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Preparation and application of bio- pigments based on waste dyed cotton fabrics

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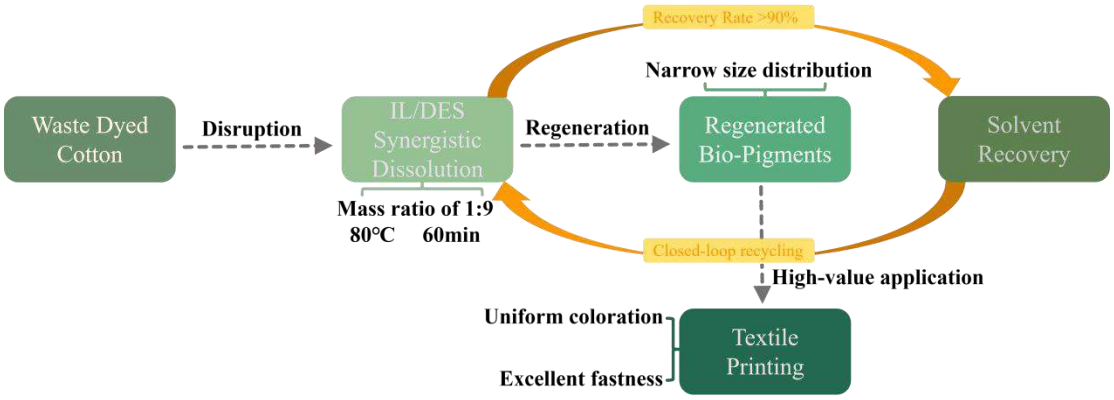
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Abstract The exponential accumulation of textile waste necessitates innovative, closed-loop recycling paradigms to mitigate its severe environmental impacts. A sustainable closed-loop strategy is presented for valorizing waste dyed cotton fabrics by directly converting them into bio-based pigments using a hybrid ionic liquid/deep eutectic solvent (IL/DES) system. Under optimized conditions (1:9 IL/DES mass ratio, 80°C, 60 min), the fabric is deconstructed into regenerated cellulose powder with a narrow, near-monodisperse size distribution. Mechanistic studies via ^{1}H NMR and FTIR reveal that synergistic IL/DES interactions effectively disrupt cellulose's hydrogen-bonding network. Formulated as printing paste, the bio-based pigment exhibits desirable shear-thinning behavior and excellent printability, achieving uniform coloration with rubbing and soaping fastness grades of 4–5. Crucially, the IL/DES solvent demonstrates outstanding recyclability, maintaining over 90% recovery across three cycles without compromised dissolution capability. This work establishes a zero-waste pathway for dyed cellulosic waste valorization, strongly aligned with circular economy and sustainability imperatives.

Keywords: Waste cotton fabric, Ionic liquid (IL), Deep eutectic solvent (DES), Bio-based pigment, Pigment printing, Closed-loop recycling

Graphical abstract



1. Introduction

The textile industry, as one of the world's most significant manufacturing sectors, is also a major contributor to resource consumption and environmental pollution(Farhana et al., 2022) . Among the various environmental challenges, the generation and disposal of vast quantities of textile waste have become increasingly prominent. It is estimated that over 92 million tons of textile waste are generated globally each year, with cotton-based materials constituting a significant proportion(Leal et al., 2019; Yu et al., 2018). These discarded cotton textiles, particularly dyed and finished colored cotton fabrics, present significant challenges for conventional recycling methods due to the complex binding between their primary component (natural cellulose) and impurities (dyes, auxiliaries), resulting in low efficiency and limited recovered value(Dissanayake and Weerasinghe, 2021). The predominant disposal methods remain landfilling and incineration(Moazzem et al., 2021),which not only lead to the severe loss of valuable biomass resources but also pose persistent ecological risks due to the release of dyes and chemicals into the environment(Lin et al., 2023). Therefore, from the perspective of a circular economy and sustainable development, developing a technology capable of directly and efficiently converting waste colored cotton fabrics into high-value-added functional materials represents a critical challenge with both environmental urgency and engineering scientific significance.

The high-value recycling of waste cotton fabrics faces two core scientific challenges. First, how to efficiently and greenly disrupt the highly ordered hydrogen-bond network and crystalline structure of cellulose to achieve its efficient dissolution and structural reconstruction(C. L. Huang et al., 2025). Second, how to maximally preserve and stabilize the original color of the fabric during the deconstruction and reconstruction processes, thereby avoiding resource downgrading(Gan et al., 2019). Existing technical approaches exhibit significant limitations in addressing this issue. Physical-mechanical methods, while operationally simple, produce shortened fibers with compromised performance, relegating them to low-grade filling applications and failing to achieve synchronous recovery of both chromatic and material value(Abtew et al., 2025). Even when waste cotton is mechanically milled into fine powder, conferring pigment functionality still necessitates subsequent chemical dyeing processes, which inherently fails to circumvent the consumption of dyestuff resources and auxiliary chemicals(Baloyi et al., 2024). While effective in cellulose depolymerization, conventional chemical routes (e.g., the viscose process(Sayyed et al., 2019)or acid/alkali hydrolysis(W. C. Liu et al., 2019)) operate under severe processing conditions, generate substantial pollutant discharge, and cause extensive breakdown of dye chromophores, leading to significant color loss. The recently developed enzymatic hydrolysis process(Freestone, 2023),while promising, suffers from low efficiency, high sensitivity to dyes, and elevated costs. Therefore, there is a pressing need for a novel green chemical strategy capable of simultaneously achieving cellulose dissolution and "in-situ fixation" of dye molecules under mild conditions.

