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Investigation of Combined Alkaline Pretreatment and Mechanoenzymatic Processing for Improved Valorization of Post-Consumer Textiles

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ABSTRACT

The continuous increase in textile waste, coupled with insufficient recycling and reuse strategies, has led to a growing issue of underutilized raw materials. Mixed textiles account for approximately 50% of global textile waste and commonly consist of tightly woven cotton blended with synthetic fibers, which complicated material separation and recovery. This study presents an integrated approach that combines mechanochemical pretreatment with subsequent mechanoenzymatic processing to valorize such complex waste streams. Using sodium hydroxide during milling enables higher solids loadings and supports efficient processing under moderate conditions. The mechanochemical approach yields glucose recoveries of up to 80.57% after 24 h. Furthermore, coupling mechanochemical and mechanoenzymatic treatment increases glucose yield by approximately 30% within 6 h, compared with conventional treatment strategies. This combined approach offers a scalable and sustainable pathway for converting mixed textile waste into valuable products. The process facilitates the selective recovery of polyethylene terephthalate (PET) while preserving its essential properties, enabling its reuse in subsequent applications or incorporation into established recycling pathways. Beyond the recovery of cellulose as glucose, this technology may enable the selective extraction of PET fibers, providing the basis for more efficient biotechnological processes and promoting enhanced circularity in PET fiber recycling.

1 | Introduction

The conversion of textile waste into valuable raw materials is part of a broader strategy by the European Union [1–3]. This strategy is fundamental in promoting resource efficiency and in minimizing the environmental consequences of waste production [2, 4]. Global textile production has increased significantly in recent years, mainly due to shorter product lifespans [5] and the faster pace of design and production in the fashion industry [6]. In 2020, approximately 109 million tons of textiles were produced globally [6], and around 124 million tons by 2023 [7]. This growth has led to a substantial rise in textile waste, which is predominantly landfilled or incinerated [5, 8, 9]. In light of the associated ecological challenges, the development of innovative and environmentally friendly recycling technologies is becoming increasingly urgent to enable sustainable waste utilization and conserve resources [10].

Globally, less than 1% of textile waste is currently recycled [7, 10, 11], primarily due to the complex composition of many textile products [11]. Polycotton (PC) fabrics, made from polyester and cotton, are especially challenging because the fibers are tightly bonded and difficult to separate [12]. This structural complexity hinders both mechanical and chemical recycling processes. Around 50 wt% of textile waste worldwide consists of PC blends, including cotton mixed with polyethylene terephthalate (PET) [5, 8, 13]. Nevertheless, given their high availability and limited recyclability, cotton-PET blended textiles represent a promising feedstock for the development of alternative pathways for glucose production in the chemical industry [14].

The enzymatic degradation of cellulose has attracted increasing scientific interest in recent years, mainly due to the high substrate specificity of cellulases [11] and their ability to function under mild reaction conditions [9]. One of the main challenges in enzymatic hydrolysis of cotton is its high crystallinity, which restricts enzyme accessibility. Consequently, an effective pretreatment is required to enable efficient enzymatic conversion [8].

A widely used approach to increase enzyme accessibility is alkaline treatment with sodium hydroxide [15], commonly referred as mercerization [16]. This treatment causes swelling of the crystalline regions and enhances enzymatic degradability [14]. It induces a structural transformation of native cellulose into alkali cellulose and subsequently into cellulose. The latter exhibits reduced crystallinity and increased reactivity toward cellulases, thereby significantly improving enzymatic degradability [9].

Mechanoenzymatic treatment has shown promise for processing both end-of-life (EOL) and pure cotton textiles [17]. In practice, textile waste is rarely homogeneous, as fibers are often blended to enhance durability [12]. Due to the differing chemical properties of cotton and polyester, enzymatic hydrolysis selectively degrades cotton while leaving PET mainly unaffected, enabling theoretical separation of both components [5, 11, 18]. The primary aim of this study is to establish a scalable process that enables the selective separation of cotton from PC blends in a controlled and targeted manner. Subsequently, the next step involves recovering pure components, enabling PET to be reintegrated into downstream processes [14] or repolymerized within established recycling workflows [19].

To enable such a selective and scalable separation process, this study extends previous work on mechanical pretreatment [20] and mechanoenzymatic systems [17] toward mechanochemical processing strategies. In this context, mechanical processing in a wet rotor mill is constrained by process-related factors, including rotor–stator gap width and solids loading, while further improvements in glucose conversion may remain limited beyond a certain energy input [20]. For this purpose, sodium hydroxide is integrated into the milling process to enhance substrate disintegration and thereby improve conversion efficiency and product yields. This approach may additionally facilitate the separation of polyester fractions, thereby contributing to the valorization of mixed textile waste streams.

To explore the applicability of these processes, two approaches were investigated. The first combined sodium hydroxide pretreatment with mechanical milling, aiming to identify optimal milling parameters such as solids loading, energy input, and rotor–stator gap width. Based on the optimized pretreatment conditions, the second approach applied mechanoenzymatic hydrolysis to white cotton (WC), PC, and EOL cotton textiles. For clarity and consistency, the term dyed cotton waste is used throughout this study to describe (EOL) cotton textiles.

This investigation focuses on mechanochemical and mechanoenzymatic processing methods. For the pretreatment step, a sodium hydroxide concentration was selected based on previous literature indicating that such conditions do induce minor alterations in PET [21, 22]. Consequently, a comparatively moderate concentration of sodium hydroxide (8 wt%) at ambient temperature was chosen, despite previous reports indicating that these conditions do not result in complete cellulose conversion of approximately 43.7% after 24 h [15].

