

Preparation and characterization of cellulose nanocrystals (CNCs) from *Pennisetum purpureum* (PP) fibers via ammonium persulfate oxidation method

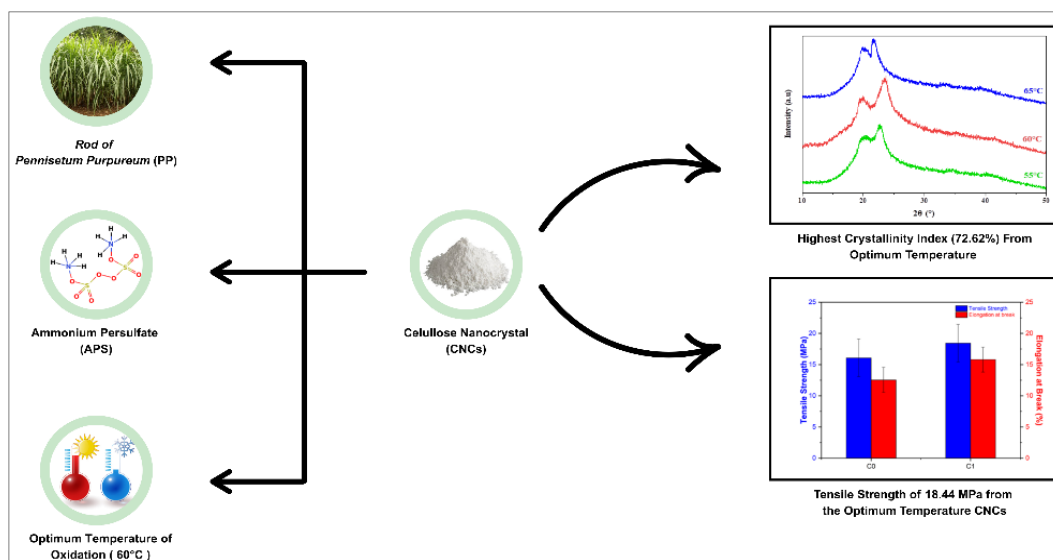
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Highlights:

- CNCs were successfully produced from *Pennisetum purpureum* fibers using the ammonium persulfate (APS) oxidation method, an energy-efficient and eco-friendly process.
- APS oxidation effectively functionalized the CNCs surface, enhancing their solubility and dispersibility, crucial for their integration into nanocomposite materials.
- The study demonstrates the potential of agricultural waste, such as *Pennisetum purpureum*, as a renewable resource for sustainable material production.
- These findings pave the way for developing green materials with superior performance while reducing environmental impact.

Abstract

The growing demand for sustainable and high-performance materials emphasizes the need for more efficient production methods for cellulose nanocrystals (CNCs). Traditional CNC synthesis, however, often requires significant energy input and produces harmful by-products, which undermine its environmental and economic viability. In response to this challenge, this study explores the use of an eco-friendly ammonium persulfate (APS) oxidation method to produce CNCs from *Pennisetum purpureum* fibers. The findings reveal that CNCs synthesized at a temperature of 60 °C exhibited the highest crystallinity index (72.62%), optimal surface functionalization, and exceptional mechanical properties, including a tensile strength of 18.44 MPa. The structural, chemical, and mechanical properties of the CNCs were comprehensively evaluated using X-ray diffraction (XRD), Fourier-transform infrared spectroscopy (FTIR), scanning electron microscopy coupled with energy-dispersive X-ray spectroscopy (SEM-EDX), and tensile strength testing. These results highlight the effectiveness of APS oxidation in producing high-quality CNCs from readily available agricultural biomass. By utilizing a sustainable approach, this research not only advances

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the production of eco-friendly materials but also demonstrates the potential for agricultural waste to be repurposed in nanotechnology applications. The study thus makes a significant contribution to sustainable material science, providing insights into improving CNCs production while minimizing environmental impact, ultimately supporting the transition towards a more sustainable and circular economy.

