10,16]. Yousef et al. [16] reported that 77 wt% of cotton and 16.2 wt% of PET was recovered from polycotton jeans (PET:cotton = 20:80) after a dye leaching process via nitric acid, followed by PET dissolution using DMSO to break the van der Waals bonds of the organic molecules at 50 °C for 6 h, and finally bleaching using sodium hypochlorite and diluted hydrochloric acid to obtain pure cotton fibers. Nevertheless, the recycling of waste polycotton textiles using these chemical approaches is still challenging due to several limitations, such as scalability to industrial capacity [9,18] and requirement for large amounts of chemicals.

Mechanical recovery is more scalable and costs less than bio-based or chemical means [9]. The textile waste are first shredded into fibers, and then re-spun into new yarn or threads, followed by knitted or weaved into recycled clothing [19], with the chemical molecular structure remaining intact [7]. However, the varied material composition contained in textile waste remains as a key challenge for mechanical recycling [8,11,19]. Furthermore, the extensive fiber entanglement in blended fabrics adversely affects the fibers separation efficiency [14].

Targeted at improving textile waste sorting, our aim was to devise and optimize a rapid physical separation approach for separating cotton and PET fibers from polycotton textile wastes. Leveraging the different natures of cotton and PET, specifically, the relatively higher hydrophilicity of cotton, it was conceived that a two-phase liquid-liquid mixture can be used to segregate the two fiber types. The polycotton were first milled to physically separate the textiles before being added to the liquid-liquid mixture (composed of de-ionized (DI) water, mineral oil

and ethanol (ETH)). The effects of post-mill fiber length and liquid composition were evaluated. The fiber separation using a liquid-liquid system made up of mineral oil, water, and ethanol is a straightforward and efficient method that demonstrates a high effectiveness in separating polycotton fibers. Additionally, the solutions from the liquid-liquid system can be reused multiple times, making it a cost-effective process.

2. Experimental

2.1. Materials

Three types of waste textiles were evaluated, namely, polyester-cotton blend (WPC; 40 % Cotton and 60 % PET; green in color), polyester (100 % PET; black in color) and cotton (pure 100 % cotton; white in color).

Mineral oil (Bluewater Chemgroup), distilled water, ethanol (99.9 % absolute, Merck) and isopropyl alcohol (IPA, Tech grade, Aik Moh) were used in various compositions to create the two-phase liquid-liquid mixture to separate the polycotton fibers physically.

Liquid nitrogen was used during cryo-milling. Filter paper (55 mm in diameter, Whatman) was used during filtration to collect the recovered fibers after the separation.

2.2. Fibers milling

Fig. 1 overviews the steps involved in the cotton-PET separation process. The waste textiles first need to be shredded into fibers. Specifically, they were cut into smaller square pieces using scissors (approximately 3 cm by 3 cm in dimension), then blended (Waring Xtreme) for 1 minute per cycle (total 5 cycles), followed by milling (using either the cryogenic grinder (SPEX 6875) or cutting miller (Fritsch; 1 mm sieve)).

The milling step is important to disentangle the fibers as well as increase the contact surface between the fibers and liquid during the subsequent separation step. For cryogenic grinding, 6 g of the post-blender sample was loaded into a vial and an impactor was placed into the vial for grinding. The textile sample was precooled for 15 min in the grinder, and the grinding rate was set at 12 cps (cycle per second) for 4 cycles. The grinding process took about 25 min at -196 °C (cooled by

Table 1
Summary of the Liquid Mixture (E = ethanol, W = water, I=IPA).

Label	Volume of ethanol (mL)	Volume of DI water (mL)	Volume of mineral oil (mL)	Volume of IPA (mL)
E-0	0	30	30	0
E-10	10	30	30	0
E-30	30	30	30	0
E-50	50	30	30	0
W-40	0	40	30	0
W-60	0	60	30	0
W-80	0	80	30	0
I-30	0	30	30	30

liquid nitrogen), after which the vial was removed from the grinder and left at room temperature for at least 10 min. For the cutting miller with a 1.0 mm sieve size installed, about 15 g of the post-blender sample was milled for 3 min at room temperature.

2.3. Physical separation

After the waste textiles were milled into fibers, they were added to a liquid mixture composed of DI water, mineral oil, and ethanol or IPA, which forms a liquid-liquid mixture with oil-rich and water-rich phases respectively at the top and bottom (Fig. 1). Different liquid mixture and different compositions were assessed, as listed in Table 1.

0.05 g of the milled waste textile (i.e., either 0.05 g of the green WPC blend, or a mix of 0.02 g of pure white cotton and 0.03 g of pure black PET) were used each time. The fibers were added to the liquid mixture and stirred for 10 min at 200 rpm using a magnetic stirrer. After that, the mixture containing the fibers was left to naturally phase-separate for 30 min, resulting in the preferential segregation of the different fibers to either the top liquid phase (TLP) or bottom liquid phase (BLP).

2.4. Performance evaluation

Prior to use, the filter papers were placed in the oven overnight at 80 °C to rid of moisture, and then weighed to get the initial weight, M_i (g). The top liquid phase (TLP) and bottom liquid phase (BLP) were

separated via vacuum filtration. Acetone was used to rinse off any liquid residue on the recovered fibers. The filter papers with the recovered fibers (Fig. 1) were placed in the oven overnight at 80 °C to remove the water and acetone residuals, then weighed (M_f). The mass of fibers, M_R (g), collected from each of the two phases was the difference between M_f and M_i .

