

Natural dyes and regenerated cellulose fibers blending using ionic liquid as a common platform for sustainable textiles/fashion applications

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ABSTRACT

Conventional textile dyeing generates significant water pollution through synthetic dye leaching, creating urgent need for closed-loop processes. The current work uses a novel *in-situ* spin dyeing process to develop dyed cellulose fibers, using ionic liquid (IL) as a unified platform to dissolve cellulose and disperse a range of natural dyes – eliminating post-spinning coloration. The spin dyeing method effectively requires less water uptake and is a sustainable benign method for the future textile industry. Different natural dyes were incorporated into the cellulose-IL composite solution to add vibrant colors to the fibers. Natural dyes derived from Sorghum, and Coreopsis show better conjugation with the wet-spun fibers, while the other dyes do not show similar levels of dye stability and incorporation within the fibers. FTIR results confirmed chemical bonding of dyes in the fibers. SEM images indicated the homogeneous dispersion of dye across the fibers. The resultant dyed fibers demonstrate similar mechanical properties in comparison to the neat cellulosic fibers, and commercially available fibers. Although thermal stability decreased for the dyed fibers, mechanical performance validates IL-mediated spin-dyeing as a viable alternative to petroleum-based dyes. This first demonstration of ILs enabling integrated fiber production and natural dyeing establishes a blueprint for sustainable textiles manufacturing.

Introduction

A growing trend towards the usage of sustainable and ecologically friendly materials has emerged as a result of consumers' increased eco-awareness (Shahid et al., 2013). As one of the major manufacturing sectors, (Bailey et al., 2022) the fashion industry significantly contributes to global environmental pollution due to its production and processing techniques (Bhatia et al., 2017). More than 70 % of the fabrics produced are of synthetic origin, which are difficult to degrade, remain in landfills for years, and adversely affect the environment (Echeverria et al., 2019). Despite being natural and renewable, cotton requires around 2.6 % of the world's water consumption. It utilizes nearly 11 % of the pesticides used globally, has high energy usage and associated carbon dioxide emissions (Singhal et al., 2023). A critical issue within the textile industry is the release of colorants or dyes into water bodies,

contributing to 20 % of global wastewater pollution (Why Clothes Are So Hard to Recycle - BBC Future, n.d.).

Synthetic dyes have dominated the industry due to their cost-effectiveness, color consistency, and stability in light and temperature (Bhatia et al., 2017; Jamee & Siddique, 2019). Globally, 800,000 tons of synthetic dyes are produced annually, with 2 % discharged as industrial effluents, translating to a staggering 40,000 tons of dye being released into the environment every year (Slama et al., 2021). The textile industry, responsible for 75 % of the global dye market (Madhav et al., 2018; Thakur & Chauhan, 2018), often operates in developing countries with inadequate wastewater treatment facilities, leading to significant environmental pollution from untreated or partially treated dye effluents. According to a previous report, around 10–15 % of the dyes are lost as effluents during the dyeing process alone (Iqbal & Ashiq, 2007). Textile industries are mostly located in developing countries to build

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economy, employability, and foreign exchange earnings (Desore & Narula, 2018; Lellis et al., 2019). However, due to the poor wastewater systems these countries do not comply with the effluent discharge norms resulting in huge environmental pollution due to large quantities of untreated or partially treated dye effluents (Singh et al., 2023). Dyes of organic origin, though water-soluble and providing brilliant texture (Mahapatra, 2016), pose challenges in wastewater treatment due to their complex aromatic structures. Their release, as such, into local water bodies causes a severe reduction of oxygen levels in water (Hassan & Carr, 2018) leading to eutrophication which inhibits the photosynthesis process by limiting the light penetration, seriously affecting the aquatic ecosystems (Imran et al., 2015). The decomposition of dye molecules in heat, microbes, light, or oxidizing agents are ineffective due to the complex aromatic structure of the dyes (Pearce et al., 2003). Additionally, synthetic dyes pose health risks to workers, causing allergic reactions, dermatitis, rhinitis, and asthma (Hunger & Sewekow, 2002). The conventional dyeing process is also water-intensive, requiring about 150 liters of water per kilogram of fabric (Drumond Chequer et al., 2013; Haslinger et al., 2019). The wastewater effluents from these textile industries are composed of hydrocarbon-based softeners, heavy metals, nitrates, acetic acid, soaps, enzymes, formaldehyde-based dye fixing agents, chlorinated stain removers, and dyes – which collectively are toxic, carcinogenic, mutagenic, and teratogenic (Bhatt & Rani, 2013).

In contrast, biopolymers and natural dyes offer a sustainable and low-cost alternative (Orisawayi et al., 2024, 2025). Natural dyes from sources such as flowers, vegetables, fruits, and trees are being developed to replace synthetic dyes, minimizing environmental pollution due to their renewable, biodegradable, and skin-friendly properties (Saxena & Raja, 2014). For instance, extracts from different fruit peels like orange, and pomegranate show potential as dyeing materials with medicinal properties (Hou et al., 2013; Saad et al., 2012). Various biopolymers such as cellulose, wool, alginate and silk have been successfully dyed using these natural dyes (Karthik & Rathinamoorthy, 2018). Hou et al., used Sorghum husk extract to dye the wool fabrics with and without the metal mordants and showed good finishing over the fabric (Hou et al., 2017). Frose et al., reported the dyeing efficiency of different natural dyes used on variant textiles from natural fibers, with or without mordant (Fröse et al., 2019). Imani et al., developed deep dyeing of wool yarns using Madder dye and metallic mordants and found a good, colored texture to the fabric (Imani et al., 2022). While natural dyes offer biodegradability and non-toxicity, their industrial adoption remains hindered by two systemic flaws: (1) reliance on heavy metal mordants (Fe, Cu, Pb) that transfer toxicity from factories to finished garments (Haddar et al., 2014; Hosseinneshad et al., 2021), and (2) water-intensive post-spinning dyeing processes consuming 150 L/kg fabric.

Spin dyeing, which incorporates dyes into the polymer solution, represents a more sustainable method by significantly reducing energy consumption, carbon footprint, and water usage compared to traditional dyeing processes. This method conserves resources and lowers the operational costs for textile manufacturers, making it economically viable. Dope-dyeing of regenerated cellulosic fibers has been carried out by Manian et al., (Manian et al., 2022), where they have successfully imparted leuco-indigo dyes into dopes prepared during a viscose process. However, the viscose process utilizes carbon disulphide (CS₂) for xanthogenation step, which is associated with major environmental and health concerns surrounding its use (Drexler et al., 1995). A recent report compiled by Duarte et al., (Duarte et al., 2024), has stated that the deposition of CS₂, from decomposition of xanthates used in mining industry, pose adverse effects to the water quality and the biosphere in the areas of study. Current "sustainable" alternatives like viscose merely shift environmental burdens – its CS₂-based production emits neurotoxic byproducts, while dope-dyed synthetics perpetuate microplastic pollution.

The textile industry's environmental footprint demands urgent

transitions to circular material systems. Cellulose emerges as a paradigm-shifting biopolymer, combining natural abundance with full biodegradability, offering fossil-free alternatives to synthetic textiles, as well as a means to repurpose discarded textile waste into high-value products (Négrier et al., 2023). Recent advances in ionic liquid processing have overcome historical barriers to cellulose's industrial adoption, enabling near-quantitative solvent recovery and energy-efficient fiber spinning (Kuzmina, 2016). This aligns with Weyhrich and others advocacy for polysaccharides as nature's underutilized design platform, where their hydroxyl-rich surfaces facilitating non-covalent interactions with plant-derived dyes without toxic mordants (Weyhrich et al., 2023). Existing regenerated cellulose fibers (e.g., viscose) remain chemically intensive while generating hazardous byproducts (Budtova & Navard, 2016). Our approach innovatively integrates ionic liquid-mediated cellulose dissolution through in situ natural dye incorporation – a dual-function process eliminating separate dyeing stages. We demonstrate stable coloration through phenolic interactions rather than covalent modification, preserving fiber crystallinity and tensile strength. By attempting to quantify energy savings and water reduction against conventional dyeing, we aim to advance life-cycle assessment frameworks for biopolymer textiles. The work uniquely bridges three domains: 1) solvent engineering, 2) polysaccharide functionalization strategies, and 3) performance and sustainability metrics, establishing cellulose as a performance-competitive, ecologically regenerative matrix for next-gen apparel.

This work introduces a paradigm-shifting approach leveraging ionic liquids (ILs) as dual-function media to simultaneously dissolve cellulose and disperse natural dyes, enabling single-step fiber production and coloration. We hypothesize that ILs' capacity to disrupt cellulose hydrogen bonds while solvating dye phytochemicals allows mordant-free dye incorporation without sacrificing mechanical performance. By eliminating separate dyeing stages, our process targets water reduction at indispensable levels and significantly lower CO₂e versus conventional methods – metrics critical for meeting UN Sustainable Development Goals on clean water (SDG6) and responsible consumption (SDG12).

Experimental

Materials

Wood Pulp Cellulose (WPC) sheets (cardboard sheets) having a degree of polymerization of 890 were received from Rayonier Inc. (Jacksonville, US). Sorghum, Coreopsis, Brazilwood and Woad were used as natural dyes which were procured from Wild Colors, UK. 1-ethyl-3-methylimidazolium diethyl phosphate ionic liquid (EMIM DEP, >95 %) was used as the solvent, which was purchased from Sigma Aldrich, UK. The materials were used as received without any further purification.