Keywords: Cellulose nanocrystals (CNCs); *Pennisetum purpureum* (PP); Ammonium persulfate oxidation (APS); Sustainable materials

1. Introduction

Pennisetum purpureum (PP), also known as elephant grass, is a plant from the Poaceae family commonly found in tropical regions such as Indonesia [1], [2]. This plant is recognized for its rapid growth and high productivity, reaching 100–200 tons/ha/year according to data from the Indonesian Institute of Sciences (LIPI) in 2021. To date, the utilization of PP stems has been mostly limited to animal feed, with the remainder becoming biomass waste [3]. Despite its abundance, this waste has not been optimally utilized in industry. With its high cellulose content, PP stems offer significant potential as raw material for cellulose-based products such as Cellulose Nanocrystals (CNCs) [4].

Research on CNCs has grown rapidly in recent years due to their excellent characteristics, including high rigidity, thermal stability, and biodegradability [5], [6]. Due to these distinctive properties, CNCs have been extensively utilized in various fields, such as reinforcement agents [7], [8]. However, the most used CNCs synthesis methods, such as acid hydrolysis, face several challenges. Acid hydrolysis generates hazardous waste and requires time-consuming and costly pre-treatment [9], [10]. Alternative methods, such as TEMPO, also have drawbacks, including long processing times, complex pH control, and environmentally harmful waste production [11], [12]. Previous studies employing APS oxidation have demonstrated great potential for improving CNCs efficiency and quality, but the results still exhibit fluctuations in parameters such as crystallinity index and thermal stability, requiring further optimization [13], [14].

In addition, conventional methods such as sulfuric acid hydrolysis are burdened by significant weaknesses in terms of cost and energy efficiency. The use of concentrated acids generates large amounts of effluents that require expensive neutralization processes, while their corrosive nature increases equipment and maintenance costs [15]. From an energy perspective, conventional synthesis demands high heating input and extended washing steps, leading to excessive water and energy consumption [16]. Furthermore, these methods often result in low CNC yields (17–20%) and reduced thermal stability due to the introduction of sulfate ester groups on the CNC surface, which restricts their applicability in high-temperature processing [17], [18]. Such limitations underscore the need for alternative approaches that are more sustainable, cost-effective, and energy-efficient, motivating the exploration of APS oxidation in this study [19].

This study offers a solution using APS oxidation for CNCs synthesis from PP stems. APS oxidation has several advantages over other methods, including no pre-treatment requirement, relatively low temperatures, economical process costs, and reduced waste production [20], [21]. Additionally, this method not only enhances the purity and crystallinity of CNCs but also introduces carboxyl groups on their surface, improving solubility and potential for further modifications [22]. With this approach, the study contributes to the development of more environmentally friendly CNCs synthesis methods while adding value to abundant and often underutilized local biomass [23], [24].

The objective of this research is to develop an efficient and environmentally friendly CNCs production process using APS oxidation with *Pennisetum purpureum* as the raw material. The method involves optimizing oxidation temperature parameters within a narrower range (55 °C, 60 °C, 65 °C) to produce CNCs with the highest crystallinity index and improved thermal stability. The findings are expected to contribute to the development of biomass-based green materials for sustainable applications such as nanocomposites and electronics. Additionally, this study provides economic benefits by optimizing the use of local resources while reducing the environmental impact of biomass waste.

2. Material and Methods

2.1. Material

The main raw material, *Pennisetum purpureum* (PP), was sourced from Tulungagung, East Java, Indonesia. Stems, rich in cellulose (46%), hemicellulose (34%), and lignin (20%) [25], were

harvested from 4–5 month-old plants. The chemical reagents, ammonium persulfate (APS) and sodium hydroxide (NaOH, ≥99%), were purchased from Sigma-Aldrich, USA.

2.2. Extraction of PP

The preparation process started with manually separating 20 kg of stems from leaves PP. The stems were crushed using a hammer to produce smaller fragments. These fragments were soaked in water for 7 days to remove impurities and loosen the fibers. Extracted fibers were air-dried for 2 days and subsequently oven-dried at 80 °C for 6 hours. The dried fibers were milled using a grinder (Fomac, model FGD-Z300) and sieved to a uniform size of 50 mesh. A schematic representation of the fiber preparation process is illustrated in Figure 1.