The chemical compositions of the recovered fibers from the top and bottom liquid phases were examined using Fourier Transform Infrared Spectroscopy (FTIR; PerkinElmer Frontier). A calibration curve of textile fibers with different cotton and PET concentrations was generated using FTIR, as shown in Fig. S1. Specifically, the y-axis values are calculated by the ratio of the difference between the peak heights corresponding to cotton (namely, 3333 and 3700 cm^{-1}) to that of PET (namely, 1713 and 1890 cm^{-1}).

2.5. Water absorption test

Different types of textiles were cut into smaller square pieces (2 cm by 2 cm) and immersed in DI water for up to 40 h. Prior to the absorption experiment, the initial mass of the sample was taken (W_i , g). For the first 10 min of immersion, the sample mass (W_f , g) was taken at every 30 s interval and the percentage change in mass recorded.

2.6. Characterization

The lengths of milled fibers were analyzed using a microscope (Olympus, BX51) and the ImageJ software. At least 100 fibers from each sample type were measured to yield an average length, as well as the fiber length distribution.

The morphologies of the recovered fibers were observed using field emission scanning electron microscope (FESEM; JEOL, JSM-7600F). Prior to FESEM observation, platinum coating was applied (JEOL, JFC-1600) to prevent overcharging during scanning.

The chemical compositions of the milled fibers and recovered fibers were analyzed using FTIR, whereby the spectra were recorded in the 600–4000 cm^{-1} wavenumber range using the KBr pellet method.

To investigate the thermal degradation of fibers, the Thermogravimetric Analyzer (TGA,TA instruments, Q500) was used in the

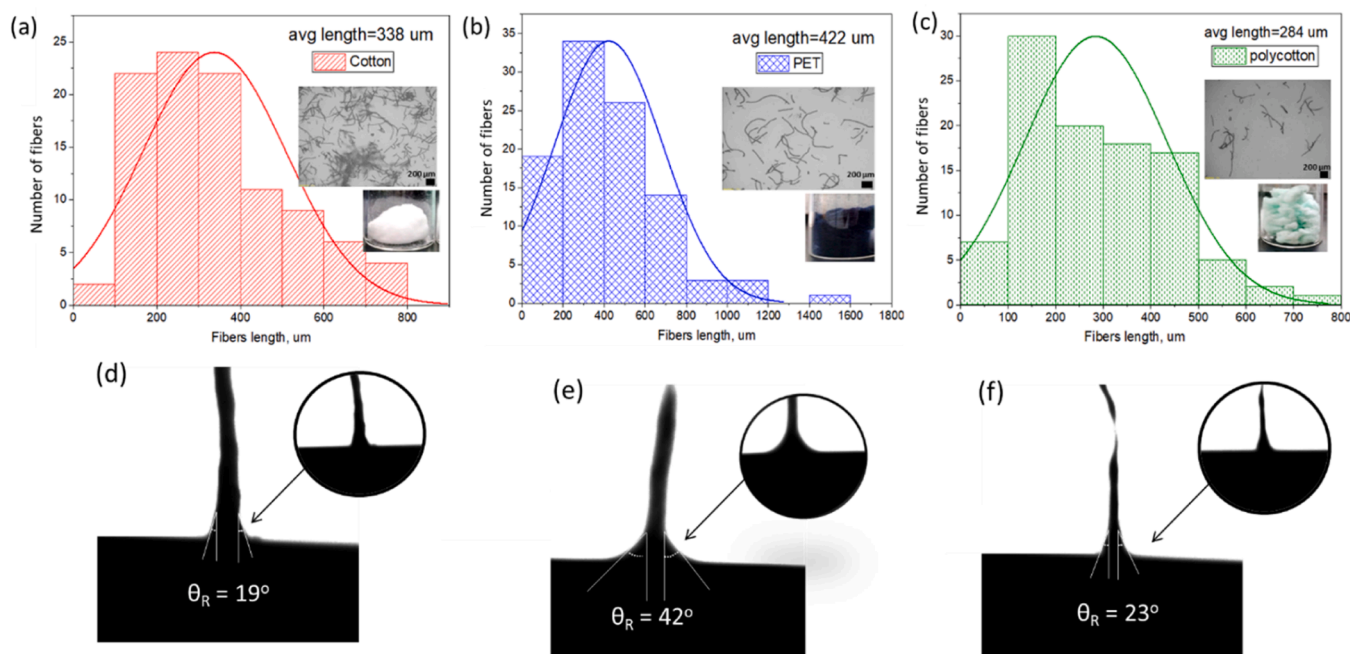


Fig. 2. Fiber length distributions of waste textiles after the cutter-miller step: (a) cotton, (b) PET, and (c) polycotton. Water contact angles of waste textiles fibers (each image is composed of several fibers): (d) cotton, (e) PET, and (f) polycotton.