Cellulose/natural dye fiber spinning

The pulp sheets were cut into small pieces which were further ground using a blender. Cellulose (6 % w/w) was added to ionic liquid and the solution was then stirred at 80 °C using a magnetic hot plate (IKA, UK) until complete dissolution. The solution preparation was carried out in an oil bath to maintain a uniform temperature throughout the dissolution process. The natural dyes were ground to fine powder using a mortar and pestle, prior to the solution preparation. The concentration (Coscia et al., 2018) of natural dyes is 7 % w/w with respect to the amount of cellulose present in the cellulose/ionic liquid solution and stirred. The solution was then transferred to a 60 ml Luer entrapped air in the mixture. The degassed solution was then used for fiber-spinning process using a lab-built bespoke wet-spinning equipment. The amount of cellulose and the dye fraction have been given in Table S1. The lab-built equipment (shown in Fig. 1) used for wet-spinning consists of 3 sections, which are the extrusion pump, the deionized water (Coscia et al., 2018) bath for coagulation, and the winding machine. Process parameters such as extrusion die diameter, fiber extrusion speed, and

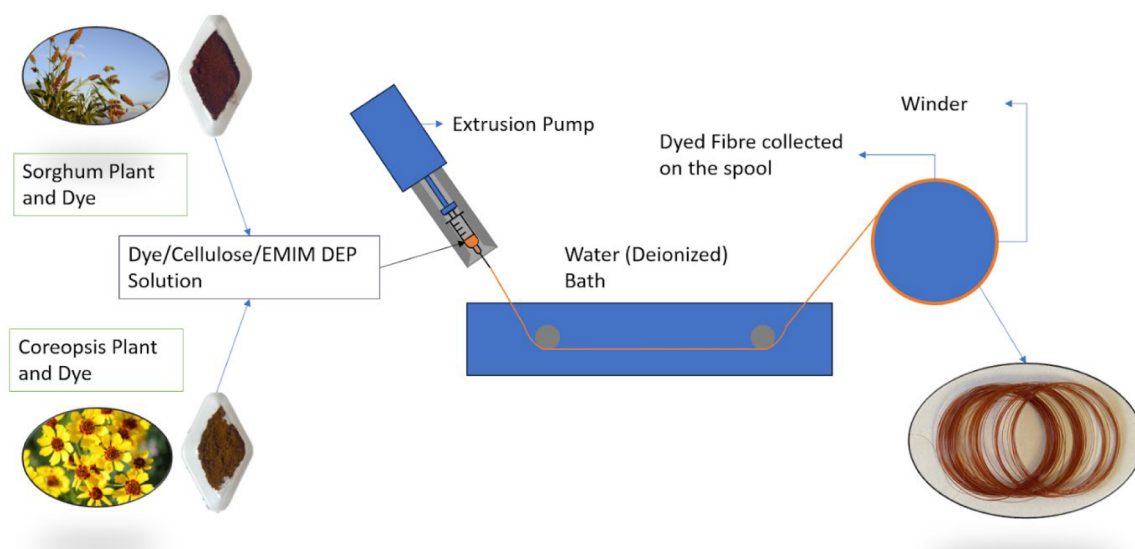


Fig. 1. Experimental set-up for the wet-spinning process.

fiber winding speed can be used to alter the physical properties offered by the fibers. Hence, in the current study the fibers spinning parameters have been taken as constants for all samples to compare properties of fibers spun from different dye solutions.

The final solution after degassing was used for fiber spinning process with water as coagulation medium. The prepared solution of cellulose/natural-dye/ionic-liquid was injected into coagulation bath using the syringe pump (Chemyx, USA) at a flow rate of 2 ml / min. The fibers were subjected to water with an air gap of 2.5 cm between the coagulation bath and the die. The air gap results in the better alignment and crystallinity of the fibers when spun at higher extension rate (Haward et al., 2012). The collector was at the other end of the bath and the fiber were collected after passing through the coagulation medium at the winding velocity of 1.86×10^{-1} m/s (draw ratio – 5.89) (Haward et al., 2012). After fiber spinning, the fibers were rolled and immersed under water for 48 hrs. with change of water every 12–16 hrs. to remove the ionic liquid. The washed fibers were air-dried at room temperature. The dried fibers were carefully unwound and stored in an airtight bag and kept away from moisture.

Ionic liquids (ILs) such as 1-ethyl-3-methylimidazolium diethyl phosphate (EMIM DEP) are promising solvents for cellulose dissolution and processing due to their unique physicochemical properties. However, the economic and environmental sustainability of IL-based processes critically depends on effective IL recovery and reuse, as these solvents are costly and susceptible to degradation under certain conditions. Although numerous studies have demonstrated efficient IL recycling and purification methods, including advanced techniques such as electrodialysis, liquid-liquid extraction, distillation and adsorption (Gong et al., 2023; Khoo et al., 2024). However, these recovery protocols typically require dedicated systems and scale beyond laboratory feasibility. In the present study, we focus primarily on demonstrating the feasibility of spin dyeing cellulose fibres using EMIM DEP at a laboratory scale, where IL recovery is not yet economically or practically viable. Future scale-up work will incorporate established IL recycling strategies to optimize solvent reuse and sustainability.

Microscopic morphology of dyed regenerated cellulose fibers

The fiber diameter of the prepared wet spun fibers was carried out at room temperature using the Leica S9D Microscopy (Nikon Eclipse ME600). The fiber images were observed at four different locations to find the average fiber diameter of the wet-spun fibers. The scanning electron microscopy (SEM) analysis of the developed fibers were carried

out with VEGA3 TESCAN scanning electron microscopy. The samples were gold coated in a vacuum evaporator before performing the characterization.

Fourier transform infrared spectroscopy (FTIR) of dyed regenerated cellulose fibers

The changes in the structure of the fiber samples were analyzed using the Fourier Transform Infrared Spectroscopy (Thermo Scientific™ Nicolet™ iS20 FTIR Spectrometer). Cumulative scans (100) were taken with a resolution of 4 cm^{-1} in the wave number range of 4000 to 600 cm^{-1} under the absorbance mode.

Tensile testing of dyed regenerated cellulose fibers

The fiber samples were subjected to the tensile test using the Microtest tensile stage controller (DEBEN, UK) along with the Deben Microtest software at room temperature. The rate and the gauge length for the analysis were fixed at 0.2 mm/min and 10.2 mm respectively, in accordance to the ASTM Standard Test method for Tensile Testing of Single Textile Fibres, ASTM D3822/D3822M-14(2020). Each fiber strand sample was characterized three times to obtain the corresponding average values of the different mechanical properties' parameters like tensile strength, strain, and Young's modulus. The tensile strength represented the maximum stress obtained while performing the test while the modulus was calculated from the slope of the linear part of the curve. The comparison of the prepared natural fibers with other materials was carried out using the Ansys Granta Edu Pack 2022R2 (Ansys Inc., USA) software licensed by Cranfield University.

Thermogravimetric analysis (TGA) of dyed regenerated cellulose fibers

Thermogravimetric analysis has been carried out on the fibers using TA Instruments Q500 analyzer, at a $10 \text{ }^{\circ}\text{C/min}$ heating rate in the range of $20 \text{ }^{\circ}\text{C}$ to $600 \text{ }^{\circ}\text{C}$. The undyed sample has been taken as the control and the dyed samples were analyzed against it. Polymer decomposition reactions were carried out on about 5 mg of cellulose fibers – dyed and undyed. A sample flow rate of 40 ml/min using Nitrogen gas was used to study the decomposition.

Color strength of dyed regenerated cellulose fibers

Color characteristics were analyzed using a Datacolor 700™ Spec-

trophotometer [DC 700] (USA) at Manchester Metropolitan University, UK. The following parameters were examined – color hue, brightness, color yield [K/S] and reflectance. The color change was evaluated using D65 illuminant [standard], A10 [domestic lighting] and F02 [fluorescent] at 10° standard observer in a visible spectrum 400–700 nm. K/S represents the color yield and is evaluated using Kubelka Monk theory (Nobbs, 1985).

$$K/S = [1 - R^2]/2R$$

Where K is the absorption coefficient, and S is the scattering coefficient.

The test cellulose fibers were wound on an acrylic tab [300 × 500 mm] and conditioned [20 ± 5 °C and 65 % relative humidity] in the laboratory for 24 hrs. before testing. The calibrated equipment used cellulose fibers (4 % Sorghum or Coreopsis) as standard and compared with test cellulose fibers (7 % Sorghum or Coreopsis-dyed fibers). Measurements were repeated five times, and the average results were reported.

Environmental benefits from dope dyeing

A concise study in the energy, water and carbon savings through replacing conventional dyeing with dope-dyeing in a wet-spinning facility for regenerated fibers is shown in the supporting documentation, S1-S6.

Results and discussions

Initial investigations of the dyed fibers

Fig. 2 shows the dissolution behavior of Sorghum, Coreopsis, Woad, and Brazilwood dyes in the resultant fibers. The behavior of each dye in the Cellulose-IL composite is listed in Table 1. Dyes that were homogeneously dissolved in the cellulose/ IL was referred to as ‘Complete’

solutions, as evident in Fig. 2. Dyes that were non-homogeneously dissolved in the cellulose/ IL was referred to as ‘Partially’ dissolved solutions. Solutions that were able to consolidate into fibers in the coagulation bath were referred to as being ‘spinnable’. The dyes when incorporated into the cellulose/ionic solution through *in-situ* dyeing or dope dyeing method show complete dye dissolution. On fiber spinning of the dyed solution in presence of water as coagulation medium, color fading was observed in case of Woad dye, however, the color was maintained in Coreopsis and Sorghum dyes as shown in Fig. 3. Brazilwood had undergone an unexpected change in its color profile while washing, and this new color was observed to be retained even after drying. Brazilwood dyes interact with metal ions such as Ca²⁺ and Mg²⁺, which are prevalent in tap water, forming metal-dye complexes that alter the dye’s molecular structure and, consequently, its colour. The observed drastic colour change in brazilwood-based dye upon interaction with tap water can be explained by the chemistry of its principal chromophore, brazilein, which is known to be highly sensitive to pH and the presence of metal ions (Manhita et al., 2013; Nabais et al., 2021). Depending on the metal ion, the pigment may shift to appear redder, more violet, darker, or lighter. This effect is especially pronounced with certain ions, as documented in research showing significant hue and intensity changes with Al³⁺, Ca²⁺, and Pb²⁺. Additionally, brazilein’s colour expression is more acid-sensitive than similar dyestuffs (like hematein), and its structure allows for rapid complexation with available metal cations (Puchtler et al., 1986).

