Influence of Processing Conditions on the Colloidal Stability of CNCs from Coconut Fiber

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The production of cellulose nanocrystals (CNCs) from agro-industrial residues represents a sustainable and technically feasible alternative, promoting the reuse of low-cost materials with high cellulose content. This study evaluated the effect of two bleaching routes of coconut fiber — using buffered sodium hypochlorite (PC) and alkaline hydrogen peroxide (PP) — on the colloidal properties of CNC suspensions obtained by sulfuric acid hydrolysis. Four different reaction conditions were tested, combining acid concentration, time, and temperature, in the following configurations: 40_25_35, 60_25_35, 40_40_35, and 60_40_35. The samples were characterized by dynamic light scattering (DLS) and zeta potential analysis, assessing average particle size (Z-Average), polydispersity index (PdI), and colloidal stability. The results showed that sample PC60_40_35 had the best performance, with a reduced size (255.4 nm), a moderate PdI (0.412), and a high absolute zeta potential (−32.36 mV), indicating good electrostatic stability and a homogeneous particle size distribution. Samples such as PP40_25_35 and PP60_25_35 also exhibited intermediate particle sizes (≈530 nm), but with lower stability. Overall, the PC bleaching route, combined with higher acid concentration and longer reaction time, favored the production of CNCs with more suitable colloidal properties, demonstrating the combined impact of treatments on suspension quality. These data have practical relevance and can be used to optimize future industrial processes for producing nanocellulose.
Keywords: Cellulose Nanocrystals. Coconut Fibers. Bleaching. Acid Hydrolysis. Colloidal Stability.

Growing concerns over the environmental impacts of the exploitation of non-renewable resources have driven a global shift toward more sustainable production models, notably the circular economy. This approach emphasizes the valorization of waste and by- products as feedstocks for new processes, thereby promoting resource recovery and minimizing waste generation. Within the agro-industrial sector, the utilization of lignocellulosic residues has gained prominence as a strategic pathway to develop renewable materials, such as cellulose nanocrystals (CNCs), which offer advanced functional properties and potential applications across a wide range of industrial sectors [1].

Green coconut fiber, abundantly available in Brazil, is often discarded after the consumption of the water and inner pulp, thus becoming a significant environmental residue. Although it exhibits an intermediate cellulose content — ranging from 23% to 43%, depending on factors such as soil conditions, maturation stage, and prior treatments — this fiber has been investigated as an alternative source for the production of cellulose nanocrystals (CNCs), primarily due to its wide availability and low cost [2,3]. CNCs are particles characterized by high crystallinity, rigidity, and colloidal stability, offering significant potential to replace synthetic materials in composites, packaging, coatings, and colloidal systems [4].

The production of cellulose nanocrystals (CNCs) generally involves a sequence of steps, including cellulose bleaching and acid hydrolysis. The bleaching stage aims to remove lignin and hemicelluloses, thereby facilitating acid access to the fiber's amorphous regions. The choice of bleaching agents directly affects the composition, structure, and reactivity of the cellulosic pulp. Buffered sodium hypochlorite (PC) and alkaline hydrogen peroxide (PP) are commonly applied bleaching routes, each exhibiting distinct mechanisms of action, making it essential to understand their specific effects on nanocrystal extraction [5].

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In addition, sulfuric acid hydrolysis requires strict control of variables such as concentration, time, and temperature, as these factors directly influence the morphology, degree of sulfation, and stability of the final product. Improperly adjusted parameters may result in excessive cellulose degradation or the formation of unstable particles with poor colloidal quality [6]. Therefore, the careful selection of hydrolysis conditions, combined with the choice of pretreatment, is crucial to obtaining CNCs with physicochemical properties tailored to their intended applications.

The first colloidal analyses performed in aqueous suspension provide key indicators of the quality of CNC suspensions. Dynamic light scattering (DLS) allows for the measurement of the average particle size (Z-Average) and the polydispersity index (PdI), which indicate the degree of uniformity of the suspension. Zeta potential measurements, in turn, provide information on the electrostatic stability of particles in liquid media, being essential for predicting material behavior in formulations and during storage. In general, absolute zeta potential values greater than 30 mV are associated with more stable suspensions [7]. Therefore, the integrated analysis of bleaching routes, hydrolysis parameters, and colloidal properties in suspension is essential to optimize the CNC production process and ensure materials with suitable quality for technological applications.