2 | Results and Discussion

The following sections present data indicating that the application of mechanochemical pretreatment and subsequent mechanoenzymatic processing of textile substrates significantly enhances the efficiency of enzymatic hydrolysis. To assess the performance of the processing approaches, a range of analytical methods was employed. These included near-infrared (NIR) spectroscopy, evaluation of crystallinity, analysis of particle size distribution, rheological testing, and measurement of glucose concentrations resulting from enzymatic hydrolysis. Detailed information on wet rotor mill operation is reported in the Supporting Information and in the prior publication [20].

3 | Determination of Digestible Cellulose Fraction

NIR spectroscopy revealed a cellulose content of 94 wt% in both cotton textile waste and WC, consistent with findings reported in previous study [20]. For the PC, the composition was determined to be 61 wt% cotton and 39 wt% polyester, with a standard deviation of 3 wt%.

To support accurate mass balance estimations during enzymatic hydrolysis and to enable consistent comparison of process outcomes, reference cellulose contents were defined for the substrate materials. For cotton textile waste and WC, a cellulose content of 94 wt% was used. For the PC fraction, a cellulose content of 60 wt% was applied. These reference values were employed to calculate the glucose yields reported in the following sections.

4 | Influence of Milling and Enzymatic Hydrolysis on Particle Size and Physical Properties

To develop a sustainable pretreatment strategy using sodium hydroxide (T1–T4), several process approaches were evaluated (see the Methods section Alkali treatment and Table 1) to identify the most effective one. Sodium hydroxide was either added directly during milling (combined treatment) or applied after (separated treatment). The milling experiments were conducted at room temperature to ensure comparability between samples. Nevertheless, localized temperature increases due to high energy input cannot be excluded. The process options were tested under

TABLE 1 | Experimental process configurations of mechanochechemical treatment (A, B).

Process option	Energy input, kWh/t	Solids load, wt%	Rotor–stator gap widths, μm	Trial time, h
A	40, 55, 85, 135, 200	5	350	24
B	40, 55, 85, 135, 200	5, 7.5	100, 350	24

varying solids loads and rotor–stator gap widths. Subsequently, samples were collected at defined energy input levels, washed, dried, and subjected to enzymatic hydrolysis under standardized laboratory conditions, maintaining consistent solids load, temperature, and agitation frequency. The resulting glucose yields after 24 h, as influenced by the applied energy input during suspension processing, are presented in Figure 1.

Previous studies have shown that even low energy input during wet rotor milling significantly enhances glucose yield [20, 23]. The desired surface roughening and particle size reduction induced by the milling process are evidenced by the increased glucose yield observed when sodium hydroxide treatment is applied. Without alkaline treatment, a technical limitation was observed at a gap width of 100 μm with 2.5% solids loading and at 350 μm with 5% solids loading. When sodium hydroxide was applied, these limits were extended to 5% at 100 μm and to 7.5% at 350 μm. Previous studies have reported that higher solids loadings combined with smaller gap widths generate greater shear forces, thereby improving comminution efficiency [24]. Comparable trends were observed in the present study under alkaline treatment. When sodium hydroxide was applied after the milling process (T2), the glucose yield increased from 59.5% (no NaOH, 350 μm, 5 wt%, 200 kWh/t) [20] to 71.59% under identical milling conditions. The treatment combining mechanical milling with sodium hydroxide application (T1), resulted in a slighly higher glucose yield of 75.75%. However, increasing the solids load proved to be even more advantageous. A maximum glucose yield of 80.57% (T3) was achieved at 7.5% solids loading and 350 μm gap width.

The data show that glucose yield increases with increasing energy input. However, as described in the literature [20], a plateau forms after a certain energy input. This indicates that higher energy inputs do not necessarily lead to improved enzymatic digestibility. This effect may be due to reduced crystallinity. While it improves hydrolysis at first, it eventually restricts further breakdown. Alkaline treatment improves cotton fiber accessibility and promotes enzymatic hydrolysis [5]. However, further improvement is strongly dependent on milling efficiency.

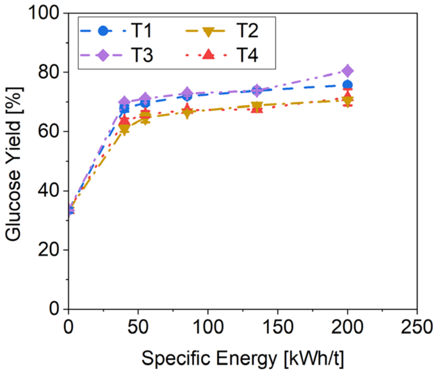

FIGURE 1 | Influence of mechanochemical pretreatment on enzymatic hydrolysis of dyed cotton waste (25 FPU/g, 2.5 wt%, 24 h) under different mechanical treatment conditions: T1, combined treatment (350 μm, 5 wt%); T2, separate treatment (350 μm, 5 wt%); T3, combined treatment (350 μm, 7.5 wt%); T4, combined treatment (100 μm, 5 wt%). 8 wt% sodium hydroxide concentration for the liquid phase was applied in all treatments.

In contrast to previously reported methods employing higher sodium hydroxide concentrations, glucose yields of 56.67% were obtained under conditions of 15 wt% NaOH at 121°C following 24 h of enzymatic hydrolysis [25]. Under ambient conditions with 20% (w/v) NaOH, a maximum yield of 77.4% was observed, which remained unchanged even at temperatures of 100°C [15]. Prior studies have demonstrated that optimal pretreatment conditions for cotton and cotton-blend textiles involve the use of 7% (w/v) NaOH and 12% (w/v) urea at −20°C [5, 21, 22, 25], resulting in glucose yields of up to 85% after 24 h of enzymatic hydrolysis [22]. The glucose yield of 80.57% (T3) obtained in the present study approaches these values, despite being achieved under significantly milder chemical conditions.

