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Innovative Application of Cellulose Nano-crystals from Agricultural Waste for Enhanced Pharmaceutical Wastewater Treatment through Artificial Intelligence-Driven Adsorption Modelling

Abubakar M. Hammari[1,2*], Usman D. Hamza[1], Maryam Ibrahim[1], Kabir Garba[1], Idris M. Muhammad[1]

[1]Department of Chemical Engineering, Abubakar Tafawa Balewa University, Bauchi, Nigeria.
[2]Department of Biochemistry, Gombe State University, Gombe, Nigeria.

*Corresponding Author:
Email: abhammari@gsu.edu.ng

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Abstract

The effective treatment of pharmaceutical wastewater poses a significant challenge due to the presence of complex and diverse organic and inorganic contaminants. Traditional treatment methods often fall short in completely removing these pollutants, leading to environmental concerns and potential health risks. In recent years, adsorption has emerged as a promising technique for the removal of pharmaceutical compounds from wastewater due to its efficiency and versatility. This review article comprehensively examines the utilization of cellulose nanocrystals (CNCs) derived from agricultural waste as novel adsorbents for pharmaceutical wastewater treatment, coupled with artificial intelligence (AI) modelling techniques. Cellulose nanocrystals, obtained through the hydrolysis of cellulose-rich agricultural byproducts, present exceptional adsorption properties owing to their high surface area, unique surface chemistry, and biocompatibility. These characteristics make them ideal candidates for capturing pharmaceutical contaminants. It highlights the various methods of CNC extraction, modification, and characterization, underscoring their effectiveness in removing diverse pharmaceutical compounds from wastewater streams. It also provides insights into the potential of combining cellulose nanocrystals (CNCs) and artificial intelligence (AI) modelling for pharmaceutical wastewater treatment. While the findings are promising, further research and real-world validation are needed to substantiate the claims of this approach's revolutionary impact. The combination of CNCs and AI modelling presents a sustainable and efficient solution to address the challenges associated with pharmaceutical wastewater pollution. Finally, it underscores the importance of interdisciplinary collaboration between materials science, environmental engineering, and AI-driven research for achieving effective and environmentally responsible pharmaceutical wastewater treatment solutions.

Keywords: Agricultural Waste, Cellulose Nano-Crystal, Adsorption, Pharmaceutical Wastewater and Artificial Intelligence.

INTRODUCTION

Pharmaceutical wastewater is a category of industrial effluent generated during the manufacturing, formulation, and utilization of pharmaceutical products. It is composed of a complex mixture of organic and inorganic pollutants, including active pharmaceutical ingredients (APIs), solvents, heavy metals, and various chemical by-products, which pose significant environmental and public health risks if not properly managed (Kundan *et al.*, 2022). Indeed, pharmaceutical manufacturing processes involve the synthesis and formulation of drugs, leading to the discharge of numerous chemical compounds into the wastewater stream. Since many of these compounds are designed to be biologically active at low concentrations, they can persist in the environment, leading to the contamination of water bodies, soil, and even drinking water sources (Jiri, 2008; Alistair *et al.*, 2012; and Patel *et al.*, 2019). Pharmaceuticals are substances used for diagnosing, treating, or preventing diseases and restoring organic functions. They encompass a wide variety of biological compounds employed in infections and disease treatment. Clean water is essential for both humans and wildlife, serving a critical role in sustaining life and ecological balance. Monitoring the quality of ground and surface water is vital, as they are major sources for domestic and industrial use. In recent times, water bodies have been affected by Emerging Contaminants (ECs), which can enter ecosystems and pose risks to human health and ecology (Omoyajowo *et al.*, 2022; Patel *et al.*, 2019).

Pharmaceutical compounds fall under the category of emerging contaminants, detected in low concentrations in the environment, and can have adverse effects on human health and ecosystems. Numerous studies exist on their discharge, occurrence, and impact in the environment, as well as analytical methods for detecting them in water (Julia *et al.*, 2018). Pharmaceuticals are classified based on their mechanism of action, chemical structures, and the diseases they treat, resulting in various therapeutic classes/groups.
The presence of emerging organic pollutants like pharmaceuticals has raised significant concerns due to potential effects on human and animal health (James *et al.*, 2018). Antibiotics, for example, have been widely used to treat bacterial infections and promote livestock growth (Fink *et al.*, 2012). Due to their increased demand, these pharmaceuticals have become environmental contaminants found in soil, water, sediment, and plants (Xu *et al.*, 2014). They enter wastewater streams through human and animal excretions, pharmaceutical manufacturing, and hospital effluents (Atallah *et al*, 2017). Typically, not targeted by wastewater treatment plants, these compounds reach surface waters and groundwater (Bhushan *et al.*, 2020). Despite their trace concentrations in aquatic environments, they can have severe effects on human health and ecosystems (Tchounwou *et al.*, 2012). This review aims to evaluate the potential of using CNCs derived from agricultural waste in combination with AI-driven adsorption modelling for pharmaceutical wastewater treatment. Additionally, the environmental risk of CNCs, including potential nanoparticle toxicity and biodegradability, was discussed to assess their overall sustainability

Importance of adsorbents in pharmaceutical wastewater treatment

The importance of adsorbents in pharmaceutical wastewater treatment is paramount due to the unique characteristics of pharmaceutical wastewater and the complex mixture of contaminants it contains. Adsorbents play a crucial role in the treatment process by effectively removing pharmaceutical compounds and other pollutants from wastewater (Julia *et al.*, 2018). Activated carbons are highly regarded among adsorbent materials due to their well-developed surface area, significant microporosity, and excellent adsorption capacity, making them highly efficient in removing organic contaminants even in low concentrations (less than 1 mg/L) (Synder *et al.*, 2007). In one study, Zhang *et al.* (2016) achieved remarkable removal efficiencies of up to 99.6% for 28 antibiotics found in superficial water by using powdered activated carbon (PAC). Additionally, Sotelo *et al.* (2012) explored fixed bed adsorption with activated carbon, reporting impressive removal efficiencies of 95% and 98% for diclofenac and caffeine, respectively.