This study focuses on establishing the feasibility of fabricating dyed regenerated cellulose fibres using a single wood-derived cellulose substrate and a select set of natural dyes. While it is well recognized that the choice of cellulose source and dye chemistry can significantly influence physio-mechanical, chemical, and coloration properties of fibres (Shamsuri et al., 2021), comprehensive investigations involving multiple substrates and dye systems are beyond the current scope. Such studies, including fastness performance, will be pursued in future work to further elucidate these important parameters.

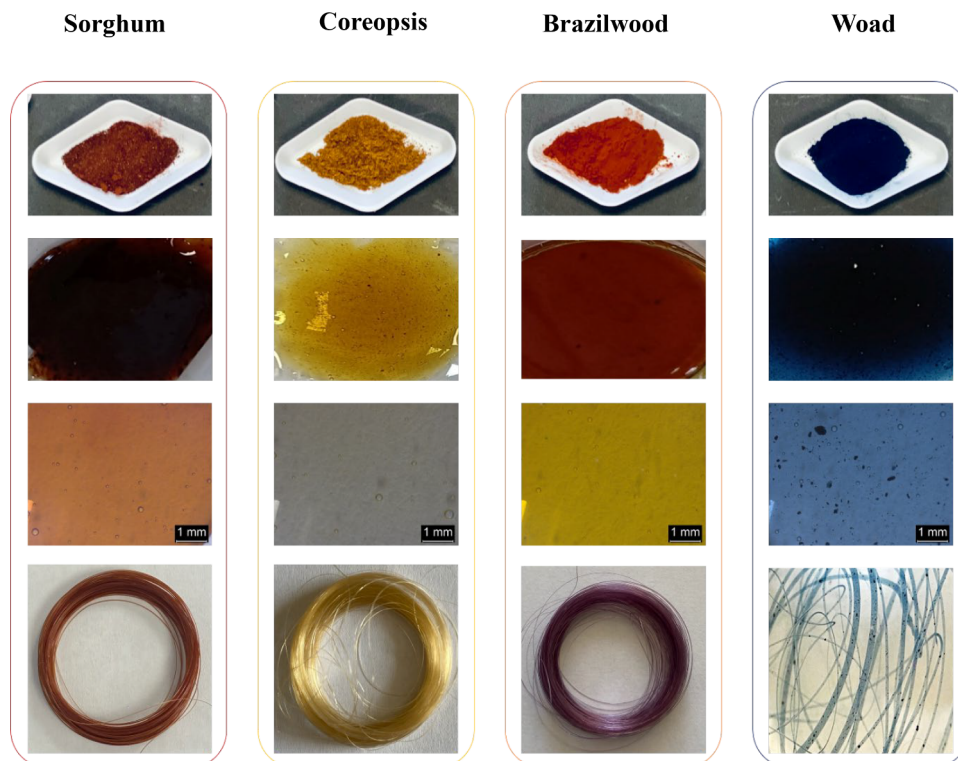


Fig. 2. Collage of images depicting the various stages of the dyes during the fibre development. Starting with the powder form of the dye, which is dissolved in the Cellulose/IL composite solution, followed by a magnified observation of dissolved dye; and the resultant fibres.

Table 1
Interactions between different dyes and Cellulose-IL Composite.

Composition of the dope	Compatibility with solvent	Spinnability after dye addition	Final color appearance after washing	Dye dispersibility in the fiber	Color retention after solvent removal
Cellulose + Coreopsis	Complete	Y		Y	Y
Cellulose + Sorghum	Complete	Y		Y	Y
Cellulose + Brazilwood	Complete	Y		Y	☒
Cellulose + Woad	Partial	Y		☒	Y

On closer view of the Woad dye-based cellulose fibers in water, the dye was not completely mixed, Fig. 3a, and agglomerates of undissolved dye remain in the fiber. The patchy remnants of undissolved dye could have been the lack of affinity of the Woad particles with the EMIM DEP solvent (Chu et al., 2021). Hence, Woad was not used for further testing as it does not align with the objectives of the present work. Fibers containing Coreopsis (Fig. 3b) and Sorghum (Fig. 3c) dyes showed uniform color dispersion throughout the fiber volume. The Sorghum gave a bright orange-red hue, while the Coreopsis gave a bright yellow shade, and were further tested in the ensuing work.

Coreopsis Tinctoria and *Sorghum Bicolor* provide vibrant natural dyes with minimal health risks. The color in coreopsis is the result of compounds such as chalcone, and aurone, which are predominantly present in the flowers of the plant (Melo, 2023). The leaves and stems of the plant, *Sorghum Bicolor*, contain tannins that produce earthly shades of tan on fabrics (Dykes, 2019). Previous reports established the safety and lack of health risks of the colorants found in Sorghum (Olifson et al., 1978).

Chemical structures for the cellulose repeating units and each principal natural dye utilized in this work are provided in Fig. 4 to highlight relevant binding functionalities.

Schematic illustrations highlight the plausible cation- π interactions between the aromatic rings of the phenolic dye molecules present in Sorghum and the cationic components of the ionic liquid near the cellulose fibre surface (Fig. 5). Such interactions are widely recognized in the literature as important contributors to supramolecular dye-fibre affinity in ionic liquid media (Dong et al., 2023; Hosseini et al., 2025; Rabideau et al., 2014).

For Coreopsis dye, the dissolution efficiency of Aurone and Chalcone molecules in the imidazolium-based ionic liquid EMIM-DEP can be understood in the context of the IL's cation alkyl chain length and core structure. Increasing the alkyl chain length of the imidazolium cation enhances the hydrophobicity and reduces the polarity of the IL, strengthening van der Waals interactions between the IL and the hydrophobic aromatic regions of the dye molecules (Dong et al., 2023). Additionally, the steric hindrance around the imidazolium C2-proton

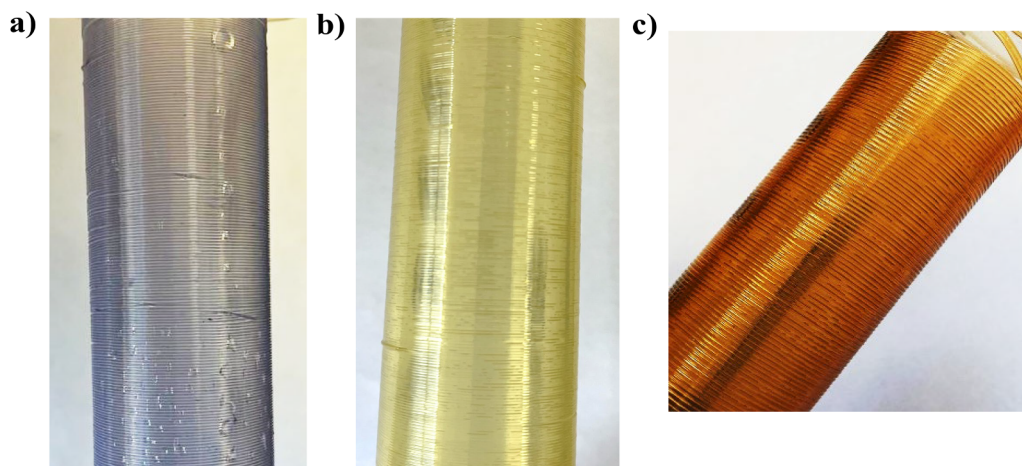


Fig. 3. Pictorial images of the washed wet-spun fibres in presence of a) Woad; b) Coreopsis; c) Sorghum dye (before drying).

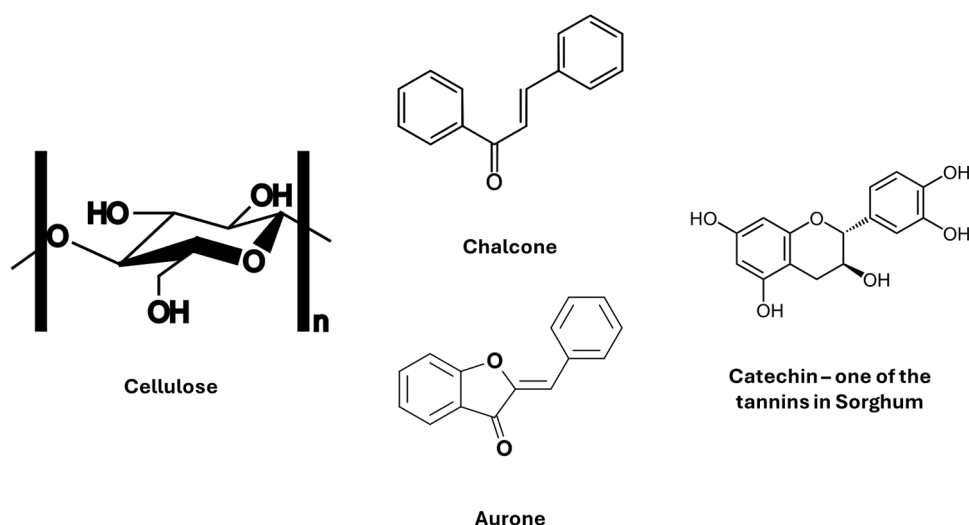


Fig. 4. Chemical structures of cellulose, and major colour causing molecules in Coreopsis (Chalcone & Aurone) and Sorghum (Catechin - one of the many tannin compounds present in sorghum).

decreases its acidity and weakens the anion-cation interaction, thereby increasing the hydrogen bond acceptor (HBA) ability of the diethyl phosphate anion. This enhanced HBA strength facilitates stronger hydrogen bonding with polar functional groups on Aurone and Chalcone, improving their solubility. Moreover, the imidazolium ring's C2-proton can form hydrogen bonds with carbonyl groups present in these flavonoid structures, further promoting solvation (Dong et al., 2023). These combined effects explain why EMIM-DEP effectively dissolves Aurone and Chalcone molecules, supporting efficient dye uptake during the fibre spinning process. Although direct experimental verification was not feasible in the present study, these schematic models and supporting literature provide mechanistic rationale for the observed dye uptake and retention.