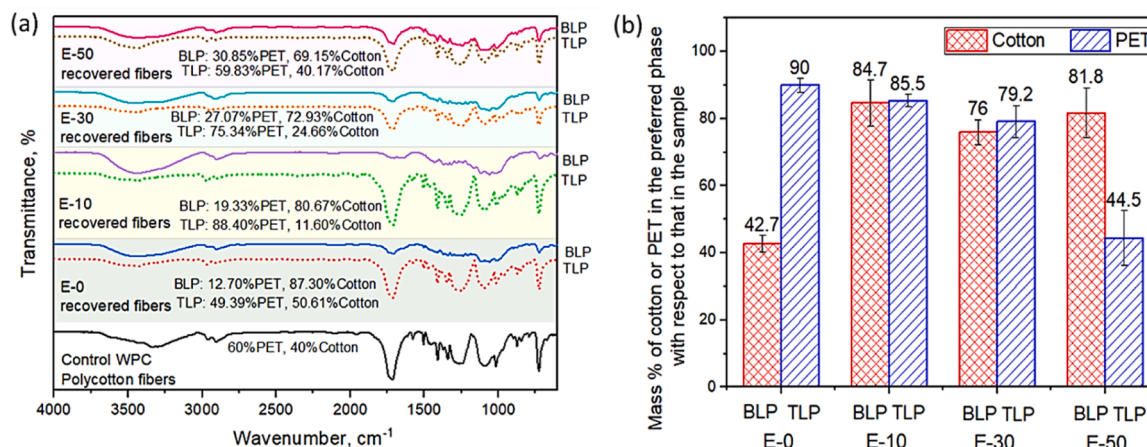


Fig. 3. Effect of different liquid compositions (Table 1): (a) FTIR analysis of recovered fibers extracted from TLP and BLP; the compositions of the fibers in each phase are indicated. (b) Mass percentage of each fiber type in the preferred liquid phase (BLP or TLP) with respect to that in the sample after subjected to different liquid mixtures (E-0, E-10, E-30, E-50). (Note: the WPC sample was cutter-milled).

temperature range of 25 °C to 600 °C (10 °C/min heating rate) and under nitrogen atmosphere at a flow rate of 40 mL/min.

The receding water contact angle of the fibers were imaged using the contact angle goniometer (OCA, Dataphysics 15) and quantified using the ImageJ software.

3. Results and discussion

3.1. Physical properties of fibers

The milled fiber length is one of the critical factors that affect polycotton separation due to the tendency of the longer ones to entangle. Therefore, it is important to understand the relationship between fiber length and fiber separation performance. Fig. 2(a)–(c) present the fiber length distributions of pure cotton, pure PET and polycotton fibers resulting from the waste textiles shredded using the cutting miller, with the corresponding optical microscope images presented as insets. The average fibers lengths ranged from 284 μm to 422 μm . On the other hand, for cryo-milling, Fig. S2 indicates shorter fibers in the range of 54.8 μm to 72.7 μm .

Figs. 2(d)–(e) compare the receding water contact angles of the different types of fibers. The average receding angles of the pure cotton and PET fibers were measured to be approximately 19° and 42°, respectively, which agree with values reported of 20° and 45° \pm 3 [20, 21]. As for polycotton, the receding angle was 23°, which is in between that of pure cotton and pure PET. Due to the abundance of -OH groups and thus hydrophilic nature of cotton, it exhibits higher affinity towards the water molecules via hydrogen bonding and tends to absorb more water, leading to a lower receding contact angle when in contact with water [22]. On the contrary, PET, with moderate hydrophobicity, exhibited a higher receding angle than cotton and polycotton fibers attributed to the lower water absorption capacity [23].

3.2. Comparison of separation performance by different liquid compositions

Fig. 3(a) shows the FTIR spectrum of the cutter-milled polycotton fibers from the WPC sample (i.e., 40 % Cotton and 60 % PET) as a benchmark for that of the fibers recovered from different compositions of the liquid mixtures (Table 1). The compositions of the cutter-milled

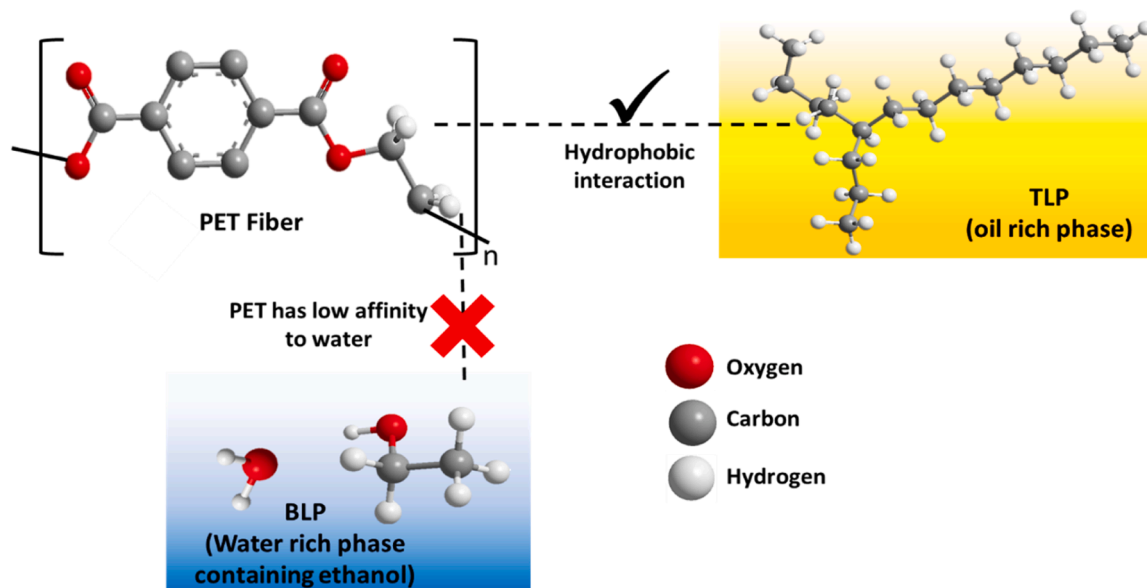


Fig. 4. Proposed interaction of the PET fibers in the TLP (oil-rich) and BLP (water-rich).

