

Pioneering microsphere-dope dyeing for sustainable cellulosic fibre colouring

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ARTICLE INFO

Keywords:

Microcapsules
Biopolymer
Sustainable textiles

ABSTRACT

The textile industry has long been criticised for its high energy consumption, significant pollution, and low efficiency, highlighting the urgent need for sustainable innovations. This study presents an advanced approach to fabric dyeing by integrating microencapsulation with dope dyeing to enhance indigo retention in cellulose fibres. Encapsulating indigo within alginate-based microspheres prevents unwanted chemical reactions and minimises environmental impact. Microscopic and SEM analyses confirm the successful incorporation of indigo within fibres, significantly reducing dye loss (0.11–0.18 %) compared to traditional methods. Eliminating the need for reducing agents and oxidants simplifies wastewater treatment, while the fibres exhibit enhanced mechanical properties and exceptional wash durability, retaining 99.76–99.95 % of their weight after 20 cycles—comparable to pure cellulose fibres. A 48-h washing test further demonstrates excellent colour fixation, with minimal indigo detected in the washing solution (0.0042–0.0053 mg/L). By significantly reducing dye loss and chemical usage, this method offers a promising solution to minimising the environmental footprint of textile dyeing, particularly in cellulosic fibre applications. These findings highlight microsphere-assisted dope dyeing as a viable, sustainable alternative to conventional techniques, aligning with global sustainability goals while maintaining high-quality fibre production.

1. Introduction

Textiles have historically played a critical role in global economies, consistently expanding in scale and diversity [1]. Nevertheless, the sector is under increasing scrutiny due to its significant environmental impacts, including substantial resource consumption, pollutant emissions, and inefficiencies [2,3]. As societal priorities shift towards low-carbon and sustainable development, transforming textile production processes has become increasingly imperative [4,5]. A life-cycle assessment of textiles identifies printing and dyeing as the primary environmental burdens, accounting for approximately 44 % of impacts, followed by fabric manufacturing (28.85 %) and finishing processes (15.38 %) [6]. Consequently, dyeing techniques and fibre production represent critical areas for enhancing sustainability. This research explicitly addresses these two vital aspects through developing innovative, eco-friendly textile technologies.

The dyeing sector is recognised for its significant environmental

challenges, primarily due to extensive water, chemical, and energy usage, which results in substantial wastewater generation. Indeed, textile dyeing processes alone contribute to nearly 20 % of global industrial wastewater. Indigo dye, historically valued for its distinctive deep-blue colouration and robust performance, particularly in denim production, exemplifies these challenges [7,8]. Despite their cultural and practical significance, traditional indigo dyeing techniques are resource-intensive and environmentally harmful, prompting research into sustainable alternatives capable of preserving indigo's desirable properties while minimising environmental harm [9–11].

Various sustainable dyeing methods have emerged, including electrochemical reduction, supercritical CO₂ dyeing, microbial dyeing, foam dyeing, and dope dyeing [12–19]. However, these alternative methods have limitations, such as high energy requirements and restricted colour ranges [9,20–23]. Among these techniques, dope dyeing stands out for its efficiency and simplicity. This method significantly reduces chemical and energy usage by incorporating dyes directly into fibres during

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<https://doi.org/10.1016/j.ijbiomac.2025.146007>

Received 12 December 2024; Received in revised form 4 July 2025; Accepted 12 July 2025

Available online 16 July 2025

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spinning [24]. For example, in 2021, Lenzing Ltd. introduced a Modal fibre produced through dope dyeing [25]. Laboratory data indicate that fibres manufactured by this method can reduce water usage, electricity consumption, and wastewater generation by up to 99 % compared with traditional dyeing techniques. Furthermore, the process obviates the need for heating and diminishes chemical usage by >80 % [26].

Regarding fibre production, synthetic fibres, predominantly polyester, dominate the global textile market, representing 55–60 % of total fibre production [27–30]. Despite economic advantages, their petroleum-based nature contributes to significant environmental issues, including pollution, non-biodegradability, and microplastic contamination [31,32]. Synthetic fibres release microplastics during laundering, adversely affecting marine ecosystems and highlighting the need for sustainable alternatives such as natural, semi-synthetic, and recycled fibres [33–37].

This study explicitly explores cellulose fibres—a subgroup of semi-synthetic fibres offering significant environmental benefits [38–42]. The history of cellulosic fibres dates back to the early twentieth century with the advent of rayon derived from wood pulp [43]. Textiles from these fibres are inherently biodegradable, decomposing naturally without harming ecosystems [44–46]. Moreover, they do not generate microplastics during laundering, mitigating one of the primary environmental concerns associated with synthetic fibres [47–49]. The integration of cellulose fibres into textile production thus holds the potential to yield high-performance, durable products that contribute to a sustainable and environmentally friendly future [50].

Industrial production of solution-dyed cellulose fibres predominantly relies on viscose technology or *N*-methylmorpholine *N*-oxide (NMMO) solutions [46,51,52]. These complex processes present notable safety concerns, given that NMMO solutions can be prone to explosion under elevated temperatures. In this context, ionic liquids have emerged as the next generation of environmentally benign solvents, offering a promising alternative [53]. For instance, Nicole Nygren et al. developed a vat dye dope dyeing technology utilising the Ioncell process, which involves dissolving cellulose in ionic liquids with specific vat dyes directly incorporated into the solution [54]. The resultant fibres, spun into dyed artificial cellulose, exhibit excellent mechanical properties and colour fastness, demonstrating the potential and environmental benefits of employing ionic liquids in solution dyeing. However, similar phenomena were observed in their experiments, where the reducing dyes reacted with the ionic liquids employed, resulting in a noticeable deviation in colour from the intended sample.

Several challenges, particularly concerning the selection of appropriate ionic liquids and dye types, necessitate further investigation. In our previous experiments, cellulose fibres were solution-dyed using 1-ethyl-3-methylimidazolium diethyl phosphate (EMIM DEP) and indigo vat dye. At 85 °C, it was observed that the indigo dye reacted with the ionic liquid, leading to an undesirable shift in colour from its characteristic blue to a yellow-green hue over 48 h. Even when the spinning process was completed before this colour change occurred, subsequent rinsing in deionised water resulted in significant colour loss, yielding a near-colourless fibre. This phenomenon underscores the complexities of utilising indigo within ionic liquid-based dyeing processes and highlights the need for further research to optimise conditions for maintaining the integrity and stability of indigo during dyeing.

To address these challenges, this study presents an eco-friendly indigo dyeing approach by integrating microencapsulation with ionic liquid technology. By encapsulating indigo in alginate microspheres, we successfully incorporated the dye into cellulose/ionic liquid spinning solutions while protecting it from undesirable interactions. This innovative method overcomes two major limitations: it expands material compatibility by preventing dye-ionic liquid reactions, and eliminates the environmental pollution associated with conventional dyeing processes. The use of biodegradable alginate ensures full process sustainability. Our investigation specifically examined (i) microcapsule integration with the spinning solution, (ii) fibre colour uniformity and

Table 1

AIMs formulations.

Sample name	Indigo (w/w)	Alginate (w/w)	F-127 (w/w)
AIM-1	1 %	4 %	1 %
AIM-2	1.5 %	4 %	1 %
AIM-3	2 %	4 %	1 %

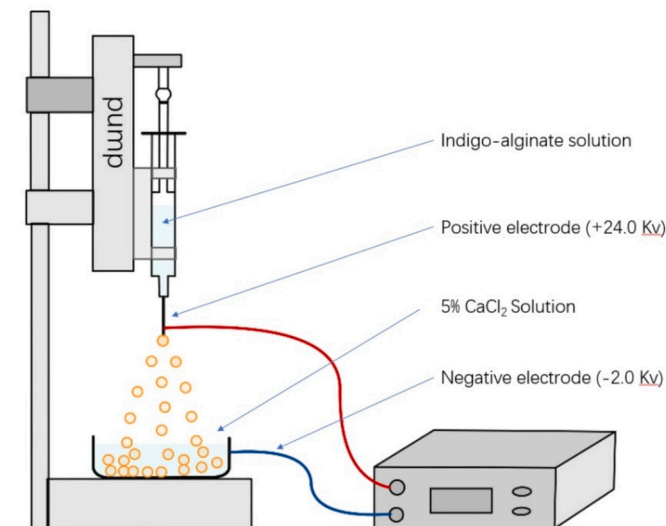


Fig. 1. Schematic diagram of electrospray device for microsphere manufacturing.

mechanical strength, and (iii) thermal and structural characteristics of the composite fibres. The developed method offers a sustainable alternative to traditional textile dyeing, with potential to transform manufacturing practices while supporting global environmental objectives.