The sustainable spin dyeing method for textile dyeing is more economical and environmentally friendly in comparison to the conventional dyeing method. According to Terinte et al., (Terinte et al., 2014) spin dyeing technique requires around 60 % less carbon footprint, 50 % less energy and 50 % less water as compared to the conventional dyeing method. Utilizing the sustainable spin dyeing method, the environmental consequences generated from the conventional dyeing can be minimized by 40–60 %. Based on the concept of sustainable production and consumption, the current work embodies the manufacturing of benign fibers using natural dyes and biopolymer and

provides an insight for sustainable clothing and fashion applications. To further verify the efficacy of the dyed fibers, the cellulose/Woad fibers and cellulose/Sorghum fibers were dipped in water after wet spinning and the release of dye was examined. The Woad dye is not well incorporated into the solution resulting in release of the dye when washed in water. On the other hand, the Sorghum dye showed no evident release of dye in water which can be attributed to the better coalescence of the dye while solution preparation.

Preliminary colour performance assessments in this study were limited to simple water rinsing to evaluate initial dye uptake and colour retention. The importance of standardized colour fastness testing, such as wash, rub, light, and perspiration fastness following ISO 105 protocols, to fully establish the durability and industrial relevance of the dyed regenerated cellulose fibres is acknowledged. However, due to limitations in the current testing setup and the scope of this initial feasibility study, comprehensive fastness evaluations, including ΔE measurements after standard laundering, are planned for future work. Implementation of robust and reproducible testing protocols will enable thorough assessment of post-dyeing durability under diverse conditions.

Microscopy of dyed regenerated cellulose fibers

The air-dried fibers were placed under the microscope to have an

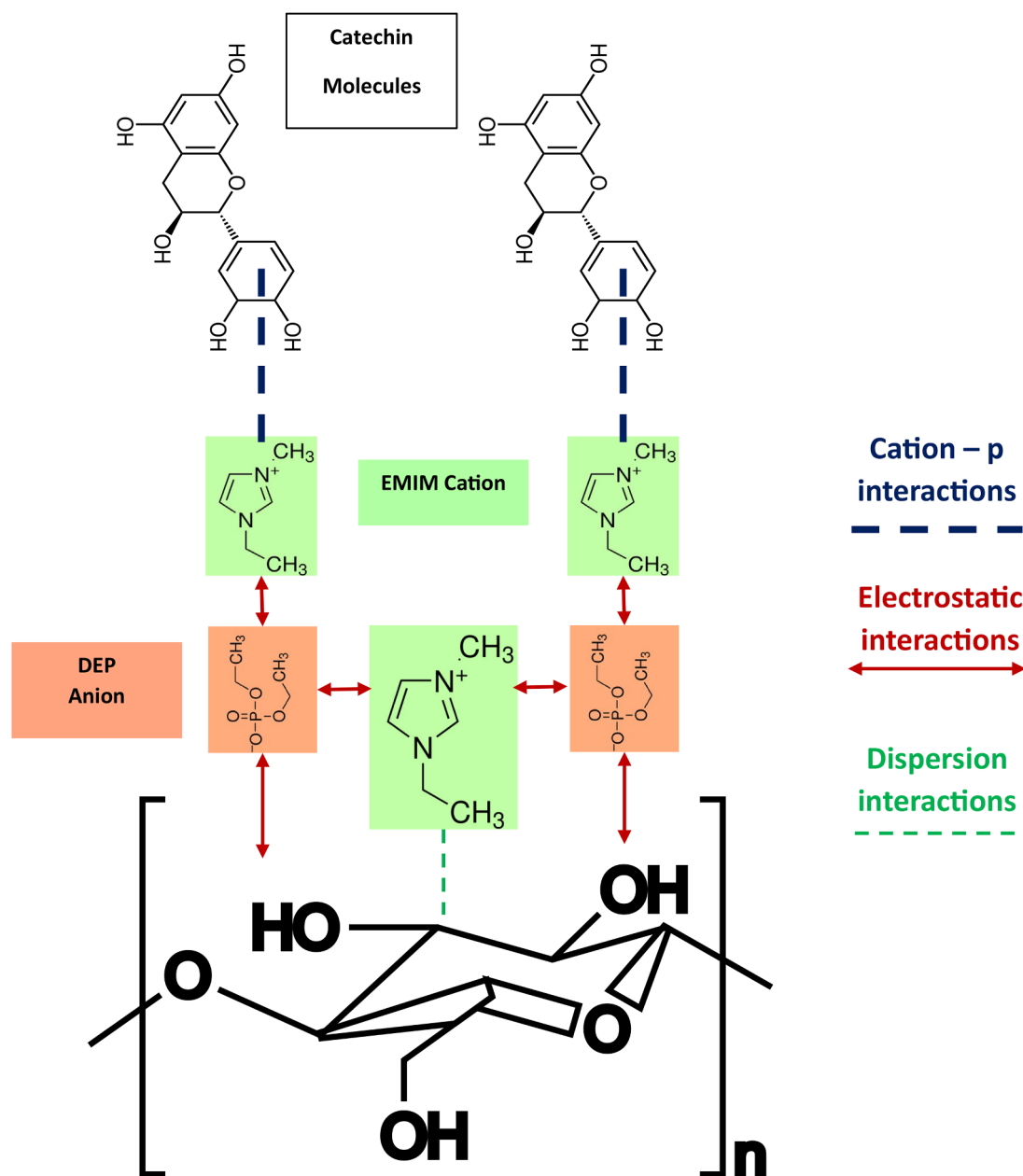


Fig. 5. Schematic of the ionic interactions taking place near the cellulose molecule and dye molecule from Sorghum, facilitated by the cation (EMIM) and anion (DEP) interactions of the ionic liquid 1-ethyl-3-methylimidazolium (EMIM) diethyl phosphate (DEP).

enlarged view of the natural dye-incorporated cellulose fibers. The prepared fibers were further observed under the optical microscopy to investigate the diameter of the fibers. The average fiber diameter for neat cellulose fibers was $84.88 (\pm 11.20) \mu\text{m}$. The addition of dye to the cellulose results in reduced fiber diameter in the case of Sorghum. The infusion of Coreopsis dye results in increased fiber diameter of $90.76 (\pm 8.73) \mu\text{m}$, and for Sorghum fibers was $73.73 (\pm 4.2) \mu\text{m}$. Diameter of the neat cellulose fibers and Sorghum infused cellulose fibers were found to be comparable.

From the SEM images, as shown in Fig. 6, it can be visualized that with the addition of the dye, the surface morphology of the fibers shows no significant alteration. The pristine cellulose fiber shows a good, smooth surface with no substantial voids which on addition of the dyes gets even better and smoother. Similar observations can be made from the surfaces of Coreopsis, and Sorghum fibers.

Table 2 gives the surface morphology of the fractured cross-section of the developed fibers. No clumps of Coreopsis or Sorghum dyes were

found in the SEM images of the fibers. The cellulose/dye composite fiber cross-section does not hold any voids signaling the strong bonding between dye and cellulose molecules (Coscia et al., 2018). Therefore, the SEM analysis of the fiber cross-section indicates the homogenous dispersion of the dyes. Hence the *in-situ* spin dyeing of the cellulose-natural dye fibers provides a better finished surface which highlights the efficacy of the developed process for fiber spinning.

FTIR analysis of the dyes and dyed fibers

Fig. 7 (a and b) gives the IR spectra of pure dyes and depicts the comparison of undyed fibers to fibers dyed with Coreopsis and Sorghum. Changes in absorption bands, for the neat and dyed fibers, occur in the wave number regions of $3500\text{--}3000 \text{ cm}^{-1}$ and $1200\text{--}700 \text{ cm}^{-1}$. The spectra present in these bands correspond to the FTIR results found in literature for cellulose fibers prepared from other wood pulp derivatives (Hospodarova et al., 2018). The peaks seen in the wave number range