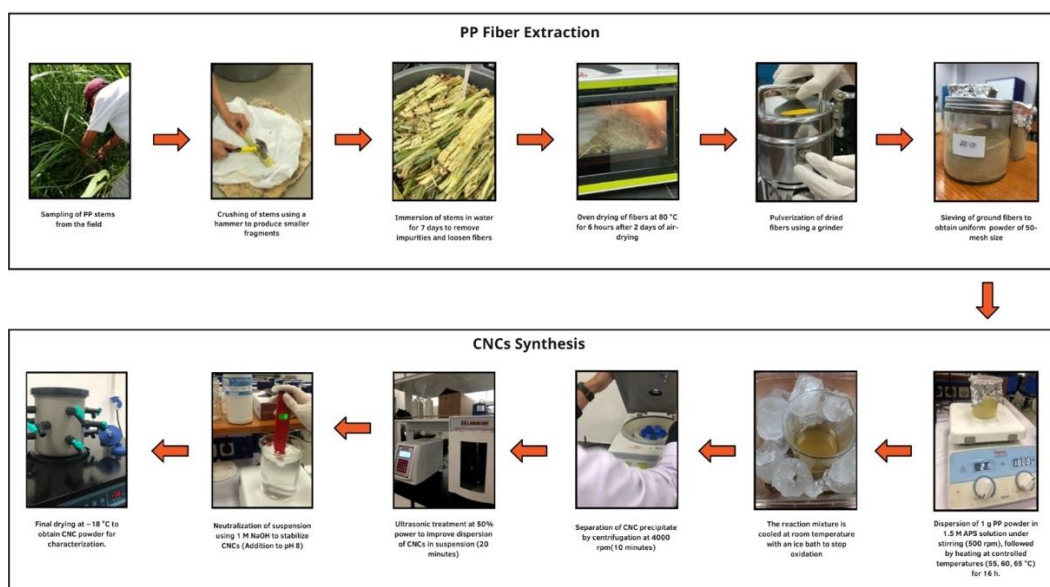


Figure 1.
Extraction of PP
fiber and synthesis
of CNCs

2.3. Synthesis of CNCs

CNCs were synthesized from PP fibers using a modified APS oxidation method [20] as illustrated in Figure 1. Initially, 1 g of fiber powder was dispersed in APS solution (1.5 mol/L) using a magnetic stirrer (Thermo Scientific) set at 500 rpm, and oxidation was performed at three temperature variations (55 °C, 60 °C, and 65 °C) for 16 hours.

The reaction medium was maintained in the acidic range (pH 2–3), generated naturally by APS dissociation, to stabilize sulfate radical formation throughout the process (Liu et al., 2023). The pH was periodically monitored to avoid neutralization, as loss of acidity can suppress radical activity and lower CNC yield. The chosen oxidation temperatures were based on previous studies where APS oxidation at 60 °C produced CNCs with the highest crystallinity and superior morphology compared to 70 and 80 °C [26]. To validate this optimum condition, 55 °C was selected as a lower boundary to observe the effect of reduced energy input, while 65 °C was chosen as an upper boundary to assess the onset of structural degradation prior to the ranges (70–80 °C) already reported to decrease crystallinity and thermal stability.

After the reaction, the samples were cooled, centrifuged at 4000 rpm for 10 minutes (Beckman Coulter, Allegra X-30R), and washed with deionized water. To improve dispersion, the samples were sonicated (Labocon, model LUH-103) at 50% power for 20 minutes, followed by pH adjustment to 8 using 1M NaOH. The CNCs suspensions were then filtered using Whatman filters, freeze-dried at -18 °C for 24 hours, and characterized for structural and chemical properties.