Although the combination of a 350 μm gap width and 7.5% solids loading during mechanochemical treatment of dyed cotton waste yielded the highest glucose yield, the operational parameters were maintained at 350 μm and 5% solids loading. This was done to prevent issues that occur when working close to technical limits at higher loadings. For further processing, an energy input of 55 kWh/t was selected. At this level, the yield curve reached a plateau where minor improvements occur, ensuring process efficiency.

5 | Influence of Mechanoenzymatic Processing

After selecting the optimal process conditions (55 kWh/t, 5 wt%, 350 μm, combined treatment), the study focused on evaluating the influence of mechanoenzymatic processing. Results can be seen in Figure 2. Two distinct process configurations involving dyed cotton waste, PC, and WC were evaluated over a 6-h experimental period. The results demonstrate that mechanoenzymatic treatment under low energy input conditions (RA 2) consistently yields the highest glucose recovery, independent of the textile composition. The maximum glucose yields achieved after 6 h were 43.91% for WC, 47.51% for dyed cotton waste, and 56.86% for PC. As sodium hydroxide/urea pretreatment of cotton has been identified as one of the most effective methods for enhancing enzymatic hydrolysis efficiency [5], these results were compared with literature data. Reported glucose yields for this pretreatment are approximately 25% after 8 h [26] and after 6 h [27] indicating that the present findings represent an improvement of roughly 30% PC. Enzyme concentrations were calculated based on the total mass of textile material. Given that PC contains

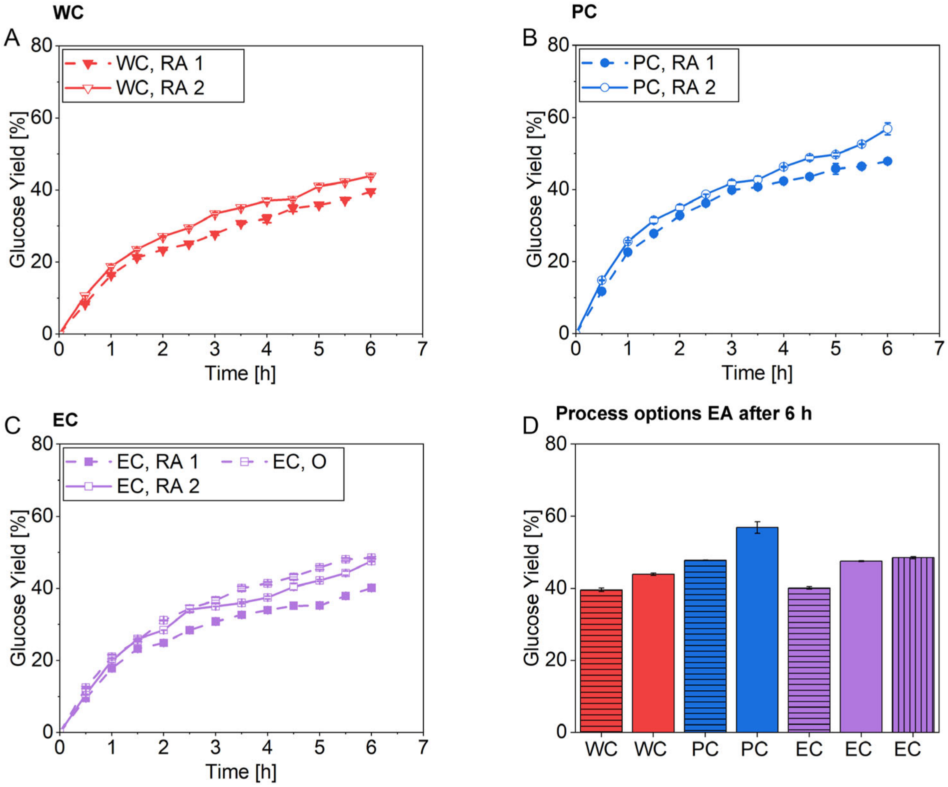

FIGURE 2 | Effect of mechanochemical pretreatment (T1, 55 kWh/t) and subsequent mechanoenzymatic processing (25 and 40 FPU/g), with 40 FPU/g indicated by marker “O” in plot C and by vertical lines in plot D, at 5 wt%, on the conversion of cellulose into glucose in textile fiber suspensions. Two RAging protocols were applied: RA1 (horizontal lines) and RA2 (solid fill with vertical lines). Analyses were performed on three textile waste types: A WC, B PC, and C dyed cotton waste (EC).

60% cotton, a higher effective cellulase concentration was applied to the cotton component, which may explain the increased glucose yield. However, this interpretation contrasts with findings reported in the literature [22], which observed no significant improvement in glucose yield when the enzyme loading was increased to 40 FPU/g textile. To examine this effect under comparable conditions, the same enzyme concentration was applied to EOL textile. The results are shown in Figure 2C for the EC curve and are indicated by the marker “OW.” Increasing the enzyme loading to 40 FPU/g resulted in a glucose yield of 48.52%, indicating only a marginal improvement compared to 47.51%. In contrast, the yields obtained from PC were notably higher and could not be matched by dyed cotton waste. One plausible explanation is the mechanical impact of milling, particularly its role in promoting liquefaction. This phenomenon predominantly affects the cellulose fraction, resulting in a marked reduction in viscosity (see Figure 6) [28]. Given that PC contains approximately 40% PET, this effect remains moderate, whereas an increased solids loading appears to exert a beneficial influence on comminution. During enzymatic hydrolysis, cotton fibers may remain physically entangled with polyester fibers. This structural interlocking [5] can expose the cotton component to increased mechanical stress during processing, particularly under conditions of high solids loading [24]. This can lead to increased surface area, reduced crystallinity, and enhanced enzymatic accessibility [29]. This hypothesis is supported by scanning electron microscopy (SEM) (see Figure 4) and particle size distribution analysis (see Figure 3), both of which reveal significant structural changes. The milling process promotes continuous mixing of textile material and enzymes, which is essential for efficient bioconversion [30]. Fourier-transform infrared (FTIR) spectroscopy further confirms a substantial reduction in cellulose content following treatment (see Figure 5).