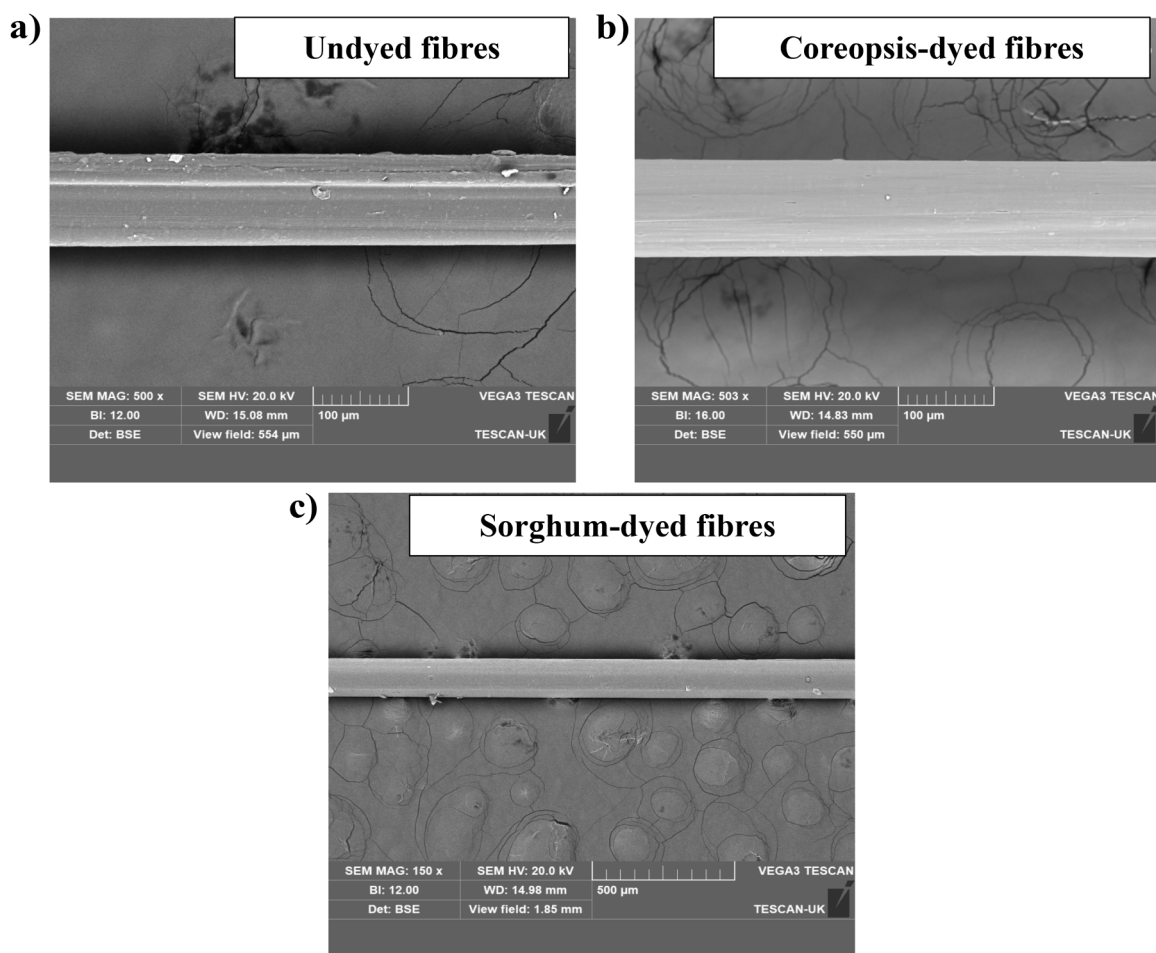


Fig. 6. SEM morphology of wet spun fibres from a) undyed cellulose; b) Coreopsis-dyed and c) Sorghum-dyed regenerated cellulosic fibres.

from 3500–3000 cm^{-1} correspond to the stretching vibrations of O—H and C—H bonds in polysaccharides. The broad peak at 3330 and 3285 cm^{-1} in Fig. 7 (a and b, respectively) is a characteristic of hydroxyl group bond stretching in polysaccharides. The peaks at 1020 cm^{-1} are attributed to the C—O bond stretching in the C—O—C group in the anhydro-glucose ring (Değermenci et al., 2019). It gets more pronounced on the addition of dyes with Sorghum demonstrating an exaggerated increase compared to Coreopsis. The sharp peak at 800 cm^{-1} corresponds to the glycosidic C—H deformation with ring vibration contribution and O—H bending, which is characteristic of β -glycosidic linkages between glucose in cellulose (Kadokawa et al., 2008). There is a significant shift in the peaks around 1650 cm^{-1} (refer to full spectra in Supplementary Information, Figure S1) in both the cases due to the increased C = C bending vibrations associated with addition of the dyes which are assumed to contain flavonoids (Deng et al., 2017) and tannins (Lin et al., 2021) that have shown to cause similar shifts as a result of aromatic ring presence in previous works done by Ahmed et al., and Taherkhani et al., (Ahmed et al., 2024; Taherkhani et al., 2020).

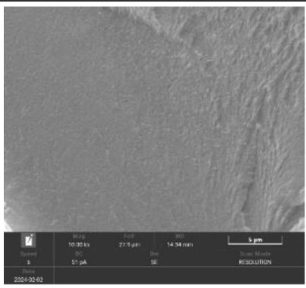
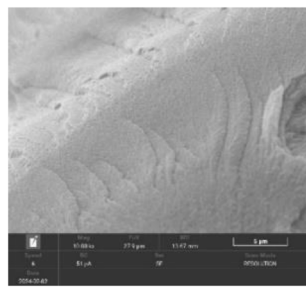
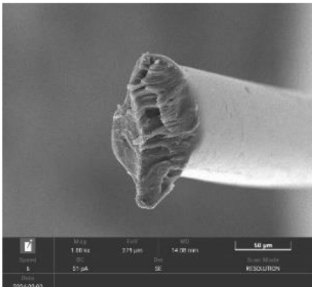
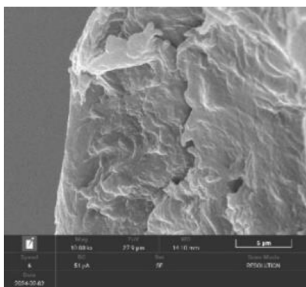
Mechanical properties of dyed regenerated cellulose fibers

The mechanical properties of the manufactured fibers are shown in the Table 3 summarizing the values obtained from the stress-strain test.

Mechanical properties of the developed fibers play a significant role for their practical applications in textile industries. Neat cellulose fibers showed average Young's modulus of 15.7 (± 1.3) GPa and tensile strength of 210.3 (± 29.6) MPa, respectively. On incorporation of Sorghum dye, the mechanical properties of the dyed cellulosic fibers are comparable with the pristine cellulosic fibers. On increasing the

Sorghum dye content further, corresponding increase in the mechanical properties of the fibers was observed. Sorghum-derived dyes are high in phenolic content, which can foster hydrogen bonding with cellulose chains, potentially reinforcing the fiber matrix at moderate concentrations. Literature suggests that such interactions sometimes improve the compatibility and adhesion between the dye and cellulose, thereby maintaining or slightly improving mechanical properties as the dye content increases (Reddy & Yang, 2007; Yulianto et al., 2020). On the contrary, deterioration of the mechanical properties was observed by increasing Coreopsis dye concentration. This could have been due to the saturation of Coreopsis molecules in the fiber structure, since Coreopsis dye may contain molecular species (e.g., flavonoids) that, when present in excess, can act as plasticizers or disrupt the intermolecular hydrogen bonding among cellulose chains. At high concentrations, dye saturation may lead to phase separation, increased amorphous regions, or weak interfaces between dye aggregates and cellulose, which can reduce the overall tensile strength. The way flavonoids interact with synthetic polymers like polyolefins (often causing phase separation due to incompatibility), as shown by Bridson et al., is directly analogous to their plasticizing behavior in cellulose-rich systems (Bridson et al., 2015). Both phenomena arise from the ability of flavonoids to disrupt the existing intermolecular interactions, sometimes beneficially (as stabilizers), but also leading to reduced mechanical properties when uniform integration is not achieved or exceeded which is expected to have occurred in this case. In a similar study, Coscia et al., have reported that the addition of curcumin beyond a certain threshold acts as an impurity in the matrix and hinders the hydrogen bonding in the cellulose matrix (Coscia et al., 2018). Costa et al., developed dyed bacterial cellulose (BC) films using plant-based natural dyes in presence of color fixer and

Table 2
SEM images of fiber cross-section at magnifications of 1000x, and 10,000x indicating the uniform dye dispersion in the fiber.

Sample	1000x	10000x
Undyed		
Coreopsis-dyed fibers		
Sorghum-dyed fibers		

softener. On comparing the mechanical properties of the prepared film, the natural dye-based BC films showed better results as compared to the BC film with synthetic dyes. The maximum elasticity obtained in the work was around 458.38 ± 5.59 MPa. From the obtained results, it was suggested that the natural dyes caused less change in the plasticity of the films in comparison to the synthetic dyes (Costa et al., 2019). Hence, addition of the dyes does not affect the fiber strength up to a certain threshold, after which the mechanical properties of the fiber deteriorate. However, this technique provides a sustainable way to develop *in-situ* spin dyed cellulose fibers minimizing the harmful impacts of the synthetic dyes & solvents used in conventional dyeing process. The mechanical properties obtained were compared with some of the commercially available materials using an Ashby chart as shown in Fig. 8.

The current dyed regenerated cellulose fiber shows promising result and better mechanical properties than some of the available fibers such as wool, coir, palm, and viscose (Enka) (Michael F. Ashby et al., 2018). Hence the approach to manufacture the sustainable naturally dyed-based cellulosic fibers using benign and cleaner route is well established based on the observation and analysis which could be potential alternate to the conventional dyeing and a viable fiber

manufacturing technique for sustainable fashion industry.

The regenerated cellulose dopes used in this study contained a relatively low cellulose concentration of 6 wt. %, yet the spun fibres exhibited high tensile strengths exceeding 200 MPa, indicating significant molecular alignment and fibre integrity. While detailed rheological characterization of the spinning dopes, such as viscosity measurements, spinneret shear rates, and draw ratio evolution, would provide valuable mechanistic insights into the fibre orientation process, the complex interactions between cellulose and natural dyes in our system induce non-Newtonian and variable rheological behaviour. This complexity complicates viscosity interpretation and reproducibility, making such studies beyond the current scope.

Instead, the high orientation and structural integrity of fibres is corroborated through complementary characterization techniques including tensile testing, FTIR, and TGA, which consistently support strong polymer chain alignment and preserved cellulose structure. Literature evidence further demonstrates that regenerated cellulose fibres with comparable mechanical properties can be produced at similar or even lower cellulose concentrations using ionic liquid-based spinning systems (Coscia et al., 2018; Rahatekar et al., 2009).