However, the widespread application of activated carbons is hindered by their high cost as conventional and commercial materials, along with the expenses associated with their regeneration (Putra *et al.*, 2019). Consequently, recent efforts have focused on finding substitutes in the form of low-cost adsorbents that remain efficient in contaminant removal. To be considered viable, these nonconventional low-cost adsorbents must meet specific criteria, such as being cost-effective, demonstrating high adsorption capacity, and exhibiting strong selectivity for various contaminant concentrations (Crini *et al*., 2006).

Cellulose nanocrystals as potential adsorbents

Cellulose nanocrystals (CNCs) have emerged as promising and environmentally friendly adsorbents for various applications, including wastewater treatment. CNCs are nanoscale particles derived from cellulose, which is the most abundant natural biopolymer found in plants. They are typically obtained from agricultural waste, such as wood, agricultural residues, or other cellulose-rich sources, through a series of chemical and mechanical processes (Jingwen *et al*., 2022). Adsorption is the phenomenon where a solid material utilizes its inherent properties to attract and retain a gas or liquid as a thin film on its surface. The integration of adsorbents with diverse polymers, such as nanocellulose and its derivatives, assumes a crucial role in enhancing the adsorption process (Padrão *et al.*, 2016).

Integration of artificial intelligence in adsorption processes

The integration of artificial intelligence (AI) in adsorption processes offers numerous advantages and opportunities to optimize and enhance the efficiency of adsorption systems. AI, particularly machine learning algorithms, can be employed to model, predict, and control adsorption processes in real-time, leading to improved performance and cost-effectiveness (Rahmad *et al*., 2021). The AI model optimized the adsorption process by adjusting key parameters, such as the initial concentration of pollutants, CNC dosage, and contact time. The ANN model's ability to accurately predict the impact of these parameters on adsorption efficiency was found to be superior to traditional isotherm models, including the Langmuir model. This optimization process highlighted the value of AI in enhancing CNC-based adsorption for wastewater treatment, making it a more effective and sustainable solution for pharmaceutical pollutant removal.

Preliminary simulations using the AI models predicted a 15% improvement in adsorption efficiency compared to conventional methods. However, further experimental validation is required to confirm these results in real-world applications" (Rahmad *et al*., 2021; Putra *et al*., 2019). Generally, the integration of artificial intelligence in adsorption processes holds significant potential to revolutionize wastewater treatment and environmental remediation efforts. By leveraging AI's capabilities in modelling, optimization, and adaptive control, adsorption systems can be enhanced to achieve higher pollutant removal efficiency, reduced costs, and increased sustainability.

Purpose of the Review and Its Importance in Environmental Management

The purpose of this review is to evaluate the potential of cellulose nanocrystals (CNCs), derived from agricultural waste, as an innovative adsorbent for treating pharmaceutical wastewater, an emerging environmental contaminant of global concern. Pharmaceutical wastewater often contains complex organic and inorganic compounds that are not fully removed by conventional treatment methods, resulting in contaminants that can persist in the environment and pose risks to public health, aquatic ecosystems, and biodiversity. This review explores the benefits of CNCs, particularly their high adsorption capacity, renewability, and low environmental footprint, as a sustainable alternative for contaminant removal.

By combining CNCs with artificial intelligence (AI)-driven modelling, the review also examines how CNC adsorption processes can be optimized for greater efficiency, lower costs, and enhanced environmental outcomes. This interdisciplinary approach has the potential to support sustainable water management practices by addressing current gaps in wastewater treatment technologies and contributing to global environmental sustainability goals. In reviewing recent advances and identifying areas for further research, this article aims to support the development of more effective, scalable, and environmentally responsible strategies for managing pharmaceutical pollutants in wastewater.

LITERATURE REVIEW

Overview of Pharmaceutical Wastewater Treatment

Sources and composition of pharmaceutical wastewater

Pharmaceutical wastewater is generated from various sources within the pharmaceutical industry and healthcare facilities. The composition of pharmaceutical wastewater can vary depending on the specific processes involved, but it typically contains a diverse range of organic and inorganic compounds. Here are the primary sources and common compositions of pharmaceutical wastewater: Pharmaceutical Manufacturing, Research and Development Laboratories, Retail Pharmacies, and Pharmaceuticals in Domestic Wastewater.

Composition of Pharmaceutical Wastewater

The composition of pharmaceutical wastewater can be complex and varies depending on the types of pharmaceuticals produced or used in healthcare facilities. Common constituents found in pharmaceutical wastewater include: Active Pharmaceutical Ingredients (APIs), Solvents and Reagents/ Byproducts and Intermediates, Heavy Metals/ Surfactants, Antibiotics and Hormones, and Residues of Personal Care Products.

Environmental impact of pharmaceutical pollutants

The environmental impact of pharmaceutical pollutants is a growing concern due to the widespread use of pharmaceuticals and their presence in various environmental compartments. Pharmaceutical pollutants, also known as pharmaceuticals and personal care products (PPCPs), encompass a wide range of chemicals used for human and veterinary healthcare, personal care, and industrial processes (Larsen *et al*., 2004). To mitigate the environmental impact of pharmaceutical pollutants, it is essential to implement proper wastewater treatment processes, encourage responsible use and disposal of pharmaceuticals, and develop more environmentally friendly pharmaceutical formulations. Additionally, ongoing research and monitoring are crucial to better understand the long-term effects of pharmaceutical pollutants on the environment and human health.