In this context, novel green solvents, particularly deep eutectic solvents and ionic liquids, offer a promising pathway to address the aforementioned challenges. ILs possess strong hydrogen-bond accepting capabilities and tunable cation-anion structures, granting them exceptional cellulose dissolution capacity(Verma et al., 2019). However, their high cost, potential biotoxicity, and complex purification and recovery processes hinder their large-scale industrial application. In contrast, deep eutectic solvents, composed of inexpensive and renewable hydrogen-bond donors and acceptors, offer notable advantages such as simple preparation, high biocompatibility, and low cost. However, single-component DES systems often suffer from high viscosity, slow dissolution rates for natural cellulose, and limited solubility(El Achkar et al., 2021). Consequently, formulating hybrid DES/ILs systems holds promise for synergistically combining the high dissolution efficiency of ILs with the low-cost, eco-friendly nature of DES. By modulating the microscopic interactions within the solvent system (such as hydrogen-bond networks, viscosity, and polarity) this strategy may yield a novel method for dissolving cotton fabrics that integrates superior solvation performance, dye stability, and process economy.

Current research primarily focuses on employing solvents to dissolve pure cellulose or undyed textiles for the regeneration of functional materials such as films(Su et al., 2022; Chen et al., 2024; Moreira et al., 2024),fibers(H. X. Liu et al., 2023; Z. Y. Huang et al., 2024; Zeng et al., 2024)or aerogels(Song et al., 2024; Sun et al., 2024; Shao et al., 2024). However, a scientifically valuable and practical application scenario has been overlooked: leveraging the inherent "color" of waste as a resource by preserving it throughout the dissolution-regeneration process to directly fabricate new materials with colorant functionality. The direct conversion of waste colored cotton fabrics into colored cellulose powder for use as a colorant in pigment printing establishes a critical link between "waste valorization" and "green manufacturing". This approach not only achieves a "closed-loop" cycle from waste to functional product, but also innovatively transforms the color attribute of the recycled material into its core functional property.

To address the dual challenges of textile solid waste accumulation and the intensive resource consumption of synthetic dyeing, this study proposes a novel closed-loop valorization strategy that transforms waste dyed cotton fabrics into high-performance bio-based pigments. We engineered a hybrid green solvent system comprising an ionic liquid ([BMIM]Cl) and a deep eutectic solvent (choline chloride/oxalic acid) to efficiently dissolve and regenerate waste cotton under mild processing conditions. By systematically optimizing the dissolution parameters, we achieved controlled micro-particulate regeneration of the dyed cellulose, and fundamentally elucidated the underlying hydrogen-bond deconstruction mechanism via spectroscopic analyses. To validate its high-value downstream applicability, the regenerated cellulose powder was utilized as a sustainable pigment in textile printing, with its rheological behavior, color characteristics, and durability rigorously evaluated. Finally, the economic viability and environmental footprint of the process were assessed through multiple solvent recovery cycles, presenting a comprehensive and scalable paradigm for the circular processing of end-of-life textiles.

2. Experimental

2.1. Materials

Fabric made of 100% bleached cotton was purchased from Textile Co., Ltd. (Hebei of China). Reactive Red X-3B were obtained from Hebei Sheng Wei Chemical Co., Ltd. (Hebei of China). Choline chloride (98%), oxalic acid (98%) and 1-butyl-3-methylimidazolium chloride ([BMIM]Cl) (≥97%) were purchased from Maicrobial Biochemical Technology Co., Ltd. (Shanghai of China). Soft binder, thickener was purchased from New Ruibei Technology Co., Ltd. (Jiangsu of China).

2.2. Dyeing of cotton fabrics

A total of 100 g of cotton fabric was weighed, and a dye bath was prepared at a liquor ratio of 1:50 containing 4 g of Reactive Red X-3B, 300 g of sodium sulfate, and 100 g of sodium carbonate. The cotton fabric was placed in a dyeing machine, and the dyeing process was started at 50 ℃. After 15 minutes, the specified amount of sodium sulfate was added to promote dye uptake, and dyeing was continued for another 15 minutes. The temperature was then raised to the fixation temperature of 60 ℃ at a rate of 2 ℃/min. The specified amount of sodium carbonate was added for fixation, and the temperature was maintained for 30 minutes. The fabric was then cooled, rinsed, and subjected to soaping (soap flakes at 2 g/L, 90 ℃, 15 minutes), followed by hot water rinsing, cold water rinsing, and finally drying.

2.3. Preparation of cellulose powder

Choline chloride (ChCl) and oxalic acid ($H_2C_2O_4$) were dried in a vacuum oven for 24 hours. After drying, they were mixed uniformly in a molar ratio of 1:2. The mixture was stirred at 80℃, and was removed once the chemicals were thoroughly blended to obtain a deep eutectic solvent (DES). The fabric was dried at 50 ℃ for 24 h, cut into 1cm × 1 cm pieces, and pulverized using a high-speed grinder at 20,000 rpm. To prevent overheating, grinding was performed in cycles of 1 minute followed by 10 minute intervals, yielding short fibers with lengths less than 3 mm. The treated fabric was placed into a solvent containing IL and DES, dissolved for a specified duration at a set temperature, and then removed. Upon removal, the solution was gradually introduced into water under ultrasonication, prior to centrifugation. This step was repeated 3–5 times to ensure complete removal of the solvent. In the final step, the regenerated cellulose was subjected to freeze-drying, producing cellulose powder.

2.4. Optimization of preparation technology for cellulose powder

Single-factor experiments were conducted focusing on the IL/DES mass ratio, dissolution temperature, and dissolution time to optimize the processing conditions. For each experimental group, all other conditions were maintained constant while only a single variable was altered, so as to accurately analyze the influence of each factor on the dissolution effect of the cotton fabric.

