

Material Separation from Polyester/Cotton Blended Fabrics Using Hydrothermal Treatment

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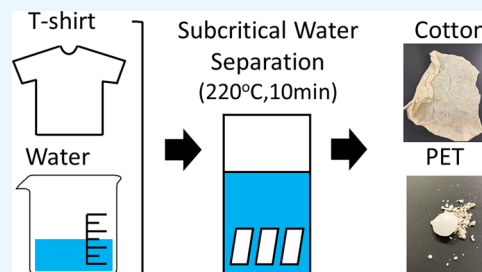
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ABSTRACT: The production of textile products is increasing annually, and most of them are disposed of after use without recycling. One of the reasons for the low recycling percentage of discarded textile products is the difficulty of recycling as a single material as these products are produced from a combination of two or more materials. Therefore, a technology to separate materials is necessary to improve the recycling percentage of textile products and to build a sustainable recycling industry. The aim of this study was to separate the most common combination of materials, such as cotton/polyester, in an environmentally friendly technique using hydrothermal treatment with only water. Herein, the optimal treatment conditions for blended fabrics in a high-pressure reactor were studied. Moreover, cotton could be separated by treating the fabrics at 220 to 230 °C for 10 min while maintaining the shape of the fabrics. Additionally, polyester showed a melting point, confirming that polyester could be separated without decomposition into monomers, unlike common chemical recycling. The strength of the separated cotton and the molecular weight of the polyester were evaluated, and a kinetic analysis of the changes due to the treatment was conducted. The activation energy obtained from the Arrhenius plot was 111.8 kJ/mol for PET, which was smaller than 142.6 kJ/mol for cotton. This indicates that the decrease in the molecular weight of PET is more likely to occur than the change in the strength of cotton, suggesting the possibility of separating the materials from the kinetic analysis.



INTRODUCTION

Textile products are extensively utilized globally, and their production rates have increased annually from 45 million tons per year in 2000 to 109 million tons in 2020. Furthermore, this value is expected to increase continuously, reaching 148 million tons per year by 2030.¹ Textiles, owing to their high production volumes, are disposed of in significant quantities and have been recognized as significant contributors to global solid waste pollution.² In 2020, 92 million tons of textile products were disposed of, and by 2030, this value is expected to exceed 134 million tons per year.³ Notably, this increase can be attributed in part to the global population growth, diversification of fashion, and shortening usage cycles. Approximately 85% of textile products are disposed of by landfilling or incineration after use, whereas 15% are recycled. Most spent textile products are employed in less-valuable applications, including as insulation or stuffing materials, and <1% is recycled into products having equal value to the original material.⁴ In addition to the high monetary costs of the landfilling and incineration processes, textile waste products can cause environmental pollution. Thus, to build a sustainable society, we must recycle these waste products must be recycled efficiently.

One of the reasons for the low recycling percentage of textile products is their multimaterial composition. Textiles are generally fabricated by blending or interweaving two or more

materials. Consequently, they exhibit better performance than products made of a single material. Blended fabrics are produced from yarns combining two or more types of short yarns, whereas interwoven fabrics are manufactured by combining two or more types of yarns. Furthermore, textile products may include buttons, fasteners, and sewing threads. As it is challenging to effectively recycle such complex mixtures, the individual materials must be separated for recycling.

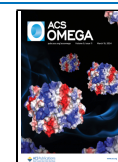
The most common combination on the market is a blend of cotton and polyester.⁵ This blend is employed in many applications in varying proportions. Both materials constitute a large proportion of the total textile products, with cotton and polyester accounting for approximately 24 and 52%, respectively.¹ Cotton is composed of cellulose, a naturally occurring polysaccharide that is employed in a wide range of industries.⁶ Polyester is a petroleum-based chemical that exhibits enhanced tensile strength, chemical resistance, and

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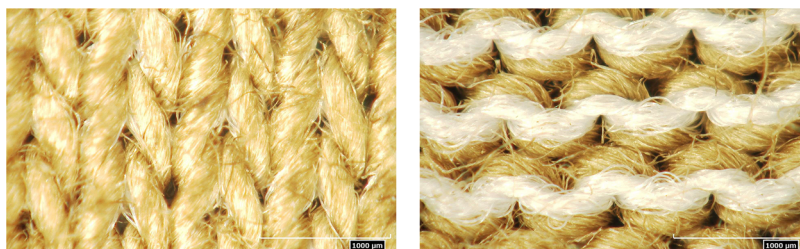


Figure 1. Digital microscopic images of the sample fabric colored by heating in an oven (the cotton yarns appear brown, and the PET yarns appear white).