Current treatment methods for pharmaceutical wastewater

Pharmaceutical wastewater treatment involves several methods to remove or reduce the concentration of pharmaceutical compounds and other contaminants before the treated effluent is discharged into the environment or further processed for safe reuse. The choice of treatment methods depends on the specific characteristics of the pharmaceutical wastewater and the desired effluent quality (Pal *et al*., 2010). Numerous methods for treating water, encompassing both conventional and unconventional approaches, have been extensively investigated to enhance the removal of pharmaceuticals from wastewater. Basic physicochemical methods like coagulation, flotation, lime softening, sedimentation, and filtration, are frequently employed at various points in water treatment processes, offering cost-effective solutions, as highlighted by Suarez *et al*. (2009). Nevertheless, existing treatments have shown limitations in effectively eliminating a significant portion of pharmaceuticals and personal care products (PPCPs), as noted by Crini *et al*. (2010). While certain compounds might react with common disinfectants like chlorine or ozone, the potential formation of hazardous oxidative byproducts raises considerable concerns. Westerhoff *et al*. (2005) conducted bench-scale experiments to replicate treatment processes within a drinking water treatment plant (DWTP) for the removal of PPCPs and endocrine-disrupting compounds. Coagulation and lime softening treatments exhibited modest reductions of these compounds, achieving a decrease of no more than 25%. In contrast, separate chlorination or ozonation displayed removal rates ranging from below 10% to over 90%. However, the oxidative mechanisms of these processes generated byproducts that could entail greater risks than the original compounds themselves. Notably, ozonation stands out as a particularly costly method due to its substantial energy consumption. Estimates suggest that this process, consuming approximately 0.1 kWh/m³, could elevate the energy demand of conventional treatment plants by 40−50%, as estimated by Kundan *et al*. (2022). Given the existing poor quality of wastewater entering water bodies, the enhancement of treatment facilities with novel end-of-pipe methodologies that exhibit superior removal efficiencies compared to conventional techniques is imperative. To address this, cutting-edge technologies have been under scrutiny, including membrane filtration, activated carbon adsorption, and advanced oxidation processes (AOPs), as discussed by Kundan *et al*. (2022). Membrane separation systems, like micro- and nanofiltration, ultrafiltration, reverse osmosis, and electrodialysis, have demonstrated potential in eliminating high molecular weight compounds. However, their applicability is hindered by demanding operational pressures and limited scalability, as highlighted by Tambosi *et al*. (2010). Although tight nanofiltration (NF) and reverse osmosis (RO) can effectively remove numerous pharmaceuticals, their outcomes are influenced by factors such as compound polarity, charge, hydrophobicity, and competition with cations and natural organic matter in solution, as elucidated by Kundan *et al*. (2022). Dolar *et al*. (2012) illustrated successful application of RO and NF in treating pharmaceutical industry effluent following coagulation and microfiltration, resulting in impressive removal rates of 94% to 100% for veterinary drugs. However, due to energy and material consumption, these methods have primarily been contemplated for wastewater reuse scenarios, according to Kundan *et al*. (2022).

Advanced oxidation processes (AOPs) such as H_2O_2/UV, Fe_2+/H_2O_2, Fe_2+/H_2O_2/UV, O_3/H_2O_2, and O_3/UV have demonstrated effectiveness in removing PPCPs while requiring minimal additional chemicals, as outlined by Sui *et al*. (2010). Nevertheless, AOPs generate oxidation intermediates (by-products), which could be more toxic than the original pollutants. Additionally, the operational complexity and substantial cost, especially concerning energy, are notable drawbacks. Alternatively, adsorption presents advantages including low energy consumption, mild operating conditions, and absence of introduced byproducts, positioning it as a promising method for pharmaceutical removal, as underscored by Kundan *et al*. (2022).

Here are some of the current treatment methods commonly used for pharmaceutical wastewater:

Cellulose Nanocrystals from Agricultural Waste

Extraction methods for cellulose nanocrystals

Cellulose nanocrystals (CNCs) are nanoscale particles derived from cellulose, the most abundant biopolymer on Earth found in plant cell walls. CNCs have unique properties, such as high strength, biodegradability, and biocompatibility, making them attractive for various applications, including in the fields of nanotechnology, materials science, and biomedical engineering (Kilole *et al.*, 2022).

There are several extraction methods for obtaining cellulose nanocrystals from cellulose sources. Here are some commonly used techniques:

Acid Hydrolysis: Acid hydrolysis is the most widely used method for producing cellulose nanocrystals in which strong acid easily neutralize the amorphous regions of whole cellulose fibers to produce CNC with reduced size (Li *et al et al*., 2022). In this process, cellulose fibers are treated with strong mineral acids, such as sulfuric acid or hydrochloric acid. The acid breaks down the amorphous regions of cellulose, leaving behind crystalline regions in the form of nanocrystals. The process involves heating the cellulose-acid mixture and then performing washing and purification steps to remove residual acid and by-products. CNC has a similar morphology to the original cellulose and has a higher degree of crystallinity. Negatively charged nanocelluloses with abundant functional groups can also be produced by acid hydrolysis; for example, the nanocellulose with sulfate groups can be stably dispersed after sulphuric acid hydrolysis aqueous solutions due to surface charge repulsion (Marakana *et al*., 2021). CNCs were extracted from lignocellulosic natural fibers using sulfuric acid hydrolysis, yielding 76% CNCs with a high crystallinity of around 54-88% (Noremylia *et al.*, 2021). The sulfuric acid concentration used was between 63-64% at a temperature range of 45-60°C for 30-120 minutes, which efficiently removed the amorphous regions while preserving the crystalline parts (Noremylia *et al*., 2021). After extraction, sonication was applied to break down the aggregated CNC particles (Noremylia *et al.*, 2021). It's essential to choose the appropriate extraction method based on the desired properties and applications of the cellulose nanocrystals, as different methods can result in variations in the size, shape, and surface chemistry of the CNCs produced. Additionally, the choice of cellulose source (e.g., wood, agricultural waste, etc.) can also influence the extraction process and the properties of the resulting CNCs.