First, the IL/DES blending ratio is one of the most critical parameters. Therefore, the dissolution temperature was controlled at 75℃ and the dissolution time at 90 minutes. The blending mass ratio was varied as 1:9, 2:8, 3:7, 4:6, and 5:5 to investigate its effect on dissolution efficiency. Second, Dissolution temperature is a

crucial parameter in the dissolution process of dyed cotton fabrics. Therefore, with the IL/DES blending mass mass ratio fixed at 1:9 and the dissolution time at 90 minutes, the dissolution temperature was adjusted to 65℃, 70℃, 75℃, 80℃, and 85℃ to investigate its effect on dissolution efficiency. Finally, with the IL/DES blending mass ratio fixed at 1:9 and the dissolution temperature at 80℃, the dissolution time was adjusted to 30, 40, 50, 60, 70, 80, and 90 minutes to investigate its effect on dissolution efficiency.

2.5. Particle size of cellulose powder

Using deionized water as the dispersion medium, and a small amount of cellulose powder was added to achieve a concentration of approximately 0.1%–1%. Perform ultrasonic dispersion for 1–5 minutes to reach the required transmittance. Inject the well-dispersed suspension into the sample cell, and then analyze the collected data. The width of the particle size distribution is quantified by the Span parameter, defined as:

$$Span = \frac{D_{90}-D_{10}}{D_{50}}$$

where D_{10}, D_{50}, and D_{90} represent the particle sizes at cumulative volume fractions of 10%, 50%, and 90%, respectively.

2.6. Recovery and recycling of IL/DES system

To evaluate the sustainability of this process, the spent IL/DES mixed solvent was recovered via vacuum distillation. After regenerated cellulose powder was separated by centrifugation, the supernatant containing water, IL and DES was collected. A rotary evaporator was used to efficiently remove water under reduced pressure at 60 ℃. The chemical integrity of the recovered solvent was verified by FTIR, and the recycled solvent was reused in subsequent dissolution cycles. The recovery rate of the IL/DES mixed solvent was calculated by the mass method, and the formula is defined as follows:

$$\eta = \frac{m_1}{m_0} \times 100\%$$

Where η is the solvent recovery rate (%), m_1 is the mass of the recovered and purified IL/DES mixed solvent (g), and m_0 is the mass of the fresh IL/DES mixed solvent initially used in the dissolution experiment (g). All recovered solvents were fully dried to eliminate the influence of residual moisture before weighing.

2.7. Application of Cellulose Powder

During the preparation of the printing paste, 5% cellulose powder, 3% thickener, and 18% binder were added, with the balance made up with deionized water. The printing paste was prepared according to the formulation, and flat-screen printing was then carried out. The printed fabric was subsequently dried at 80℃ for 5 minutes, followed by curing at 150℃ for 3–5 minutes.

3. Results and Discussion

3.1. High-Value Recovery of Waste Dyed Cotton Fabrics

Single-factor experiments were conducted to optimize three key processing parameters: the IL/DES mass ratio, dissolution temperature, and dissolution time.

Among these factors, the IL/DES mass ratio exerted the most pronounced effect on particle size. As shown in Fig. S1, the minimum particle size was achieved at an IL/DES mass ratio of 1:9; the corresponding span value was 0 (Table S1), indicating an extremely narrow and nearly monodisperse particle size distribution. For dissolution temperature, the smallest particle size was obtained at 80 ℃ with a span value of 0.203 (Fig. S2 and Table S2), which reflects a narrow and uniform particle distribution. In terms of dissolution time, particle size remained approximately 450 nm from 60 to 80 min and further decreased to around 110 nm when the time was extended to 90 min (Fig. S3). Considering both energy efficiency and particle characteristics, 60 min was selected as the optimal duration, at which the span value was 0.278 (Table S3), corresponding to a moderately narrow particle size distribution.

By optimizing the IL/DES mass ratio, dissolution temperature, and dissolution time, the optimal conditions were determined to be an IL/DES mass ratio of 1:9, a dissolution temperature of 80 ℃, and a dissolution time of 60 min. Under above optimum parameters, regenerated cellulose was precipitated and then freeze-dried to obtain cellulose powder (Fig. 1a). To comprehensively evaluate the impact of the recovery process on the physicochemical properties of the material and its potential as a high-value recycled resource, the micromorphology, chemical structure, and thermal properties of the resulting cellulose powder were systematically characterized.

The micromorphological transformation was first examined by Scanning Electron Microscopy (SEM). As shown in Fig. 1b, the recovered material predominantly exists as agglomerated cellulose powder particles, confirming that the IL/DES system effectively disintegrated the original dense, highly ordered macroscopic fiber bundles. A magnified view of an individual particle (Fig. 1c) reveals its surface morphology and textural details. This transition from a continuous macroscopic fabric to a micro-particulate form is highly advantageous for its subsequent application as a pigment, as the increased specific surface area and rough surface texture promote intimate contact with the binder and enhance dispersion stability in the printing paste.

Fourier Transform Infrared (FTIR) spectroscopy (Fig. 1d) was employed to assess the chemical composition and functional groups of the materials. Both the original dyed cotton fabric and the recovered cellulose powder exhibit characteristic absorption bands of the cellulosic backbone, most notably the broad peak centered at approximately 3400 cm^{-1}, assigned to O–H stretching vibrations, and the peak at 1644 cm^{-1}, attributed to the bending vibration of absorbed water. Notably, a distinct new peak emerges at 1730 cm^{-1} in the spectrum of the recovered cellulose powder, which is entirely absent in the original dyed cotton fabric. This peak is typically associated with C=O stretching vibrations. Its appearance suggests that the IL/DES treatment may have induced mild oxidation or esterification reactions on the cellulose surface, or alternatively, it may reflect trace residual solvent components interacting with the cellulose matrix.