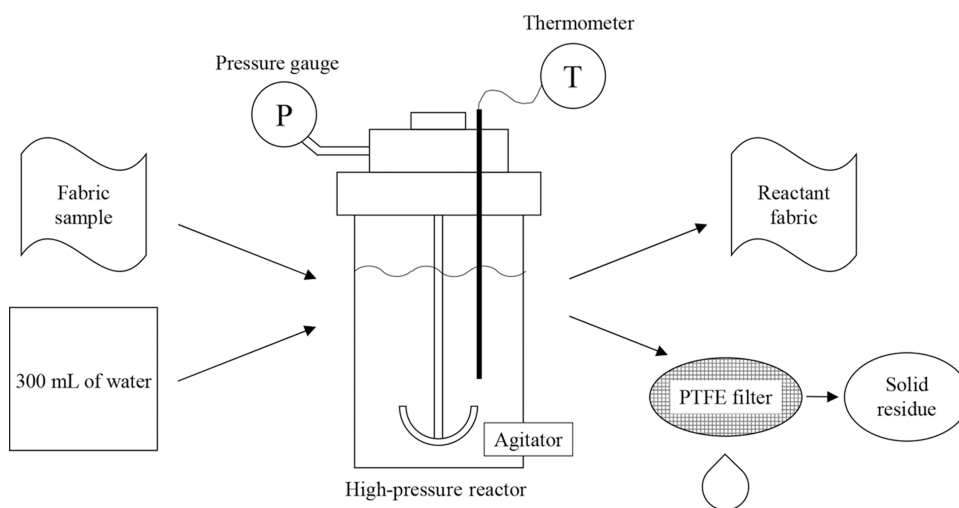


Figure 2. Schematic of the hydrothermal treatment equipment and process.

heat resistance. Notably, both materials contain valuable resources that can be recycled.

The recycling methods for polyester include thermal recycling via incineration, mechanical recycling, and chemical recycling, which breaks down the material into raw monomers.⁷ As mechanical recycling is generally feasible only for single-material wastes, chemical recycling is generally the preferred method. One of the notable technologies for chemical recycling involves the use of sub/supercritical fluids. These fluids enable the easy decomposition of polymers into monomers without a catalyst. Many studies have been conducted on the decomposition of polyesters into the constituent monomer, terephthalic acid (TPA), using sub/supercritical water for chemical recycling.^{8–11} Notably, alcohols are used as supercritical fluids, and many studies have investigated their application in the decomposition of polyesters for chemical recycling.^{12–18}

Although the recycling of single materials is relatively easy, recycling blends is generally challenging. Therefore, it is essential to separate the constituent materials. There are abundant studies on the separation and recycling of cotton and polyester. For example, Ling et al. investigated the separation of polyester/cotton blended fabrics via an environmentally friendly process using phosphotungstic acid.¹⁹ By treating the fabrics under optimal conditions, the cotton could be separated in high yield as microcrystalline cellulose and the polyester as TPA by neutral hydrolysis. Yan et al. studied the hydrolysis mechanisms of cellulose and polyester for their separation from polyester/cotton blended fabrics in subcritical water using the dispersion-corrected density functional theory and noted the preferential hydrolysis of cellulose.²⁰ Hou et al. investigated the

degradation of cotton in blended fabrics after hydrothermal treatment using dilute hydrochloric acid as a catalyst; they observed that polyester was separated and that it retained its fiber properties.²¹ Valh et al. investigated the depolymerization of waste fiber, polyethylene terephthalate (PET), into high-purity TPA via hydrothermal hydrolysis.²² Furthermore, moderate-purity TPA was successfully recovered, and a PET resin was produced via repolymerization. Islam et al. investigated the hydrolysis of PET using a sulfuric acid solution and recovered TPA in 95–98% purity.²³ Wang et al. investigated the separation of cotton/polyester fiber waste via the hydrolysis of cotton with cellulase, an enzyme that can hydrolyze glycosidic bonds, produced by submerged fermentation using cotton/polyester fiber waste as the substrate. This research suggested the possibility of recovering polyester and glucose degraded by cellulase.²⁴ Yousef et al. investigated a chemical recycling process for dissolving polyester and organic materials, such as dyes, from cotton/polyester blend fabrics using dimethyl sulfoxide. This research proposed a sustainable strategy involving extracting the dissolved polyester and regenerating the solvent and other materials.²⁵ Evidently, various technologies have been proposed thus far; however, to separate such mixtures, it is necessary to decompose them into monomers after which they are recycled, often chemically. This process is generally inefficient and complicated.

The aim of this study was attempted to separate polyester in the polymer state from a polyester/cotton blend fabric via hydrothermal treatment using only water. Thereafter, we investigated the optimum treatment conditions for achieving an efficient separation. Finally, the separated materials were

characterized, after which the separation mechanisms and kinetics were discussed.

MATERIALS AND METHODS

Fabric Sample. The utilized fabric samples were white shirts comprising 66% cotton and 34% polyester. Proton nuclear magnetic resonance (H NMR) spectroscopy revealed that polyester was composed of 100% PET and that the diol composition included 97.3 mol % ethylene glycol (EG) and 2.3 mol % diethylene glycol (DEG). These yarns were knitted into a plating stitch, with the cotton stitch facing one side (outside of the shirts) and the polyester facing the other side (inside) of the knitted fabrics. Figure 1 shows a digital microscopic image of the sample (for clarity, the cotton yarn was colored by heating it to 200 °C in an oven).

Hydrothermal Treatment. The hydrothermal treatment was performed in a high-pressure reactor (series MMJ-500, OM Lab-Tech Co., Ltd.). The sample fabric was cut into approximately 5 × 5 cm² pieces, whose weight was about 400–500 mg, and placed in the reactor containing 300 mL of pure water. Thereafter, the fabric was heated to the desired temperature (180 °C–250 °C) and treated for 10–180 min. The heating lasted for 30–50 min depending on the set temperature. Afterward, the reactor was cooled to 40 °C naturally. The reactant fabric was removed, and the solid residue in the water was recovered by filtration using a PTFE membrane filter. Subsequently, the reactant fabric and solid residue were oven-dried (Figure 2).