Material and Methods

Green coconut fiber was first washed, oven-dried (Quimis Q314M222) at 60 °C for 24 h, and milled using a Wiley-type mill (Tecnal, model R-TE- 650/1). The resulting material was sieved (Bertel, AGT.P) through a 40-mesh screen (~425 µm) and subjected to a mercerization step, using 60 g of fiber in a 2% (w/v) sodium hydroxide solution under constant stirring (IKA, C-MAG HS4) at 80 °C. This procedure was repeated for 4 cycles to partially remove hemicelluloses and promote cell wall swelling, thereby facilitating access to the cellulosic constituents.

Following this step, the mercerized pulp was divided and subjected to two distinct chemical bleaching routes:

PC route: treatment with 1.7% (v/v) sodium hypochlorite buffered with acetate solution (pH ≈ 4.45), carried out at 80 °C for 6 h under constant stirring (IKA, C-MAG HS4).

PP route: treatment with 16% (v/v) hydrogen peroxide buffered with 5% (w/v) sodium hydroxide, carried out at 55 °C for 2 h under constant stirring (IKA, C-MAG HS4).

In both routes, the post-bleaching material was filtered, dried (Quimis, Q314M222), and milled (Ariete, Universal PRO Grinder).

Acid hydrolysis was carried out in a water bath at 35 °C, using 2 g of bleached pulp in 40 mL of sulfuric acid (H_2SO_4), with variations in concentration and reaction time as indicated in Table 1. At the end of the reaction time, 40 mL of cold water was added to quench the reaction, and the hydrolyzed solution was placed in an ice-water bath until it reached 20 °C. The sample was then centrifuged (Hettich, Rotina 380R) at 4400 rpm for 10 min, with 5 mL of distilled water added to the Falcon tubes after each cycle until a final volume of 20 mL was reached. The supernatants were dialyzed against a cellulose membrane (model D9402, Sigma-Aldrich) until neutral pH was reached.

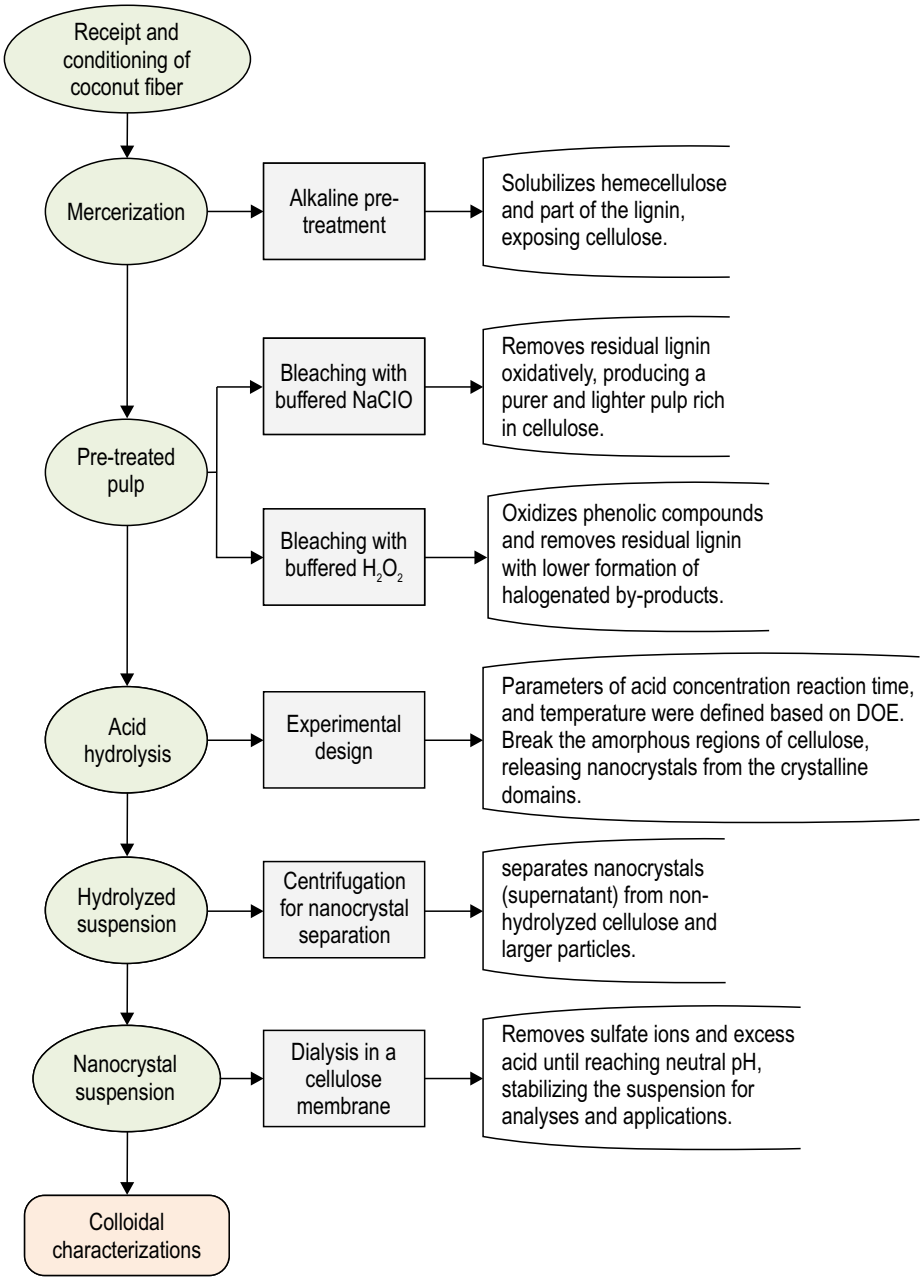
A flowchart (Figure 1) of the process for obtaining the nanocrystals is provided below.