Further results provide additional insight into the role of dyes and their impact on enzymatic hydrolysis. Additionally, investigation showed that sodium hydroxide pretreatment followed by a washing step resulted in comparable glucose yields for WC and dyed cotton waste under both RAging conditions. This finding suggests that pretreatment has likely enhanced enzymatic degradability by removing or reducing dyes and other residual contaminants, thereby improving substrate accessibility and enabling more uniform hydrolysis across different textile types. An alternative explanation could be the mechanochemical treatment, which may have increased fiber accessibility for both materials in the same way. This hypothesis appears particularly plausible, as similarly high yields were observed for EOL material subjected to the same pretreatment conditions. However, the exact causes cannot be definitively determined from the present data.

6 | Influence of Milling and Enzymatic Hydrolysis on Particle Size and Physical Properties

Accurate fiber size determination remains challenging. Microscopic imaging offers resolution and flexibility but is resource-intensive [31–33]. Conventional fiber analyzers cover

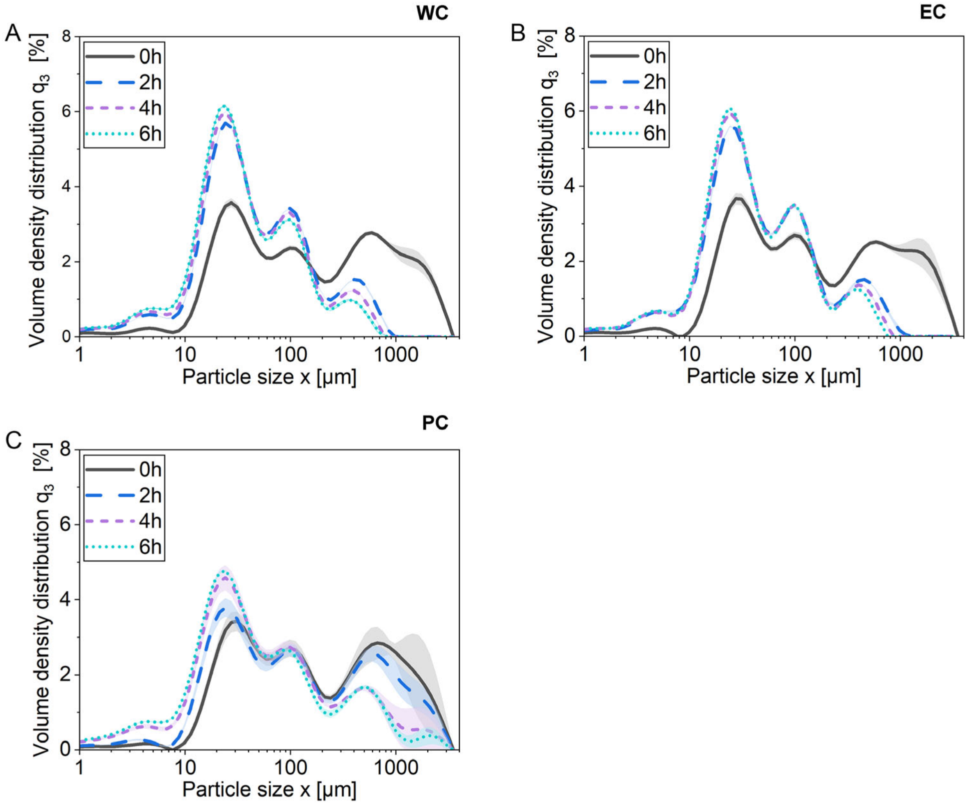

FIGURE 3 | Influence of mechanochemical pretreatment followed by mechanoenzymatic processing (25 FPU/g, 5 wt%, RA2) at different time intervals (0, 2, 4, 6 h) on particle size distribution. Three distinct cotton-based textile waste materials were evaluated: A WC, B dyed cotton waste (EC), and C PC. The table below the graph compares particle size distributions at 0 and 6 h of treatment, based on $x_{50,3}$ (µm) and $x_{90,3}$ (µm) values.

0.2–5 mm, requiring alternative methods for finer ranges [34]. Laser diffraction was therefore selected for its speed and broad applicability. Due to inherent variability [35], results were interpreted relatively, though trends are consistent with complementary data.

The impact of sequential mechanochemical and mechanoenzymatic treatments on particle size and associated physical properties was systematically investigated. The results are presented in Figure 3 and Table 2. Three textile types, including dyed cotton waste, WC, and PC, were subjected to identical processing protocols (see Figure 7C) and subsequently evaluated based on their particle size distributions. Particle size analysis was performed during mechanoenzymatic hydrolysis in order to isolate the effects of mechochemical pretreatment. The parameter $x_{90,3}$ represents the particle size below which 90% of the total particle volume is present, whereas $x_{50,3}$ corresponds to the volume-based median particle diameter. A distinct decrease in particle size after 6 h of mechanoenzymatic processing is presented in Table 2. The median particle sizes ($x_{50,3}$) were measured at 31.91 µm for WC, 35.14 µm for dyed cotton waste, and 38.90 µm for PC, indicating a comparable level of fragmentation across the different fiber types. A similar trend is evident in the $x_{90,3}$ values. Initially, the PC exhibited an $x_{90,3}$ of 1372.67 µm, which decreased to 506.74 µm following enzymatic degradation. Despite this reduction, the final particle size remains approximately threefold higher than that of WC and dyed cotton waste, which exhibited $x_{90,3}$ values of 160.74 and 164.57 µm, respectively.