MEASUREMENT

Microscopic Image. The dried reactant fabric was observed under a digital microscope (RH-2000, HIRX Co., Ltd.) and SEM (VE-8800, KEYENCE Co. Ltd.). The images observed from both sides were employed to visually evaluate the feasibility of separation.

Identification of Residues. The residues after treatment were identified by FT-IR/ATR spectral analysis (Cary 660, Agilent Technologies).

Separation Efficiency. The efficient separation of cotton/polyester is crucial for the recycling process. Thus, the separation efficiency was evaluated based on the proportion of PET in the reactant fabric sample, calculated from the H NMR spectra obtained by using AVANCE-NEO 600 (Bruker).

A solution of the weighed fabric sample in a solvent (d-chloroform/hexafluoro isopropanol-d = 90/10 vol %) containing 2 mg of dimethyl isophthalate (DMI) as a standard sample for quantification was used as the measurement sample. The integrals of each peak was indicative of the polyester components (EG, DEG, and TPA), water, and DMI in the sample were determined from the H NMR spectra. From eq 1, the relative amount of substance $n_{\text{rel.}}[X]$ was calculated from the integral value I_X and the number of protons H_X of the corresponding peak, and from eq 2, the relative weight $W_{\text{rel.}}[X]$ was calculated from $n_{\text{rel.}}$ and molecular weight M_X .

$$I_X/H_X = n_{\text{rel.}}[X] \quad (1)$$

$$n_{\text{rel.}}[X] \times M_X = W_{\text{rel.}}[X] \quad (2)$$

Regarding PET, the sum of the $W_{\text{rel.}}$ of each component is represented by $W_{\text{rel.}}[\text{PET}]$. The weights of PET in the sample ($W[\text{PET}]$) and water ($W[\text{water}]$) were determined from the ratio of $W_{\text{rel.}}[\text{DMI}]$ to the actual weight $W[\text{DMI}]$

$$W_{\text{rel.}} \times W[\text{DMI}]/W_{\text{rel.}}[\text{DMI}] = W(\text{mg}) \quad (3)$$

The percentage of PET in the reactant fabric sample (X_e) was calculated from the weight of the fabric sample calculated from $W[\text{PET}]$ and the difference between the weights of the sample (weighed at the first) and water

$$X_e(\%) = W[\text{PET}]/(W[\text{sample}] - W[\text{water}]) \times 100 \quad (4)$$

Characterization of PET. The separated solid residue or yarns unraveled from the reactant fabric were used as samples for the PET evaluation. The thermal property of PET was evaluated via differential scanning calorimetry (DSC) (DSC214 Polyma, NETZSCH). Approximately 5 mg of the sample was measured in the dry state from 25 to 280 °C, at a heating rate of 10 °C/min using an aluminum pan. To evaluate the thermal properties during hydrothermal treatment, DSC measurements were performed with water. Employing a pressure-resistant pan, ~2 mg of water was added to 7 mg of the fabric sample. The measurement conditions were the same as those employed in the dry state.

The molecular weight of PET was measured via gel permeation chromatography (TOSOH CORPORATION). For the measurement solution, 2 mg of the sample was dissolved in 2 mL of hexafluoro isopropanol, after which 2 mL of chloroform was added. The main peak of the measurement data was analyzed to obtain the number-average molecular weight (M_n). The value of M_n divided by that of the sample before treatment was used as the molecular weight retention rate, which served as a degradation index for PET.

Characterization of Cotton. The yarns unraveled from the reactant fabric were used as samples for characterizing cotton. The tensile strength of the cotton yarns was evaluated using a Technograph TG-200NB (MinebeaMitsumi Inc.). The yarn with a length of 5 mm was tensile at a rate of 10 mm/min. The test force at break was determined, and the ratio of the post-treatment to pretreatment values was calculated as the strength retention using the index of cotton-yarn degradation.

RESULTS AND DISCUSSION

Separation Treatment. The effects of varying treatment temperatures were compared. Figure 3 shows the microscopic

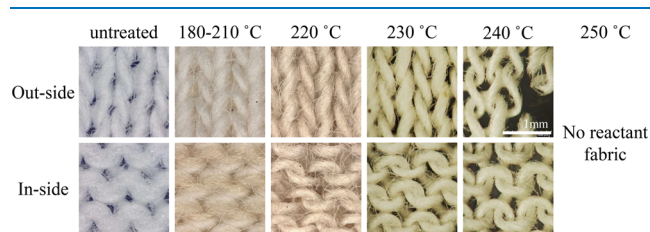


Figure 3. Digital microscopy images of the reactant fabrics. The PET yarns were not observed in the samples treated at temperatures over 220 °C.

images of the reactant fabrics after 10 min of treatment at 180–250 °C. The cotton and PET yarns were present as they were before the treatment at temperatures up to 210 °C. However, at 220 °C, small, colored pieces entangled in the fabric sample were observed instead of the PET yarns. At 230 °C, the mesh of the cotton yarns remained, whereas no PET yarns could be observed. The surface conditions of the mesh fabric after treatment were compared with those of raw