Characterization techniques of cellulose nanocrystals

Characterization techniques are crucial for understanding the properties and behavior of cellulose nanocrystals (CNCs). These techniques help researchers analyze the physical, chemical, and structural characteristics of CNCs, enabling them to tailor the material for specific applications (Yunfeng *et al.*, 2020). Here are some common characterization techniques used for cellulose nanocrystals: Transmission Electron Microscopy, Scanning Electron Microscopy, X-ray Diffraction, Fourier Transform Infrared Spectroscopy and Thermogravimetric Analysis.

These characterization techniques, used individually or in combination, provide valuable insights into the properties and behavior of cellulose nanocrystals, aiding researchers in optimizing their applications in various fields.

Advantages of using agricultural waste-derived cellulose nanocrystals

Using agricultural waste-derived cellulose nanocrystals (CNCs) offers several advantages, making them a promising and sustainable material for various applications. Here are some key advantages: Abundant and Renewable Resource, Cost-Effectiveness, Environmental Benefits, Biocompatibility Light Weight and High Strength, Non-Toxic and Safe and Potential for Circular Economy (Aziz *et al*., 2022)

MATERIALS AND METHODS

Literature Search Strategy

To conduct a comprehensive review of cellulose nanocrystals (CNCs) and their applications in pharmaceutical wastewater treatment, a systematic literature search was performed using academic databases, including Google Scholar, PubMed, and ScienceDirect. Keywords used in the search included "cellulose nanocrystals," "CNCs," "pharmaceutical wastewater," "adsorption modelling," "artificial intelligence in adsorption," and "wastewater treatment." The search focused on studies published within the last 10 years to capture recent advancements in CNC research and AI-driven optimization of adsorption processes.

Selection Criteria

Articles were selected based on specific inclusion and exclusion criteria to ensure relevance and quality. Inclusion criteria consisted of:

- Peer-reviewed studies published mostly between 2013 and 2022.
- Research articles focusing on CNCs as adsorbents, specifically in wastewater treatment applications.
- Studies exploring artificial intelligence (AI) techniques, such as machine learning or predictive modelling, to enhance adsorption efficiency.

Exclusion criteria were applied to omit:

- Studies not available in English.
- Articles focused on non-pharmaceutical wastewater applications unless they contributed essential insights on CNCs or AI in adsorption.
- Research not directly related to CNCs, AI integration, or adsorption mechanisms.

Screening and Review Process

After the initial search, studies were screened for relevance by reviewing titles and abstracts. Full-text articles of potentially relevant studies were then reviewed in detail. Each study was analyzed and categorized into key themes, including the effectiveness of CNCs in pharmaceutical wastewater adsorption, comparative performance with other adsorbents, and AI-driven modelling applications for optimizing CNC adsorption. Articles were also reviewed for insights into challenges, such as regeneration potential, cost, and scalability, providing a foundation for identifying gaps in current research and areas needing further exploration. This structured approach enabled a critical synthesis of recent developments in CNC-based wastewater treatment and AI-enhanced adsorption technologies.

RESULTS AND DISCUSSION

Adsorption Mechanism and Models

Fundamental principles of adsorption

Adsorption is a process by which molecules or ions from a fluid (gas or liquid phase) adhere to the surface of a solid material. It is a fundamental phenomenon with widespread applications in various fields, including catalysis, separation processes, water purification, gas storage, and many more (Ruthven, 1984).

Understanding the fundamental principles of adsorption is essential for optimizing adsorption processes, designing efficient adsorbents, and developing innovative applications in various industries. It allows scientists and engineers to tailor adsorption systems to specific needs, leading to improved efficiency and performance (Babel *et al*., 2023).

Adsorption isotherms

Adsorption isotherms play a crucial role in the design and evaluation of any adsorption process. These isotherms delineate the connection between the amount of adsorbate (measured in milligrams) removed from a liquid phase and the mass of the adsorbent (measured in grams) used, all at a constant temperature. The Langmuir, Freundlich, and Temkin isotherm models are among the most commonly employed models for characterizing dye adsorption. To determine the suitability of these isotherm equations, their applicability is often assessed by examining the correlation coefficient, denoted as R^2, as discussed by Atef and Waleed (2017).

CNCs offer several advantages over traditional adsorbents, such as high surface area and renewability. However, their adsorption capacity may be lower in certain cases compared to conventional adsorbents like activated carbon. Further optimization is needed to improve CNCs' efficiency in specific applications" (Putra *et al.*, 2019; Bimová *et al.*, 2021).

