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Fabrication of nanocellulose-EDTA composite from oil palm trunks for cadmium removal from aqueous solutions

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Abstract

The development of nanocellulose-based materials with enhanced properties has garnered significant interest among scientists. Oil palm trunks are a promising source of nanocellulose due to their high cellulose content, excellent adsorption capacity, and abundant availability. Cadmium, a toxic heavy metal, poses serious risks to environmental and human health. It accumulates in fish and plants, entering the human body through the food chain. Since cadmium resists natural degradation, effective removal methods are crucial to mitigate its hazardous effects. In this study, a functionalized nanocellulose composite (NCE) was successfully synthesized using ethylene-diaminetetraacetic acid (EDTA) as a chelating agent to enhance cadmium adsorption in aqueous solutions. Analysis with FTIR confirmed the reaction between nanocellulose and EDTA, with characteristic bonds appearing at wavenumber 1100, 1172, and 48 cm^{-1}. Particle size analysis revealed polydisperse nanoparticles, with average sizes of 411.5 nm for nanocellulose and 665.3 nm for NCE. Microscopic imaging showed distinct morphological changes, indicating successful EDTA incorporation into the nanocellulose structure. NCE exhibited a high surface area (2.792 m^2 g^{-1}). Atomic absorption spectroscopy showed a decrease in cadmium concentration, from 1 ppm to 0.2–0.3 ppm, indicating the adsorption ability of NCE. These findings highlight the potential of NCE for heavy metal remediation in water.

[Keywords: adsorption, cadmium, EDTA, membrane, nanocellulose]

Introduction

Biomass is typically derived from plant and animal sources such as sewage and solid waste. Oil palm biomass specifically comes from replanting, milling, and pruning activities. There are significant challenges in converting this biomass into valuable products with diverse applications. Analysis of oil palm trunk biomass reveals high chemical components including carbon (51.41%), nitrogen (0.17%), hydrogen (11.82%), oxygen (51.16%), and no sulfur. Additionally, these materials contain high levels of cellulose (34.44%), hemicellulose (23.94%), and lignin (35.89%) (Onoja et al., 2019). Given the high cellulose content, there's a practical opportunity to convert oil palm trunks into valuable products such as nanocellulose.

Cadmium is a toxic heavy metal that readily accumulates in plants and animals. This metal can be absorbed into the human body through the food chain. Because cadmium is persistent, toxic, and environmentally harmful, it's crucial to reduce this pollutant to maintain environmental stability, particularly in aquatic ecosystems. Addressing the negative effects of this heavy metal requires highly efficient technology. Consequently, various treatments have been employed to remove cadmium from contaminated waters, including chemical electrodeposition, precipitation, membrane filtration, and adsorption (Akl et al., 2015). Water quality regulations regarding cadmium set the maximum limit at 0.05 ppm for waste and drinking water, according to the World Health Organization and Indonesian environmental regulations (Kementerian Lingkungan Hidup Republik Indonesia, 2014).

Adsorption is frequently used due to its eco-friendly nature, ease of operation, low cost, high efficiency, rapid pollutant removal rate, and reversibility. Various adsorbent materials such as zeolite, activated carbon, clay, and apatite have been used for metal adsorption. Despite the existence of materials with good adsorption performance, the search continues for more cost-effective, environmentally safe, and excellent adsorbents. Currently, nanocellulose materials are emerging for

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potential use in water treatment due to numerous advantages including low cost, renewability, biodegradability, and easy functionalization capability (Lim, et al., 2016; Ayob et al., 2021).

Most cellulose-based adsorbents reported in literature focus on grafting amino and carboxyl groups. For instance, Saberi et al. (2023) investigated cadmium adsorption using carboxymethyl cellulose (CMC) bridged chlorapatite. Their results demonstrated that these materials can effectively remove cadmium with a capacity of 150.2 mg g^{-1}. Chen et al. (2019) successfully modified cellulose adsorbent with multi-functional groups for cadmium removal in water, achieving a maximum adsorption capacity of 277 mg/g (Ablouh et al., 2022; Faisal et al., 2022). Hubbe et al. (2011) also successfully removed dye pollutants from aqueous systems using cellulosic substrates. While cellulose materials show considerable effectiveness in removing pollutants, attention must be paid to enhancing their physical and chemical properties.

In its native form, cellulose has a small specific surface area and uncharged structure, which limits its adsorption capacity. Larger surface are of nanocellulose makes it a better alternative for metal ion adsorption (Rahman et al., 2023). Nevertheless, research has been widely focused on improving the chemical and adsorption properties of native cellulose by impregnating active functional groups onto its chemical bonding. This modification allows for better immobilization of heavy metal ions (Yang & You, 2023).