The crystalline structure was further investigated by X-ray Diffraction (XRD) (Fig. 1e). Both samples display characteristic diffraction peaks at 2θ values of approximately 15°, 16.5°, and 22.5°, corresponding to the signature reflections of

native Cellulose I. The retention of these peaks indicates that the mild IL/DES dissolution conditions achieved structural disintegration into powder form without triggering a complete polymorphic transformation to the Cellulose II allomorph. However, the visibly reduced peak intensities observed for the cellulose powder signify a decreased degree of crystallinity, a direct consequence of the solvent system penetrating the fibers and disrupting the extensive inter- and intra-molecular hydrogen bond networks during treatment.

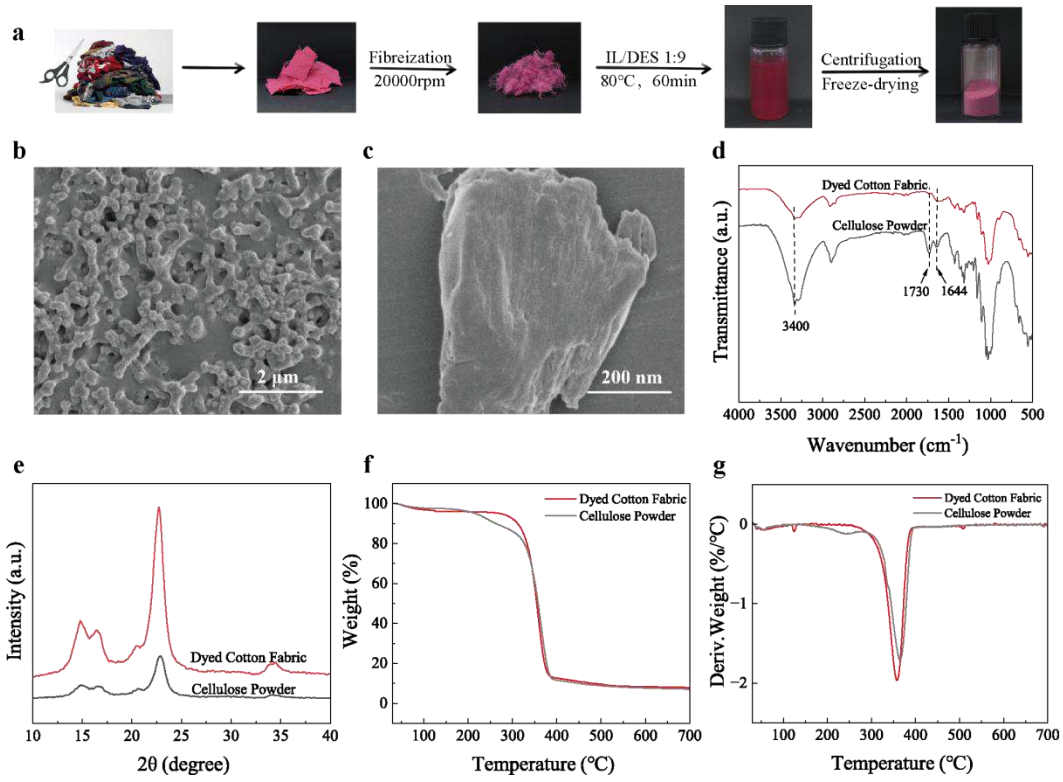


Fig. 1 Preparation and physicochemical characterization of regenerated cellulose powder from waste dyed cotton fabrics. (a) Schematic illustration of the preparation process of regenerated cellulose powder from waste cotton fabrics. (b) Low-magnification SEM image of the regenerated cellulose powder. (c) High-magnification SEM image of the regenerated cellulose powder. (d) FTIR spectra of the dyed cotton fabric and the regenerated cellulose powder. (e) XRD patterns of the dyed cotton fabric and the regenerated cellulose powder. (f) TGA curves of the dyed cotton fabric and the regenerated cellulose powder. (g) DTG curves of the dyed cotton fabric and the regenerated cellulose powder.

Finally, the thermal stability of the recycled material was evaluated by Thermogravimetric Analysis (TGA) and Derivative Thermogravimetry (DTG) (Fig. 1f and 1g). The degradation profiles reveal that the primary mass loss for both materials occurs within the temperature range of 300–400 °C. The cellulose powder exhibits a slightly lower onset degradation temperature compared to the original dyed cotton fabric. This earlier mass loss is primarily attributable to the aforementioned reduction in crystallinity and the particulate morphology of the sample, both of which promote heat transfer and accelerate initial thermal decomposition. Nevertheless, the DTG curves confirm that the maximum degradation temperature remains relatively stable. Overall, the thermal stability of the cellulose powder is largely preserved, with

its degradation profile well exceeding the typical drying (80–100 ℃) and curing (150 ℃) temperatures employed in textile pigment printing, thereby confirming its suitability as a bio-based colorant in practical printing processes.

3.2. Dissolution mechanism of the ILs/DES system

To gain in-depth insight into the efficient dissolution mechanism of waste cotton fabrics in the IL/DES hybrid solvent system, the synergistic interactions between the solvent components and the solvent-cellulose interactions were systematically investigated by proton nuclear magnetic resonance (^{1}H NMR) and Fourier transform infrared (FTIR) spectroscopy. The results reveal that the fundamental nature of the dissolution process lies in the establishment of new non-covalent interactions between solvent molecules and cellulose, which disrupt and replace the original strong hydrogen-bonding network within the cellulose matrix.