Langmuir isotherm

The Langmuir isotherm model revolves around the concept of a monolayer with a maximum adsorption capacity for the adsorbent. This implies that each active binding site can host only one molecule, assuming a consistent adsorption energy across all such sites. Furthermore, the model postulates that the adsorbed molecules remain stationary on the surface and do not undergo movement or interaction with neighbouring molecules, as explained by Inyang *et al.* (2016). This model specifically characterizes monolayer adsorption and assumes a uniform energy of adsorption, envisioning a single homogeneous layer of solute adsorbed at a steady temperature, a concept outlined by Enenebeaku *et al.* (2015). The Langmuir isotherm is applicable when monolayer adsorption occurs on a surface comprising a finite number of identical sites. The model presumes a uniform distribution of adsorption across the surface, with no lateral translocation within the plane of the surface, as noted by Atef and Waleed (2017). The Langmuir equation, often expressed in a linear form, captures these principles.

$$\frac{C_e}{q_e} = \frac{1}{q_m k_l} + \frac{C_e}{q_m} \qquad (1)$$

The Langmuir isotherm model quantifies the adsorption process by considering the equilibrium amount of dye adsorbed (qe) in relation to the maximum adsorption capacity (qm) when saturation is reached. Ce denotes the equilibrium dye concentration, and kl represents the Langmuir constant, which is associated with the strength of dye binding to the adsorbent surface.

Freundlich isotherm

Moving to the Freundlich isotherm, this model is an empirical equation that proves highly effective in describing the partitioning of solutes between solid and aqueous phases at saturation. It operates under the assumption of an exponential variation in site energies on the adsorbent and a logarithmic decline in heat of adsorption, as outlined by Hammari *et al.* (2020).

The linearized form of the Freundlich equation can be expressed as:

$$Log q_e = Log K_f + \frac{1}{n} Log C_e \qquad (2)$$

In this equation, Kf represents the Freundlich constant, a measure of adsorption capacity, while n signifies the intensity of adsorption. The value of n holds particular significance:

n = 1 corresponds to linear adsorption
n < 1 implies a chemical process
n > 1 indicates a physical process (Kumar *et al.*, 2013).

Adsorption Kinetics

The prediction of adsorption kinetics is imperative for the effective design of adsorption systems. Kinetic parameters, which offer insight into adsorption rates, are crucial for process modelling and design. The successful utilization of adsorption methods necessitates the development of cost-effective, readily available, and abundant adsorbents characterized by well-known kinetic parameters and sorption traits, as emphasized by Atef and Waleed (2017). The analysis of adsorption kinetics provides a forecast of how dye adsorption advances over time until equilibrium is achieved. Furthermore, understanding the adsorption mechanism holds significance for design considerations. To unravel the adsorption mechanism of anionic dyes, pseudo-first-order, pseudo-second-order, and intra-particle diffusion models have been employed, as explored by Intidhar *et al.* (2017).

Pseudo-First-Order kinetic model

Beginning with the pseudo-first-order kinetic model, Lagergren's kinetics equation, which was among the first to describe liquid-solid system adsorption based on solid capacity, has been termed pseudo-first order due to its reliance on adsorption capacity rather than solution concentration. The linear representation of this model is:

$$\mathrm{Ln}(q_e - q_t) = lnq_e - k_1 t \qquad (3)$$

Here, q_e and q_t signify the amount of pollutant adsorbed per unit mass on the adsorbent at equilibrium and various time intervals t, respectively. The pseudo-first-order adsorption rate constant is denoted as k_1 (min^-1). By plotting ln(q_e - q_t) against time t, k_1 and calculated q_e can be obtained from the slope and intercept, respectively (Hammari *et al.*, 2020).

Pseudo-Second-Order kinetic model

The pseudo-second-order kinetic model is expressed by: (Enenebeaku *et al.*, 2015)

$$\frac{t}{q_t} = \frac{1}{k_2 {q_e}^2} + \frac{1}{q_e} \qquad (4)$$

Here, k2 represents the pseudo-second-order adsorption rate constant (g/mg·min), and qe is the amount of dye adsorbed (mg/g) at equilibrium. The initial adsorption rate, h (mg/g·min), is given by:

$$h = k^2 q_e \qquad (5)$$

A linear relationship between t/qt and t enables the computation of k^2, h, and calculated qe. The choice of applicable models is typically based on the correlation coefficient (R^2) and the degree of agreement between experimental and calculated qe values, as detailed by Hammari *et al.* (2020).

Thermodynamic Parameters

Investigating the thermodynamics is crucial to determine whether the adsorption process is favorable. The thermodynamic parameters—free energy (ΔG°), enthalpy (ΔH°), and entropy (ΔS°) play a crucial role in assessing heat changes during the adsorption process. These parameters are calculated using the following equations: (Ali *et al.*, 2017)

$$k_e = \frac{qe}{C_e} \qquad (6)$$

$$lnk_e = \frac{\Delta S°}{R} - \frac{\Delta H°}{RT} \qquad (7)$$

$$\Delta G° = \Delta H° - T\,\Delta S° \qquad (8)$$

Here, 'Ke' stands for the equilibrium constant, 'qe' represents the amount of pollutant adsorbed in mg on the adsorbent per dm^3 of the solution at equilibrium, 'Ce' is the equilibrium concentration of pollutant in the solution (mg/L). 'R' is the universal gas constant (8.314 J/mol K), and 'T' is the temperature in Kelvin. 'ΔH°' and 'ΔS°' parameters are calculated from the slope and intercept of the plot of ln(ke) versus 1/t."

These are some of the commonly used adsorption models. Researchers choose the appropriate model based on the type of adsorption (e.g., physisorption or chemisorption) and the specific characteristics of the adsorption system being studied. Fitting experimental data to these models helps in understanding the adsorption behavior and obtaining important parameters like adsorption capacity, surface area, and affinity of the adsorbate-adsorbent system (Richardson *et al.*, 2005).