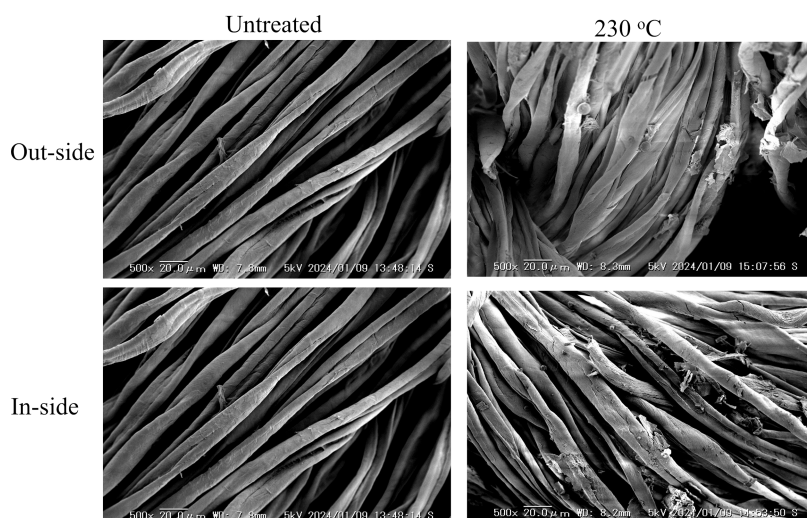


Figure 4. SEM images of the mesh fabric after treatment at 230 °C and raw material ($\times 500$).

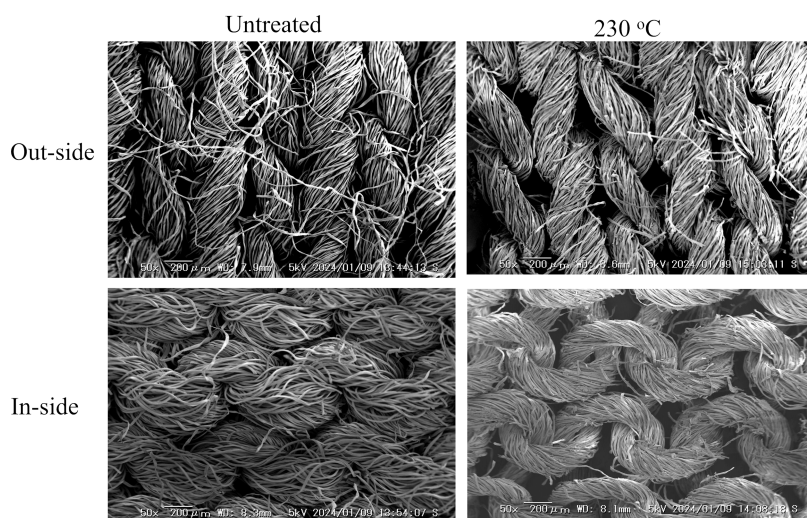


Figure 5. SEM images of the mesh fabric after treatment at 230 °C and raw material ($\times 50$).

material using SEM images, as shown in Figures 4 and 5. SEM images show that the cotton fiber state is maintained after treatment. However, it can also be seen that the fibers are slightly damaged after treatment at 230 °C. It is suggested that the subcritical water treatment has progressively degraded the cotton fibers. The microscopic image of residues is also shown in Figure 6. The small pieces and powdery solid collected after treatment were confirmed to be PET and the mesh fabrics were confirmed to be cotton by FT-IR/ATR analysis (see Figure 7). These results confirm that the subcritical water treatment separates nonfibrous solids and that the solid residue is PET by FT-IR/ATR spectra. Furthermore, the IR analysis results of the fabric after treatment suggest that most of the PET can be desorbed and separated. At 240 °C, part of the knitted cotton yarns collapsed. At 250 °C, no reactant fabric sample remained, and the solid residue recovered from the water after the treatment exhibited many short fibers. The solid residue from the 200 °C treatment, with no change in the appearance of the fabric, was 3 mg, whereas from the 230 °C treatment, when no PET yarns were observed, around 50 mg of solid residue could be recovered.



Figure 6. Microscopic image of residues obtained after treatment at 230 °C ($\times 50$).

Figure 8 shows the percentage of PET in the reactant fabric sample (X_e) as a function of the treatment temperature. At temperatures up to 210 °C, X_e was almost unchanged from the state before treatment, i.e., $\sim 34\%$. However, at 220 °C, the X_e value was approximately halved and close to 0% at higher

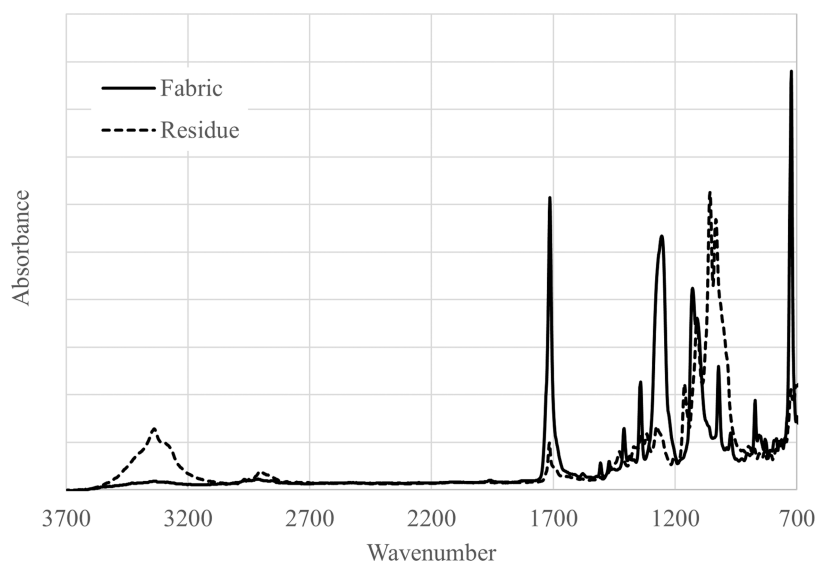


Figure 7. FT-IR/ATR spectra of the solid residues and mesh fabric after treatment at 230 °C.