Various research has been conducted on nanocrystalline cellulose from different sources as biosorbents for metal adsorption. These materials have been derived from bacteria, oil palm empty fruit bunch, orange peels, cotton, as well as modified cellulose nanocomposite hydrogels and cross-linked nanocrystalline cellulose aerogels. This work specifically uses oil palm trunks as raw material for adsorbent production. Oil palm trunks have a relatively high cellulose content of 40% and are abundantly available in Indonesia.

Phosphorylated cellulose, phosphate groups, and metal-binding compounds have also been used as adsorbents. Phosphate groups have the ability to reduce cellulosic materials (Odenigbo & Micheal, 2023). Additionally, cellulose has been functionalized with commercial chelating agents such as EDTA, deferasirox, D-penicillamine, and meso-2,3-dimercaptosuccinic acid. Cellulose modification through esterification with carboxylic acid anhydride has also been explored. These materials are used in various applications to address numerous pollution issues (Suppapruek et al., 2021). However, this concept has not yet been fully demonstrated for optimized removal of cadmium from polluted water. The purpose of modifying cellulose structure is to improve its physical properties and chemical resistance. For example, succinyl mercerized cellulose modified with EDTA for cadmium adsorption has demonstrated a capacity of 192.3 mg/g (Akl et al., 2015; Yang & You, 2023).

The characteristic EDTA-like cellulose derivative functionalized with multidentate N/O atom donors provides adsorptive properties for cadmium (Yang & You, 2023). In this study, EDTA was used to modify cellulose membranes because Na_2EDTA can form stable complex compounds with heavy metals such as cadmium.

Material and Methods

Preparation of nanocellulose crystal membrane from oil palm trunks

Oil palm trunk waste collected from Capari village, Cilacap, Indonesia, was processed through mechanical grinding to achieve 60-mesh particle size. The oil palm trunks were prepared through sequential drying, grinding, and screening processes. The resulting powder underwent a three-stage chemical treatment protocol consisting of alkali pretreatment, bleaching, and acid hydrolysis.

In alkali pretreatment, 5 g of oil palm trunk powder was treated with 4% (w/v) NaOH solution at a solid-to-liquid ratio of 1:25. The suspension was stirred at 300 rpm for 2 h at 80°C. The treated material was washed with distilled water and dried at 105°C.

The alkali-treated sample was subjected to bleaching using 1.7% sodium chlorite ($NaClO_2$) solution (100 mL) and acetate buffer (110 mL, pH 5). The mixture was stirred at 300 rpm for 2 h at 80°C, followed by thorough washing to neutral pH and drying at 60°C.

Cellulose hydrolysis was performed using 50% H_2SO_4 at a cellulose-to-acid ratio of 1:20. The reaction proceeded at 40°C for 45 min and was terminated by ten-fold dilution with distilled water. Excess acid was then removed by centrifugation at 700 rpm for 15 min.

Nanocellulose-EDTA (NCE) composite synthesis

The composite was prepared by mixing 52 g of EDTA (Merck) with 1 g of nanocellulose under continuous stirring at 300 rpm for 20 min. The mixture was ultrasonicated for 30 min to ensure homogeneous dispersion, followed by freeze-drying at -40°C for 24 h.

Characterization of NCE

Functional group analysis was conducted using fourier transform infrared spectroscopy (FTIR, PerkinElmer) over the wavenumber range of 4000-400 cm^{-1}. Surface area and porosity were determined by nitrogen adsorption-desorption isotherms at 77 K using Brunauer-Emmett-Teller (BET, Quantachrome Novatouch LX4) analyzer following degassing at 120°C for 6 h. Morphological characterization was performed using field emission scanning electron microscopy (FESEM, Hitachi Regulus 8220) and transmission electron microscopy (TEM, JEOL JEM-1400). Particle size distribution was analyzed using a particle size analyzer (PSA, Malvern).

NCE adsorption test for heavy metal cadmium (Cd)

Batch adsorption experiments were conducted using 0.04 g of NCE in 1 L of Cd solutions with initial concentrations of 1 mg L^{-1} and 60 mg L^{-1}. The suspensions were agitated at 200 rpm, and samples (20 mL) were collected at predetermined time intervals (20, 60, and 100 min) for Cd quantification using atomic absorption spectroscopy (AAS, Shimadzu AA7000).

Results and Discussion

Alkali pretreatment effectively removed some hemicellulose and lignin in powdered oil palm trunk, as evidenced by the enhanced surface roughness and ordered structure. The NaOH treatment facilitates the removal of pectin, waxy substances, and natural oils while increasing the accessibility of hydroxyl functional groups. Subsequent bleaching with sodium chlorite removed residual lignin, yielding purified cellulose fibers (Lim et al., 2016) rich in hydroxyl functional groups (-OH). The acid hydrolysis process oxidizes and cleaves ester bonds linking lignin-hemicellulose and lignin-carbohydrate complexes, facilitating a complete lignin removal and exposing crystalline cellulose domains (Nafi'ah & Primadevi, 2020). The synthesized nanocellulose and NCE exhibited sheet-like morphology with smooth surfaces (Figure 1).