For WC and EC, the coarse fraction decreases significantly after 2 h, while PC exhibits a comparable reduction only after 4 h (see Figure 3). This delay is likely due to the composite structure of PC, which requires more time for the disintegration of agglomerates and associated fiber fractions. The reduction in the coarse fraction is achieved through both enzymatic hydrolysis and mechanical refining [36, 37]. Literature indicates that increased refining intensity promotes continuous particle size reduction [29]. As processing time increases, the energy input into the material rises, and this effect is further enhanced by enzymatic

TABLE 2 | Influence of mechanochemical pretreatment followed by mechanoenzymatic processing on characteristic particle sizes ($x_{50,3}$ and $x_{90,3}$) after 0 and 6 h.

	A: WC		**B: EC**		**C: PC**	
	$x_{50,3}$, µm	**$x_{90,3}$, µm**	**$x_{50,3}$, µm**	**$x_{90,3}$, µm**	**$x_{50,3}$, µm**	**$x_{90,3}$, µm**
0 h	137.54	1455.29	132.40	1527.32	144.86	1372.67
6 h	31.91	160.74	35.14	164.57	38.90	506.74

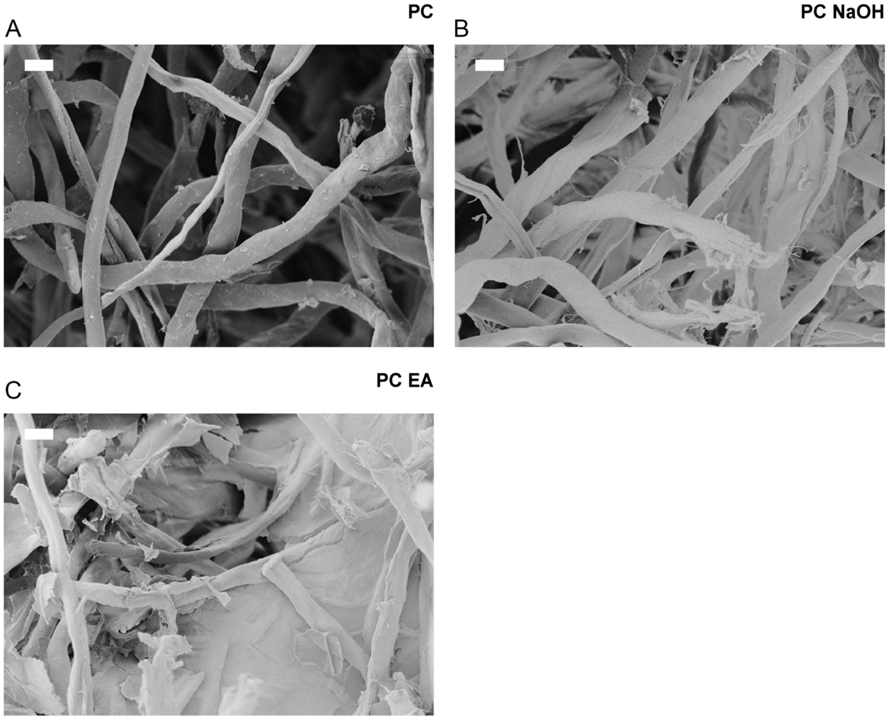

FIGURE 4 | Influence of mechanochemical pretreatment (combined treatment 5%, 350 µm) and mechanoenzymatic processing (25 FPU/g, 5 wt%, RA2) on morphology of PC. SEM images are presented as follows: A crude textile (PC), B NaOH treatment (NaOH), C mechanoenzymatic processing (EA). Scale bar: 20 µm.

hydrolysis. Across all materials, a consistent increase in the fine fraction is observed.

In PC, mechanical forces contribute to the fragmentation of both PET and cotton fibers. However, the synergistic effect of enzymatic hydrolysis and mechanical energy applies exclusively to the cotton fraction. As a result, enzymatic hydrolysis exerts a comparatively minor effect on PC relative to pure cotton fractions. In comparison to the other two materials, the increase in the fine fraction is less pronounced. This suggests that enzymatic hydrolysis may be limited by the presence of nondegradable cellulose and PET components.

Focusing on the PC, only minor differences are observed between the untreated fibers and those subjected to sodium hydroxide treatment, indicating limited structural alteration at this stage. With enzymatic hydrolysis, SEM images (see Figure 4) show partial detachment of individual fiber layers, with loosely connected fibrillar structures emerging on the surface. Furthermore, clear signs of delamination and fibrillation are visible [38], indicating progressive degradation of the fiber matrix. Since the image was taken after 6 h of enzymatic hydrolysis, it is likely that the defibrillated fibers are mainly cotton. In contrast, intact fibers were also observed, which are most likely attributable to the PET component of the blend, given their undamaged morphology and resistance to enzymatic degradation [22]. The morphological alterations observed through SEM analysis align with the trends identified in particle size measurements and are further supported by complementary characterization techniques.

FTIR spectroscopy was additionally used to assess the purity of the polyester fibers and to monitor the crystallinity reduction content during processing. This technique allows for a more detailed analysis of cellulose decomposition over time [14]. Figure 5 presents FTIR spectra, whereas Table 3 provides crystallinity data for PC samples in three conditions: untreated, sodium hydroxide–treated, and after 6 h of enzymatic processing. Details on the calculation of the adsorption ratio are described in the Methods section.