2.4. Characterization

2.4.1. X-ray Diffraction (XRD)

The crystalline structure of the CNCs was examined using XRD (Cu-K α radiation, λ = 0.154 nm) at 40 kV and 30 mA. Samples were scanned at a rate of 0.1°/min within the 10°–50° range. The crystallinity index (CI) was calculated using Segal's empirical method [27], as given by Eq. (1):

$$CI = \frac{(I_{200} - I_{am})}{I_{200}} \times 100\% \quad (1)$$

where I_{200} is the peak intensity at $2\theta \approx 22^\circ$ (crystalline region) and I_{am} at $2\theta \approx 18^\circ$ (amorphous region). Calculation of nanocrystal size can be done with Eq. (2) using Bragg's Law.

$$D_V = \frac{(K\lambda)}{(\beta \cos \theta)} \quad (2)$$

2.4.2. Fourier Transform Infrared Spectroscopy (FTIR)

FTIR spectra were recorded using a Shimadzu IR Prestige 21 spectrometer to identify functional groups. CNCs samples were mixed with KBr in a 1:10 ratio and scanned in the 4000–450 cm^{-1} range at a resolution of 4 cm^{-1} .

2.4.3. Scanning Electron Microscopy (SEM) with Energy Dispersive X-ray (EDX)

The morphology of the CNCs powder was observed using SEM (FEI Inspect-S50), and elemental analysis was performed using EDX. Samples were mounted on carbon tape and coated with gold under an argon atmosphere prior to imaging.

2.4.4. Tensile Strength Test

The tensile strength of CNCs films was tested using a Universal Testing Machine (UTM, JTM) in compliance with ASTM D882. CNCs suspensions were cast onto glass plates, dried for 3 days, and cut into strips (200 × 50 × 1 mm). Samples were conditioned at 23 °C and 50% relative humidity before testing at a strain rate of 10 mm/min.

3. Results and Discussion

3.1. XRD

The XRD analysis confirmed that APS oxidation effectively produced CNCs with high crystallinity. As shown in Figure 2, the XRD diffractograms revealed the characteristic peak of crystalline cellulose at $2\theta \approx 22^\circ$ (plane 200) and an amorphous region near $2\theta \approx 18^\circ$ [28]. The crystallinity index (CI), calculated using Segal's method, indicated that CNCs synthesized at 60 °C exhibited the highest crystallinity (72.62%), followed by those at 65 °C (70.17%) and 55 °C (69.29%) (Table 1).

The CI values obtained (69.29–72.62%) fall within the range reported in recent CNC studies, such as 68–72% for maize stalk CNCs [29], 74.5% for corncob-derived CNCs [30], and 70–75% for APS–microwave-assisted CNCs [31]. These consistencies indicate that APS oxidation under moderate thermal input preserves crystalline order while efficiently removing amorphous regions. This contrasts with sulfuric acid hydrolysis, which often results in CNCs with lower thermal stability due to sulfate ester introduction [32].

The highest CI observed at 60 °C is attributed to optimal oxidation conditions, where energy input is sufficient to selectively remove amorphous regions (hemicellulose and lignin) without

degrading the crystalline structure of cellulose [33]. At 55 °C, insufficient energy likely hindered the removal of amorphous components, leading to a lower CI [34]. Conversely, at 65 °C, excessive thermal energy may have caused partial degradation of crystalline regions, reducing overall crystallinity [35]. This aligns with prior studies that emphasized the delicate balance between energy input and structural integrity in CNCs synthesis [36], [37].