Analysis with FTIR spectroscopy confirmed the successful modification of nanocellulose with EDTA (Figure 2). In the absence of EDTA, the oil palm trunk-derived nanocellulose showed stronger absorption bands at 2900-2800 cm^{-1} which are characteristic of C-H stretching in cellulose derivatives (Babaei-Ghazvini & Acharya, 2023). A broad absorption around 3487 cm^{-1} and a sharper peak at 1560 cm^{-1} indicated strong O-H stretching while a peak at 1050 cm^{-1} indicated C-O bonds in both nanocellulose samples. The absence of peaks around 1253 cm^{-1}, characteristic of C-O-C stretching vibrations of aromatic ether linkages, indicated complete lignin removal during alkali treatment.

The NCE composite displayed additional absorption band at 1172 cm^{-1}, corresponding to C-N stretching vibrations characteristic of EDTA incorporation (Adsul et al., 2012; Naihi et al., 2021). This spectroscopic evidence confirms the successful functionalization of nanocellulose with EDTA, creating new binding sites for metal ion coordination.

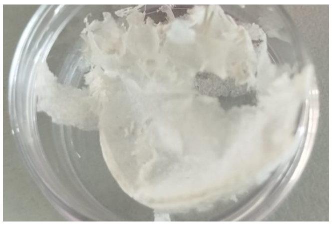

Figure 1. Sheet-like morphology of nanocellulose and NCE synthesized from ground oil palm trunks

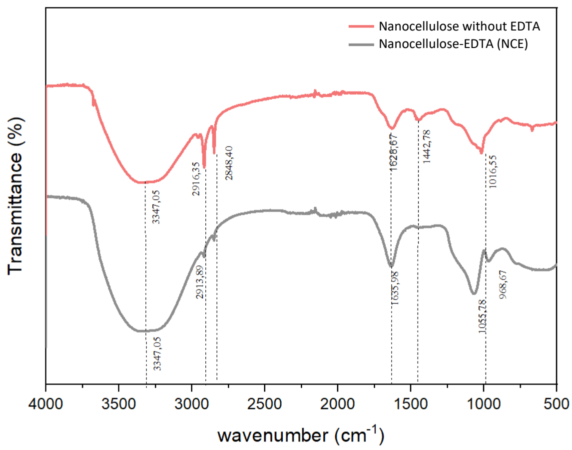

Figure 2. FTIR spectra of oil palm trunk-derived nanocellulose with (NCE, grey) and without EDTA (red).

Microscopic analysis with TEM revealed that NCE consists of quasi-spherical nanoparticles with nanofibrillar structures extending from the particle surfaces (Figure 3). The morphology resembles that of nanocrystalline cellulose (NCC), which is typically rod-like with lengths ranging from 50 to 500 nm (Balea et al., 2021) and is commonly produced via acid hydrolysis using concentrated sulfuric acid to remove amorphous regions and enhance crystallinity. In this study, the NCC-like structure of NCE supports the use of sulfuric acid hydrolysis during synthesis. The morphological differences between the smooth surface of EDTA-free nanocellulose and the nanofibrillar extensions observed in NCE provide visual confirmation of EDTA integration into the cellulose matrix.

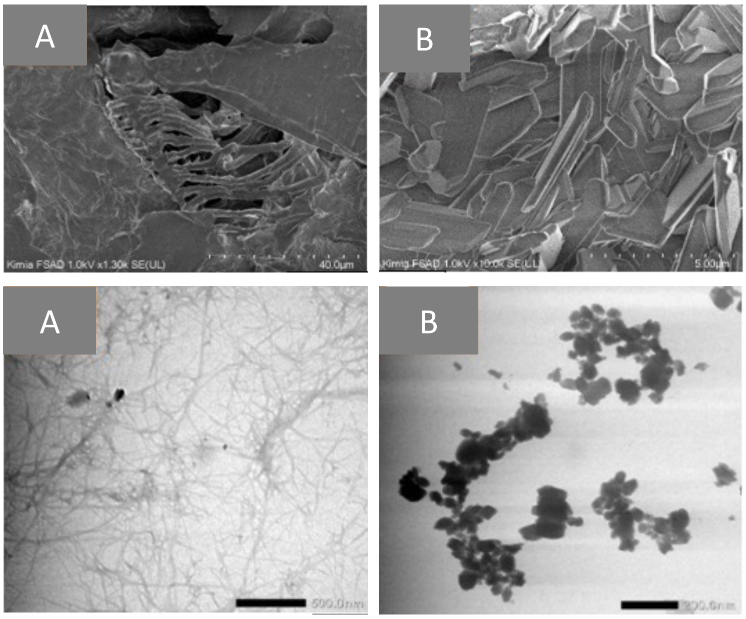

Figure 3. Images of FESEM (upper) and TEM (lower) of EDTA-free nanocellulose (A) and NCE (B)