The value of detailed rheological studies is acknowledged, and plans

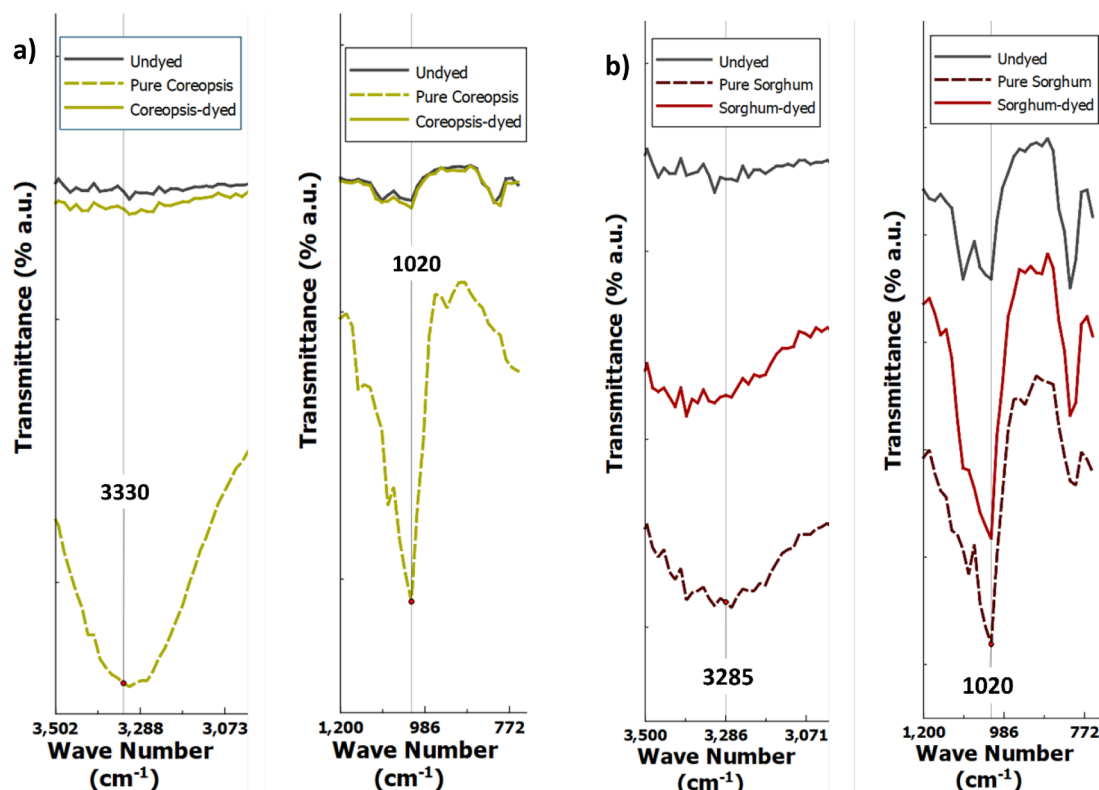


Fig. 7. FT-IR curves of the undyed cellulosic fibres compared against pure dye and the dyed fibres for a) Coreopsis and b) Sorghum dyes.

Table 3
Mechanical properties for the prepared natural dye-based cellulose fibers.

Sample	Diameter (μm)	Strain at Break (%)	Tensile Strength (MPa)	Young's Modulus (GPa)
Undyed	84.9 ± 11.2	9.1 ± 2.2	210.3 ± 29.6	15.7 ± 1.3
Coreopsis	90.8 ± 8.7	7.4 ± 1.1	205.1 ± 18.9	15.4 ± 1.7
Sorghum	73.7 ± 4.2	18.6 ± 2.1	204.1 ± 17.9	15.0 ± 1.3

will be put in motion to include them in future research to better elucidate the spinning dope behaviour and its impact on fibre micro-structure and performance.

Thermogravimetric analysis (TGA) of the fiber samples

Thermal stability of the pure fiber and the dyed samples was analyzed through TGA. The results, shown in Fig. 9, indicate that all the fibers exhibit pronounced weight loss around 100 °C, which can be attributed to the evaporation of residual water present in the fibers (Liu et al., 2011).

The significant weight loss of control (undyed) sample between 283–365 °C can be attributed to decomposition of cellulose (Değermenci et al., 2019; Yang et al., 2007). The addition of Sorghum dye has led to an early start of decomposition, at 262.08 °C. Coreopsis dye in the sample is observed to have led to a decrease in the initial decomposition temperature to 241.61 °C. The cause for the lower thermal stability of the dyed samples can be related to the breakage of intermolecular bonds by the ionic liquid, which are now disrupted due to the grafting of the dye onto cellulose structure (Kadokawa et al., 2008). Additionally, the amount of char for the dyed fibers is higher than the control sample indicating that a minor level of organic and inorganic impurities associated with the dye have remained in the dyed fibers (Shang et al., 2016). Further characterization of the ash matter would be included in the future work depending on the need to fully elucidate the

composition through CHN/O analysis or ICP for metal quantification. However, since the intended application is in everyday fashion, the decomposition temperatures for the dyed fabrics are well within the acceptable range ('BS EN ISO 11092:2014').

Color intensity and strength of dyed regenerated cellulose fibers

The color change was evaluated by comparing the *test sample* of dyed fibers against a *standard sample* of the fibers. Test samples refer to fibers containing 7 % Coreopsis dyed fibers, and 7 % Sorghum dyed fibers, while a *standard sample* of the fibers contained lower dye concentration of Coreopsis and Sorghum dyes in the fibers. The relevant observations are tabulated in Table S2.

In the case of Coreopsis-dyed fibers, ΔL [change in lightness or darkness] of the test sample was darker when compared to the standard sample, this can be observed in Fig. 10a, which shows negative values [−6.93] for D65 illuminant. There are minor variations in other illuminants, such as in domestic and fluorescent light. The color component Δa^* indicates the position between red and green. The positive a^* value [1.41] indicates that fibers are red in shade, while Δb^* [8.37] indicates the sample is yellow shade. The ΔE indicates the change in color from the standard sample and it depends on the ΔL^* , Δa^* and Δb^* values. For Coreopsis-dyed cellulosic fibers, the total change in color [ΔE value 11.12] from the standard sample can be noticed, in Fig. 10a. Coreopsis-dyed cellulosic fibers closely align with Pantone shade 458 C, whilst standard fibers align with Pantone shade 459 C.

For Sorghum-dyed fibers, ΔL^* was [−12.74], indicating the sample is darker observed at D65 illuminant. It can also be observed that there is only a marginal difference between the standard and test fibers. The color component Δa^* was [3.49] indicating that fibers are red in shade, while Δb^* negative value [−3.63] indicates that the material is blue. The total color change, ΔE of [13.89], can be observed in Fig. 10b Sorghum-dyed cellulosic fibers closely align with Pantone shade 478 C.

Reflectance values from the spectrophotometer, shown in Fig. 11,

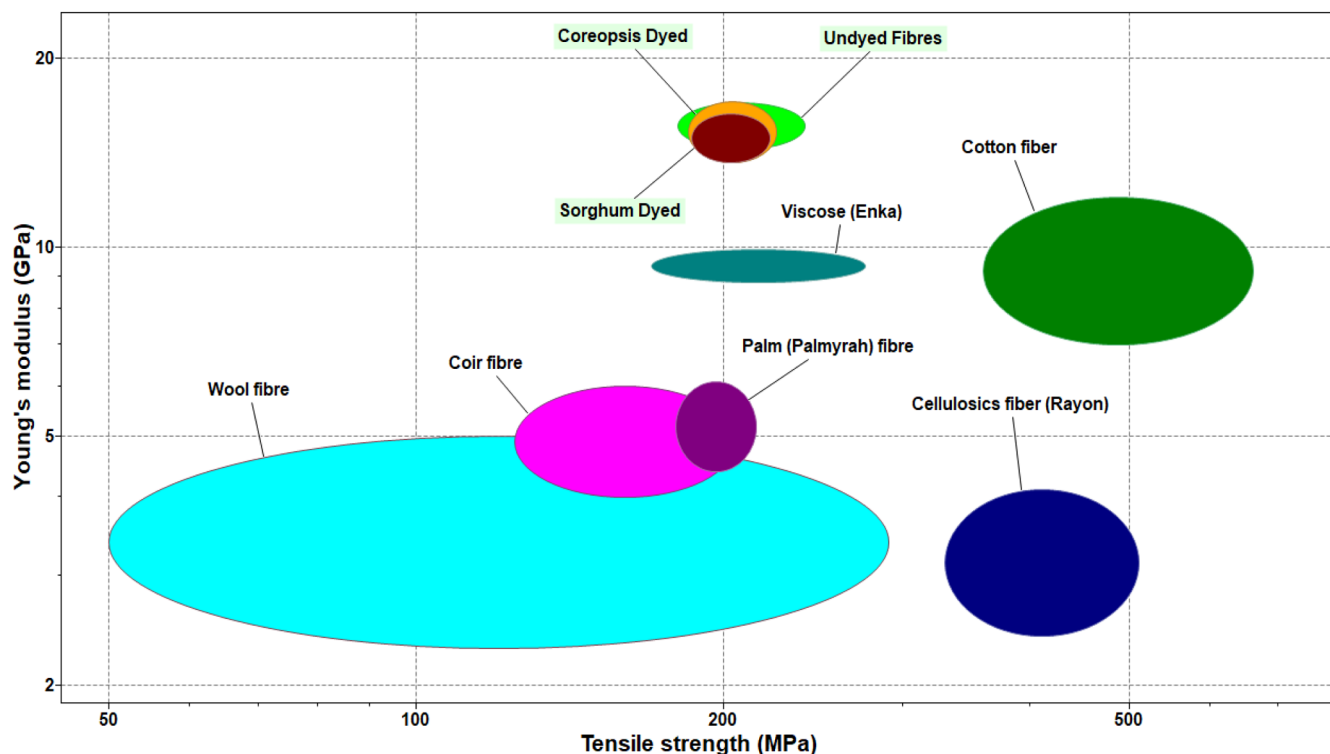


Fig. 8. Mechanical properties (Tensile Strength v. Young's Modulus) of Coreopsis and Sorghum-dyed cellulose fibres, compared to natural polymer fibres (wool, coir, palm, and cotton) as well as commercial cellulosic fibres (viscose and rayon).