2. Material and experiments

2.1. Materials

The primary dye used in this experiment, indigo, was supplied by Wildcolours Ltd. To prepare the microspheres, Pluronic® F-127 (P2443-250G, Sigma-Aldrich), a surfactant, was added to the indigo-alginate solution. The microspheres consisted of sodium alginate (W201502-1KG, Sigma-Aldrich) as the shell, indigo as the core, and calcium chloride (≥93.0 %, Sigma-Aldrich) as the cross-linking agent. Cellulose (GreatArt Ltd) was dissolved using 1-ethyl-3-methylimidazolium diethyl phosphate (EMIM DEP, ≥98 %, Sigma-Aldrich). For UV spectrophotometric analysis, indigo was extracted from the aqueous solution using chloroform (≥99.8 %, Sigma-Aldrich). And the laundry detergent and disinfectant, used for washing tests, were procured from a mainstream supermarket.

2.2. Sample preparation

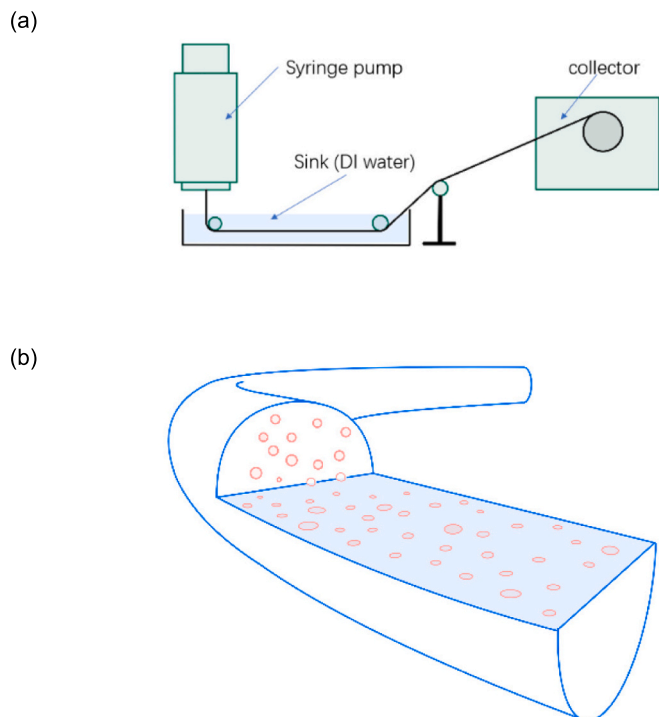
2.2.1. Microspheres preparation

The alginate-indigo solutions for microsphere preparation, with varying indigo concentrations (1–2 wt%), were prepared by adding sodium alginate powder (4 wt%), Pluronic® F-127 powder (1 wt%) as a non-ionic surfactant, indigo powder, and deionised water into a beaker, according to the formulation stated in Table 1. The mixture was stirred using a magnetic stirrer at 200 rpm for six hours at room temperature to ensure homogeneity. The alginate-indigo microspheres were produced via the electrospray procedure, as illustrated in Fig. 1. For a typical

Table 2

Parameters of textile samples.

Sample	Cellulose(w/w)	Percentage of AIM (wet) (w/w)
AIM-CF-1	3 %	2.5 %
AIM-CF-2	3 %	5 %
AIM-CF-3	3 %	7.5 %
Cellulose Fibres (CF)	3 %	0 %

**Fig. 2.** (a) Schematic diagram of wet spinning device and (b) of composite fibre

batch, 10 ml of the alginate-indigo solution was transferred into a syringe connected to a positive electrode set to 24 kV, with an injection speed of 1 ml/h. The microspheres were collected in a petri dish containing 100 ml of 5 % calcium chloride solution, with the dish set to -2 kV. Following the electrospray procedure, the formed microspheres were separated from the solution using a vacuum-assisted filtration system with a ceramic Buchner funnel. The microspheres were rinsed three times with deionised water at room temperature (approximately 25°C), with each rinsing session lasting no less than five minutes, to ensure the removal of any surface impurities, such as calcium chloride.

2.2.2. Preparation of microsphere embedded Fibres

To prepare the microsphere fibre solution, 3 % cellulose and 97 % EMIM DEP were combined in a beaker, heated in an oil bath at 85°C , and stirred at 200 rpm for 12 h until the cellulose fully dissolved. The solution was then cooled to 70°C before adding various amounts of alginate-indigo microspheres (AIM) as specified in Table 2. After stirring the mixture for an additional 15 min, it was transferred into a syringe and placed in an oven at 60°C for two hours to remove any air bubbles. As illustrated in Fig. 2a, the syringe containing the spinning solution was connected to an automatic syringe pump equipped with a 1.37 mm needle. The needle was submerged in a 500 ml deionised water bath, which served as the coagulation medium. The pump was operated at a constant rate of 5 ml/min to ensure uniform extrusion of the solution. Upon contact with the aqueous medium, the ionic liquid rapidly diffused out of the extruded stream into the surrounding water, initiating immediate fibre solidification through phase separation. The resulting continuous fibre was collected using an automated winding system

rotating at 30 rpm, which maintained consistent tension and alignment during the collection process. Once the syringe was emptied, the fibre product was immersed in deionised water at room temperature ($\sim 25^{\circ}\text{C}$) for over 24 h, with the water being changed at least three times to ensure complete removal of the ionic liquid. Finally, the fibre was rewound onto the collector and placed at room temperature ($\sim 25^{\circ}\text{C}$) for over 48 h to ensure natural drying.

2.3. Experimental analysis methods and characterisation techniques

2.3.1. Scanning electron microscopy (SEM)

The morphology of alginate-indigo microspheres (AIM) and alginate-indigo composite cellulose fibre (AIM-CF) was examined using Scanning Electron Microscopy (SEM) (TESCAN S8000 (Kohoutovice, Czech Republic)). The samples were coated with a 10 nm-thick gold layer in a vacuum and before being subjected in SEM.

2.3.2. Microscopic analysis

The wet microsphere sample was mounted onto a microscope slide and covered with a glass coverslip to facilitate the observation of the microspheres' colour and morphological characteristics. To analyse the composite fibres of the microspheres, their colour and morphology were initially examined using an optical microscope (Leica Microsystems CH-9435). The textile samples were subsequently placed into rectangular sample holders with a width of 10.20 mm, ensuring that the fibres were maintained in a naturally extended state. Using LAS (Version 4.12.0) software, three locations on each fibre sample were randomly selected for measurement, the diameters were recorded, and the mean diameter was calculated.

2.3.3. CIELab-based colour analysis of AIM-CF

The optical properties of the manufactured fibre samples were characterised through CIELab coordinates, employing a 2° observer and standard illuminant D65 (CM-700d, KONICA MINOLTA, Japan) for AIM-CF samples. Each fibre produced is divided into four parts. CF (cellulose fibre) was the standard sample, and the rest were control samples tested by the colorimeter to intuitively compare the dyeing effect. Within the CIELab colour space, the total chromatic deviation, ΔE , quantifies the overall colour difference between samples, where ΔL^* represents the differential in lightness, Δa^* denotes the variation along the red-green axis, and Δb^* signifies the variation along the yellow-blue axis. This formulation served to determine the metric of colour variation, expressed as ΔE^*_{ab} .

$$\Delta E^*_{ab} = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2} \quad (1)$$

2.3.4. Thermogravimetric analysis (TGA)

TGA tests were conducted using a TA Q500 instrument. Approximately 5 to 10 mg of the raw materials, microspheres, and composite fibre were weighed and then transferred to a platinum sample pan with an attached hook, which was placed on the autosampler. All tests were performed in a closed heating chamber with a constant air flow rate of 60 ml/min. The samples were heated from 25°C to 800°C at a rate of $10^{\circ}\text{C}/\text{min}$. To minimise experimental error, each material was tested at least three times.

2.3.5. Analysis of mechanical properties of composite fibres

The mechanical properties of composite fibres were analysed using a DEBEN 200 N microtest tensile stage controller (Suffolk, UK) with a stretching speed set at 0.2 mm/min. AIM-CF samples were attached to a 10.2 mm sample plate to minimise the influence of the clamps. The fibre diameter was measured by optical microscopy (Leica Microsystems CH-9435). The ultimate tensile strength and strain at the breaking point of the fibres were determined, and Young's modulus was calculated from the slope of the linear portion of the stress-strain curve prior to the yield

point.

2.3.6. FTIR spectroscopy

Fourier Transform Infrared (FTIR) spectra were obtained using a Thermo Scientific Nicolet™ iS™10 FT-IR spectrometer. The IR data of raw materials, microsphere samples, and composite fibres were collected in the range of 600–4000 cm⁻¹. For each spectrum, 50 consecutive scans with a resolution of 4 cm⁻¹ were averaged; all spectra were recorded at room temperature.