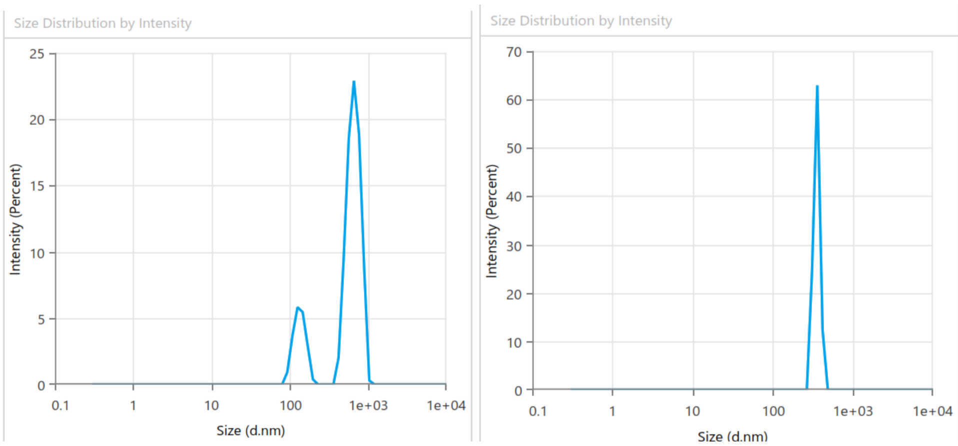

Figure 4. Particle size distribution of EDTA-free nanocellulose (left) and NCE (right)

Table 1. Cadmium (Cd) adsorption capacity of NCE

Initial concentration of Cd (mg L^{-1})	Adsorption capacity (mg g^{-1})		
	20 min	60 min	100 min
1	76	77	70
60	79.16	79.80	72.28

Particle size analysis demonstrated that EDTA-free nanocellulose exhibited an average diameter of 352 nm, while NCE showed increased dimensions of 665 nm, indicating successful composite formation through aggregation during EDTA incorporation. Both samples exhibited monodisperse size distributions within the nanoscale range (100–500 nm) (Manzoor et al., 2019; Nafisah et al., 2022), a typical feature of cellulose nanocrystals. The increase in particle size is attributed to aggregation phenomena during ultrasonication and composite formation (Brinkmann et al., 2016).

The specific surface area was determined using the Brunauer–Emmett–Teller (BET) method based on nitrogen gas adsorption isotherms at 77 K, analyzed with a surface area and porosity analyzer. The nanocellulose-EDTA (NCE) composite exhibited a surface area of 2794 m^2 g^{-1}. A higher surface area generally corresponds to greater adsorption capacity (Saef et al., 2022). The large surface area observed may be attributed to the aggregation process.

The Cd adsorption capacity of NCE was evaluated at two initial concentrations (1 mg L^{-1} and 60 mg L^{-1}) over different contact times (20, 60, and 100 min) (Table 1). The results showed that adsorption capacity increased with initial concentration and contact time, consistent with diffusion-driven mechanisms and progressive adsorbate attachment. The higher adsorption at concentration of 60 mg L^{-1} indicates that NCE has not yet reached saturation under these conditions, suggesting that equilibrium had not been fully established (Bassyouni et al., 2022).

Known for its strong chelation with transition metals, EDTA significantly enhances the adsorption performance of NCE. Using atomic absorption spectroscopy (AAS), it was observed that cadmium concentrations decreased from 1 mg L^{-1} to 0.2 and 0.3 mg L^{-1} after treatment with NCE, corresponding to a removal efficiency of approximately 73%.

The chelating ability of EDTA contributes not only to the improved adsorption capacity but also to the selectivity and stability of the composite. EDTA-based adsorbents exhibit varied physical and chemical properties, such as chemical stability, mechanical strength, and specific metal ion affinity, depending on the carrier materials and fabrication methods used (Nadhila & Titah, 2021). External factors, such as the presence of competing ions and solution composition, also influence the adsorption efficiency (Zhang et al., 2021).

Conclusion

A nanocomposite material combining cellulose and EDTA ligands (NCE) was successfully synthesized from oil palm trunks. The material was thoroughly characterized using FTIR, BET, FESEM, TEM, and PSA techniques. Cd removal analysis indicated an improved adsorption capacity of the metal ion in NCE.

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