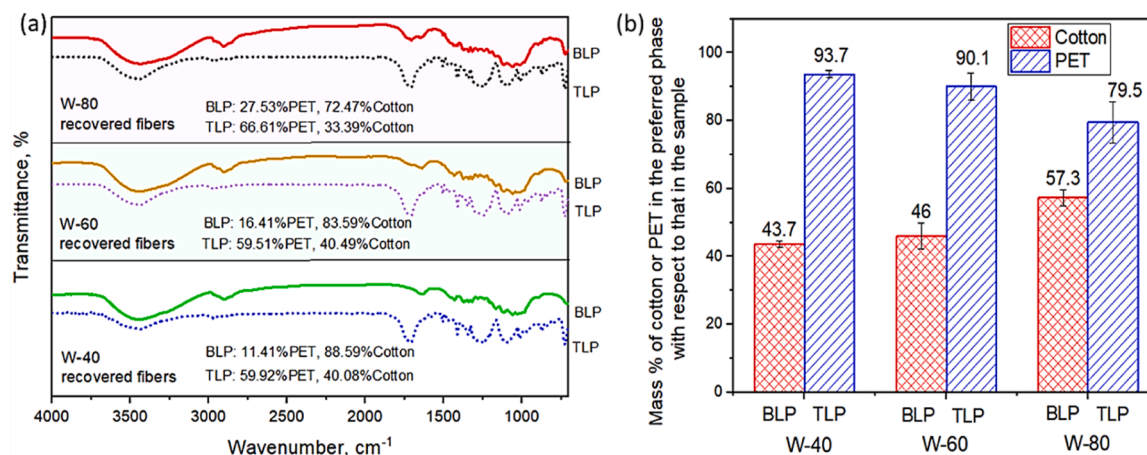


Fig. 5. Effect of different liquid compositions (Table 1): (a) FTIR analysis of recovered fibers extracted from TLP and BLP; the compositions of the fibers in each phase are indicated. (b) Mass percentage of each fiber type in the preferred liquid phase (BLP or TLP) with respect to that in the sample after subjected to different liquid mixtures (W-40, W-60, W-80) (Note: the WPC sample was cutter-milled.).

fibers retrieved from the BLPs and TLPs are also indicated in Fig. 3(a). Most of the fibers recovered from the top liquid phases (TLPs) exhibited significant peaks at 1710 cm^{-1} , 1243 cm^{-1} and 1093 cm^{-1} , corresponding to the C=O stretching, C-C-O stretching (ester group) and O-C-C stretching (ester group), respectively, of PET fibers [24–27]. On the other hand, the recovered fibers from the bottom liquid phases (BLPs) exhibited lower peak intensities at 1710 cm^{-1} , 1243 cm^{-1} and 1093 cm^{-1} , with significant O-H stretching broad bands at $3100\text{--}3700\text{ cm}^{-1}$. Also, multiple peaks in the region of $1030\text{--}1200\text{ cm}^{-1}$ that correspond to the C–O–C stretching vibration of the pyranose ring [24–27] can be found for all of the recovered fibers from the BLP. This indicates that cotton fibers preferentially segregated to the BLP compared to PET fibers, which is due to the higher wetting tendency of cotton (Fig. 2(a–c)). As the ethanol volume increased, the fiber composition in each phase changes. Of the fibers retrieved from the BLP, the mass percentage of cotton with respect to the total fibers collected from the BLP monotonically declined with ethanol volume, and correspondingly that of PET increased. One of the reasons for the significant presence of PET in the BLP is entanglement with cotton fibers. Specifically, as cotton preferentially segregates to the BLP, the entangled PET fibers are dragged along. As for the fibers from the TLP, the composition of PET increased then decreased with ethanol volume. The non-monotonic behavior of PET suggests that, other than preferential

affinity based on hydrophilicity, other mechanisms are at play.

Two plausible reasons for the effect of ethanol volume on the separation are tied to reduced surface tension and reduced differential density, both of which enhanced sedimentation of the fibers of both types to the BLP. Regarding surface tension, the water surface tension has been reported to reduce from 72.6 mN/m to 29.2 mN/m after 50 vol % of ethanol was added [28]. As for density differential between the two liquid phases, as the ethanol volume increased from 0 to 50 mL, the density of the BLP declined from 997 kg/m^3 to 856 kg/m^3 , thereby reducing the density differential between the TLP and BLP. Consequently, because sedimentation affects both fiber types particularly in view of entanglement, the separation is not only governed by preferential affinity of cotton to the water-rich BLP.

Fig. 3(b) presents the effect of liquid composition on the recovery of each fiber type in the preferred liquid with respect to the total fiber type in the sample to the preferred liquid phase. 100 % represents perfect segregation of all that fiber type in the sample to the preferred liquid phase. As the ethanol volume increased from 0 to 50 mL, the recovery of cotton enhanced while that of PET declined. Specifically, the mass percentage of recovered cotton fibers generally increased from 42.7 % to 81.8 % in the BLP, while that of recovered PET fibers collected from the TLP reduced from 90.0 % to 44.5 %. This indicates that the intermediate composition E-10 performed the best, recovering 84.7 % of cotton and 85.5 % of PET from the