The colloidal characterization of cellulose nanocrystal (CNC) suspensions was performed using Dynamic Light Scattering (DLS) and zeta potential analysis on a Zetasizer Ultra (Malvern Panalytical) equipped with a Non-Invasive Back Scatter (NIBS) system and a fixed backscatter detector angle of 173°. Samples were previously redispersed in ultrapure water (resistivity ≥ 18.2 MΩ·cm), standardized to 0.01% (w/v), and subjected to indirect sonication for 10 min in an ultrasonic bath (50/60 Hz) to disrupt agglomerates.

Table 1. Reaction conditions applied for the acid hydrolysis of bleached coconut fibers.

Sample Code	H_2SO_4 concentration (%)	Reaction time (min)	Temperature (°C)
40_25_35	40	25	35
60_25_35	60	25	35
40_40_35	40	40	35
60_40_35	60	40	35

Figure 1. Simplified flowchart of the methodology for obtaining cellulose nanocrystals and the objective of each step.



The average particle size (Z- Average), polydispersity index (PdI), and zeta potential were obtained from three independent measurements per sample at 25 °C, using disposable cells suitable for each analysis.

Results and Discussion

The colloidal evaluation of cellulose nanocrystal (CNC) suspensions obtained from coconut fiber enabled the identification of the direct effects of pretreatment steps (bleaching) and hydrolysis conditions on the material's physicochemical parameters. Table 2 presents the results for zeta potential, average particle size (Z-Average), and polydispersity index (PdI) for the eight samples obtained from the PC and PP routes.