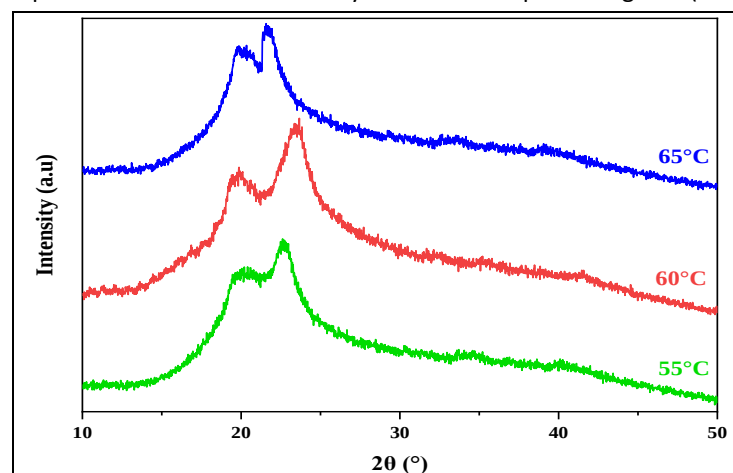


Figure 2.
XRD analysis for
CNCs with different
APS temperatures

Table 1.
CI index and crystal size

Sample	Crystallinity Index (%)	Crystal Size (nm)
55 °C	69.29	30.09
60 °C	72.62	28.71
65 °C	70.17	34.2

High crystallinity is a key parameter for CNCs applications as it correlates directly with improved mechanical and thermal properties [38], [39], [40]. Our tensile results confirm this relationship, as the highest CI at 60 °C coincided with the best tensile strength and modulus, reinforcing the view that crystallinity is the dominant microstructural factor governing macroscopic reinforcement [41].

The dominance of crystalline regions enhances molecular order, facilitating stronger hydrogen bonding within the cellulose structure [42]. This structural organization contributes to better performance in nanocomposites and barrier films, where high mechanical strength and thermal stability are required [43]. Moreover, the narrow variation in CI across the temperature range highlights the robustness of the APS oxidation method in producing consistent results [44].

3.2. FT-IR

The FTIR spectra (Figure 3) and (Table 2) revealed distinct functional group changes associated with APS oxidation. Key absorption bands, such as hydroxyl stretching (O-H, 3300 cm⁻¹) [45], C-H

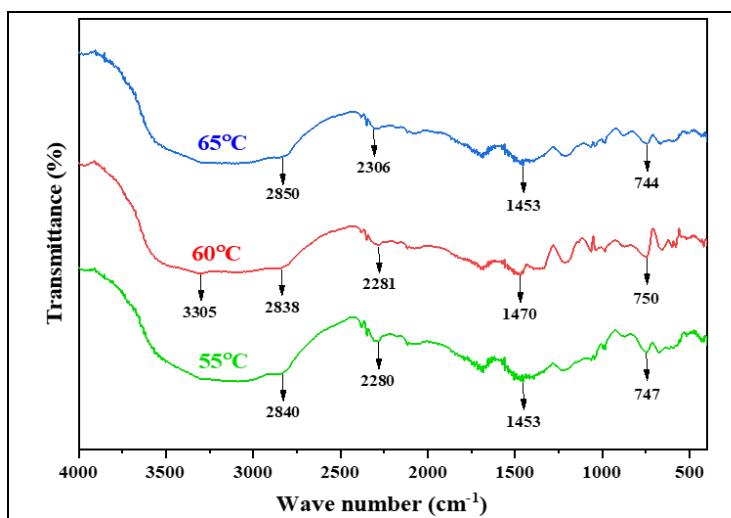


Figure 3.
MQTT explorer for
data monitoring

stretching (2900 cm⁻¹), and carboxylate stretching (-COO⁻, 1500–1600 cm⁻¹) [9], [46], confirmed successful oxidation and functionalization of the cellulose surface. CNCs synthesized at 60 °C exhibited the most intense -COO⁻ bands, indicating effective addition of carboxylate groups, while retaining strong O-H stretching signals that signify a cleaner cellulose structure [47].