A noticeable decline in cellulose-specific peaks (3350, 3000–2800, 1020 cm^{-1}) [14] is observed, while PET-related signals (1700, 1250, 1200, 1015, 720 cm^{-1}) [14] become more prominent. These findings suggest that cellulose undergoes partial degradation, whereas the polyester component remains structurally intact and can be clearly identified through its characteristic absorption bands. Furthermore, the ratios of characteristic

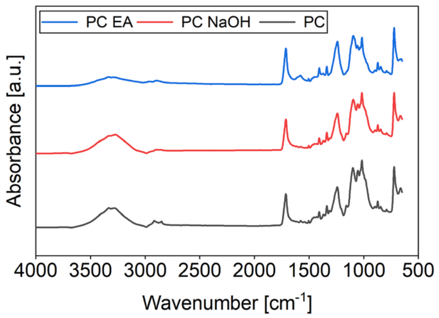

FIGURE 5 | FTIR spectra from PC are shown at three processing stages. PC, crude textile (untreated); PC NaOH, after mechanochemical treatment; PC EA, after mechanoenzymatic processing (25 FPU/g, 5 wt%, RA2).

TABLE 3 | Adsorption ratios from polycotton are shown at three processing stages.

Sample	A_{1340}/A_{1370}	A_{1471}/A_{1453}	A_{848}/A_{898}	A_{972}/A_{1043}
PC EA	1.46 ± 0.03	0.89 ± 0.02	0.96 ± 0.02	0.65 ± 0.04
PC NaOH	1.62 ± 0.02	0.91 ± 0.01	1.03 ± 0.02	0.82 ± 0.01
PC	1.63 ± 0.07	0.88 ± 0.02	0.99 ± 0.04	0.77 ± 0.05

Note: PC, crude textile (untreated); PC NaOH, after mechanochemical treatment; PC EA, after mechanoenzymatic processing.

adsorption bands of PET were determined using the distinct gauche/trans conformational pairs.

Sodium hydroxide treatment resulted in no detectable reduction in PET crystallinity. In contrast, enzymatic hydrolysis led to a decrease in the gauche/trans ratio, evident across all calculated values. This indicates a moderate reduction in trans conformations in enzymatically treated PC compared to untreated material, suggesting a potential decrease in crystallinity [21, 39]. Overall, the applied process configuration appears to exert only a minimal influence on the crystallinity of PET fibers.

The rheological properties of textile suspensions in conjunction with enzymatic hydrolysis were investigated to assess the influence of material composition on glucose production. Figure 6 illustrates the reduction in viscosity observed across different process configurations involving textile fiber suspensions.

A pronounced liquefaction of the suspension was observed in the conducted experiments, consistent with previously reported behavior for corn stover pellets [28] and other lignocellulosic substrates [40]. This process is accompanied by substantial alterations in the rheological properties of the suspension [40, 41].

A distinct reduction in viscosity was observed within the first hour of processing, thereby improving enzymatic accessibility. For both WC and dyed cotton waste, viscosity decreased from approximately 3000 to 500 mPa s. A comparable trend was observed in PC, where viscosity stabilized at around 500 mPa s after 1 h. Given that cellulases predominantly hydrolyze the amorphous cellulose domains located at the fiber surface [40], the persistently elevated viscosity can be attributed largely to the remaining crystalline cellulose fractions as well as the PET component within the blended substrate. This behavior may be particularly advantageous for PC, as the tightly interwoven structure of the two fiber types facilitates more effective transmission of shear forces to the cotton fraction. The increased viscosity improves the transmission of shear forces throughout the suspension [24]. As a result, enzymatic hydrolysis proceeds more efficiently than in pure cotton, potentially explaining the higher glucose yields. That fiber size reduction becomes apparent only after 4 h of processing does not conflict with these findings. Viscosity decreases are generally known to occur prior to the initiation of saccharification [42].

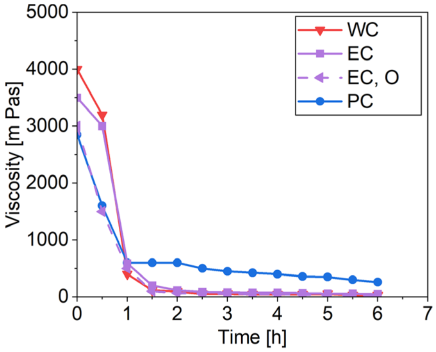

FIGURE 6 | Effect of mechanochemical pretreatment (combined treatment 5%, 350 μm) and subsequent mechanoenzymatic processing (25 and 40 FPU/g (O) at 5 wt%) on the rheological properties of textile fiber suspensions. One RAging protocol was applied: RA2. The analysis was performed on three textile waste types: WC, dyed cotton waste (EC), and PC.

7 | Conclusion

This study demonstrates that tailored pretreatment strategies can significantly enhance the enzymatic hydrolysis of PC blends, facilitating the recovery of PET component and glucose. The most effective approach combined sodium hydroxide treatment with mechanical processing, achieving glucose yields of up to 80.60% after 24 h of enzymatic hydrolysis for dyed cotton waste and enabling milling at a solids load of 7.5 wt%.