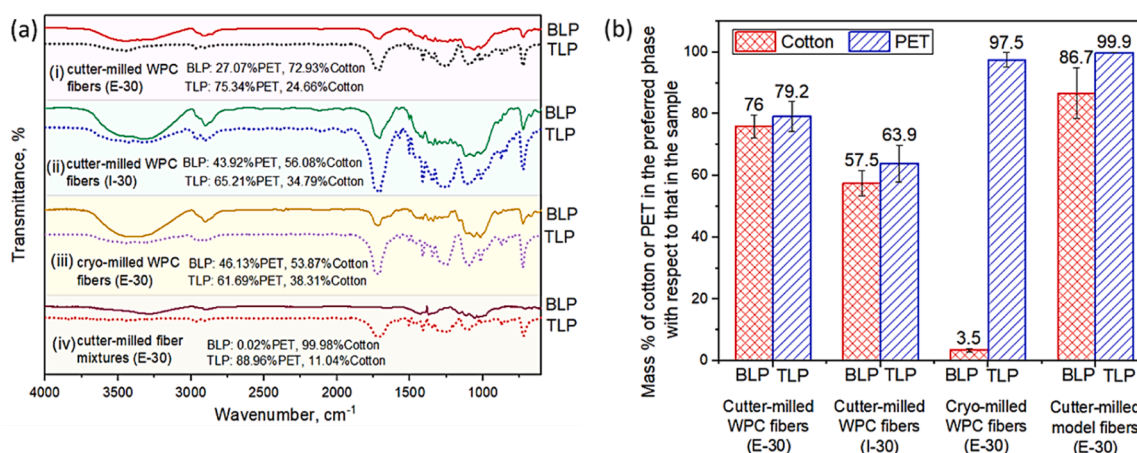


Fig. 6. Effects of ethanol versus IPA and cryo-mill versus cutter-mill: (a) FTIR analysis of recovered fibers extracted from TLP and BLP; the compositions of the fibers in each phase are indicated. (b) Mass percentage of each fiber type in the preferred liquid phase (BLP or TLP) with respect to that in the sample. Note: For the cryo-milled fibers, the data were based on 68 % of total cryo-milled fibers collected from both BLP and TLP, because 32 % of cryo-milled fibers were lost due to their adhesion to the inner walls of the bottle.

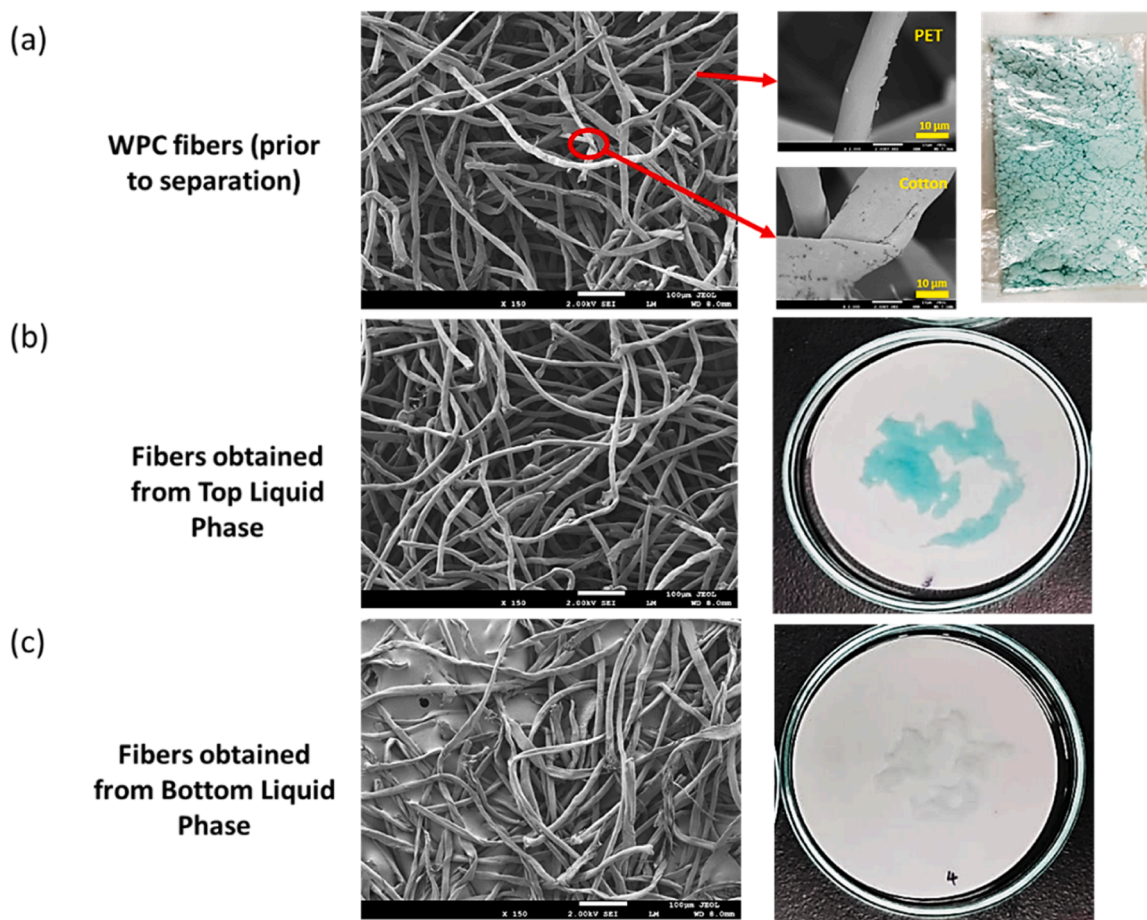


Fig. 7. (a) FESEM images of cutter-milled waste polycotton (WPC) textile fibers, along with zoomed in view of the entanglement and photograph of the milled sample; (b) and (c) FESEM images of fibers obtained from the TLP and BLP, along with the photographs of the recovered fibers on petri dishes. The liquid mixture was E-10.

samples. It is worth noting that PET fibers mostly were retained at the oil-rich TLP, which is attributed to the lower affinity of the PET fibers towards the water-rich BLP [29], and also the hydrophobic interaction between oil-rich TLP and PET fibers [30,31], as illustrated at Fig. 4.