Table 2.
The peak positions and
assignments of chemical
groups of CNCs from PP

Wavenumber range (cm ⁻¹)	Types of samples	Source/Possible assignment
3600–3000	3305	O-H stretching (Hydroxyl groups in cellulose)
3000–2800	2850, 2838	C-H stretching (Alkyl groups in cellulose & hemicellulose)
2400–2200	2306, 2281, 2280	C≡N stretching (Nitrile groups), C≡C stretching (Alkyne groups)
1600–1400	1470, 1453	C-H bending (Lignin & cellulose)
800–500	750, 747, 744	C-H out-of-plane bending (Aromatic groups in lignin)

Carboxyl content in APS-oxidized CNCs has been reported at 0.25–0.32 mmol/g [31], values that correlate directly with enhanced elongation and modulus when incorporated into polymer matrices. The dual chemistry, retained hydroxyl groups and introduced carboxylates provides both hydrogen bonding capacity and reactive sites for further modification [48]. In this study, the presence of strong COO⁻ and OH signals at 60 °C confirms that APS oxidation simultaneously preserves crystallinity and enhances surface functionality, an advantage over acid hydrolysis which often reduces interfacial reactivity.

The higher intensity of the -COO⁻ band at 60 °C is indicative of optimal reaction conditions that promote the addition of carboxyl groups to the cellulose backbone [49]. This enhances the hydrophilic properties of CNCs, making them highly dispersible in aqueous media and suitable for applications requiring surface functionalization [50]. At 55 °C, the lower intensity of -COO⁻ bands suggest incomplete oxidation, likely due to insufficient energy for generating free radicals [51]. Meanwhile, at 65 °C, a reduction in O-H and C-H intensities indicates possible thermal degradation, where excessive heat disrupts cellulose molecular structure and leads to loss of functional groups [52].

Enhanced functionalization at 60 °C not only improves dispersibility but also opens pathways for further chemical modifications, such as esterification or grafting reactions. This explains why CNCs obtained here demonstrated superior dispersion and reinforcement capability, aligning with findings that carboxylated CNCs improved interfacial adhesion and elongation by more than 20% in PVA and PLLA composites [53]. These functionalized CNCs are particularly valuable in reinforcing

polymer matrices, as the carboxyl groups increase interfacial bonding, thereby improving mechanical properties [54]. Furthermore, the preservation of O-H groups at 60 °C ensures CNCs compatibility with hydrophilic systems, expanding their versatility in applications such as drug delivery or environmental remediation [55], [56].

3.3. SEM-EDX

The SEM images (Figure 4a) revealed significant differences in CNCs morphologies across synthesis temperatures. CNCs produced at 60 °C displayed smooth, rod-like structures with minimal agglomeration, indicating successful isolation of nanoscale cellulose [20], [57]. The homogeneous surface of the CNCs indicates that the oxidation reaction at 60 °C proceeds optimally, allowing the formation of a more ordered crystalline structure without significant thermal degradation that could occur at higher temperatures [58]. However, SEM imaging alone cannot fully describe dispersion stability. Recent works demonstrate that CNC suspensions often re-agglomerate upon drying, but redispersibility can reach up to 95% when gelatin is incorporated during dehydration [59]. Similarly, lignin-modified CNCs maintained stable dispersions for more than six months. The relatively uniform rod-like morphology observed here suggests that APS oxidation produces CNCs inherently resistant to agglomeration, a feature consistent with the improved colloidal stability reported for carboxylated CNCs at temperatures up to 80 °C, whereas sulfated CNCs destabilize above 50 °C [60].

Furthermore, the presence of uniform particles suggests that the free radical reaction of APS is sufficiently selective in attacking the amorphous regions without damaging the crystalline areas, which is consistent with the highest crystallinity value achieved at this temperature [61]. Sodium can interact with carboxyl and hydroxyl groups, providing additional stability to the CNCs in the form of dispersion [48]. The presence of sodium can also act as a chemical buffer to stabilize the negative charge on the surface of the CNCs due to sulfate groups, which ultimately improves the ability of the CNCs to remain dispersed in a liquid medium without forming agglomerates [62].

Elemental analysis via EDX (Figure 4b and Table 3) showed that CNCs synthesized at 60 °C contained carbon (22.2%), oxygen (46.3%), and sulfur (12.2%). The sulfur content, originating from sulfate groups introduced during APS oxidation, enhances CNCs dispersibility and stability in aqueous systems [31]. Taken together, these results align with broader findings that carboxylated CNCs not only maintain dispersion stability under variable ionic strengths but also provide enhanced compatibility in nanocomposite matrices [48].