Mechanochemical treatment followed by a mechanoenzymatic treatment with low energy input achieved the highest glucose yields, around 43.91% for WC, 47.51% for dyed cotton waste, and 56.86% for PC after 6 h of enzymatic hydrolysis. PET fibers remained largely unaffected by mechanochemical treatment, as confirmed by FTIR analysis; however, a minor reduction in PET in crystallinity was observed after enzymatic hydrolysis. Future work will concentrate on developing a scalable process for the controlled and selective separation of PET from PC blends, achieved through degradation of the cotton component followed by employing density-based separation techniques. To ensure reliable process control, the analytical framework will be expanded to include differential scanning calorimetry (DSC) and molecular weight analysis. The subsequent goal is to utilize the recovered PET in downstream applications or repolymerize it within established recycling systems. The glucose released can be further utilized in fermentation processes, holding potential for integrated biotechnological applications. Given that approximately 56.86% of the cellulose is degraded within the first 6 h, the necessity of extending the reaction time to 24 h should be critically evaluated in future studies, especially with regard to process efficiency and resource consumption.

8 | Experimental Section

Detailed information on all materials and experimental procedures is provided in the Supporting Information (Appendix: Experimental Details).

8.1 | Materials

Two representative textile blend samples were sourced from ModaRe (Caritas, Madrid, Spain). For comparative purposes, a third substrate consisting of commercially available WC (ATM Handel and Service GmbH, Winsen/Luhe, Germany) was included in the study. Before mechanical pretreatment, the material was manually cut into pieces of uniform size to ensure consistency during experimental procedures. The cellulose content of all textile samples was verified using NIR spectroscopy.

8.2 | Methods

8.2.1 | Mechanical Pretreatment

8.2.1.1 | Dry Shredding. Initial size reduction of the textile materials was performed using a Pallmann PS 3 ½ (Pallmann, Zweibrücken, Germany) and a Wanner C17.26s (Wanner Technik, Wertheim, Germany) cutting mill. The desired fiber size was achieved by employing a 2-mm Conidur sieve (Hein, Lehmann GmbH, Krefeld, Germany).

8.2.1.2 | Wet Milling. Further mechanical fiber processing was performed using a Fryma MZ 80 wet rotor mill (FrymaKoruma, Rheinfelden, Germany). A stirrer was installed in the feed tank of the mill, operating at a controlled speed of 150 rpm, regulated via the IKA Eurostar 60 control system (IKA, Staufen im Breisgau, Germany). The gap width was set to 350 µm, and solids concentrations of 5 wt% were investigated. Temperature regulation of both the feed tank and the feed pipe was ensured using a MAGIO MS BC 4 circulation thermostat (Julabo, Seelbach (Schütter), Baden-Württemberg, Germany).

8.2.1.3 | Alkali Treatment. In chemical treatment step, 250 g of each textile fraction was initially soaked in 3990 g of water for 30 min. Subsequently, 760 g of a 50 wt% sodium hydroxide solution (AnalytiChem, Oberhausen, Germany) was added, yielding a final NaOH concentration of 8 wt% in the liquid phase. Samples were taken during milling at specific energy inputs of 40, 55, 85, 135, and 200 kWh/t. Each experimental condition was evaluated at solids concentrations of 5.0 and 7.5 wt% and rotor–stator gap widths of 350 and 100 µm. The 2-h exposure to sodium hydroxide began concurrently with the start of the milling process. All steps were carried out at ambient temperature. Additionally, an alternative process configuration was investigated in which milling was performed prior to the addition of sodium hydroxide. In this setup, the textile material was first mechanically processed, followed by the introduction of NaOH and a soaking period of 2 h under ambient conditions. Following chemical treatment, the fibers were thoroughly rinsed with distilled water until a neutral pH of 7 was reached. The washed samples were pressed to remove excess water and then stored in a refrigerator to keep them stable.

8.2.2 | Process Configuration

Process configurations are illustrated in Figure 7, and process parameters are summarized in Tables 1 and 4. To investigate the effects of mechanochemical and mechanoenzymatic treatment, a range of processing conditions was applied to dyed cotton waste, PC, and WC. Initial trials evaluated mechanochemical treatment with different parameters in combination with sodium hydroxide and milling using separated treatment (see Figure 7A) and combined treatment (Figure 7B) for dyed cotton waste. Best suitable pretreatment (Figure 7B, 5 wt%, 350 µm, 55 kWh/t) was selected for subsequent mechanoenzymatic treatment (Figure 7C). The experiment started by adding enzymes and proceeded according to an RAging protocol with two cycle types: one involving 1 min of milling followed by 29 min of resting (RA 1, see Figure 7C), and another comprising 0.16 min of milling followed by 29.84 min of resting (RA 2, see Figure 7C).

8.2.3 | Analysis of Product Properties

8.2.3.1 | Crystallinity Measurement. FTIR spectroscopy was employed to characterize gauche and trans conformations within the cotton polyester matrix [43]. Spectral measurements were conducted in triplicates for each sample, and the resulting data were averaged to ensure accuracy. To investigate structural variations between crystalline and amorphous regions of PET, specific gauche/trans band pairs were analyzed, as these reflect the relative abundance of amorphous (gauche) versus crystalline (trans) conformations [39, 43, 44]. Quantification of this ratio was achieved by calculating intensity ratios of selected absorption bands, such as 1471 cm^{-1}/1466 cm^{-1}, corresponding to the bending vibration of the glycol group [45]. Additional ratios utilized include 1043 cm^{-1}/972 cm^{-1} for gauche/trans-related C–O stretching vibrations of the glycol unit [43], 898 cm^{-1}/848 cm^{-1} for CH_2 rocking modes [45], and 1340 cm^{-1}/1370 cm^{-1} for CH_2 wagging vibrations associated with the glycol group [43].

8.2.3.2 | Particle Size Distribution Analysis. The particle size distribution of the textile suspensions was determined using laser diffraction analysis, performed with the wet dispersion module of the Mastersizer 3000 (Malvern Panalytical, UK). Measurements were conducted in at least three replicates to ensure statistical reliability. To prevent sedimentation or phase separation within the aqueous dispersant, continuous stirring at 2000 rpm was applied throughout the measurement process. Sample concentration was adjusted based on the obscuration parameter, which was maintained within the optimal range of 5%–20%. Particle size distribution was calculated using the Mastersizer 3000 software employing Fraunhofer scattering theory.