2.3.7. Analysis of indigo concentration in liquids during the production processes using UV spectrophotometric detection

The quantification of dye utilization and loss during the production processes was conducted through several key steps. Indigo loss in the post-cross-linking and post-spinning solutions was calculated by determining the concentration of indigo and multiplying it by the total volume of the respective solutions. The indigo mass used in the fabrication of microspheres was determined, followed by the calculation of the indigo quantity incorporated into fibre-added microspheres. This involved multiplying the indigo mass in the mixture by the mass of microspheres added to the fibre and dividing by the total number of microspheres produced. The dye loss rate was calculated as the ratio of indigo lost during production to the weight of indigo used for fibre production, using specific equations provided in the supplementary information. The concentration of indigo in various solutions during and after manufacturing was investigated by comparing the UV absorption of test solutions to reference solutions of indigo in chloroform with known concentrations. The reference solutions were placed in an ultrasonic bath for 10 min to ensure complete dissolution of the dye before their absorbance values were recorded at a wavelength of 604 nm. The limit of detection (LoD) and limit of quantification (LoQ) for this method are 0.02 mg/L and 0.08 mg/L, respectively [55].

2.3.8. Determination of indigo in the calcium chloride solution for microsphere synthesis

A 20 ml sample of the calcium chloride solution, post-microsphere synthesis, was transferred from the petri dish to a separatory funnel using a pipette, ensuring the stopcock at the bottom was closed. This solution was then mixed with 5 ml of chloroform. The funnel was sealed and gently shaken 3–4 times, periodically opening the top stopper to release any gas generated. This shaking process was repeated for 10 min. Afterward, the funnel was left undisturbed on a metal stand for 3–4 min to allow for complete separation of the layers. The stopcock was then carefully opened to drain the chloroform into a beaker, ensuring that only chloroform was collected without any water. Subsequently, 2–3 ml of the chloroform layer was transferred to a cuvette and analysed using a UV spectrophotometer (Jenway 6715 UV/Vis). The absorbance result was compared to a reference to determine the indigo concentration. The procedure was repeated three times for each of the three AIM solutions.

2.3.9. Analysis of indigo in post-spinning solutions

A 100 ml sample of the post-spinning solution from AIM-CF-1, AIM-CF-2, and AIM-CF-3 was mixed with 5 ml of chloroform and extracted using the method described above. Each test was performed three times, and the indigo concentration was calculated.

2.3.10. 48-Hour Washing Test

20 ml of laundry detergent was mixed with 20 ml of laundry disinfectant and then diluted to 2 l with deionised (DI) water. A 0.2 g sample of fibre was fully immersed in 40 ml of this washing solution. The solution was heated to 45 °C and stirred at 300 rpm with a magnetic stirrer for 48 h. After the washing process, 20 ml of the solution was extracted with 5 ml of chloroform. As a reference, 20 ml of the detergent solution was also extracted using the same procedure. The indigo concentration was calculated based on the reference. The test was repeated three times for each of the AIM-CF-1, AIM-CF-2, and AIM-CF-3 samples.

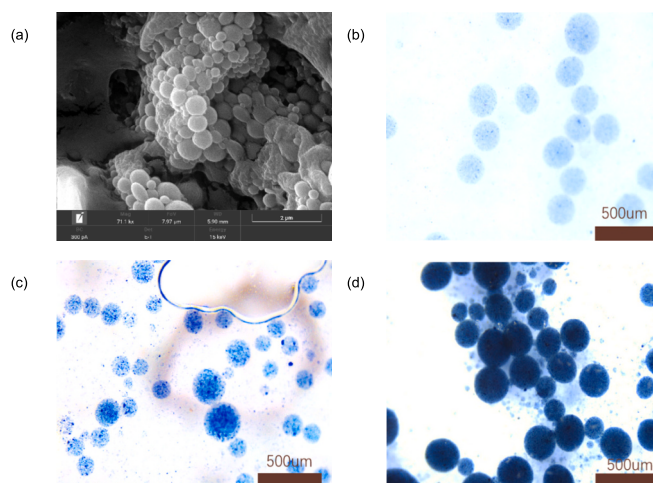


Fig. 3. Microscopic images of (a) dry and wet AIMs with (b) 1 wt% (c) 1.5 wt% and (d) 2 wt% indigo.

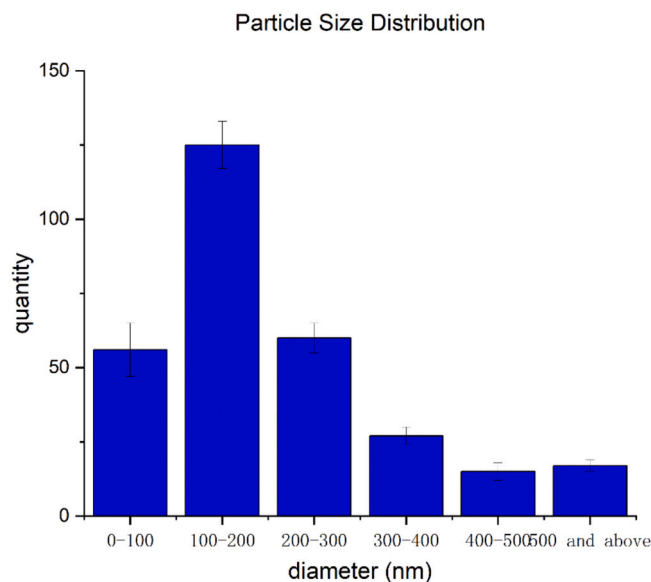


Fig. 4. Particle Size Distribution of Dry AIM Samples.

2.3.11. Fibre weight retention assessment post multiple laundering cycles

Fibre samples of CF, AIM-CF-1, AIM-CF-2, and AIM-CF-3 (~0.2 g each) were dried in a vacuum oven at 50 °C for two hours and weighed using an analytical balance. Each sample was then immersed in 40 ml of washing solution, heated to 45 °C, and stirred at 300 rpm with a magnetic stirrer for 48 h. The washing and rinsing process was repeated 20 times. Subsequently, the samples were rinsed with deionised water three times and dried in a vacuum oven for two hours. After drying, the fibres were weighed again to determine weight retention. Fibre weight retention was calculated by comparing each sample's post-laundering weight to its pre-laundering weight using the equation:

$$F_w = \frac{F_B}{F_A} \times 100\% \quad (2)$$

Where: F_w is the fibre weight retention (in percentage), F_A is the weight of the fibre after washing, and F_B is the weight of the fibre before washing.