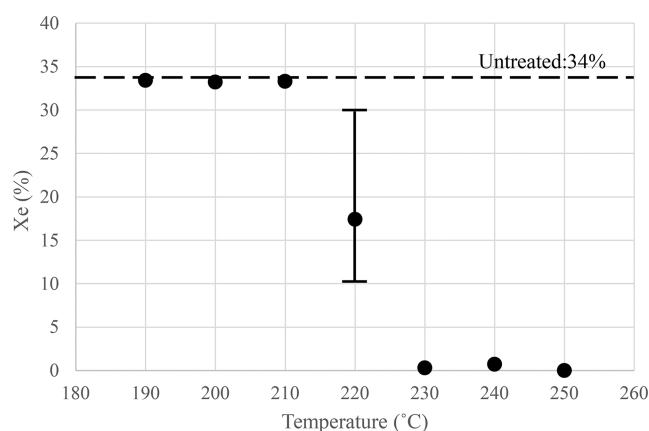


Figure 8. Percentage of PET in the fabric sample (X_e).

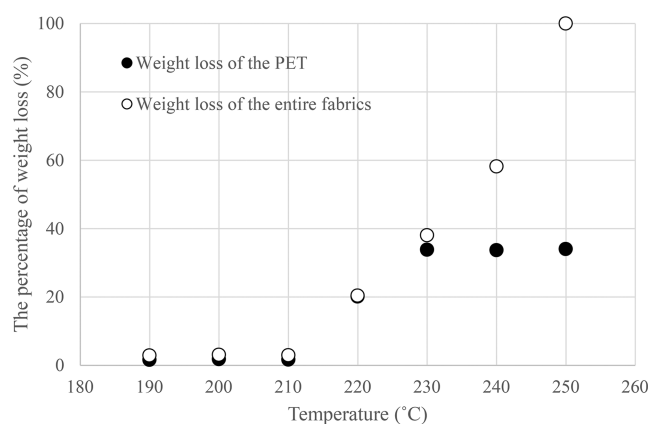


Figure 9. Percentage weight losses of PET and the entire fabric samples.

temperatures. The small pieces entangled in the sample that was treated at 220 °C were derived from PET and exhibited a wide range of X_e values, depending on the cleaning process after the treatment.

The weight loss of the reactant fabric sample was measured before and after the treatment. Using eqs 5 and 6, the percentage weight loss of the entire fabric sample $X_{\text{loss}}[\text{fabric}]$ and that attributed to PET $X_{\text{loss}}[\text{PET}]$ were calculated, respectively

$$X_{\text{loss}}[\text{fabric}] = (W_{\text{before}} - W_{\text{after}})/W_{\text{before}} \times 100 \quad (5)$$

$$X_{\text{loss}}[\text{PET}] = (W_{\text{before}} \times 0.34 - W_{\text{after}} \times X_e/100)/W_{\text{before}} \quad (6)$$

where W_{before} and W_{after} are the weights before and after the treatment, respectively. Figure 9 shows the percentage weight losses attributable to the entire fabric sample and PET against the treatment temperature. As the PET content before treatment was 34% of the sample, the maximum weight loss attributable to PET was 34% of the sample. There was almost no discrepancy between the weight losses of the sample and PET up to 230 °C. However, at 240 °C and above, the weight loss of the entire sample was larger than that of PET, indicating that the weight loss of the cotton was also reduced from the

sample. This result is consistent with the observation that a part of the knitted cotton yarns collapsed at ≥ 240 °C.

According to the above results, the most efficient separation could be achieved at 230 °C for 10 min.

The PET-derived solid residue was evaluated. Figure 10 shows the DSC heat flow of the untreated fabric sample and the solid residue treated at 220 °C for 10 min. The untreated sample displayed endothermic peaks around 75 and 255 °C. The peak around 75 °C was attributed to cotton, whereas the peak at 255 °C indicated the melting point (T_m) of PET. The endothermic peak was also observed in the result of the solid residue. This indicates that the solid residue was derived from PET and was not decomposed into monomers TPA, EG, and DEG because of its T_m . Thus, polyester and cotton were efficiently separated by hydrothermal treatment at 230 °C for 10 min. The cotton retained its cloth shape, and PET could be recovered without decomposing into monomers.

Next, the separation mechanism of PET from the blended fabric was discussed. To schematically reproduce the behavior during the hydrothermal treatment, the thermal properties of PET when heated with water were evaluated. Figure 11 shows the measured DSC heat flow for PET only and PET with water in a pressure-resistant pan. Without water, PET exhibited a T_m of 255 °C. However, with water, it exhibited a T_m of 228 °C,

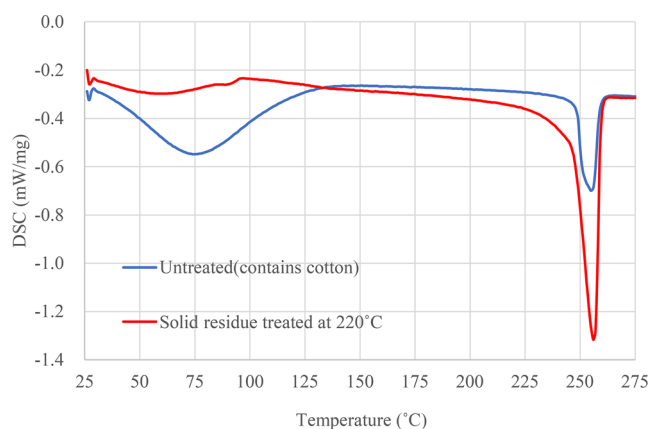


Figure 10. DSC heat flow of the untreated sample (containing cotton) and solid residue at 220 °C.