8.2.3.3 | Viscosity Measurements. Viscosity measurements of textile suspensions were performed at 30-min intervals over a total duration of 6 h during enzymatic hydrolysis. A rotational viscometer (HAAKE Viscotester 3, Thermo Fisher Scientific, Karlsruhe, Germany) was utilized to quantify shear stress under deformation at a constant shear rate of 62.5 min^{-1}. Measurements were conducted at ambient temperature (21°C) using measurement cup no. 3 (internal diameter,

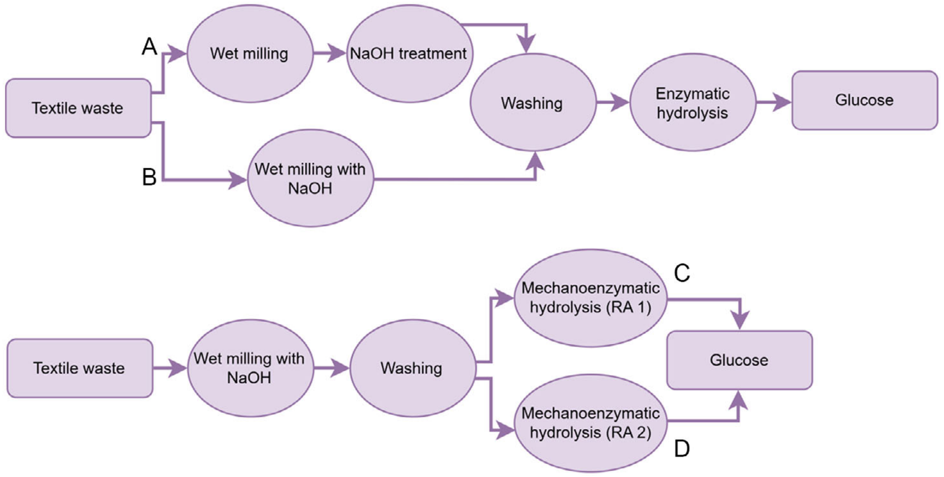

FIGURE 7 | Schematic workflow of the mechanochemical (A,B) and mechanoenzymatic treatment (C,D) procedures.

TABLE 4 | Experimental process configurations of mechanoenzymatic treatment (C, D).

Process option	Material	RAging cycle, RA	Trial time, h
C: RA 1	Cotton textile waste White cotton Polycotton (60/40)	1 min/29 min	6
D: RA 2	Cotton textile waste White cotton Polycotton (60/40)	0.16 min/29.04 min	6

90 mm; height, 75.5 mm) in combination with rotor no. 3 (diameter, 45.1 mm; height, 47 mm).

8.2.4 | FTIR Analysis

FTIR spectroscopy was performed using a Vertex 70 spectrometer (Bruker Optik GmbH, Ettlingen, Germany), equipped with a single-reflection diamond attenuated total reflectance (ATR) accessory (MIRacle, Pike Technologies) and a liquid-cooled mercury cadmium telluride (MCT) detector. To minimize interference from atmospheric moisture and carbon dioxide, the Vertex 70 was continuously purged with dry, CO_2-free air. Spectral data were collected over the range of 4000–6000 cm^{-1}, with 64 scans per spectrum, using the OPUS 7.0 software package (Bruker Optik GmbH). All measurements were conducted under dry sample conditions. Each spectrum underwent baseline correction and area normalization. To account for temporal baseline drift, a background spectrum was recorded against ambient air immediately before sample measurement.

8.2.5 | SEM

Following each experimental trial, fiber samples were collected and subjected to drying in a laboratory oven (Heraeus, Hanau, Germany) at 50°C for a period of 3 days to eliminate residual moisture. SEM was employed to investigate the morphological characteristics of textile fibers using a Zeiss Supra 55 VP instrument. All images were acquired at an accelerating voltage of 3 kV. A qualitative assessment of the SEM images was conducted to evaluate microstructural alterations induced by the mechanoenzymatic pretreatment.

9 | Enzymatic Hydrolysis

The enzymatic hydrolysis of textiles was carried out using the commercial cellulase formulation Cellic CTec2 (Novonesis; supplied by Sigma–Aldrich, SAE 0020–50 mL). Reactions were performed in a 50 mmol/L citrate buffer (pH 4.8) with 1 g/L polyethylene glycol (PEG 6000, AnalytiChem, Eschborn, Germany) at 50°C within a feed tank connected to a wet rotor mill (FrymaKoruma, Rheinfelden, Germany), enabling simultaneous mechanical and enzymatic treatment. An enzyme loading of 25 FPU/g was applied in all experiments, with 40 FPU/g used in one case. Textile suspensions were prepared with a solid content of 5 wt%.

To evaluate the effect of NaOH pretreatment, additional enzymatic hydrolysis experiments were performed in an orbital incubator (INFORS Multitron, Bottmingen, Switzerland) set to 150 rpm with a shaking orbit of 25 mm. Reactions were carried out with a textile substrate loading of 2.5 wt% and an enzyme concentration of 25 FPU/g substrate. Enzymes were added at the beginning of each reaction. Aliquots of 400 µL were withdrawn at 0, 3, 6, and 24 h for subsequent analysis. Glucose concentrations were determined using a Cedex Bio Analyzer 3D Lab (Roche, Basel, Switzerland) equipped with software version 3.0.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section.The authors have cited additional references within the Supporting Information [20, 46–51]