In the hybrid solvent system, the IL and DES components do not simply undergo physical mixing; rather, strong interactions occur between them, leading to a reconstruction of the solvent hydrogen-bonding network. As shown in the FTIR spectra (Fig. 2b), compared with pure IL and pure DES, the IL/DES hybrid system exhibits pronounced changes in both the peak shape and absorption intensity of the O–H and N–H stretching vibrations in the 3000–3500 cm^{-1} region, as well as the C=O stretching vibration near 1700 cm^{-1}. These alterations in infrared absorption characteristics confirm the formation of a new, more complex intermolecular hydrogen-bonding network between the IL (imidazolium ring and chloride ions) and the DES components (carboxyl groups of oxalic acid, hydroxyl groups of choline chloride). Further evidence for these solvent-solvent interactions was provided by ^{1}H NMR spectroscopy (Fig. 2a and Fig. S4a). A comparison between the pure IL and the IL/DES hybrid reveals that the protons on the imidazolium ring of the IL (primarily in the range of 7.6–9.5 ppm, corresponding to positions a, b_1, and b_2 in Fig. 2a) undergo a distinct upfield shift (lower ppm values) upon the addition of DES (as shown in the top panel of Fig. S4a, where the peak at approximately 9.41 ppm shifts leftward). This upfield shift indicates an increase in electron density (enhanced shielding effect) around the imidazolium ring protons. The underlying reason is that the hydrogen bond donors in DES (oxalic acid) form strong hydrogen bonds with the free Cl^- ions in the IL, thereby weakening the originally strong cation-anion (imidazolium-Cl^-) electrostatic interaction network within the IL. This endows the hybrid solvent system with higher fluidity and enhanced penetration capability, laying the foundation for the subsequent attack on the crystalline structure of cellulose.

The primary component of waste cotton fabrics is highly crystalline cellulose, whose structure is maintained by strong intramolecular and intermolecular hydrogen-bonding networks. To investigate how the solvent system deconstructs this network, the ^{1}H NMR spectra before and after cellulose dissolution were compared (Fig. 2c and Fig. S4b). Upon dissolution of cellulose in the IL/DES system, the proton signals in the 3.4–4.0 ppm region (primarily corresponding to the choline backbone protons in DES and the alkyl chain protons attached to the nitrogen atom in IL) exhibited a downfield shift (higher ppm values) or peak broadening. This downfield shift is a typical deshielding effect, directly demonstrating that strong interactions

occur between the solvent molecules (particularly the hydroxyl and carboxyl groups serving as hydrogen bond donors/acceptors, as well as the cations and anions) and the hydroxyl groups (–OH) on the cellulose chains, leading to the formation of new hydrogen bonds.

