

Production of nanofibrillated cellulose from agrowaste residues and its application in phosphate removal from wastewater



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By

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CHAPTER 6

CONCLUSIONS AND FUTURE PERSPECTIVES

This study investigated an integrated and sustainable strategy for biomaterial development by converting agricultural residues, specifically sugarcane bagasse, into nanofibrillated cellulose. Through targeted modification techniques, the potential of the synthesized NFC for efficient phosphate removal from wastewater was systematically evaluated. In this chapter, key findings and the conclusions from previous chapters are summarised and compiled to give an overview of the thesis. In addition, future directions following the current work are discussed.

(a) Employing a two-stage pretreatment process involving acidic NADES, acetosolv and HIU, the method promotes green chemistry principles, waste valorisation, and efficient resource utilization. Among the NADES synthesized and characterized via FTIR and NMR, Gly-CA (glycerol–citric acid) exhibited the most effective removal of hemicellulose, while acetosolv pretreatment achieved efficient delignification with minimal loss of cellulose. The integrated process yielded an impressive 84.4% cellulose recovery, with subsequent HIU-assisted defibrillation resulting in a high NFC production yield of $\geq 90\%$. The structural and morphological integrity of the resulting NFC was confirmed through extensive characterizations including FT-IR, DLS, SEM, TEM, XRD, SEM-EDX, and TGA/DTG. These results were in strong agreement with literature-reported values, affirming the consistency and quality of the developed NFC.

Beyond the production of biomaterial, this thesis explored value-added applications of the produced NFC in environmental remediation. Two functionalized NFC-based adsorbents were developed and evaluated for their phosphate removal potential:

- (b) Cationic NFC (CTAB@NFC): Synthesized via a straightforward quaternization technique using cetyltrimethylammonium bromide (CTAB), this adsorbent achieved a maximum phosphate adsorption capacity of 21.78 mg/g. Adsorption followed the Sips isotherm model, indicating surface heterogeneity, and the kinetics conformed to the pseudo-second-order model, suggesting chemisorption as the dominant mechanism. Hydrogen bonding and electrostatic interactions were identified as key drivers of phosphate capture. Notably, CTAB@NFC showed excellent stability and reusability over three adsorption-desorption cycles, retaining its performance and structural integrity in aqueous environments.
- (c) Magnesium-doped NFC/BN Composite (Mg@NFC/BN): Engineered by incorporating Mg^{2+} ions and bentonite clay into the NFC matrix, this composite demonstrated a slightly lower yet competitive adsorption capacity of 19.2 mg/g. The Langmuir isotherm model best fit the data, indicating monolayer adsorption on a uniform surface, and the process was governed by pseudo-second-order kinetics. Thermodynamic analyses confirmed the spontaneous and exothermic nature of the adsorption. Additionally, the phosphate-loaded composite showed potential for reuse in soil water retention, offering dual environmental benefits.

This work shed the light on the required properties in adsorbents for phosphate removal, which were identified by the investigation of the phosphate adsorption mechanism in both adsorbents. The presence of quaternary ammonium groups in the phosphate adsorbent material (CTAB@NFC) increases the overall positive surface charge,

resulting in the enhancement of the adsorption performance. The positively-charged surface of the adsorbent materials contributes to the phosphate adsorption via electrostatic attraction. Also, in metal doped composite adsorbent (Mg@NFC/BN) the surface hydroxyl groups of NFC/BN composite attached to the metallic species become protonated, where phosphate is then chemisorbed via surface complexation. While both adsorbents demonstrated high efficiency, CTAB@NFC outperformed Mg@NFC/BN in terms of adsorption capacity and reusability, making it the more effective candidate for phosphate removal from wastewater. Its superior performance is attributed to the high surface area of NFC and the strong electrostatic attraction provided by the cationic quaternary ammonium groups. However, the Mg@NFC/BN composite offers a multifunctional advantage by combining pollutant removal with soil enhancement capabilities, thus supporting circularity and agricultural sustainability.

Summary and Future Perspectives

Building on the findings of this study, several promising avenues for further research and application can be proposed. For example, although CTAB proved effective for quaternization, it is a synthetic surfactant with limited biodegradability. Future studies should explore green and biodegradable cationic agents, such as green surfactants, chitosan, quaternized starch, or amino acid-based compounds, to replace CTAB and enhance the environmental safety profile of NFC-based adsorbents.