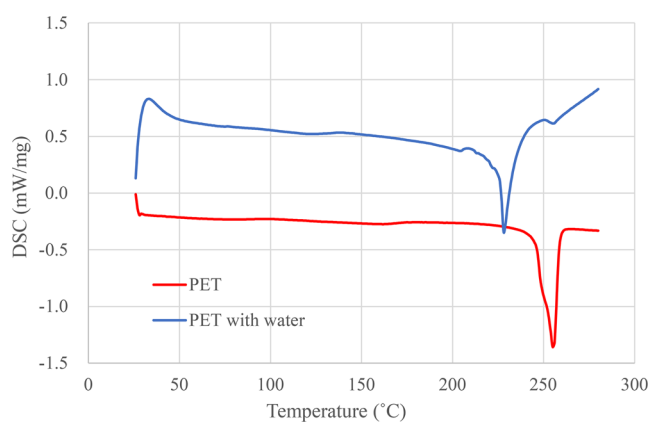


Figure 11. DSC heat flow of PET only and PET with water. PET with water exhibited a comparatively low melting point.

which was nearly 30 °C lower. The T_m of PET with water was almost equal to the temperature range at which PET could be separated from the blended fabric sample.

The lower T_m with water and the fact that PET was not decomposed into monomers suggested that the melting of PET via hydrothermal treatment contributed to the improved separation efficiency.

Kinetic Analysis. Kinetic analyses were carried out to investigate the changes in the properties of the cotton and PET obtained after the treatment. The samples were treated at 180, 200, and 230 °C around the T_m with water, for 10–180 min, and the retention of the molecular weight of PET and the strength of the cotton yarns were evaluated. Table 1 presents the results. Both retention rates decreased with time, and the molecular weight of PET changed more rapidly than the strength of cotton.

The degradation-rate constant of PET was calculated using Mn, as shown in eq 7:²⁶

$$kt = 1/Mn_t - 1/Mn_0 \quad (7)$$

where Mn_0 is Mn before the treatment, and Mn_t is Mn after the treatment for t min. According to eq 7, the inverse of Mn ($1/Mn$) was plotted against time, as shown in. Linear approximation was applied using the inverse of the molecular weight before treatment, Figure 12 as the intercept, and the rate constant (k) at each temperature was obtained from the slope. The results at 180 and 230 °C plotted linearly against

Table 1. Retention Rates of Mn and Strength

temperature (°C)	time (min)	retention rate of Mn (%)	retention rate of strength (%)
180	10	80.4	87.0
	60	23.7	114.5
	120	15.7	71.3
	180	11.3	27.8
200	10	35.2	94.1
	30	8.9	69.0
	60	3.8	4.8
	120	4.4	0.8
230	10	1.6	5.1
	15	1.4	2.3
	20	1.4	1.3
	60	collapse	collapse

time following the equation. On the other hand, the coefficient of determination R^2 for 200 °C treatment was 0.7, which was slightly lower. At 200 °C, the small change in molecular weight at 60 and 120 min suggests that the decrease in molecular weight is saturated by 60 min at that temperature. Therefore, the plot at 120 min was considered to deviate from a linear approximation, resulting in a lower R^2 .

The degradation-rate constant of cotton was calculated using the tensile-strength retention of the cotton yarn, as expressed in eq 8:²⁷

$$\log(SR_0/SR_t) = kt \quad (8)$$

where SR_0 and SR_t are the tensile-strength retentions before (100%) and after treatment for t min. According to the equation, the ordinary logarithm of the strength retention versus time was plotted (Figure 13). The plot was linear at each temperature, as expressed in the equation. A linear approximation was applied using 0 as the intercept, and the rate constant (k) at each temperature was obtained from the slope. The degradation-rate constant was confirmed to be high at high temperatures.

An Arrhenius plot of the natural logarithm of the rate constant (k) obtained from each equation plotted against the inverse of the temperature is shown in Figure 14. The k values of both PET and cotton changed with the temperature according to the Arrhenius equation. The difference in k value between the materials was not significant because of the difference in the equations employed to calculate k .

Although the degradation-rate constants of PET and cotton could be calculated individually, they could not be simply compared by using their numeric values because of the different formulas and indices used. To compare the rates of the two materials, the equation for calculating the strength retention of the cotton yarn (eq 8) was also applied to calculate the retention of the molecular weight of PET and calculated as k for the change in the retention rate. The calculated k values and coefficients of determination of the change at each temperature R^2 are presented in Table 2, and the Arrhenius plot using this value is shown in Figure 15. Comparing the activation energies obtained from the slope of this plot, the value of cotton is larger, indicating that the decrease in the strength retention of the cotton yarn was less likely to occur than the decrease in the molecular-weight retention of PET.