Figure 4.
(a) CNCs morphology;
(b) Element results for
APS temperature 60 °C

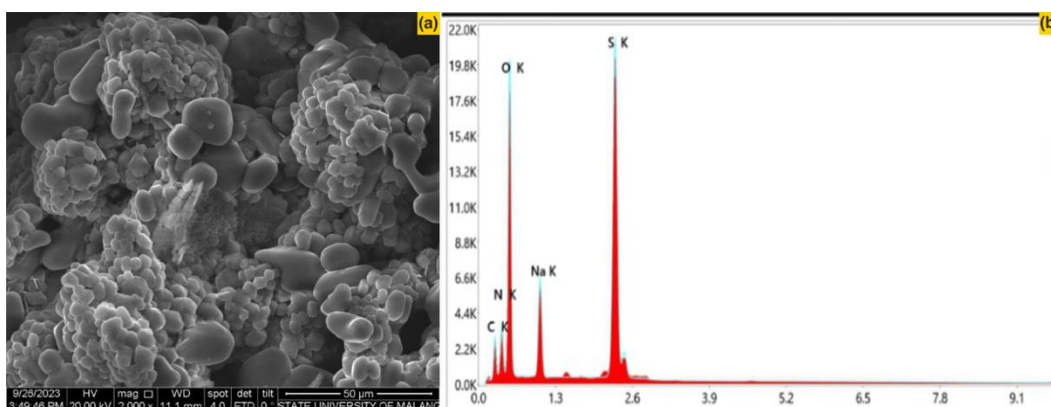


Table 3.
CNCs elements with APS
temperature 60 °C

C	S	O	Na	N
22.2	12.2	46.3	6.1	13.2

3.4. Mechanical Properties

The tensile properties of CNCs-reinforced films were evaluated to determine the mechanical reinforcement capabilities of the synthesized CNCs [63]. As shown in Figure 5 and Table 4, the addition of CNCs significantly improved both tensile strength and elongation at break compared to the neat polymer matrix [64]. Films reinforced with CNCs synthesized at 60 °C exhibited the highest tensile strength (18.44 MPa) and elongation at break (15.8%).

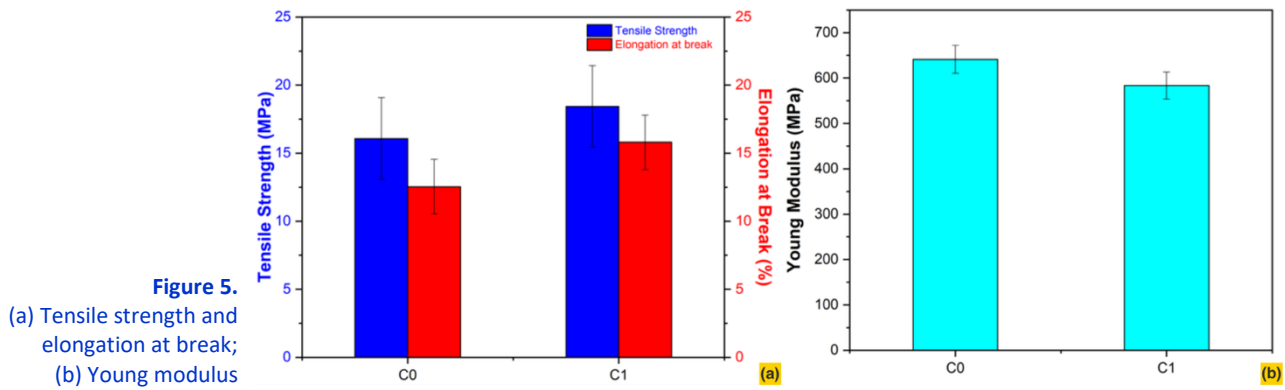


Table 4.
CNCs film properties

Sample	Tensile Strength (Mpa)	Elongation at Break (%)	Young Modulus (Mpa)
C0 (Without CNCs)	16.08 ± 3	12.54 ± 2	641.48 ± 32.07
C1 (With CNCs)	18.44 ± 3	15.8 ± 2	583.33 ± 29.18

This combination of high strength and flexibility is notable because CNCs often improve modulus at the expense of elongation. The balanced outcome here mirrors recent findings where CNCs with moderate crystallinity and carboxylated surfaces enhanced both stiffness and ductility [30]. In comparison, films with CNCs produced at 55 °C and 65 °C showed lower tensile strengths of 16.08 MPa and 17.02 MPa, respectively, with corresponding elongations of 12.54% and 13.96%.