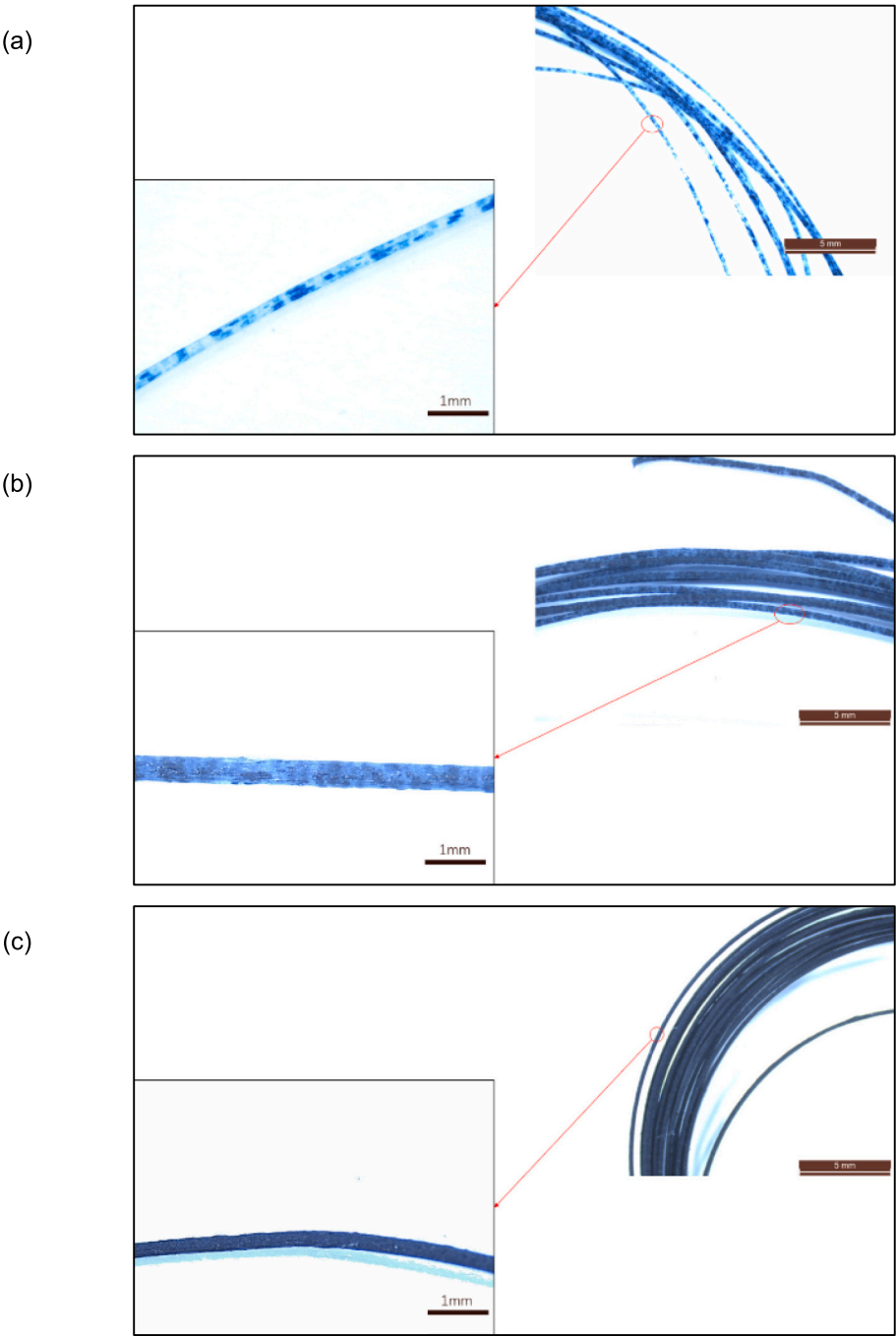


Fig. 5. Microscopic images of AIM-CF with (a) 2.5 %, (b) 5 % (c) 7.5 % wet AIM.

3. Results and discussion

3.1. Microscopic analysis and colour control of indigo-encapsulated microspheres in Fibres

Fig. 3(a) presents SEM micrographs illustrating the microstructure of desiccated microspheres. The microspheres exhibit remarkable uniformity in both morphology and dimensional characteristics, displaying smooth spherical architectures. The complete absence of surface indigo particulate matter confirms successful encapsulation within the alginate matrix, validating our microencapsulation protocol. This effective encapsulation was achieved through optimised electrospray parameters [56], complemented by rigorous magnetic agitation (≥ 30 min duration) and stabilisation employing Pluronic F-127 surfactant [57,58]. Figs. 3

Table 3
Colour Coordinates and Colour Difference of Dyed Fibre Samples.

Sample	L^*	a^*	b^*	ΔE^*_{ab} (vs CF)
AIM-CF-1	41.84 ± 1.4	-1.60 ± 0.23	-10.80 ± 0.41	52.69
AIM-CF-2	30.74 ± 1.2	0.35 ± 0.14	-4.96 ± 0.22	62.35
AIM-CF-3	23.43 ± 0.84	1.10 ± 0.11	-5.18 ± 0.31	69.64
CF	92.50 ± 0.71	0.03 ± 0.12	3.61 ± 0.17	0.74*

* Note: The ΔE^*_{ab} value of CF (0.74) reflects internal variation, not in comparison to itself.

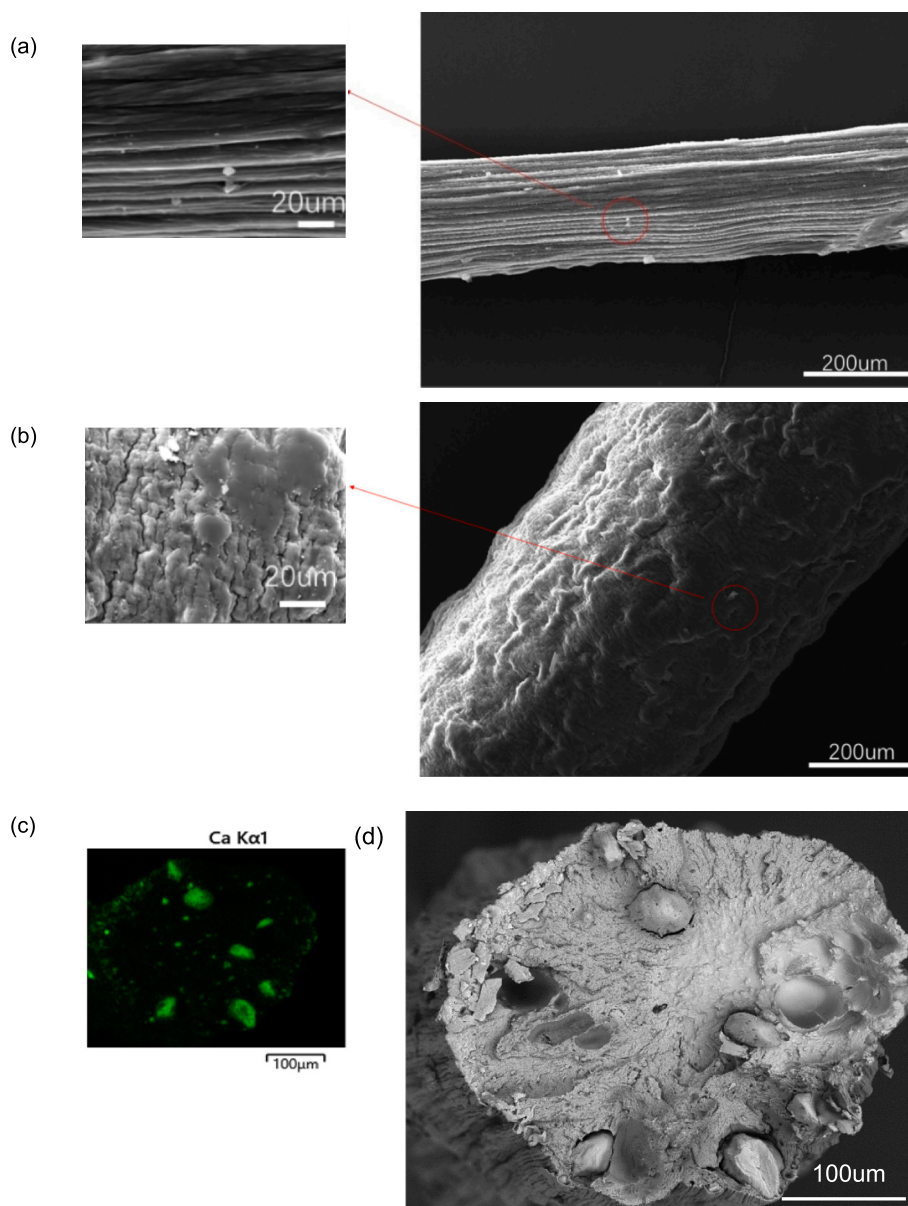


Fig. 6. SEM images of cellulose fibres with (a) 1 % and (b) 2 % AIM content, (c) EDS calcium element map, and (d) SEM image of the fracture surface of the 2 % AIM fibre.

(b)-(c) show microscopy images of aqueous indigo microspheres (AIMs) in solution. The wet microspheres display uniform colour at each concentration. Their colour darkens progressively with increasing indigo content. Notably, the hydrated microspheres ($10\text{--}10^2\text{ }\mu\text{m}$) are much larger than their dry counterparts ($10^{-1}\text{ }\mu\text{m}$) (See Fig. 4). This size reduction during drying significantly deepens their colour. The effect is strongest at high concentrations, making AIM-3 appear nearly black. For textile applications, we chose AIM-1 (1 % indigo) as the base material. This selection matches commercial colour standards while ensuring practical usability. Importantly, the microspheres' colour intensity depends directly on their indigo concentration during synthesis.

Fig. 5 presents micrographs of cellulose fibres embedded with varying concentrations of indigo microspheres to illustrate the dyeing effect. Table 3 provides corresponding data on these composite fibre samples' colour coordinates and colour differences, detailing the final dyeing outcomes across different microsphere contents. Together, these visual and quantitative data reveal how microsphere content and distribution influence the colour intensity and uniformity of the fibres. As the microsphere content increases, the fibre colour deepens and

becomes more uniform, shown by a decrease in the brightness parameter (L^*) from 48.84 to 23.43 in Table 3. Additionally, the red-green axis (a^*) values shift from -1.60 to 1.10 , while the yellow-blue axis (b^*) values change slightly from -10.80 to -4.96 , reflecting a subtle colour shift consistent with microscopic observations.