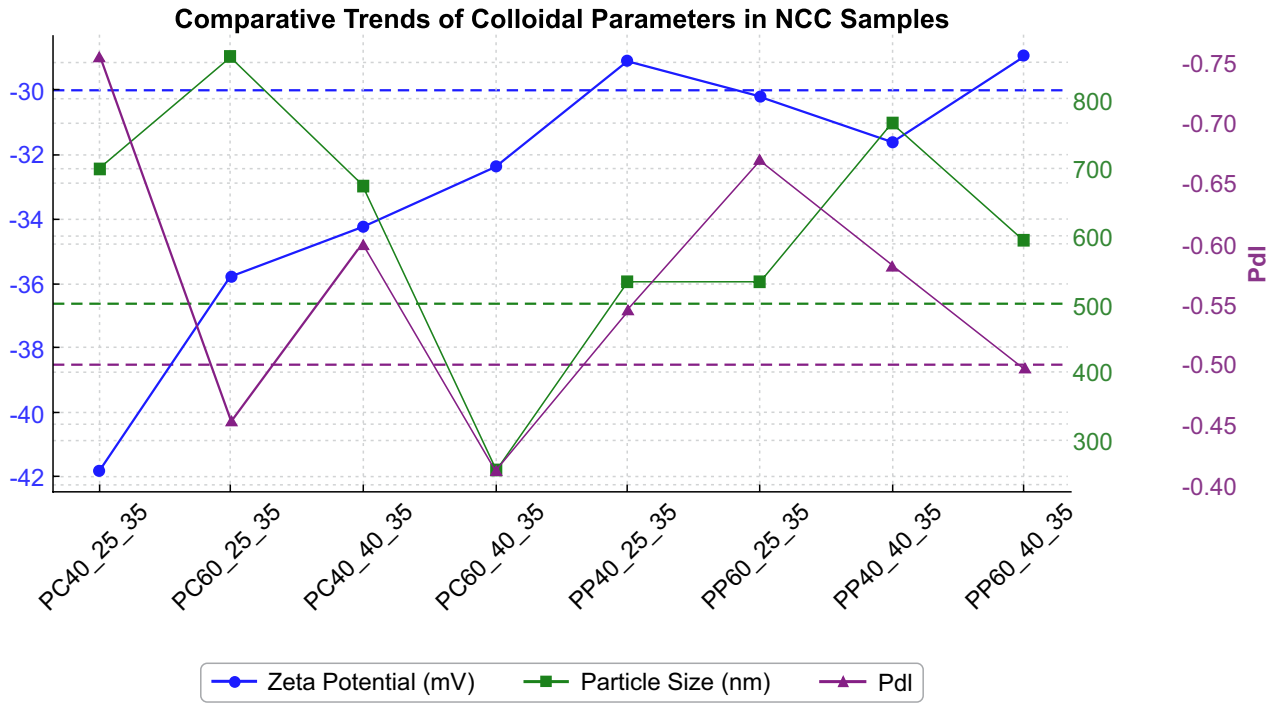
According to the literature, the ideal criteria for CNCs in suspension indicate that absolute zeta potentials greater than 30 mV indicate good electrostatic stability [7]. At the same time, average particle sizes of 100-500 nm suggest effective degradation of amorphous regions while preserving the integrity of crystalline domains [8]. Additionally, PdI values below 0.3 indicate monodispersion; however, materials obtained from agro-industrial residues often exhibit values up to 0.5, which are still considered acceptable [3]. Figure 1 presents an integrated comparison of the three main colloidal parameters analyzed: zeta potential, particle size, and polydispersity index (PdI). The combined analysis facilitates the identification of the sample with the best performance in terms of stability, uniformity, and hydrolysis efficiency (Figure 2).

In this context, the PC60_40_35 sample exhibited superior performance, with a zeta potential of –32.36 mV, a particle size of 255.4 nm, and a PdI of 0.412. This profile suggests a stable suspension, with relatively homogeneous particles and dimensions comparable to those reported for sisal residue, corn husk, and rice husk fibers, which have a lignocellulosic composition similar to that of coconut fiber. When compared to CNCs obtained from rice straw [9], which presented significantly smaller dimensions (11.2–30.7 nm in width and 117–270 nm in length, as determined by microscopy), the CNCs produced in this study (255.4 nm hydrodynamic diameter, –32.36 mV zeta potential, PdI = 0.412) exhibited particle sizes closer to the reported lengths but considerably larger widths, which is expected given that DLS measures hydrodynamic diameters of aggregates in suspension. In relation to nanocellulose derived from sugarcane bagasse [10], which was reported to have particle sizes within the nanorange by DLS without specific values in the abstract, the CNCs from coconut fiber showed a tendency toward larger hydrodynamic sizes, possibly due to residual lignin and hemicelluloses or the specific hydrolysis conditions employed. Nevertheless, the zeta potential obtained (–32.36 mV) indicates good

Table 2. Results of colloidal stability, particle size, and dispersion analyses of nanocrystals in suspension.