To understand the influence of ethanol on the separation, only water and oil were used to form the two phases. Similar to Fig. 3(a), Fig. 5(b) shows that the mass percentage of cotton with respect to the total fibers in the BLP decreased monotonically with water volume. This is due to the greater affinity of cotton to water (Fig. 2(a)–(c)). As for PET, compared to W-40 and W-60, the PET composition was slightly higher for W-80 in the TLP. More importantly, Fig. 5(b) shows that cotton recovery suffers without ethanol. The highest cotton recovery from the sample was 57.3 % without ethanol (Fig. 5(b)), which is significantly lower than that ranging from 76.0 to 84.7 % in the presence of ethanol (Fig. 3(b)). This demonstrates the importance of ethanol in enhancing cotton separation, which has been attributed to the role of amphiphilic solvents like ethanol in facilitating separation in binary mixtures [32].

3.3. Comparison between IPA and Ethanol

Armed with the results on the beneficial role of ethanol in enhancing cotton separation, another solvent, namely, IPA, was assessed. Instead of ethanol, IPA was added (Table 1). Fig. 6(a) shows better enrichment of cotton in the BLP and PET in the TLP by ethanol compared to IPA, indicating the superior performance of the former. Furthermore, Fig. 6 (b) indicates 76.0 % and 79.2 % recovery of respectively cotton and PET from the samples with ethanol, as opposed to the significantly lower 57.5 % and 63.9 % with IPA. The better separation induced by ethanol

can be tied to the higher polarity index of 5.2, relative to that of 3.9 for IPA [33].

3.4. Comparison between cryo-milled and cutter-milled fibers

Milling of the textile fibers into smaller sizes milling process is a critical step to reduce fiber entanglement and thus enhance the cotton-PET separation. As evidenced in Fig. 2(a–c) and Fig. S2, the cryo-miller resulted in much smaller sizes (i.e., 338, 422 and 284 μm for cotton, PET and polycotton, respectively) compared to the cutter-miller (i.e., 54.8, 139.5 and 72.7 μm for cotton, PET and polycotton, respectively). Unfortunately, due to the much smaller resulting particle sizes post-cryo-milling, only approximately 68 % of the initial fiber mass can be retrieved for analysis, while about 32 % by mass remained adhered to inner wall of the bottle. On the contrary, most of the cutter-milled fibers were retrievable. Comparing the different milling means, Fig. 6(b) shows that the overall recoveries of cotton and PET were better for cryo-milling at respectively 76.0 and 79.2 %. On the other hand, for cryo-milling, the recovery of PET was excellent at 97.5 %, whereas that for cotton was dismal at 3.5 %. The sizes of the milled fibers affect the buoyancy, which has been shown to influence the tendency to segregate to the different liquid phases [34].

3.5. Comparison of waste polycotton (WPC) textile blends versus mixtures of pure cotton and pure PET

The separation of the waste polycotton (WPC) textile blends was compared against that of a mixture of pure cotton waste textile and pure