The micrographs in Fig. 5 show that higher microsphere concentrations lead to a denser distribution within the fibre matrix, resulting in a richer, more even colour that resembles conventional dyeing methods. This effect is quantitatively supported by the decreasing colour difference parameter (ΔE) in Table 3 as microsphere content increases, highlighting improved colour uniformity. In contrast, fibres with lower microsphere content exhibit a sparser distribution, producing a more discontinuous dye pattern and a distinctive visual texture.

These findings suggest a practical approach to customising fibre colouration by adjusting microsphere content, presenting an innovative method for textile dyeing with considerable potential in fashion applications. The ability to modulate hue intensity and colour consistency through controlled microsphere distribution during fibre production underscores this technique's potential for creating unique, visually

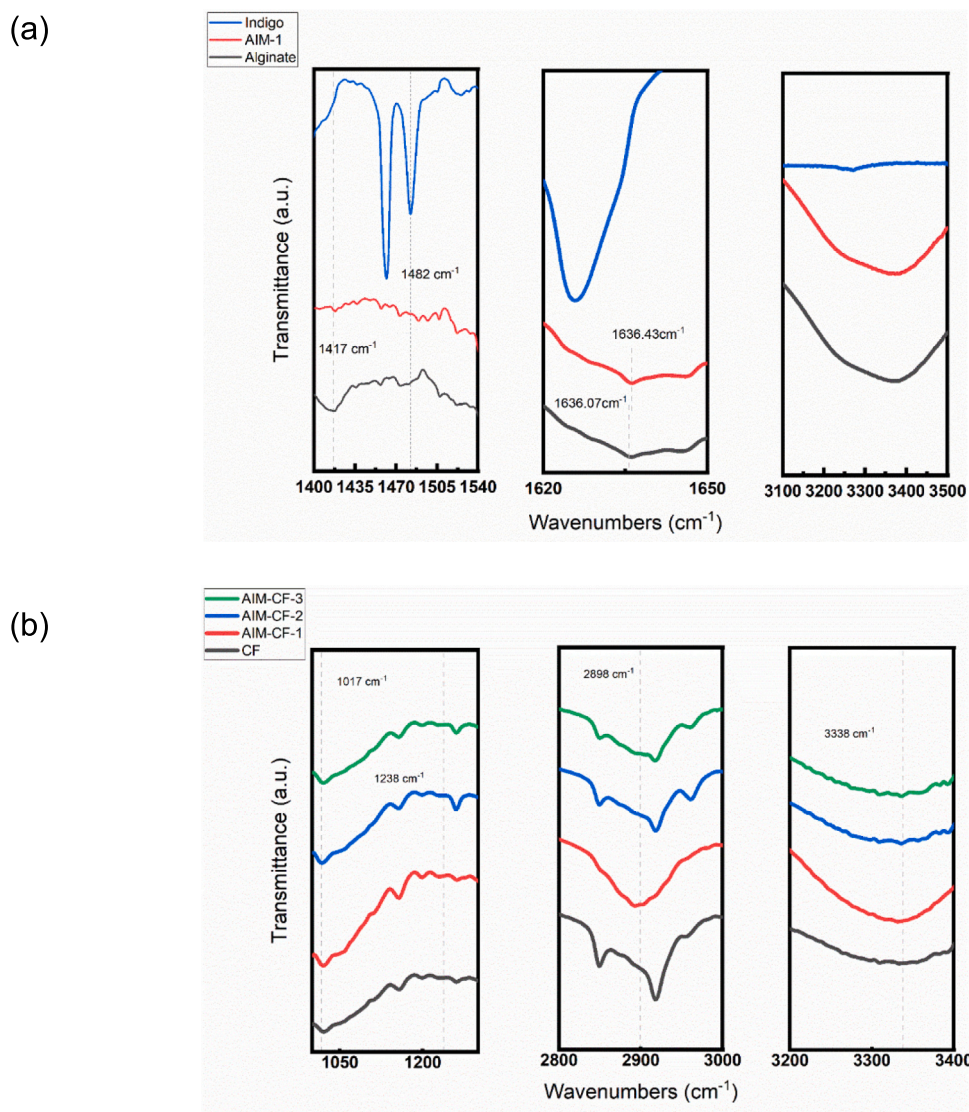


Fig. 7. FTIR spectrum: (a) indigo, alginate, AIM-1 (b) AIM-CF-1, AIM-CF-2, AIM-CF-3, CF.

striking textile products.

3.2. Fibre microstructure

Fig. 6 illustrate the morphologies of the continuous and fractured composite fibres. It is evident from Fig. 6 (a) to (b) that there is a change in the structure of the fibre surface of the AIM-CFs. As the concentration of the microspheres increases the surface of the fibre's changes from a more continuous linear distribution to a more conjugated structure. Also, individual spherical microspheres are visible on the fibre surface. Moreover, in the cross-sectional images of the fractured fibres shown in Fig. 6 (d), there are some large particles inside the fibres, which could result from microspheres aggregating during the wet spinning process. The energy dispersive X-ray spectroscopy (EDS) analysis, shown in Fig. 6 (c), further supports this finding. The EDS calcium (Ca) element map indicates significant signals on the spherical particles. This is consistent with the composition of the microspheres, whose outer shell is made of sodium alginate cross-linked with calcium chloride. As cellulose does not contain calcium, only the microspheres exhibit calcium signals in the calcium image. Meanwhile, there are visible voids adjacent to some large particles suggesting a lack of chemical bond connection between the sodium alginate and the cellulose. This finding is significant as it could impact the mechanical strength of the cellulose fibres [59].

Strategies such as evenly dispersing the microspheres before spinning (e. g., using ultrasonic vibration or a high-speed disperser) could mitigate this issue.

3.3. Chemical analysis (TGA /FT-IR)

The supporting data encompasses comprehensive infrared spectra of AIM-1, sodium alginate, indigo, pure cellulose fibre (CF), and composite fibres. The infrared spectrum of indigo displays characteristic vibrations, including C=O stretching at 1625 cm^{-1} , C=O rocking at 1069 cm^{-1} , and C=O wagging at 698 cm^{-1} , along with contributions from C—N, C—H, and N—H groups near 1698 cm^{-1} . Characteristic five- and six-membered ring vibrations appear between 1298 and 697 cm^{-1} , consistent with literature reports for indigo [60]. Sodium alginate exhibits broad O—H and N—H stretching vibrations (3100–3520 cm^{-1}), C=C stretching at 1636 cm^{-1} , and characteristic polysaccharide fingerprint modes between 1000 and 1100 cm^{-1} [61,62].

In the FTIR spectrum of AIM-1 (Fig. 7 (a)), the predominant features reflect the alginate and cellulose matrices. A careful examination of the 1400–1500 cm^{-1} region reveals a weak shoulder near 1428 cm^{-1} that may tentatively correspond to the aromatic C=C stretching of indigo [63], though this signal is significantly attenuated compared to pure indigo. This attenuation likely results from two factors: first, the ATR-IR