Applications of Cellulose Nanocrystals in Wastewater Treatment

Adsorption of pharmaceutical pollutants using cellulose nanocrystals

The adsorption of pharmaceutical pollutants using cellulose nanocrystals (CNCs) has gained attention as a promising and sustainable approach for water treatment and environmental remediation. Cellulose nanocrystals, derived from renewable sources like agricultural waste or wood, possess unique properties that make them effective adsorbents for various pollutants, including pharmaceutical compounds. (Reshmy *et al.*, 2021)

The adsorption process for pharmaceutical pollutants using CNCs involves bringing the contaminated water in contact with the CNCs, allowing the pollutants to adsorb onto the surface of the nanocrystals. The process can be influenced by factors such as pH, temperature, contact time, and the initial concentration of pollutants in the water (Arunachalam and Rajarathinam, 2020).

Generally, the use of cellulose nanocrystals for adsorbing pharmaceutical pollutants presents a promising solution for the removal of these contaminants from water sources. The combination of their unique properties, renewability, and low cost makes CNCs a potential game-changer in the field of water treatment and environmental protection. However, it's essential to conduct further research and optimization to explore the full potential of CNCs as adsorbents for pharmaceutical pollutants and to ensure their safe and effective implementation in practical applications.

Comparative analysis with other adsorbents

When comparing cellulose nanocrystals (CNCs) with other adsorbents, several factors should be considered to determine their suitability for specific applications. Here's a comparative analysis between CNCs and other common adsorbents (Asim *et al.*, 2020):

i. Specific Surface Area: CNCs typically have a high specific surface area, which provides numerous active sites for adsorption. This property is particularly advantageous when compared to larger particles or granular adsorbents with lower surface areas.
ii. Pore Size and Distribution: The nanoporous nature of CNCs allows for efficient adsorption of small molecules and ions. Some other adsorbents, such as activated carbon, may have a wider range of pore sizes, making them suitable for adsorbing a broader spectrum of pollutants.
iii. Surface Chemistry: CNCs can be functionalized or modified to enhance their affinity for specific pollutants through surface chemistry alterations. This flexibility provides an advantage over some other adsorbents with fixed surface properties.
iv. Regeneration and Reusability: CNCs often exhibit good regeneration potential and can be reused for multiple adsorption cycles. In comparison, some other adsorbents may lose their effectiveness or structural integrity after repeated usage (Sadare *et al.*, 2022).

It is essential to assess the specific requirements of the adsorption process, the target pollutants, and the operational conditions when choosing the most suitable adsorbent. While CNCs have several advantageous properties, other adsorbents may excel in different aspects based on the specific application needs. Consequently, a comprehensive evaluation of performance, cost, sustainability, and ease of implementation is necessary to make an informed decision regarding the choice of adsorbent for a particular adsorption process.

Artificial Intelligence in Adsorption Processes

Overview of artificial intelligence applications in environmental engineering

Artificial intelligence (AI) has found numerous applications in environmental engineering, offering innovative solutions to address complex environmental challenges. AI techniques leverage data-driven models, machine learning algorithms, and intelligent decision-making systems to analyze, predict, and optimize various environmental processes. Artificial intelligence (AI) modelling techniques have gained popularity in various scientific and engineering fields, including adsorption processes. AI models offer the advantage of being data-driven and capable of handling complex relationships between adsorption variables, making them valuable tools for understanding and predicting adsorption behaviour (Mingyi *et al.*, 2018). AI's integration into the adsorption process enables real-time optimization and predictive modelling, improving efficiency by 20% in preliminary tests. The AI models allow for the adjustment of process parameters, such as adsorbent dosage and contact time, to achieve optimal pollutant removal" (Soltani *et al.*, 2021; Jianxun *et al.*, 2021)

Machine learning algorithms for adsorption modelling

The utilization of machine learning algorithms holds significant promise in the realm of adsorption modelling. This is attributed to their adeptness in deciphering intricate data patterns, providing precise forecasts, and fine-tuning adsorption procedures. Numerous machine learning algorithms are prevalent for the purpose of adsorption modelling, each possessing distinct advantages and compatibility contingent on the adsorption scenario and the availability of data. Machine Learning algorithms are widely used in adsorption modelling to analyze large datasets and extract patterns and trends. ML models can be trained on experimental adsorption data to predict adsorption capacities, isotherms, and kinetics for new adsorbates and adsorbents. Some popular ML algorithms for adsorption modelling include support vector machines (SVM), decision trees, random forests, and neural networks (Gulza *et al.*, 2021).

The choice of machine learning algorithm depends on the specific adsorption modelling task, the size and complexity of the dataset, and the level of interpretability required. Researchers often perform comparative analyses of different algorithms to identify the most suitable model for a particular adsorption system. The AI techniques used in this study include neural networks and decision trees to optimize adsorption parameters such as adsorbent dosage, contact time, and pH (Rahmad *et al.*, 2021; Soltani *et al.*, 2021). These models were trained on datasets from CNC-based adsorption experiments and optimized using cross-validation methods to predict optimal conditions for maximum pollutant removal. The dataset for training the AI models was sourced from previous experimental studies on CNC-based adsorption and was validated using cross-validation techniques to ensure the reliability of the predictions (Rahmad *et al.*, 2021; Soltani *et al.*, 2021). These techniques ensured that the model could generalize across different scenarios, improving the accuracy of the adsorption optimization.

AI modelling in adsorption processes can significantly improve our understanding of adsorption behavior, aid in the design of better adsorbents, optimize operational conditions, and facilitate the development of efficient and sustainable adsorption systems for various applications, including water treatment, gas separation, and pollutant removal. The AI model used was based on an artificial neural network (ANN), designed with two hidden layers. Each layer applied different activation functions, including linear, TanH, and Gaussian, to optimize the removal of ions from water (Syah *et al.*, 2021). This approach allowed the model to capture the non-linear relationships between the adsorption process and influencing factors, which significantly improved predictive accuracy over conventional adsorption models.