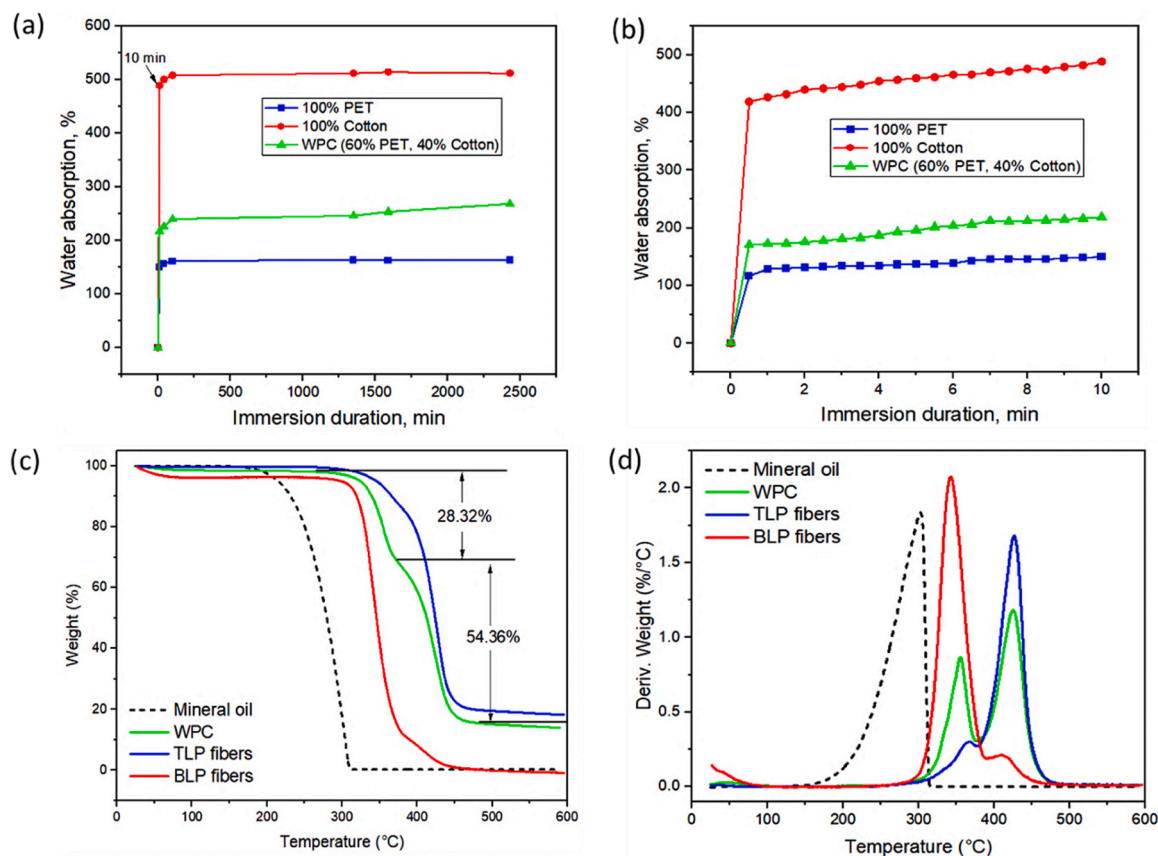


Fig. 8. Water absorption tendency (in terms of percentage change in mass) of different waste textiles (namely, pure cotton, pure PET and WPC (i.e., 40 % cotton and 60 % PET)) for different durations: (a) 0–2500 min, and (b) 0–10 min; Thermogravimetric - Derivative Thermogravimetric analysis (TGA-DTG) curves of the mineral oil and fibers (the TLP and BLP was from E-10 in terms of (c) weight % versus temperature, and (d) derivative weight per °C versus temperature).

PET waste textile at the same composition (i.e., 60 % PET and 40 % cotton). For the fiber mixture, the BLP contained almost pure cotton while the TLP consisted of almost 90 % of PET (Fig. 6(a)), giving recoveries with respect to the initial sample of 86.7 % of cotton from the BLP and 99.9% of PET from the TLP. The outstanding recoveries of the cotton and PET from the mixture compared to that of the blend (i.e., WPC) highlight the entanglement of the cotton-PET fibers in the textile blends. While the interwoven design of cotton and PET in textiles confers multiple benefits like comfort and strength [35], the resulting entanglement is challenging for recovery via this method and thus detrimental for circularity.

4. Discussion

Fig. 7 displays the FESEM images of the cutter-milled WPC fibers prior to liquid-liquid separation, as well as the resulting fibers retrieved from the TLP and BLP using the E-10 mixture. Agreeing with earlier studies on the morphologies of the different fiber types [36–38], Fig. 7 (a) shows that the PET fibers have smooth surfaces, while the cotton fibers are flatter with rougher surfaces. Clearly, the two fiber types are very much intertwined even after the milling step to reduce them to smaller sizes. After liquid-liquid separation (Fig. 7(b) and (c)), the fiber lengths appear intact, and the dominance of PET and cotton in respectively the TLP and BLP is visually discernible in both the FESEM and camera images.

Fig. 8(a) and (b) show the water absorption tendency (in terms of percentage change in mass) of the different waste textiles (namely, pure PET, pure cotton and WPC (i.e., 40 % cotton and 60 % PET)) as functions of time. Clearly, pure cotton exhibited the highest water absorption tendency, reaching more than 400 % within the first minute then

plateauing off to 510 % after 10 minutes till up to 2500 min. In contrast, the water absorption plateaued at 270 % for WPC and 160 % for pure PET. The significantly higher water absorbability of pure cotton is due to its inherent hydrophilic nature due to the -OH functional groups [39], as evident in the FTIR spectrum in Fig. S3(a). This agrees with the lower water contact angle in Fig. 2(d).

Thermogravimetric - Derivative Thermogravimetric (TGA-DTG) analysis was employed to evaluate the thermal stability of the cutter-milled WPC fibers, and the fibers recovered from the TLP and BLP. The TGA curves in Fig. 8(c) indicate slight weight losses for all three fiber samples between 30 °C to 100 °C, which results from the evaporation of the absorbed water [40]. In this temperature range, the greatest weight loss was by the fibers from the BLP, which is predominantly cotton, whose amorphous structure tending to absorb more water [41]. For the WPC fibers, Figs. 8(c) and 8(d) show two major decomposition phases. The first weight loss phase was at about 265 °C to 370 °C, giving an approximate weight loss of 28.3 %, which corresponds to the thermal decomposition of cotton in the WPC. The second weight loss phase continued from 370 °C till 480 °C, resulting in a weight loss of 54.4 %, which is attributed to the thermal decomposition of PET. For the recovered BLP fibers, Fig. 8(d) indicates significant decomposition at 343 °C, with a weight loss of 84 %, indicating cotton pyrolysis [42,43]. Also, a slight decomposition peak appeared at 416 °C, which corresponds to the thermal decomposition of PET. As for the recovered TLP fibers, a higher PET thermal decomposition peak exists at 425 °C (weight loss = 66.8 %), while a smaller cotton decomposition peak appears at 365 °C, which agrees with the higher content of PET in the TLP. The cotton thermal decomposition temperatures of WPC and the recovered BLP fibers were slightly different at 353 °C and 343 °C, respectively. This is due to the higher composition of PET in the former conferring cotton

Table 2
Comparison of fiber separation methods.