The development of NFC composites capable of simultaneously adsorbing multiple contaminants (e.g., nitrates, heavy metals, dyes, antibiotics) would significantly broaden the scope of water purification applications. Synergistic formulations,

combining NFC with graphene-based composites, biochar, zeolites, or MOFs, could be tailored for specific environmental targets.

Most adsorption studies are performed under controlled laboratory conditions. To advance the practical implementation of NFC-based adsorbents, future work must focus on pilot-scale studies using real industrial or municipal wastewater, considering factors such as pH variability, competing ions, flow rates, and regeneration cycles.

The phosphate-laden Mg@NFC/BN composite presents an opportunity for slow-release fertilizers and soil conditioning, integrating waste remediation with sustainable agriculture. Similarly, CTAB@NFC composites may be investigated for their antimicrobial properties, potentially serving in medical textiles, wound dressings, or food packaging due to the inherent antibacterial and antifungal activity of CTAB.

While numerous advanced materials have been proposed for environmental remediation, their large-scale application often faces limitations due to high synthesis costs and energy requirements. The preparation of Mg@NFC/BN composite and CTAB@NFC in this study was intentionally designed to minimize such drawbacks by employing low-cost and abundantly available precursors, including sugarcane bagasse-derived nanofibrillated cellulose and naturally occurring bentonite clay, both of which are industrial by-products or locally sourced materials. The only chemical reagent used in significant quantity during synthesis was $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$, which is commercially inexpensive (approximately 0.25–0.35 USD kg^{-1} in bulk). Based on current laboratory-scale synthesis, the estimated material cost of Mg@NFC/BN is roughly 0.9–1.2 USD per kg, which is significantly lower than many synthetic phosphate adsorbents such as metal–organic frameworks or lanthanum-based oxides (5–20 USD kg^{-1}). Moreover, the

composite's dual functionality, its reuse potential as a phosphate-enriched soil conditioner, eliminates post-treatment waste disposal costs and provides an added-value pathway for agricultural applications. Thus, despite involving multi-step synthesis, the Mg@NFC/BN composite remains economically feasible for practical implementation, particularly in decentralized wastewater treatment and nutrient recovery systems. Future work should include life cycle cost assessments and regeneration studies to further evaluate the long-term economic and environmental benefits of this material at pilot scale. While in another study that uses CTAB for modification, the overall preparation of CTAB@NFC remains economically feasible due to the small quantity of CTAB required for surface modification. Here, the optimal CTAB concentration was 1 mM, which results in a negligible cost contribution (CTAB ≈ 10 USD kg⁻¹). Considering that the main substrate i.e., nanofibrillated cellulose, it is derived from agro-waste sugarcane bagasse, and the synthesis process involves only mild alkaline treatment and ambient conditions, the total material cost was estimated at approximately 1.0–1.3 USD kg⁻¹. Furthermore, CTAB imparts stability and reusable characteristics to the modified NFC, enhancing its durability and enabling potential reuse in multiple adsorption cycles. Hence, despite the incorporation of CTAB, the developed CTAB@NFC adsorbent remains cost-effective and practically applicable for phosphate removal from wastewater. In addition, this work aligns closely with global efforts toward a circular bioeconomy, where biomass waste is transformed into high-value, reusable materials. Future collaborations with industries and policymakers could facilitate the integration of such technologies into existing biorefinery frameworks, strengthening supply chains and promoting sustainable development goals (SDGs),

particularly SDG 6 (Clean Water), SDG 12 (Responsible Consumption and Production), and SDG 13 (Climate Action).

Thus, the research successfully bridges biomass valorization, green chemistry, and environmental engineering by converting sugarcane bagasse into nanocellulose-based materials for high-value applications. The NFC-based adsorbents, particularly CTAB@NFC, have demonstrated promising capabilities in phosphate removal, offering cost-effective, efficient, and sustainable solutions for water treatment. By leveraging agro-waste, green solvents, and functional modification strategies, this study contributes significantly to the development of sustainable materials and clean technologies that support environmental stewardship, resource circularity, and the broader vision of a sustainable future.