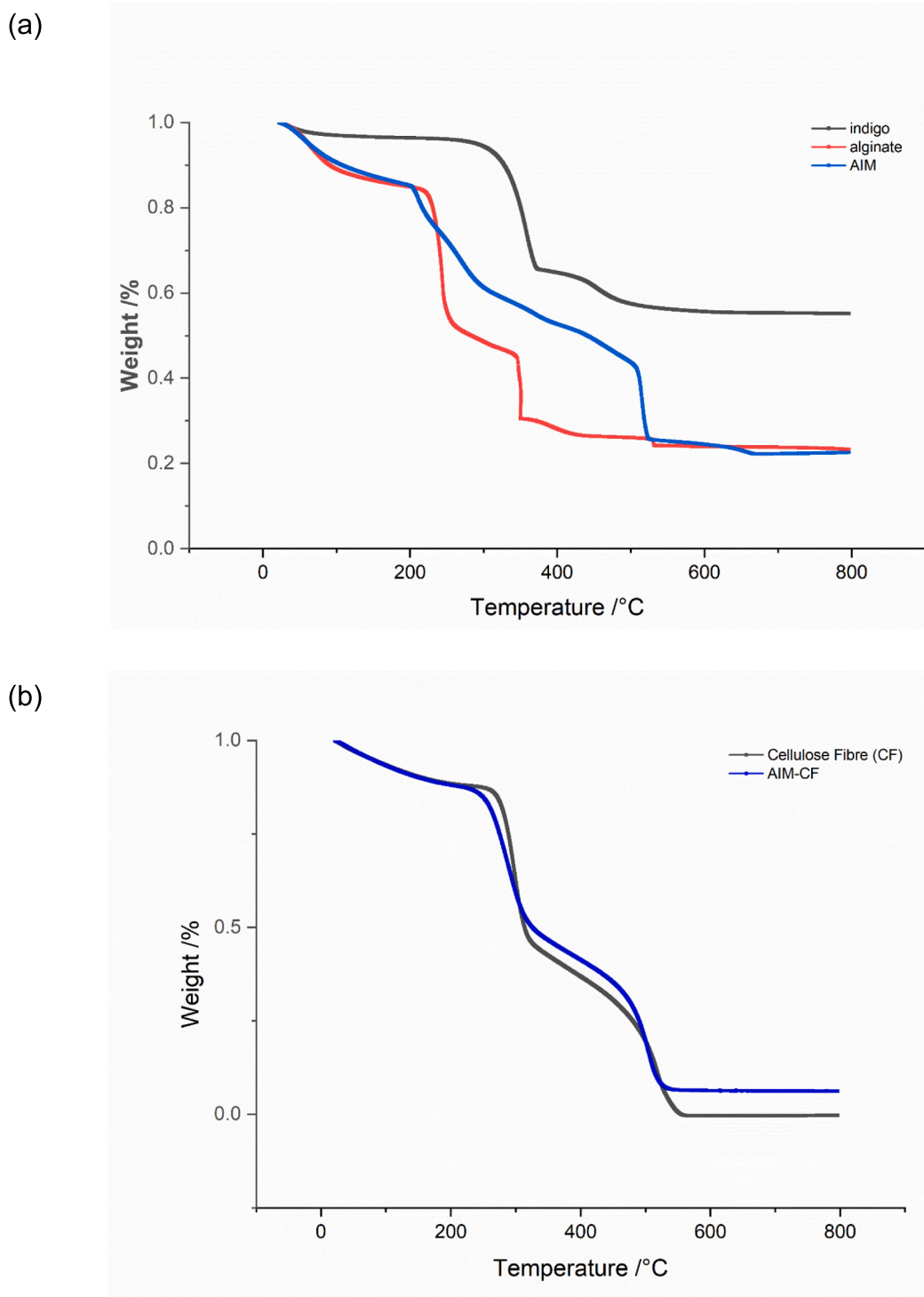


Fig. 8. TGA thermogram of (a) indigo, alginate and AIM (b)CF and AIM-CF-2.

technique probes only surface composition, and since most indigo is encapsulated within alginate microspheres, its spectral contribution is limited; second, the strong overlapping signals from the cellulose and alginate matrices may obscure the weaker indigo peaks. The observed shift in the alginate-associated band from 1417 to 1422 cm^{-1} in AIM-1 could potentially indicate interactions with indigo or effects from Ca^{2+} cross-linking, though definitive assignment remains challenging due to signal overlap and the complexity of the composite system [64,65].

The infrared spectra (Fig. 7 (b)) of both the composite fibre and pure cellulose fibre show characteristic cellulose peaks, including O—H

stretching at 3338 cm^{-1} , C—H stretching at 2897 cm^{-1} , and C—O vibrations at 1017 cm^{-1} , confirming the presence of cellulose in the composite fibre. The complete absence of a characteristic peak at 1238 cm^{-1} , which would correspond to the P=O bond of the emim DEP solvent, indicates successful removal of the solvent during fibre processing [66].

Given the inherent limitations of FTIR spectroscopy for detecting encapsulated pigments in complex matrices, the evidence for indigo incorporation in AIM-1 is further supported by complementary techniques. These include the characteristic UV–Vis absorbance at 604 nm,

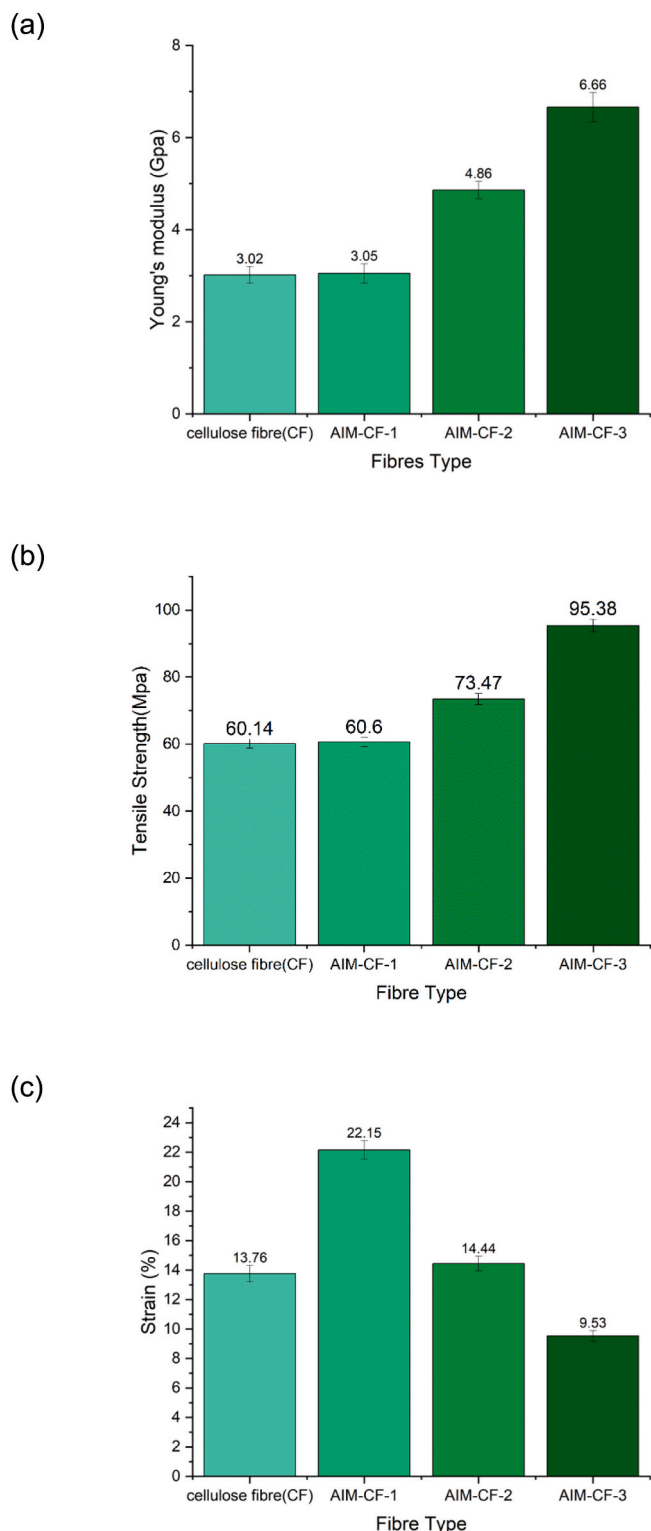


Fig. 9. Mechanical properties of fibres (a) young's modulus of fibres (b) tensile strength of fibres (c) strain of fibres.

colorimetric analysis (ΔE values), and consistent microsphere incorporation observed during the fibre spinning process. Together, these multiple lines of evidence provide a more robust confirmation of indigo presence than could be obtained from FTIR analysis alone [67–69].

Fig. 8(a) presents the TGA curves for pristine indigo, alginate, and the prepared AIM-1 microspheres. The alginate exhibits an initial weight loss between 65 and 100 °C due to moisture evaporation from its

hydrophilic structure, followed by a major decomposition step at 220–250 °C corresponding to cleavage of glycosidic bonds and other structural linkages in the alginate chain [70,71].

Indigo demonstrates a distinct thermal degradation pattern, with the primary weight loss occurring at 350–400 °C as a result of breakdown of its aromatic ring structure. The higher residual mass observed for indigo reflects its tendency for char formation through carbonization during thermal decomposition, a characteristic behavior of conjugated aromatic systems [72]. This represents a different decomposition mechanism [73].

The AIM-1 microspheres show thermal behavior intermediate between its components. The initial moisture loss (55–100 °C) mirrors that of alginate, while the main decomposition occurs across a broader temperature range (220–500 °C) due to combined degradation of both alginate and indigo. The modified degradation profile suggests interactions between the components leading to altered decomposition pathways [74].

Fig. 8(b) compares the thermal degradation of neat cellulose fibres (CF) and microsphere-embedded fibres (AIM-CF-2). Both samples show similar initial weight losses at 70–110 °C from moisture evaporation. The main decomposition temperatures remain comparable between CF (~300 °C) and AIM-CF-2, indicating that the incorporation of microspheres does not significantly alter the thermal stability of the cellulose matrix. The more complex degradation pattern of AIM-CF-2 primarily reflects the presence of additional components with different decomposition characteristics rather than substantial changes in thermal resistance.