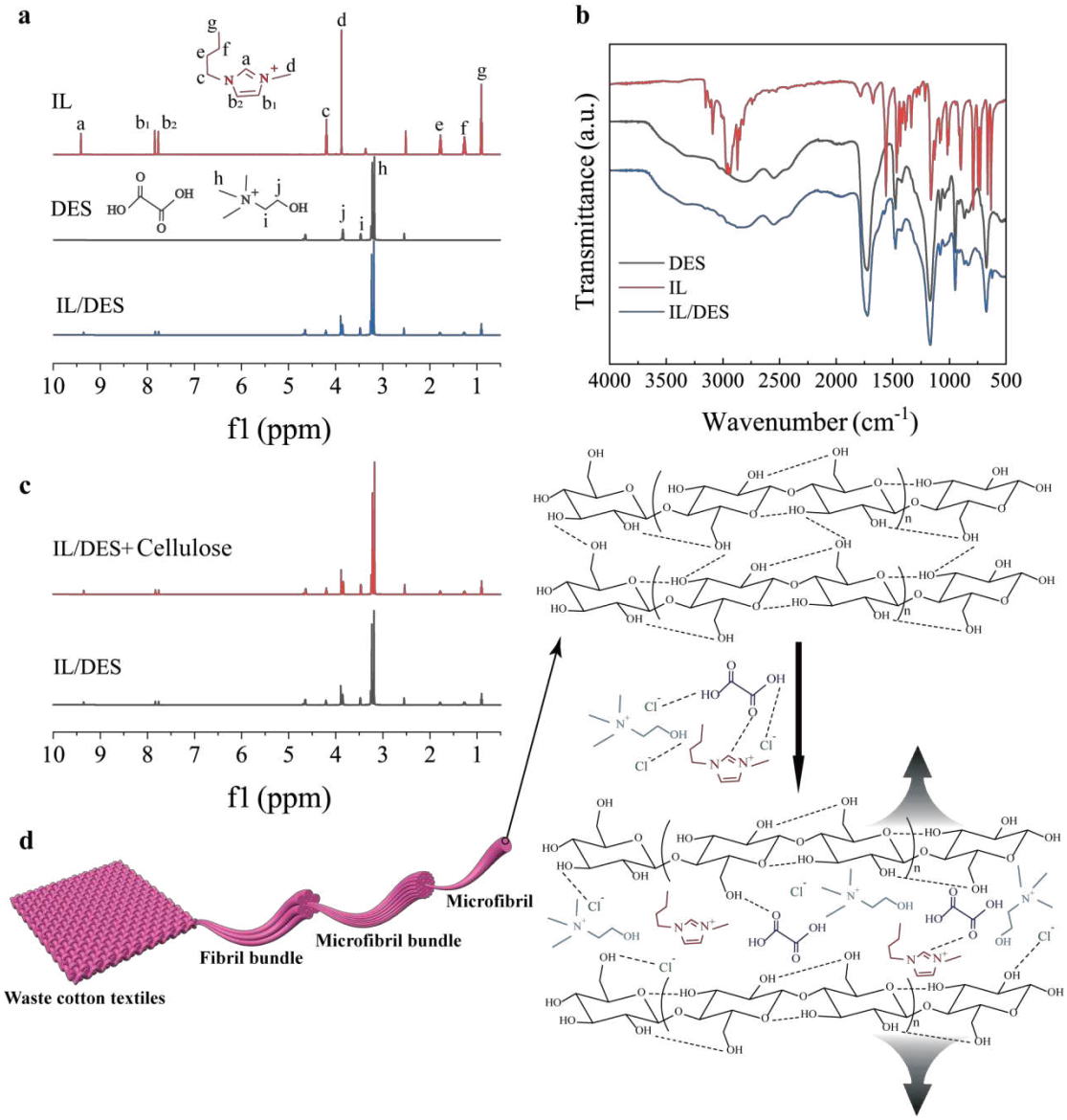


Fig. 2 Structural characterization of the IL/DES solvent system and its cellulose dissolution mechanism. (a) ^{1}H NMR of IL, DES, and IL/DES. (b) FTIR spectra of IL, DES, and IL/DES. (c) ^{1}H NMR of IL/DES before and after cellulose addition. (d) Schematic of waste cotton dissolution in IL/DES via hydrogen bond disruption.

Based on the above spectroscopic evidence, the dissolution mechanism of waste cotton fabrics was elucidated, as schematically illustrated in Fig. 2d. The abundant free Cl^- ions in the system (derived from both the IL and DES) act as strong hydrogen bond acceptors, preferentially attacking the hydroxyl protons on the cellulose chains and thereby disrupting the original intramolecular and intermolecular hydrogen bonds of cellulose. The bulky imidazolium cations, choline cations, and oxalic acid molecules subsequently insert themselves between the cellulose chains that have been

pried apart by Cl^-, through electrostatic interactions and van der Waals forces. These components function as both steric spacers and hydrogen-bonding network reorganizers, forming new hydrogen bonds with the oxygen atoms of the cellulose chains. Under the synergistic action of the anions, cations, and DES molecules, the intermolecular distances between cellulose chains are irreversibly enlarged, the native crystalline structure is thoroughly disrupted, and ultimately the cellulose macromolecules are uniformly dispersed in the IL/DES hybrid solvent as random coils, achieving the efficient and homogeneous dissolution of waste cotton fabrics.

3.3. Application of the Regenerated Cellulose Powder as a Bio-Based Pigment in Textile Printing

Building upon the successful recovery of the cellulose material, its practical viability for high-value downstream applications was evaluated. Specifically, the material was utilized to prepare bio-based pigment printing paste to explore its potential in sustainable textile processing.

As shown in Fig. 3a, the prepared bio-based printing paste exhibits typical shear-thinning (pseudoplastic) behavior, with its viscosity decreasing sharply as the shear rate increases. This rheological profile is highly desirable for textile screen printing: it allows the paste to flow readily under the shear forces of the squeegee and penetrate the screen mesh, while rapidly recovering its viscosity at rest to prevent capillary bleeding and ensure sharp print definition.The macroscopic and microscopic effects of the printing process were examined by optical photography and SEM (Fig. 3b). Macroscopically, the pristine white cotton fabric was successfully transformed, displaying a vibrant and uniform pink coloration. Microscopically, whereas the original cotton fibers exhibit relatively smooth longitudinal surfaces, the SEM images of the printed fabric reveal that the bio-based pigment particles are evenly distributed and firmly anchored onto the fiber surfaces. This indicates excellent compatibility and film-forming capability, without significant agglomeration or structural damage to the underlying cellulose matrix.

To systematically evaluate the print quality and color levelness, reflectance and color strength (K/S) spectra were acquired from five randomly distributed sampling positions (A–E) on the printed fabric (Fig. 3c and 3d). The spectral curves across all positions are highly superimposable, exhibiting a pronounced absorption maximum (corresponding to the lowest reflectance and highest K/S values) within the 530–550 nm wavelength range. The remarkable consistency across different sampling points robustly confirms the excellent color uniformity and printability of the formulated bio-based pigment system.

The colorimetric data (Tables S4 and S5) rigorously substantiate the uniform deposition and exceptional color consistency of the bio-based pigment system. As detailed in Table S4, the printed fabric demonstrated a significant color yield, with the L^* values across the five diverse positions stabilizing within a narrow range of 70.293 to 71.821, and the a^* values (redness) remaining tightly grouped between 36.333 and 38.445. The highly consistent distribution of these metrics across the substrate demonstrates the excellent coloring capacity of the bio-based pigment on cellulosic fibers.

Spatial color levelness is a critical parameter in textile printing. The pairwise color difference matrix (ΔE) presented in Table S5 definitively confirms the minimal variation among the sampling positions. The ΔE values for several spot pairs (e.g., A vs. C: 0.324; A vs. B: 0.561) fall entirely below the threshold of human visual perception (ΔE < 1.0). The maximum deviation recorded across the entire sampled area occurred between positions D and E, with a ΔE of merely 2.235, which remains strictly within the commercially acceptable tolerance limits for textile color matching and print uniformity. This constrained matrix of microscopic color differences robustly demonstrates that the bio-based pigment can achieve homogeneous dispersion and uniform deposition on the fabric surface, thereby delivering a premium-quality printed finish.