The superior mechanical performance at 60 °C can be attributed to the high crystallinity and uniform morphology of the CNCs (as observed in Figure 4), which facilitated efficient stress transfer from the polymer matrix to the reinforcing phase [34], [65]. Additionally, recent work has emphasized that excessive milling of cellulose prior to hydrolysis can disrupt crystalline domains, reducing modulus despite increased surface area [66]. Our results suggest that APS oxidation at 60 °C achieves reinforcement without over-disruption, as reflected in the consistent modulus values.

Furthermore, the optimized surface functionalization of CNCs at 60 °C, as evidenced by FTIR analysis (Figure 3), likely enhanced interfacial adhesion between the CNCs and the polymer matrix. In contrast, CNCs synthesized at 55 °C, with lower crystallinity and incomplete oxidation, exhibited weaker interfacial bonding, leading to reduced mechanical properties [67]. Similarly, CNCs from 65 °C, although more functionalized, suffered from structural irregularities that compromised stress transfer efficiency [68].

The elongation at break data provides further insight into the role of CNCs in balancing stiffness and flexibility in composites [69]. The higher elongation values at 60 °C suggest improved load distribution and compatibility between the CNCs and the matrix, reducing stress concentrations that could lead to premature failure [70]. The observed trends align with previous studies, where CNCs with high crystallinity and uniform functionalization demonstrated superior reinforcement capabilities in nanocomposites [71]. The scalability of APS oxidation cannot be overlooked. Microwave-assisted APS treatments have reduced reaction times from 16 h to 90 min [31] and life-cycle assessments indicate up to 62% lower environmental burden than sulfuric acid hydrolysis [32]. This places APS oxidation as not only a laboratory success but also a sustainable candidate for industrial CNC production.

4. Conclusion

This study successfully synthesized cellulose nanocrystals (CNCs) from PP fibers using ammonium persulfate (APS) oxidation. The reaction conditions, particularly temperature, significantly influenced the properties of the CNCs. CNCs synthesized at 60 °C demonstrated superior characteristics, including the highest crystallinity index (72.62%), optimal surface functionalization with carboxylate groups, and well-defined rod-like morphology. These properties contributed to enhanced mechanical performance, with films reinforced by CNCs at 60 °C achieving the highest tensile strength (18.44 MPa) and elongation at break (15.8%). APS oxidation is an efficient, eco-friendly method for CNCs production, offering minimal pretreatment, low energy use, and high-quality output. Using abundant PP enhances sustainability by valorizing agricultural waste. The resulting CNCs are suitable for applications in nanocomposites and advanced materials.

Future research should address process scalability, optimize conditions for various biomass sources, and assess CNCs performance in industrial applications. In addition, emphasis should be

placed on evaluating long-term colloidal stability, agglomeration tendencies, and redispersion behavior of APS-derived CNCs under realistic processing environments. Furthermore, compatibility tests with diverse polymer matrices, including thermoplastics, biopolymers, and hydrogels, are necessary to establish performance reliability in real composite systems. These directions will ensure that APS-derived CNCs transition effectively from laboratory synthesis to industrial and commercial applications.

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Authors' Declaration

Authors' contributions and responsibilities - The authors made substantial contributions to the conception and design of the study. The authors took responsibility for data analysis, interpretation and discussion of results. The authors read and approved the final manuscript.

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Availability of data and materials - All data is available from the authors.

Competing interests - The authors declare no competing interests.

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