Sample Code	Zeta Potential (mV)	Particle Size (d.nm)	Polydispersity Index (PdI)
PC40_25_35	-41.80	699.0	0.756
PC60_25_35	-35.76	865.5	0.453
PC40_40_35	-34.22	673.9	0.600
PC60_40_35	-32.36	255.4	0.412
PP40_25_35	-29.08	533.9	0.545
PP60_25_35	-30.19	534.6	0.669
PP40_40_35	-31.59	765.5	0.582
PP60_40_35	-28.93	593.6	0.496

Figure 2. Comparative trends of colloidal parameters in NCC samples.



electrostatic stability, comparable to or potentially superior to that observed for CNCs from both rice straw and sugarcane bagasse, highlighting the potential of coconut fiber as a competitive lignocellulosic source for CNC production.

The influence of the bleaching route was also evident. The PC route (buffered hypochlorite) provided greater electrostatic stability across all tested conditions. This can be attributed to more efficient lignin removal and the exposure of reactive sites on the fiber surface, which facilitate acid action during hydrolysis. In contrast, the PP route, although also oxidative, yielded zeta potential values below |30| mV, suggesting a lower density of negative surface charges. When compared to CNCs obtained from cotton via different bleaching methods followed by acid or enzymatic hydrolysis [11], the CNCs produced in this study demonstrated a level of electrostatic stability above the |30| mV threshold, indicating a high density of negative surface charges and good suspension stability. In the cotton-based study, variations in bleaching method were shown to influence hydrodynamic properties and surface charge density significantly. However, specific zeta potential values were not provided in the abstract. Considering this, the stability observed in the coconut fiber CNCs produced under the PC60_40_35 condition suggests that the chosen bleaching and hydrolysis parameters effectively enhanced the surface charge density, achieving stability levels likely comparable to or greater than those reported for cotton-derived CNCs, despite differences in raw material composition and fiber morphology.

Regarding the polydispersity index, none of the samples reached the ideal value of < 0.3. However, the values observed for samples PC60_40_35 (0.412), PC60_25_35 (0.453), and PP60_40_35 (0.496) indicate a reasonably acceptable distribution for systems derived from complex fibers. In the present study, the CNCs obtained from green coconut fiber indicate a moderately narrow particle size distribution and an acceptable level of monodispersion for CNCs derived from complex lignocellulosic fibers. In contrast, studies involving coconut shell powder or particles as fillers in polymeric matrices [12,

13] do not report quantitative PdI values; however, SEM analyses reveal agglomeration and an uneven distribution of filler particles, suggesting a high degree of polydispersity at the microscale. These observations align with the understanding that achieving low polydispersity is more feasible in nanocellulose suspensions, where surface chemistry and colloidal stabilization mechanisms can be optimized, than in particulate polymer composites, where interfacial adhesion and particle wetting dominate dispersion behavior.

Additionally, the reduction in average particle size observed for the PC60_40_35 sample suggests that the combination of a high acid concentration and a prolonged reaction time, together with effective pretreatment, promoted the selective degradation of amorphous regions without compromising the crystalline structure. Thus, the results demonstrate that the PC route combined with the 60_40_35 condition produces CNCs with superior colloidal characteristics compared to the other conditions, making it the most promising option for future applications in films, coatings, or nanocomposites. The similarity between the results obtained and those reported in the literature for lignocellulosic residues confirms the feasibility of coconut fiber as an alternative source of cellulose nanocrystals.

Conclusion

The objective of this study was successfully achieved by producing cellulose nanocrystals (CNCs) from coconut fiber, using different bleaching routes and acid hydrolysis conditions, followed by a comprehensive evaluation of their colloidal properties. The results demonstrated that the PC bleaching route, when combined with the 60_40_35 hydrolysis condition, provided CNCs with greater electrostatic stability, adequate particle size distribution, and dimensions comparable to those reported for other lignocellulosic residues.

These findings confirm the potential of coconut fiber as an alternative, sustainable source for CNC production, with promising applicability in films, coatings, and nanocomposite materials. Furthermore, future studies should focus on incorporating these CNCs into polymeric matrices to evaluate their reinforcing effect, mechanical performance, and compatibility with biodegradable polymers, thus expanding their potential for industrial applications in sustainable packaging, advanced composites, and functional coatings.

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