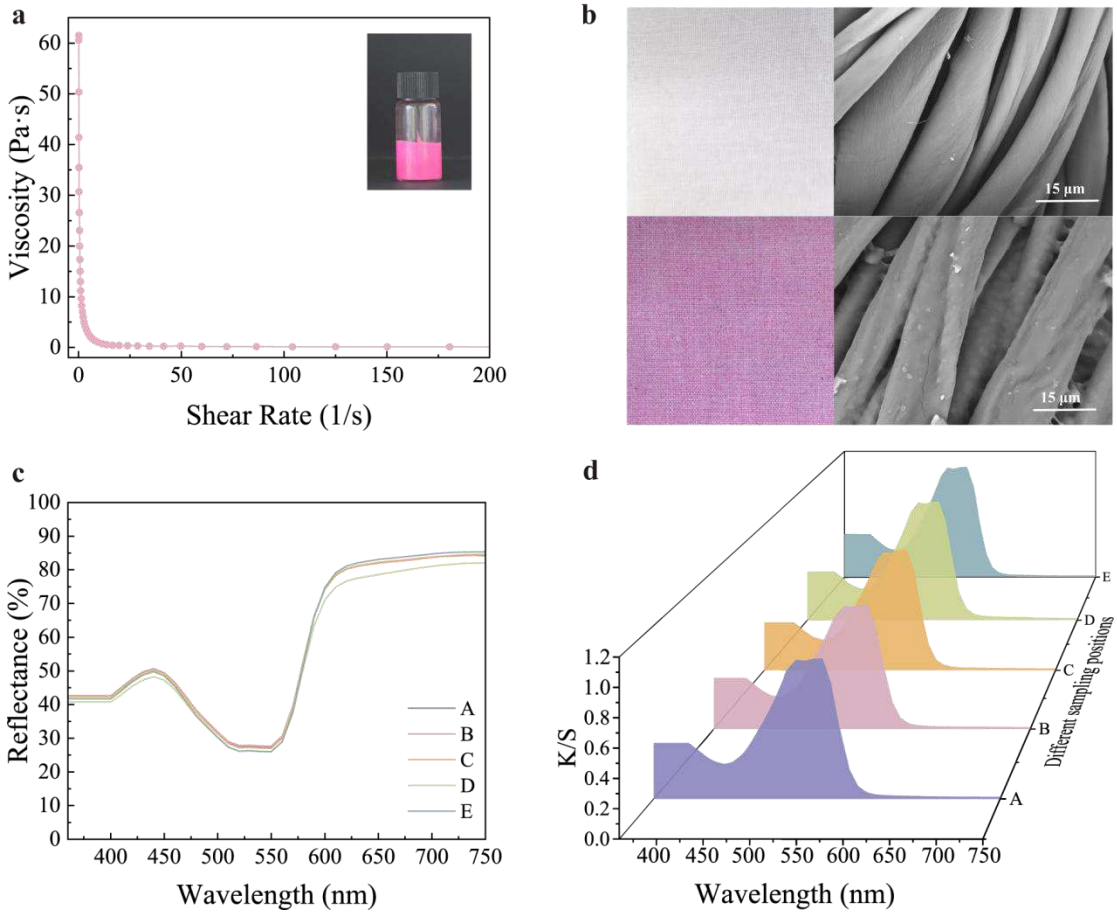


Fig. 3 Rheological behavior of the bio-based pigment printing paste and printing performance of the printed cotton fabrics. (a) Viscosity-shear rate curve of the pigment printing paste. (b) Macroscopic and microscopic morphologies of the pristine and printed cotton fabrics. (c) Reflectance spectra of the printed fabric at different positions (A-E). (d) Corresponding K/S curves of the printed fabric at different positions.

Finally, in the context of resource conservation and the circular economy, the practical lifespan of dyed/printed textiles is heavily dependent on their durability against daily wear and maintenance. Accordingly, the color fastness properties of the printed fabrics were rigorously evaluated. The samples exhibited outstanding rubbing fastness (Table S6), achieving the maximum grade of 5 for dry rubbing and a grade of 4 for wet rubbing. Furthermore, the color fastness to soaping (Table S7) was

exceptional: the color change of the printed fabric was rated at 4–5, and the staining on adjacent multifiber strips (wool, acrylic, polyester, nylon, cotton, and viscose) consistently achieved high grades ranging from 4–5 to 5.These excellent fastness results indicate robust physical and chemical adhesion of the pigment–binder network to the cellulosic fibers, confirming the practical viability of this bio-based pigment for textile printing applications.

3.4. Recovery and Reuse of the IL/DES Hybrid Solvent

To evaluate the economic viability and environmental sustainability of the proposed dissolution process, the recyclability of the IL/DES hybrid solvent was systematically investigated. Establishing a closed-loop solvent recovery system is a critical prerequisite for large-scale textile waste valorization.