3.4. Analysis of mechanical properties of fibres (tensile test)

An analysis of the mechanical property data (shown in Fig. 9) reveals a clear relationship between microsphere content and the mechanical characteristics of the composite fibre. Specifically, as the microsphere content increases, there is a corresponding rise in the Young's modulus and tensile strength, while the ultimate tensile strain decreases. This behavior can be attributed to the size distribution of the dried microspheres, which predominantly ranges from 100 to 200 nm. Since this size is comparable to that of certain nanofillers, the microspheres likely provide a reinforcing effect similar to traditional nanofillers [75]. The addition of these fillers reduces the free volume within the composite, thereby increasing the stiffness of the laminate.

While these values are comparable to natural cotton yarns, they remain below those of commercial regenerated cellulose fibres (above 200 MPa) [76]. The performance gap can be attributed to several experimental factors in our current system. The wet-spinning process without an air gap limited molecular alignment due to reduced drawing ratio [77]. Furthermore, unfiltered spinning dopes potentially contained undissolved particulates that compromised fibre uniformity, and the low cellulose concentration combined with the absence of post-spinning treatments restricted crystallinity development [78–80].

Notably, the mechanical enhancement observed with microsphere incorporation suggests promising reinforcement potential. Future process optimizations could bridge the performance gap with conventional regenerated fibres while maintaining the unique functionality of our microsphere-loaded system. Potential improvements include modifying the spinning configuration to introduce an air gap, implementing dope filtration, and incorporating post-processing treatments to enhance molecular alignment and crystallinity.

3.5. Indigo loss during the production and after washing

The UV spectrophotometric detection results (support information Fig. 1) showed that the absorbance of the calcium chloride solution used in the microsphere synthesis was below 0.2 %, similar to that of pure deionised water. This suggests that there was no indigo waste during the microsphere manufacturing process. This outcome can be attributed to

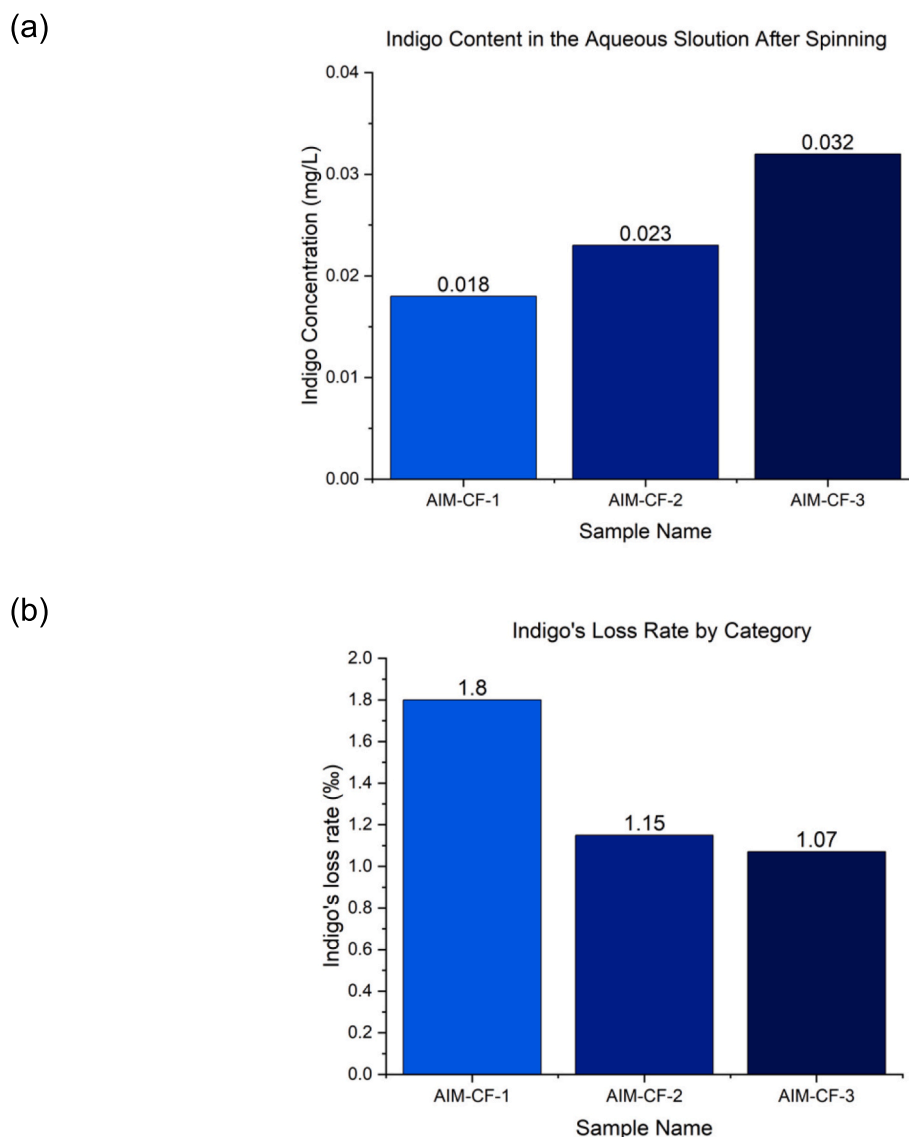


Fig. 10. (a) Indigo content in the aqueous solution after spinning (b) Indigo loss rate during production.

indigo's inherent insolubility in water. When mixed with sodium alginate, indigo forms minute droplets that sink into the calcium chloride solution due to gravity. The sodium alginate then undergoes a cross-linking reaction with the calcium chloride, creating a network of intermolecular crosslinks that restrict molecular movement, effectively encapsulating the indigo within the microspheres.

Fig. 10 (a) presents data correlating indigo concentration and absorbance in the residual solution following the wet spinning procedure. The results indicate a minimal amount of indigo into the solution post-spinning, ranging between 0.018 mg/L and 0.032 mg/L. Fig. 10 (b) further elucidates the calculated diminution of indigo throughout the spinning process, demonstrating a loss margin of merely 0.10 % to 0.18 %. This method results in significantly lower dye loss compared to traditional dyeing methods, which typically experience a 20 % dye loss. Moreover, the concentration of indigo in the solution increases as the number of microspheres rises after spinning. This is due to the dissolution of indigo from the microsphere surfaces into the ionic solution during the spinning process. However, there is a decrease in indigo loss as the AIM content increases. This may result from the tendency of microspheres to aggregate at higher concentrations. The aggregated structure provides a protective shield for the microspheres in the inner layers, reducing their risk of being dissolved by the ionic liquid.

The accompanying data in Fig. 11(a) depict the results of the comprehensive laundering test on fibres. After a 48-h washing regimen, the indigo concentration in the remaining solution was significantly low, indicating that the cellulose fibres dyed with this method have excellent washing resistance and exhibit strong colour fastness.

There are two reasons for this resistance. First, the indigo itself is not chemically modified which maintain its physical properties, hydrophobic nature and also improves its performance in wash tests [81]. At the same time, this double encapsulation dyeing technique, where indigo microspheres are encased in sodium alginate shells and then integrated into cellulose-based composite fibres, significantly enhances the chemical durability of the indigo. It also improves colour fastness in water and wash durability compared to traditional dyeing methods, where the dye is directly exposed to environmental factors. Fig. 11(b) illustrates the stability of fibre weight over multiple wash cycles. Compared with pure cellulose fibres, the mass of the composite fibres remained unchanged. This stability demonstrates the fibres' ability to resist shrinking or stretching, confirming their suitability for everyday use.