Therefore, the decrease in the strength of the cotton yarn can be controlled while the molecular weight of PET decreases,

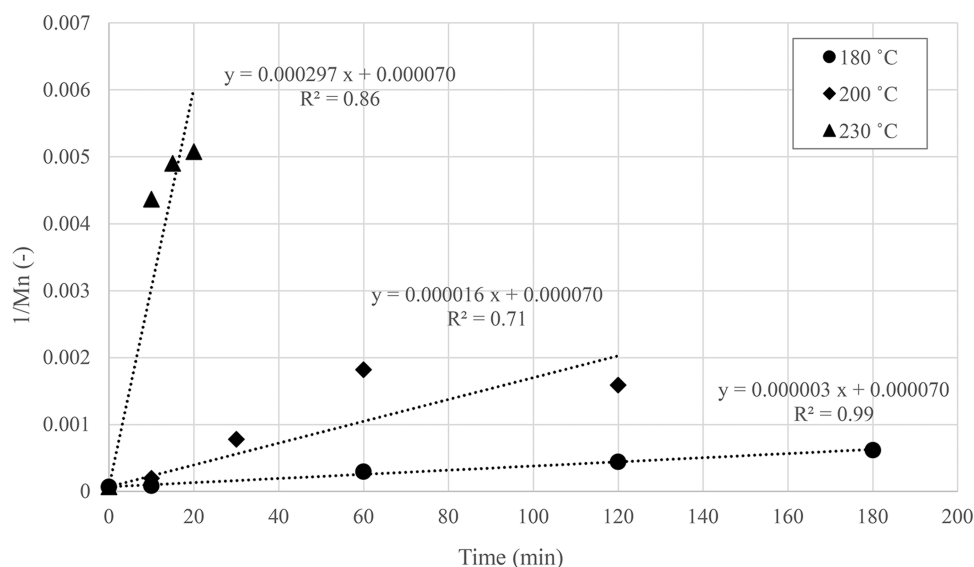


Figure 12. Inverse Mn as a function of the treatment time.

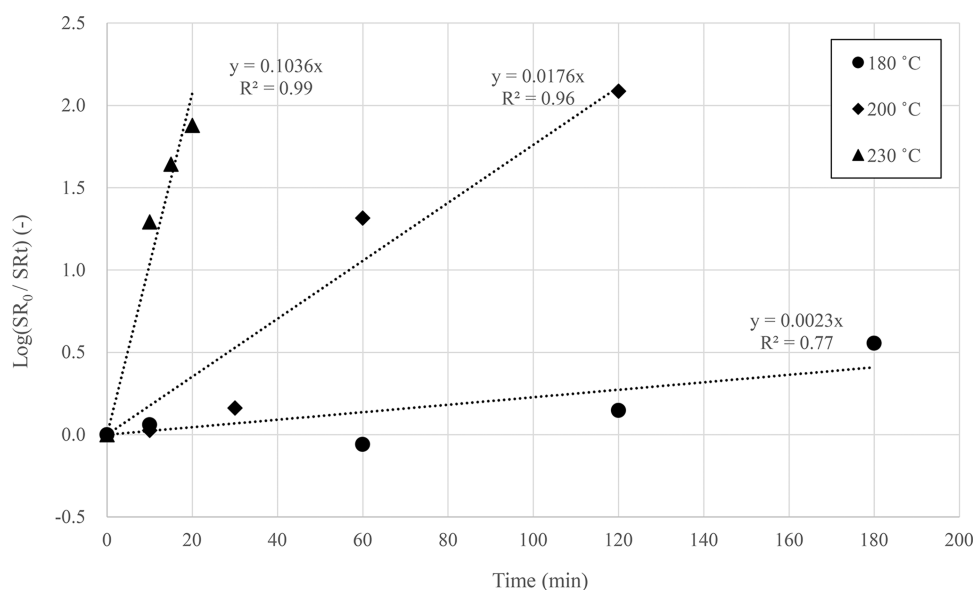


Figure 13. Ordinary logarithm of strength retention as a function of time.

and PET could be separated from the cotton yarn, retaining the same state as the fabric. Additionally, by applying treatment under optimal conditions, the separation was facilitated by the decrease in the molecular weight and melting due to the lowered T_m in water. Consequently, PET was not degraded into the constituent monomers and could be separated while maintaining its polymer characteristics. Unlike chemical recycling in which PET is decomposed into TPA and utilized or repolymerized, the separation method employed here facilitates efficient recycling because fewer processes than that for monomers are required for the polymerization. Moreover, unlike conventional technologies, cotton could be separated while maintaining the shape of the fabric. Thus, the post-treatment separation is facile, and the recovered cotton can be expected to be used in a wide range of applications because the fiber length is sufficient to maintain the shape of the yarn.

CONCLUSIONS

In this study, the application of hydrothermal treatment with only water to separate a cotton/polyester blend fabric, a common fabric product composed of multiple materials, was investigated. The study's objective was to achieve the efficient separation of each material rather than implement chemical recycling wherein the material is decomposed into monomers. The optimum conditions for the separation were obtained by varying the temperature, and the results revealed that almost all of the PET content was removed from the fabric sample by heat treatment at 230 °C for 10 min, whereas the cotton remained in the cloth state.

After treatment, the recovered PET melted at the same temperature as the PET before treatment, confirming that it did not decompose into the monomers. As the T_m of PET in the presence of water is approximately 30 °C lower than that in air, the melting of PET during the treatment was considered as the factor that promoted the efficient separation.