Advantages of AI-assisted adsorption processes

i. AI-Optimized Adsorbent Design: AI can assist in designing novel adsorbents with specific surface properties, functional groups, and morphologies tailored for efficient adsorption of target pollutants. Machine learning models can predict the adsorption performance of different materials, guiding researchers in selecting the most promising candidates for synthesis and testing.
ii. AI-Driven Adsorption System Control: AI algorithms can optimize the operation of adsorption systems in real-time. Using data from sensors and process variables, AI models can dynamically adjust parameters such as flow rates, temperature, and regeneration cycles to maximize adsorption efficiency and minimize energy consumption.
iii. AI-Based Pollutant Removal from Industrial Effluents: AI can be used to develop predictive models for adsorption processes in industrial wastewater treatment. Machine learning algorithms can analyze historical data from various industrial effluents and predict the performance of different adsorbents, enabling more effective and targeted pollutant removal.
iv. AI-Enhanced Contaminant Detection: AI can aid in detecting and monitoring pollutants in water sources. By analyzing real-time data from sensor networks or remote sensing platforms, AI models can identify emerging contaminants and assess their concentration levels, enabling timely response and appropriate treatment.
v. AI-Driven Adsorption Isotherm and Kinetics Modelling: AI can assist in developing accurate adsorption isotherm and kinetics models based on experimental data. Machine learning algorithms can fit the data to various adsorption models, helping researchers understand the adsorption behavior of specific pollutants on different adsorbents.
vi. AI-Based Predictive Maintenance: AI can be applied to predict the lifespan and performance degradation of adsorbents. By analyzing operational data and historical trends, AI models can anticipate when adsorbents need regeneration or replacement, optimizing maintenance schedules and preventing adsorption system failures.
vii. AI-Enabled Multi-Objective Optimization: AI algorithms can perform multi-objective optimization in adsorption processes, considering factors such as adsorption capacity, selectivity, cost, and environmental impact. This aids in finding the most suitable adsorbent and process conditions for a given application.
viii. AI-Assisted Adsorbent Regeneration: AI can optimize the regeneration process of exhausted adsorbents. By analyzing regeneration data and the efficiency of different regeneration

methods, AI models can determine the most effective and sustainable regeneration protocols (Soma *et al.*, 2022).

It is important to note that AI-assisted adsorption processes are continuously evolving; researchers and engineers are exploring the potential of AI to optimize and revolutionize adsorption technologies for diverse environmental and industrial applications. Although the AI models show promising results in simulations, additional experimental validation is necessary to confirm the models' performance in real-world scenarios. Initial tests indicate potential, but large-scale applications must be explored" (Gulza *et al.*, 2021; Jianxun *et al.*, 2021).

Environmental and Economic Assessment

Environmental impact of using cellulose nanocrystals in wastewater treatment

The use of cellulose nanocrystals (CNCs) in wastewater treatment is gaining attention as a potentially eco-friendly and sustainable solution. However, like any technology or material, CNCs also have potential environmental impacts that need to be carefully considered. CNCs derived from agricultural waste present significant economic benefits due to their lower production costs compared to synthetic nanofillers, such as carbon nanotubes, which are significantly more expensive (Noremylia *et al.*, 2021). Furthermore, CNCs offer high sustainability due to their biodegradability, renewability, and the use of abundant natural materials. Despite their benefits, potential environmental risks arise from the release of CNC nanoparticles into ecosystems. As with other nanomaterials, there is concern regarding their long-term ecological impact, particularly nanoparticle accumulation and the challenges associated with their degradation (Noremylia *et al.*, 2021). Therefore, proper environmental risk assessments and lifecycle analyses are essential to fully understand and mitigate the environmental implications of large-scale CNC production and application. A full lifecycle assessment of CNCs is required to evaluate their sustainability and environmental impact. Although they offer renewable, biodegradable solutions for wastewater treatment, their long-term effects on ecosystems, particularly regarding nanoparticle release, must be studied further" (He *et al.*, 2020; Aziz *et al.*, 2021). Although CNCs are biodegradable, their release into ecosystems could result in nanoparticle accumulation, potentially leading to environmental pollution. Studies indicate that CNCs may persist under certain conditions, raising concerns about long-term ecological impacts (He *et al.*, 2020; Reshmy *et al.*, 2021).

Economic feasibility of integrating AI in pharmaceutical wastewater treatment

The economic feasibility of integrating AI in pharmaceutical wastewater treatment depends on various factors, including the specific AI applications, the scale of the treatment system, and the overall context of the wastewater treatment process. Here are some considerations that can influence the economic feasibility:

i. Upfront Investment: Implementing AI-based technologies often requires significant upfront investment in hardware, software, and expertise. The cost of acquiring and installing AI systems and the required computational infrastructure should be considered.
ii. Operational Costs: While AI can optimize processes and reduce resource consumption, there will still be ongoing operational costs, such as energy consumption, maintenance, and personnel training. The overall impact of AI on operational costs should be evaluated.
iii. Savings from Efficiency: AI can enhance the efficiency of pharmaceutical wastewater treatment by optimizing processes, reducing chemical usage, and improving resource utilization. These efficiency gains can lead to cost savings over the long term.
iv. Improved Treatment Performance: Effective use of AI in controlling and monitoring treatment processes can lead to better treatment performance, reducing the risk of non-compliance with regulatory standards and potential fines.
v. Data Acquisition and Management: AI relies on large datasets for training and continuous improvement. The cost of data acquisition, storage, and management should be considered when assessing the economic feasibility.
vi. Maintenance and Support: AI systems require regular maintenance and updates to ensure they operate optimally. Factoring in the cost of on-going maintenance and technical support is essential.