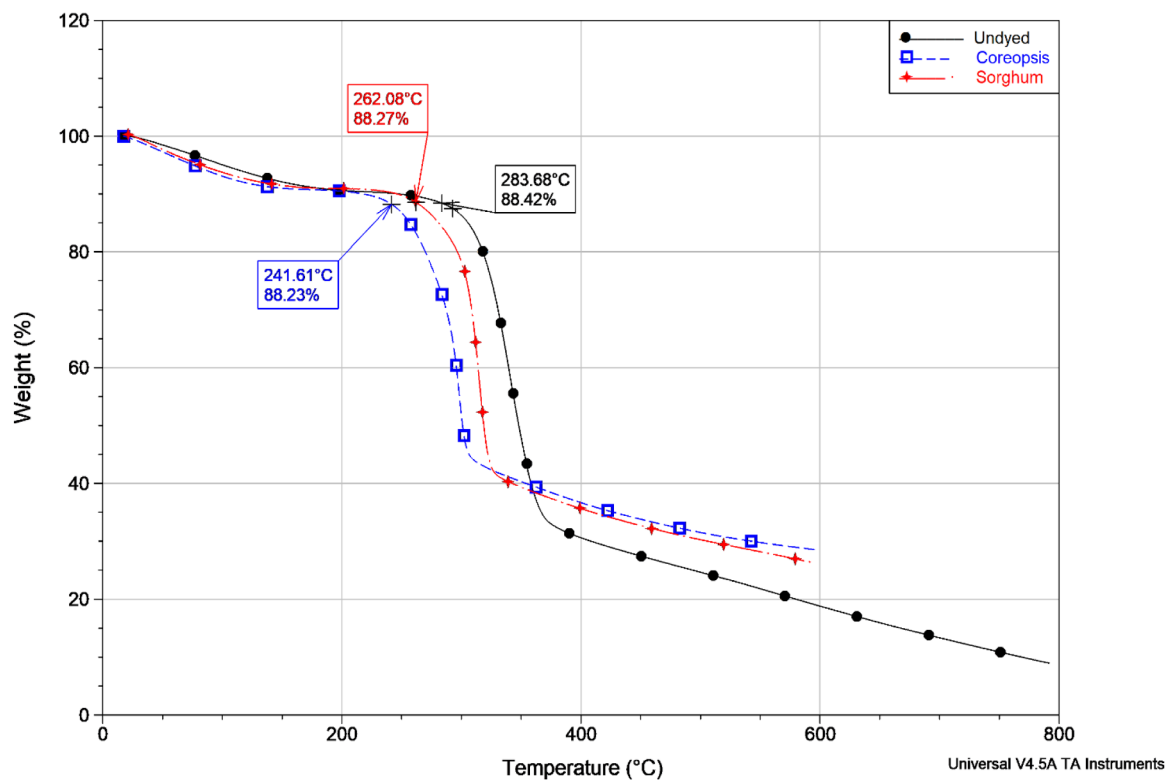


Fig. 9. TGA Curves for the undyed and dyed fibres.

provide the depth of the shade of the cellulose fibers in the visible spectrum. For Coreopsis-dyed fibers, reflectance values were similar for standard fibers and test samples, and peaks were observed between 520–700 nm. These peaks also indicate that the shade of cellulose fibers

can be pale in this region. However, the peaks for reflectance values for Sorghum-dyed fibers were observed between 670–700 nm. Since the Sorghum-dyed cellulose fibers are darker in the shade, the reflectance values were low between 360–600 nm. The overall shape of reflectance

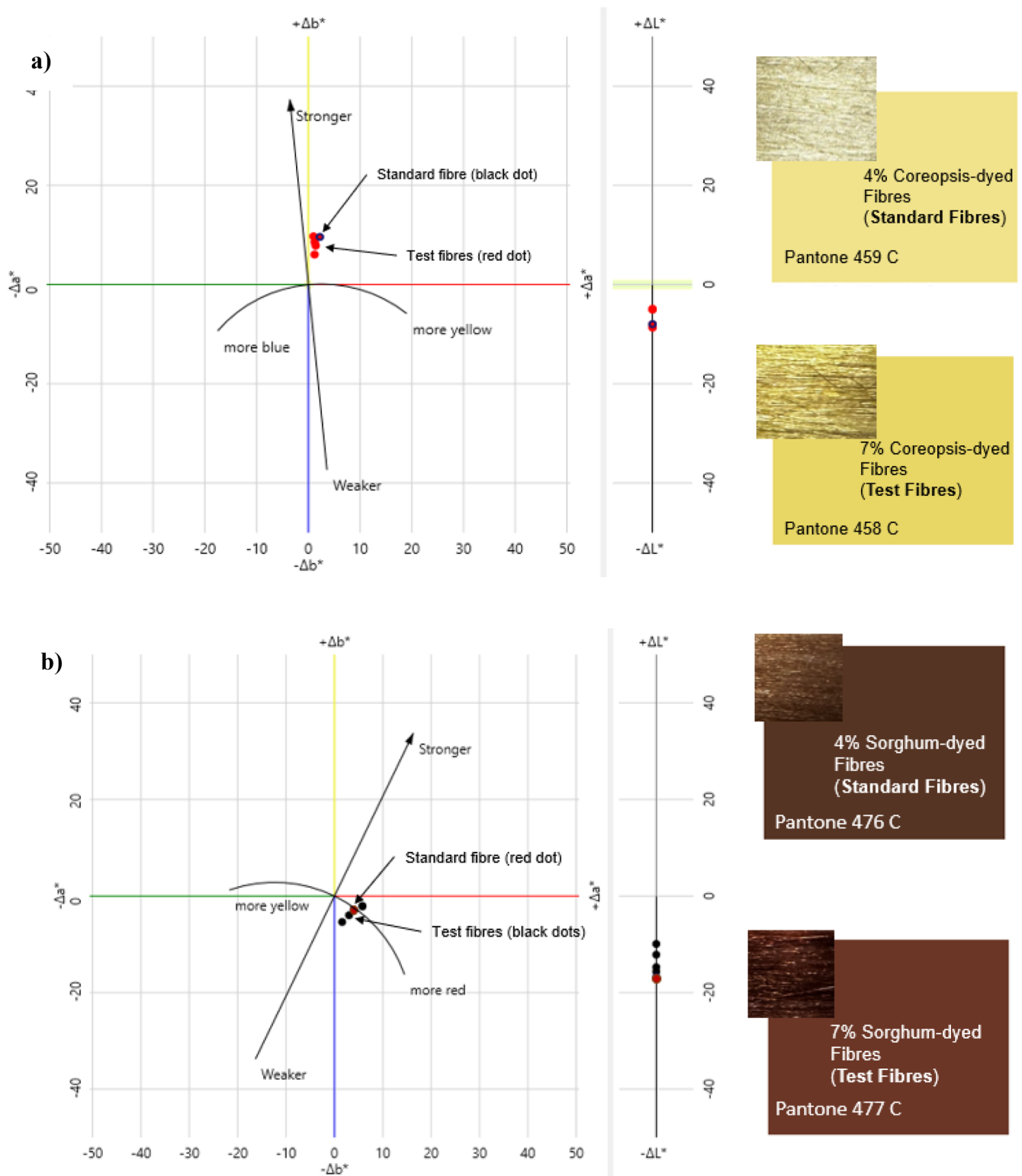


Fig. 10. Change in colour characteristics - a) Coreopsis-dyed cellulose fibres; b) Sorghum-dyed cellulose fibres [ΔL^* , Δa^* and Δb^*].

curves for standard and test samples for Sorghum-dyed cellulose fibres were similar.

Color yield, depicted in Fig. 12, for Coreopsis-dyed fibers had higher peaks of 360–420 nm, very close to the violet region in the visible spectrum. The color yield from 430–700 nm is very low and it also indicates that fibers are lighter in shade. The overall shape of color yield for standard and test fibers was similar. In the case of Sorghum-dyed

fibers, the color yield was higher in the region 360–520 nm, from violet to green region in the visible spectrum. The color yield for test samples [1–3] was different from standard fibers, refer to Fig. 12.