Separation method	Polycotton	Chemicals & condition	PET recovered, %	Cellulose recovered, %	refs
Chemical fibers separation					
Cellulose dissolution	30/70 PET-cotton	<ul style="list-style-type: none"> 1-allyl-3-methylimidazolium chloride (AmimCl) and bleach mixture heated to 80 °C for 3 h 	92.0–96.0	55.0–65.0	[10]
Cellulose dissolution	65/35 PET-cotton	<ul style="list-style-type: none"> deep eutectic solvent (DES) consist of zinc chloride, water, and phosphoric acid in molar ratio of 1:3:0.5 mixture stirred at 600 rpm 	Close to 100	-	[44]
Hydrothermal treatment of cellulose	PET-cotton	<ul style="list-style-type: none"> Organic acid catalyst needed mixture heated to 180 °C for 3 h 	> 95.0	≥ 80.0	[45]
Glycolysis of PET	PET-cotton	<ul style="list-style-type: none"> ethylene glycol and zinc acetate mixture heated to 196 °C for 1 h 	86.0	-	[46]
Acidic hydrolysis of cellulose	50/50 PET-cotton	<ul style="list-style-type: none"> acidic aqueous solutions mixture refluxed for 7 h 	-	26.0	[14]
Enzymatic hydrolysis	40/60 PET/cotton	<ul style="list-style-type: none"> mixture containing sodium citrate buffer, 0.5 g/L sodium azide, 30FPU cellulase and 60 IU β-glucosidase per gram of cellulose hydrolyzed at 45 °C, pH 4.8 for 72 h 	51.5	-	[15]
Alkaline pretreat + enzymatic hydrolysis	40/60 PET/cotton	<ul style="list-style-type: none"> sodium hydroxide was used in pretreatment process at –20 °C for 1 h. sample treated with sodium citrate buffer, 0.5 g/L sodium azide, 30FPU cellulase and 60 IU β-glucosidase per gram of cellulose and hydrolyze at 45 °C, pH 4.8 for 72 h 	98.0	-	[15]
Acidic hydrolysis of cellulose	Ratio 1:19 for PET/cotton	<ul style="list-style-type: none"> 8 % H₂SO₄, bath ratio 1:25 at 95 °C for 30 min followed by 10 min grinding time microcrystalline cellulose structure 	-	80.4	[47]
Physical fibers separation					
Liquid-liquid system for waste polycotton textile	60/40 PET-cotton	<ul style="list-style-type: none"> liquid-liquid mixture containing water, mineral oil and ethanol. stirred for 10 min at room temperature and 30 min for phase-separation no chemical reaction 	85.5	84.7	This Study
Liquid-liquid system for mixture of waste cotton and waste PET textiles	60/40 PET-cotton	<ul style="list-style-type: none"> separation occurs in a liquid mixture system containing water, mineral oil and ethanol. stirred for 10 min at room temperature and 30 min for phase-separation no chemical reaction 	99.9	86.7	this Study

with higher thermal stability [42]. Notably, the thermal decomposition peak of the mineral oil used ($T_{\max} = 302$ °C) was not found in the fibers retrieved from either the BLP or TLP, indicating the complete removal of the oil with the acetone rinse. Further FTIR analysis in Fig. S3 confirmed the absence of the mineral oil from the recovered and rinsed fibers. This negates concerns regarding remnant mineral oil persisting in the recovered fibers, though the acetone rinse also poses as chemical waste that has to be treated.

Table 2 compares the cotton and PET recovery efficiency of reported studies vis-à-vis the best-performing separation results in this study from the E-10 liquid mixture. It is worth noting that the fibers recovery process here can be performed in minutes, without requiring heating or a lot of chemicals, which represent significant advantages compared with other findings. Moreover, our method gave comparable fibers recovery efficiency with other findings that are mainly based on chemical means. Nevertheless, a single stage appears insufficient, and staging of a few of such units in series is needed for complete segregation of the two fiber types.

5. Conclusion

In this study, a simple, environmentally friendly (i.e., few chemicals, ambient condition), and rapid separation method was devised to physically separate cotton and PET from waste polycotton textiles. The waste textile had to be milled down into smaller fibers first. Then, three liquids (namely, water, mineral oil, and ethanol or IPA) were mixed to form a two-phase liquid-liquid system, which induced the preferential segregation of cotton to the water-rich bottom liquid phase due to the intrinsic higher hydrophilicity. While oil and water alone would cause two liquid phases, ethanol was pivotal in enhancing the separation and outperformed IPA.

The best-performing mixture (E-10) consisted of 14 vol%, 43 vol%

and 43 vol% of ethanol, DI water and mineral oil, respectively. Regarding the separation performance of the cutter-milled waste polycotton (WPC) textile (i.e., 40 % cotton and 60 % PET), 84.7 % and 85.5 % of the cotton and PET fibers were retrieved from the bottom liquid phase (BLP) and top liquid phase (TLP), respectively. The less than perfect segregation is due to the interwoven nature of the two fiber types (i.e., entanglement issues). When the entanglement issue is circumvented through a prepared mixture of pure cotton and pure PET textile wastes, the separation was significantly improved, with 99.9 % of PET and 86.7 % of cotton recovered. While the interwoven design of cotton and PET in textiles confers benefits like comfort and strength, the resulting entanglement is detrimental for circularity. Although cryomilling can reduce fiber entanglement, the resulting smaller fiber sizes led to inefficient separation compared to the cutter-milled fibers.

Compared to previously reported methods, the fiber recovery efficiencies achieved here are similar. However, this approach offers several clear advantages: it is significantly faster (on the order of minutes), eliminates the need for heating or extensive use of chemicals, and operates under simple ambient conditions.

CRedit authorship contribution statement

Jia Wei Chew: Writing – review & editing, Supervision, Conceptualization, Methodology, Project administration. **Xiao Hu:** Writing – review & editing, Supervision, Project administration. **Yen Nan Liang:** Visualization, Validation. **Ying Siew Khoo:** Writing – original draft, Methodology, Formal analysis, Data curation, Conceptualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence

the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.jece.2024.114281](https://doi.org/10.1016/j.jece.2024.114281).

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