vii. Scaling and Adaptability: Consideration should be given to the scalability of AI applications in wastewater treatment. Will the AI technology be adaptable to different treatment plants and varying wastewater compositions?
viii. Regulatory Environment: Compliance with environmental regulations and discharge standards is critical for any wastewater treatment facility. AI can help in ensuring adherence to these standards, but the costs associated with maintaining compliance should be evaluated.
ix. Risk Mitigation: AI can help identify potential issues and anomalies in real-time, enabling proactive responses to potential threats. This can help reduce the risk of operational disruptions and improve overall plant reliability.
x. Research and Development: Depending on the level of innovation and novelty, some AI applications may require additional research and development efforts. Consideration of these costs is essential.

Generally, while integrating AI in pharmaceutical wastewater treatment may involve some initial investment and ongoing operational costs, the potential benefits in terms of improved efficiency, performance, and risk management can outweigh the expenses. Conducting a thorough cost-benefit analysis and considering the long-term advantages of AI implementation will be crucial in determining the economic feasibility for each specific case. Additionally, as AI technologies advance and become more mainstream, costs may decrease, further enhancing the economic viability of their integration into wastewater treatment processes.

Integration of cellulose nanocrystals and AI in pharmaceutical wastewater treatment

The integration of cellulose nanocrystals (CNCs) and artificial intelligence (AI) in pharmaceutical wastewater treatment holds great promise for addressing the challenges associated with the removal of pharmaceutical pollutants from wastewater. By combining CNCs' unique adsorption properties with AI-driven optimization and decision-making capabilities, more efficient and sustainable treatment solutions can be developed. Here's how CNCs and AI can be integrated in pharmaceutical wastewater treatment:

i. Enhanced Adsorbent Design: AI can assist in designing CNC-based adsorbents tailored to the removal of specific pharmaceutical pollutants. Machine learning models can predict the adsorption behaviour of CNCs with different functionalization, surface chemistries, and morphologies, leading to the development of adsorbents with high selectivity and adsorption capacity for targeted pharmaceutical compounds.
ii. Real-Time Monitoring and Control: AI-driven sensor networks can continuously monitor the concentration of pharmaceutical pollutants in wastewater. By integrating this data with AI algorithms, the treatment system can be dynamically adjusted to optimize the dosage of CNCs, flow rates, and other operational parameters in real-time, ensuring efficient pollutant removal.
iii. Predictive Modelling for Adsorption Kinetics: AI can be used to model the adsorption kinetics of pharmaceutical pollutants onto CNCs. Machine learning algorithms can analyze experimental data and predict the adsorption rates under varying conditions, helping to optimize contact times and achieve rapid and effective pollutant removal.
iv. Adaptive Regeneration Strategies: AI can optimize the regeneration process of CNC-based adsorbents. By analyzing data on adsorption capacity, exhaustion levels, and regeneration methods, AI models can determine the most effective regeneration protocols, minimizing energy and chemical usage and extending the adsorbents' lifespan.
v. Multi-Objective Optimization: AI algorithms can perform multi-objective optimization in pharmaceutical wastewater treatment. By considering factors such as adsorption efficiency, regeneration costs, and environmental impact, AI can identify the most sustainable and economically viable treatment strategies.
vi. Early Warning Systems: AI can help predict potential breakthroughs of pharmaceutical pollutants in the treatment process. By analyzing historical data and pollutant behavior, AI models can provide early warnings and trigger countermeasures to prevent the release of pollutants into the environment.

vii. Decision Support Systems: AI can provide decision support for treatment operators and engineers. AI-driven analytics can process vast amounts of data, identify trends, and recommend optimal operational strategies, improving overall treatment performance and resource utilization.

viii. Scaling-Up Adsorption Processes: AI can assist in scaling up lab-scale adsorption experiments to industrial wastewater treatment. Machine learning models can extrapolate adsorption behavior, predict system performance, and recommend optimal CNC-based adsorbent configurations for large-scale applications.

By integrating CNCs and AI in pharmaceutical wastewater treatment, it is possible to develop more efficient, cost-effective, and environmentally friendly solutions for removing pharmaceutical pollutants from wastewater. This interdisciplinary approach has the potential to revolutionize pharmaceutical wastewater treatment and contribute to a more sustainable and cleaner environment.

CONCLUSION

Cellulose nanocrystals (CNCs) derived from agricultural waste offer a sustainable and eco-friendly alternative for pharmaceutical wastewater treatment. These materials demonstrate effective adsorption capabilities for pharmaceutical compounds, making them promising candidates for addressing wastewater pollution. Additionally, artificial intelligence (AI) modeling, particularly machine learning algorithms, can optimize the adsorption process by predicting optimal conditions and dosages, enhancing the efficiency of CNCs. Research into the adsorption kinetics and mechanisms further illuminates the effectiveness and capacity of CNCs as adsorbents.

Furthermore, investigating methods for regenerating and reusing CNCs not only increases their cost-effectiveness but also minimizes waste generation, contributing to a more sustainable treatment process. The environmental benefits of using CNCs are significant, as they help reduce pharmaceutical contaminants in wastewater, thus mitigating their potential impact on ecosystems. The integration of AI, agricultural waste valorization, and CNCs holds great promise for improving pharmaceutical wastewater treatment, advancing sustainability, and fostering resource efficiency in the wastewater treatment industry. However, while the results are promising, further empirical data and large-scale studies are necessary to validate these findings, with future research focusing on real-world applications to fully realize the potential of this approach.

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