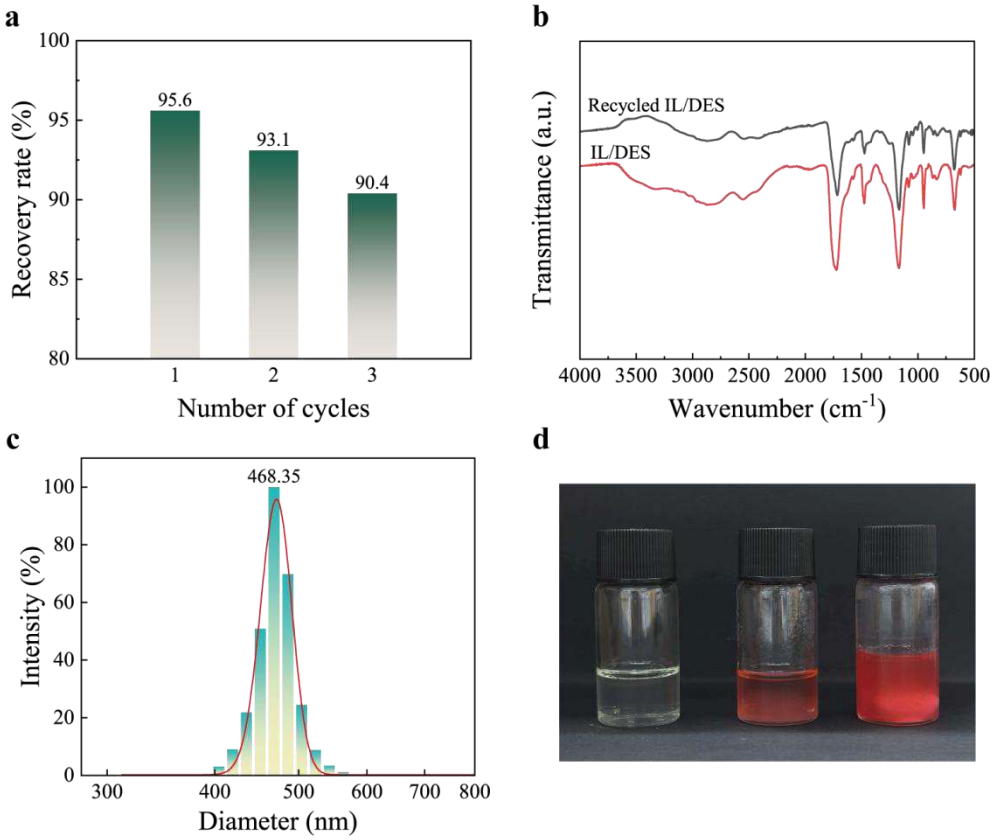


Fig. 4 Recyclability and characterization of the IL/DES hybrid solvent. (a) Recovery rates of the IL/DES solvent over three consecutive cycles. (b) FTIR spectra of the IL/DES solvent before and after recovery. (c) Particle size distribution of cellulose powder regenerated using the recovered IL/DES solvent. (d) Photographs comparing the IL/DES solvent before and after recovery, along with the dissolution behavior of cotton fabric in the recycled solvent.

As shown in Fig. 4a, the recovery efficiency of the IL/DES solvent remained consistently high over three consecutive dissolution-regeneration cycles. The recorded recovery rates were 95.6%, 93.1%, and 90.4% for the first, second, and third cycles, respectively. Although a marginal decline was observed, this can be attributed primarily to inevitable physical mass losses during filtration, washing, and transfer steps. Nonetheless, the solvent recovery rate was maintained above 90% throughout. This demonstrates the robust cyclability of the IL/DES system, highlighting its

potential to substantially reduce virgin solvent consumption and operational costs in practical applications.

The chemical integrity of the recovered solvent is paramount to its sustained performance. Fig. 4b compares the FTIR spectra of the original and recycled IL/DES solvent. The spectrum of the recovered solvent is virtually identical to that of the original solvent, with no emergence of new absorption peaks or significant shifts in the characteristic functional group bands. This verifies the excellent chemical stability of the IL/DES system, confirming that the solvent undergoes a purely physical dissolution process with the cotton fabric and does not suffer from irreversible chemical degradation or decomposition during the heating and regeneration stages.The practical efficacy of the recycled solvent was further validated by employing it for subsequent cotton fabric dissolution and cellulose regeneration. Fig. 4c displays the particle size distribution of the cellulose powder regenerated entirely using the recycled IL/DES. The resulting cellulose powder exhibited a particle size distribution centered at approximately 470 nm, demonstrating that the recycled solvent retains a dissolution capacity comparable to that of the original solvent.

Furthermore, the photographic comparison in Fig. 4d illustrates the changes in solvent appearance after recycling. Although the recycled solvent effectively dissolves the cotton fabric, a distinct color change is observed relative to the initially transparent solvent. Such discoloration is a common phenomenon in textile recycling and is typically caused by the accumulation of residual dyes or finishing agents extracted from the waste cotton. Combined with the FTIR and particle size results in Fig. 4b and 4c, these findings indicate that the trace colored impurities do not compromise the intrinsic thermodynamic properties of the solvent or its capacity to disrupt the hydrogen-bonding network of cellulose. Overall, the IL/DES hybrid system demonstrates considerable potential as a sustainable and highly recyclable green platform for the continuous processing of cellulosic waste.

4. Conclusion

In summary, a sustainable and closed-loop process was successfully developed for upcycling waste dyed cotton fabrics into high-value bio-based pigments. The hybrid IL/DES solvent system demonstrated excellent dissolution capability under mild conditions (80 °C, 60 min), yielding regenerated cellulose powder with a narrow particle size distribution. Spectroscopic analyses confirmed that the dissolution is driven by the synergistic disruption of the native cellulose hydrogen-bonding network, facilitated by the abundant Cl^- acceptors and the steric insertion of solvent cations and molecules. Notably, when applied as a textile printing pigment, the recycled particulate material produced vibrant, uniform coloration with favorable rheological compatibility and robust physical–chemical adhesion, with both rubbing and soaping fastness consistently reaching grades of 4–5.

Crucially, the proposed process exhibits high environmental sustainability and economic feasibility. The IL/DES hybrid solvent can be efficiently recovered via vacuum distillation, maintaining a recovery rate above 90% over three consecutive cycles while preserving its structural integrity and native dissolution efficiency. This

work provides a highly promising, scalable, and environmentally benign pathway for the high-value recycling of dyed textile waste.

CRediT authorship contribution statement

Youchao Lian: Methodology, Investigation, Data curation, Visualization, Writing - original draft, Writing - review & editing. **Shaohai Fu:** Resources, Funding acquisition, Project administration. **Jingjing Cao:** Methodology, Supervision, Writing - review & editing. **Rong Li:** Supervision, Writing - review & editing. **Zhiguang Li:** Supervision, Writing - review & editing. **Chi-wai Kan:** Writing - review & editing. **Chang-E Zhou:** Conceptualization, Methodology, Project administration, Supervision, Writing - review & editing. **Xin Chu:** Writing - review & editing. **Qing Zhang:** Writing - review & editing. **Huixia Li:** Writing - review & editing.

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Data availability

The data supporting the findings of this study are included within the article. Additional experimental data are provided in the Supplementary Material.

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