3.6. Comparative analysis with traditional reduction dyeing

The comparative analysis of traditional reduction dyeing and an

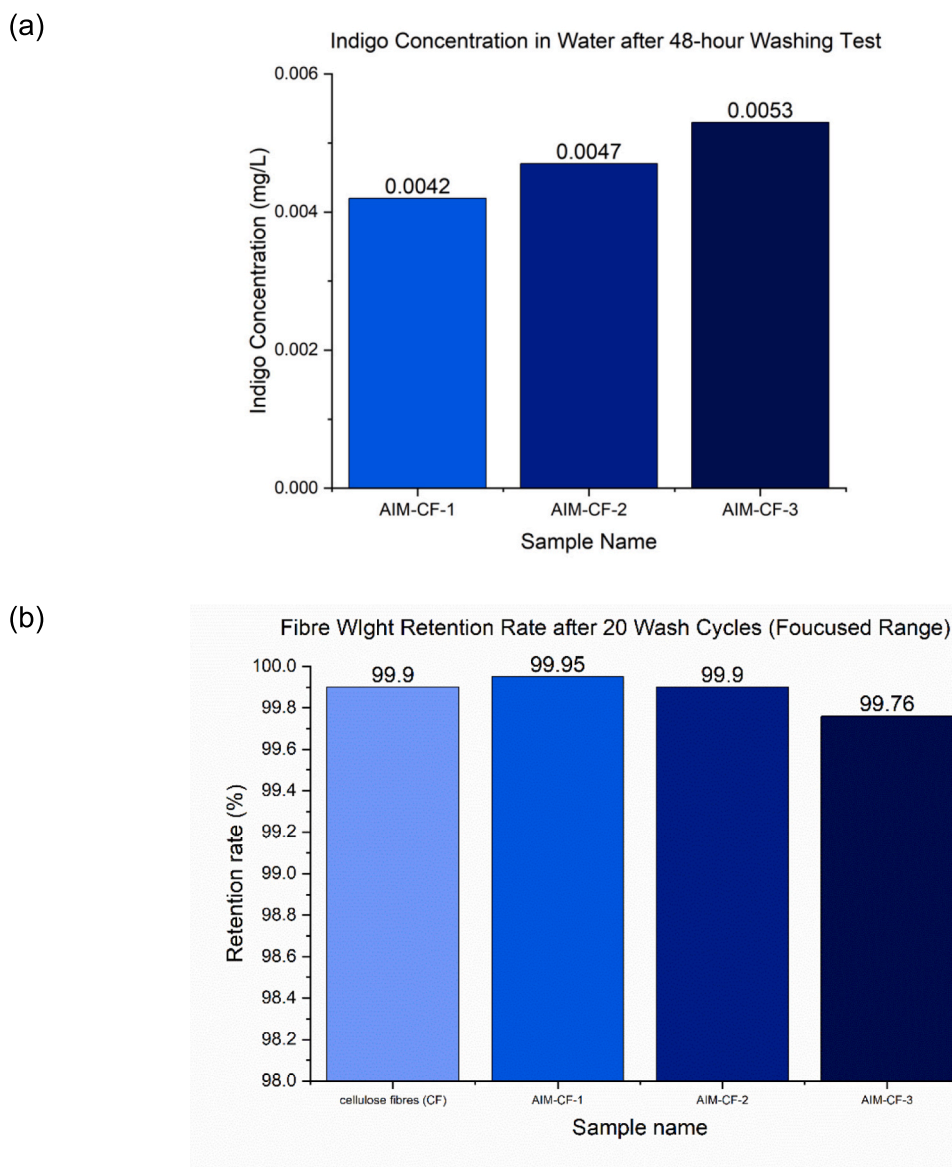


Fig. 11. (a) The content of indigo in the solution after 48 h of water washing (b) Weight retention rate of fibres after 20 cycles of water washing.

innovative dyeing method reveals substantial environmental benefits, as illustrated in Fig. 12. Traditional methods consume 170 g of chemicals, and 50-100 L of water per kilogram of fibre [82,83], generating significant wastewater and sulphur-based air emissions [27]. This high resource use depletes natural resources and contributes to water pollution and air quality deterioration. In contrast, the new method eliminates additional dyeing after fibre production, resulting in zero chemical use and drastically reducing water consumption to 7-8 L per kilogram of fibre, with minimal wastewater (2-4 L) and no air emissions. This significant reduction in dye waste during the dyeing of cellulose fibres, along with the promising potential for recycling the ionic liquid used in fibre production, offers a cleaner, more resource-efficient approach that contributes to sustainability and potential cost savings for manufacturers. Additionally, reducing chemical use mitigates specific health risks for workers, such as respiratory issues and skin conditions, and lessens the ecological impact of wastewater discharge, including soil contamination and water pollution. The durability and colour fixation of fibres produced using this new method have been proven superior to conventionally dyed fibres. This advancement signifies a crucial step towards sustainable textile production, aligning with global environmental goals and responding to increasing consumer demand for eco-

friendly products, thereby potentially enhancing the industry's reputation and market competitiveness.

4. Conclusion

This study introduces a novel microencapsulation technique for sustainable indigo dyeing of cellulose fibres through ionic liquid spinning. Building upon the established advantages of ionic liquids in cellulose processing, our approach effectively addresses the challenge of indigo-ionic liquid reactivity, a barrier to solution-dyeing. The alginate microencapsulation strategy successfully isolates the dye from the ionic liquid, preventing undesirable chemical interactions and resulting in exceptionally high dye fixation rates (99.8–99.9 %), representing a significant improvement over conventional dyeing methods.

The environmental benefits of this approach are substantial. It eliminates the need for toxic reducing agents, with water and recoverable ionic liquid as the only by-products. The biodegradable alginate matrix further enhances the sustainability of the technology, making it a promising alternative for textile coloration that aligns with the principles of dye stabilisation via microencapsulation.

While the tensile strength of the produced fibres (60–95.38 MPa) is

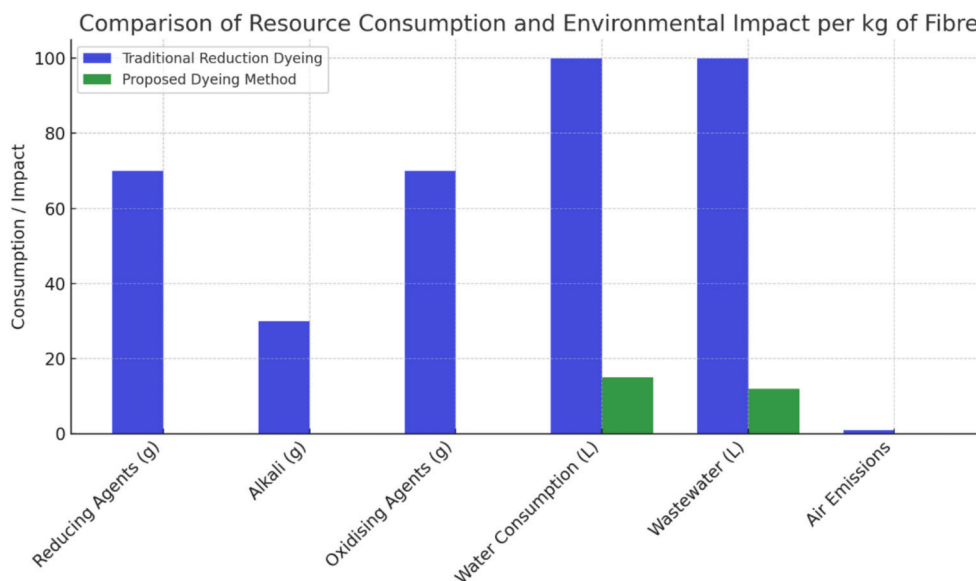


Fig. 12. The comparative bar chart of traditional reduction dyeing and an innovative dyeing method.

comparable to natural cotton, it remains below the standards of commercial regenerated cellulose (>200 MPa). This discrepancy arises from factors such as the absence of air-gap drawing, unfiltered spinning dopes, and the cellulose concentration (3 wt%), all of which are identifiable areas for further optimisation. These factors represent opportunities for improving the process rather than intrinsic limitations.

This research establishes a solid foundation for the development of sustainable high-performance fibres. However, further work is needed to enhance the mechanical properties of the fibres, particularly through process optimisation, and to assess the viability of the encapsulated dyeing approach in industrial fibre production. Future studies should focus on intensifying the process while preserving the environmental advantages. This will help solidify the potential of this technology to contribute to a more sustainable textile industry that aligns with circular economic principles. Additionally, the compatibility of this methodology with existing reinforcement techniques shows promise for meeting both ecological and industrial demands.

CRedit authorship contribution statement

Shuai Hao: Writing – review & editing, Writing – original draft, Visualization, Validation, Project administration, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Shouqi Shen:** Writing – review & editing, Visualization, Investigation, Formal analysis. **Abimbola Oluwatayo Orisawayi:** Writing – review & editing. **Shivam Tiwari:** Writing – review & editing, Formal analysis. **Ishrat J. Badruddin:** Writing – review & editing, Formal analysis. **Krzysztof Koziol:** Supervision, Resources. **Sameer S. Rahatekar:** Supervision, Resources.

Declaration of Generative AI and AI-assisted technologies in the writing process

During the preparation of this work the authors used ChatGPT in order to assist with polishing and refining the use of language. After using this tool, the authors reviewed and edited the content as needed and take full responsibility for the content of the publication.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

The first author of this article, Shuai Hao, has submitted a patent application to the China National Intellectual Property Administration related to the technology described in this work. This application does not influence the objectivity or integrity of the research presented. The authors declare no additional competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.ijbiomac.2025.146007>.

Data availability

Data will be made available on request.

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Pioneering microsphere-dope dyeing for sustainable cellulosic fibre colouring

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2025-08

Elsevier

<https://doi.org/10.1016/j.ijbiomac.2025.146007>

Hao S, Shen S, Orisawayi AO, et al., (2025) Pioneering microsphere-dope dyeing for sustainable cellulosic fibre colouring. International Journal of Biological Macromolecules, Volume 320, Part 4, August 2025, Article number 146007