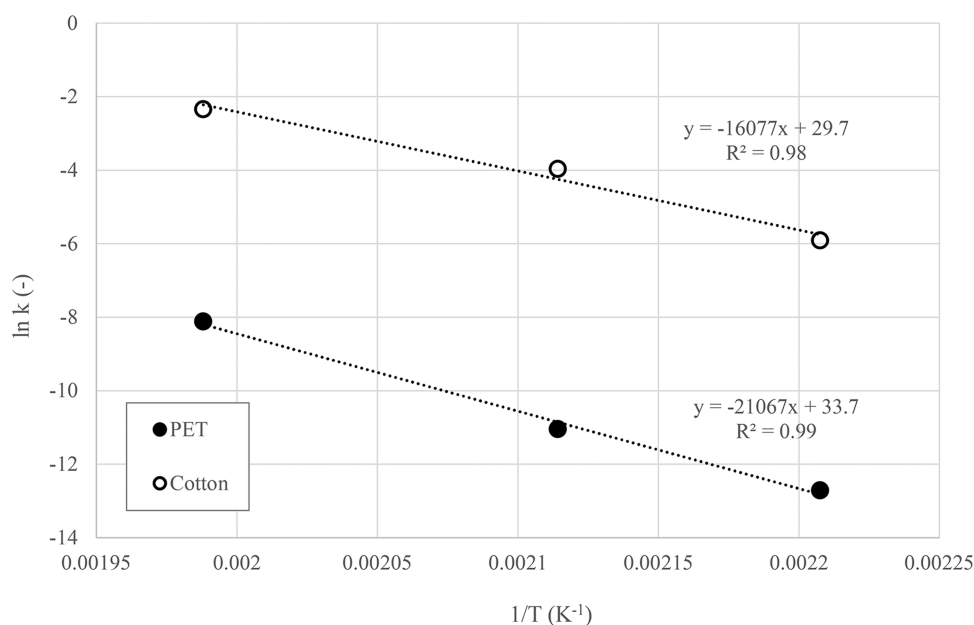


Figure 14. Arrhenius plot of the degradation-rate constants of PET and cotton.

Table 2. Rate Constant and Coefficient of Determination for the Change in the Retention Rate

temperature (°C)	PET		cotton	
	k	R^2	k	R^2
180	0.006	0.95	0.002	0.77
200	0.015	0.82	0.018	0.96
230	0.114	0.94	0.104	0.99

Kinetic analysis was carried out based on the retention rates of the molecular weight of PET and the strength of cotton yarns in samples treated for different periods at a constant temperature. For each case, the rate constant (k) was demonstrated to depend on the temperature. To compare the rate of change between PET and the cotton materials due to the treatment, we calculated the rate of decrease in the molecular weight or strength retention was calculated. The k

value for the change in the retention was larger for PET at each temperature, and the activation energy calculated from k was smaller, compared with the values for cotton. This indicated that a decrease in the molecular weight of PET was more likely to occur than a decrease in the strength of the cotton yarn. Consequently, the materials were successfully separated, with cotton retaining its fabric state. Furthermore, when treated under the optimized conditions, PET retained its polymer characteristics. The recovered PET could be efficiently converted into a high-molecular-weight material by fewer processes than those required in chemical recycling, wherein PET decomposes into monomers. As cotton retains its fabric state, it is easily separated from PET, and because of its sufficient fiber length, which enables it to retain its yarn state, it can be reused in a wide range of applications. Thus, a novel, environmentally friendly, and efficient recycling method using

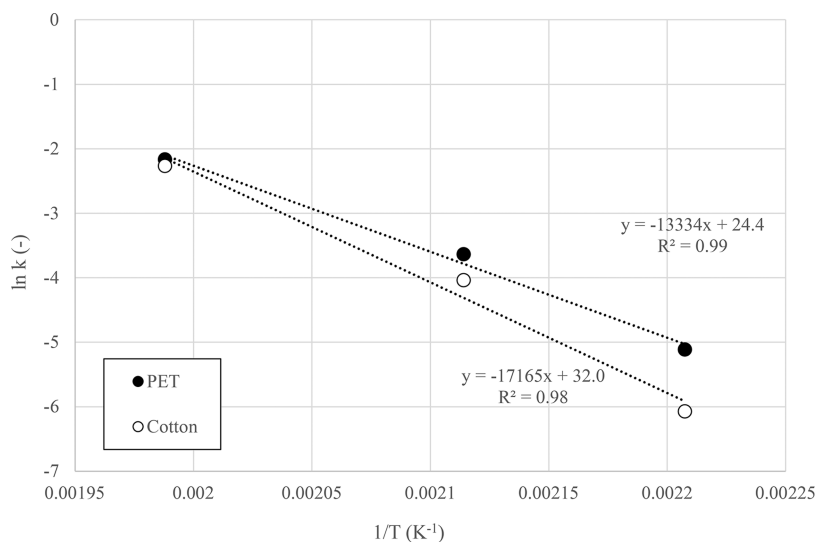


Figure 15. Arrhenius plot of (k) for the change in the retention rate.

only water was developed, and its effectiveness was demonstrated.

A major future challenge is to search for conditions that are less degrading than these reports, to investigate the effects of colored fabrics, continuous processing system design, and how to mass produce.

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Author Contributions

M.S. contributed to the management, leadership, and coordination of the research activity planning and execution, as well as the reviewing and editing of the original draft, specifically with the critical review, commentary, and revision. R.Y. and N.T. contributed to the research and investigation by specifically performing experiments and data collection. M.M. and J.I. contributed to the preparation and writing of the original draft as well as to the product analysis. K.I. contributed to the management and the reviewing and editing of the revised manuscript, specifically with the critical review and commentary.

Notes

The authors declare no competing financial interest.

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