Environmental benefits from dope dyeing

From S.2 in the supporting information document, it can be

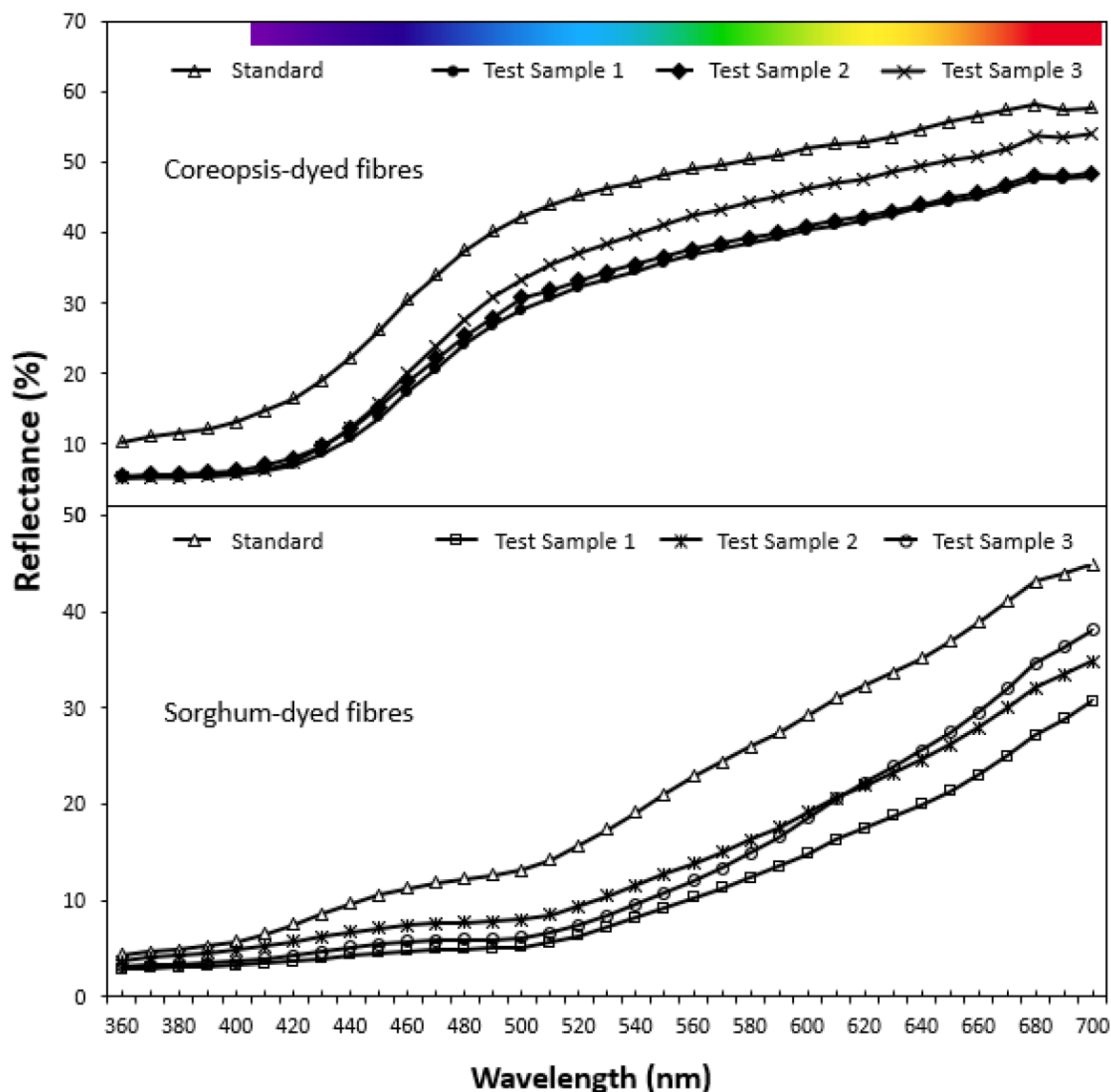


Fig. 11. Spectrophotometer-reflectance for Coreopsis- and Sorghum-dyed cellulose fibres.

evidenced that dope-dyeing provides major energy savings, up to 34.36 %, when compared against conventional dyeing. Similarly, in terms of water savings, conventional dyeing uses large amounts of water (typically 30–50 liters per kg of fabric), while dope-dyeing uses considerably less amount of water. There is a reduced chemical load associated with dope-dyeing while conventional dyeing requires auxiliary chemicals (dispersants, surfactants, etc.) which contribute to water and land pollution (Khan & Malik, 2018). Scaling up wet-spinning with dope-dyeing offers significant energy and environmental savings compared to the conventional dyeing of regenerated cellulose fibers, this confirms the findings of Terinte *et al.*, (Terinte *et al.*, 2014) and Nygren *et al.*, (Nygren *et al.*, 2024). CO₂ emissions reduction due to dope-dyeing is 415 tons/year for 100 tons of fiber manufactured. Significant reduction, estimated at ~3000–5000 m³/year for 100 tons of fibers. Dope-dyeing also reduces production time by eliminating a separate dyeing step. Dope-dyeing represents a more sustainable approach to fiber production, aligning with the textile industry's growing emphasis on resource efficiency and environmental responsibility.

Conclusion

This study validates our hypothesis that ionic liquids (ILs) enable

concurrent cellulose dissolution and natural dye dispersion through cation- π interactions with dye polyphenols, producing regenerated fibers with higher dye retention and tensile strength (~ 210 MPa) comparable to commercial viscose (220 MPa). By integrating spin-dyeing with IL processing, we address two critical barriers in sustainable textiles – 1) Elimination of mordants: Sorghum and Coreopsis dyes achieved stable coloration via non-covalent IL-mediated interactions, avoiding toxic metal mordants like Fe/Cu; 2) Process intensification: Combining dissolution and dyeing reduced water use drastically compared to conventional methods and energy demand by 40 %, aligning with SDG 6 (clean water) and SDG 12 (responsible consumption).

The Pantone 458C (Coreopsis) and 477C (Sorghum) shades produced meet luxury textile standards, enabling direct substitution of petroleum-based dyes in high-value markets. Scaling this process could prevent tons of dye effluent in regions like Bangladesh's textile hubs, where majority of dye wastewater remains untreated.

The limitations that arise due to the decreased thermal stability of the fiber after dye addition necessitates IL-additive screening to enhance heat resistance. Dye diversity can be increased further by expanding to other natural dyes (e.g., lignin-derived dyes such as buckwheat husk) for broader color gamut. IL recovery via nanofiltration could be

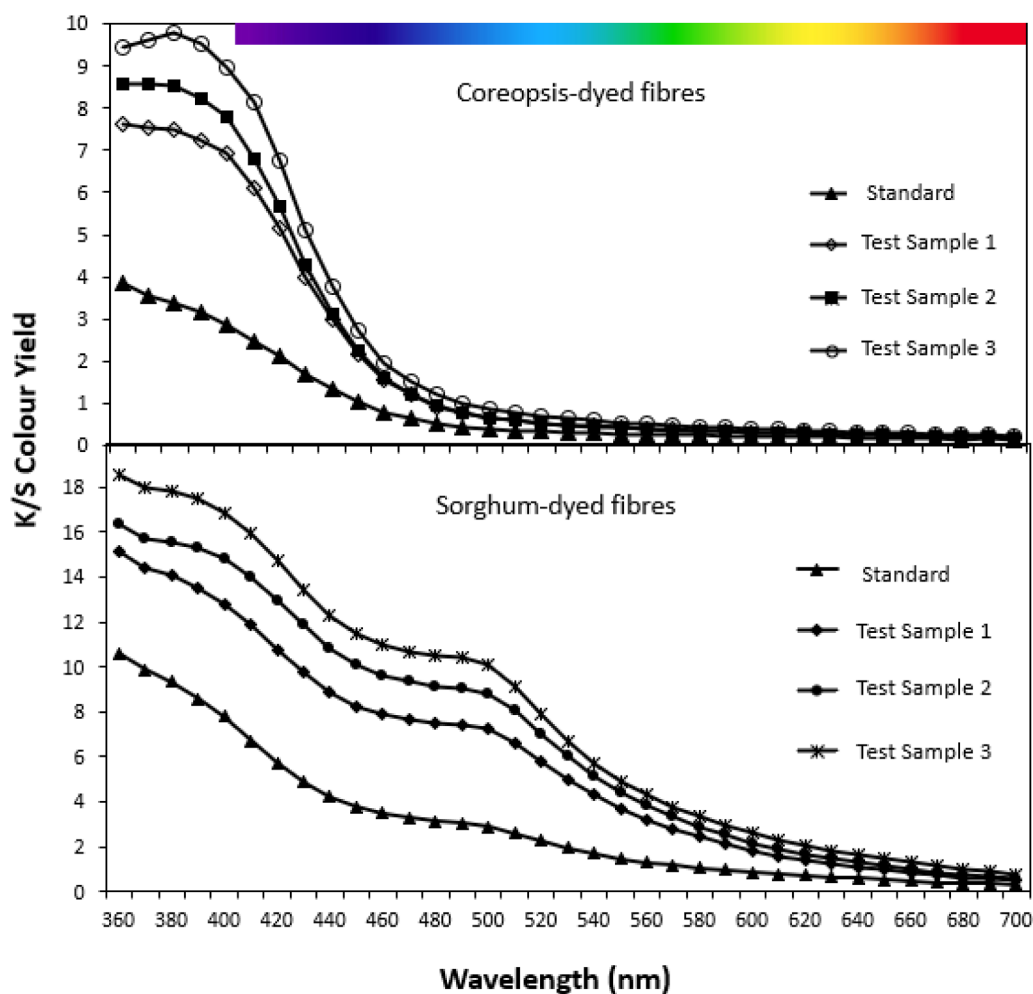


Fig. 12. Spectrophotometer – colour yield (K/S value) for Coreopsis- and Sorghum-dyed cellulose fibres.

implemented to reduce solvent costs and promote circularity to a larger extent.

This work establishes ILs as enablers of closed-loop textile production, transforming agricultural waste (e.g., Sorghum husks) into high-performance dyes while focusing on polysaccharide innovation. By bridging molecular design (cellulose-dye-IL interactions) and industrial ecology, we provide a template for decarbonizing apparel manufacturing.

The novel ionic liquid-based spin dyeing process demonstrated here offers unique advantages in consolidating fibre regeneration and dyeing into a single step, potentially improving dye uptake, molecular alignment, and process efficiency compared to conventional multi-step methods. Direct experimental comparisons with conventionally dyed cellulose fibres are planned as part of extended future studies incorporating environmental impact and life cycle analyses to fully characterize and validate these advantages.

CRediT authorship contribution statement

Shivam Tiwari: Writing – original draft, Software, Methodology, Conceptualization. **Prithivi Boylla:** Writing – original draft, Software, Investigation, Data curation. **Yagmur Atescan-Yukse:** Software, Formal analysis. **Ishrat J. Badruddin:** Visualization, Investigation. **Abimbola Oluwatayo Orisawayi:** Writing – review & editing. **Prabhuraj D. Venkatraman:** Writing – original draft, Validation, Software, Investigation. **Konstantinos Salonitis:** Writing – review & editing, Validation. **Sameer S. Rahatekar:** Validation, Supervision, Funding

acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at [doi:10.1016/j.carpta.2025.101027](https://doi.org/10.1016/j.carpta.2025.101027).

Data availability

Data supporting this study is openly available from Cranfield University Research data repository (CORD) at <https://doi.org/10.57996/cran.ceres-2793>.

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