Review Article

Exploring the Potential of Cationic Modified Microfibrillated/Nanocellulose as Slow/Controlled Release Fertilizers: A Review

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DOI: 10.14416/j.asep.2026.02.003
Received: 26 July 2025; Revised: 26 September 2025; Accepted: 12 December 2025; Published online: 5 February 2026

Abstract

Conventional controlled-release fertilizers often rely on synthetic polymer coatings that are costly, non-biodegradable, and environmentally harmful. Biodegradable alternatives such as microfibrillated cellulose and nanocellulose have attracted significant attention for use as sustainable fertilizer coatings. Their high surface area and mechanical strength enable improved nutrient retention, moisture resistance, and reduced leaching. Cationic modification further enhances coating adhesion and nutrient binding, offering superior control of nutrient release compared with unmodified cellulose. This review evaluates recent progress in the development of cationic cellulose-based coatings, including cost considerations, biodegradation behavior under soil conditions, physicochemical interactions that regulate nutrient release, and potential industrial applications. Challenges and future directions are also discussed, highlighting the role of cationic cellulose in advancing environmentally friendly fertilizer technologies.

Keywords: Cationic microfibrillated/nanocellulose modification, Nanotechnology in fertilizer efficiency, Sustainable slow/controlled release fertilizer

1 Introduction

The world population is estimated to increase to 2.3 billion by 2050. As the population grows, food demand also increases. Enhancing food production is closely related to the role of fertilizer as a provider of nutrients for plants because fertilizer is one of the critical plant nutrients commonly used by farmers to increase yields. Statistical data provided by the Food and Agriculture Organization (FAO) of the United Nations in collaboration with the International Fertilizer Association has demonstrated a continuous increase in global agricultural fertilizer consumption every year, where the average total percentage increase in the three essential fertilizers (Nitrogen (N), Phosphorus (P), and Potassium (K)) during 2001 - 2020 was around 45.21% (Figure 1), where China was the country with the highest fertilizer consumption, namely around 22% (Figure 2). The highest consumption was N, which reached 112,364 Mt and was followed by P with a total consumption of around 48,975 Mt, and K with a total consumption of about 40,495 Mt [1].

Recent literature emphasizes the critical role of fertilizers in sustainable agriculture and global food security. Fertilizers directly contribute to increased crop yields and productivity while maintaining soil fertility in the long term. Beyond serving as essential nutrient sources for plants, fertilizers also enhance nutrient absorption efficiency, enabling agriculture to meet the growing demands of a rapidly increasing global population [2], [3]. These contributions highlight fertilizers as indispensable inputs for achieving both high productivity and sustainable farming practices.

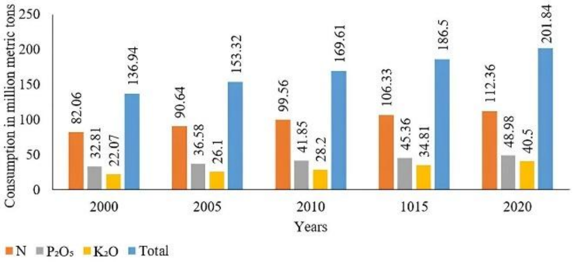


Figure 1: Fertilizer consumption in the world.

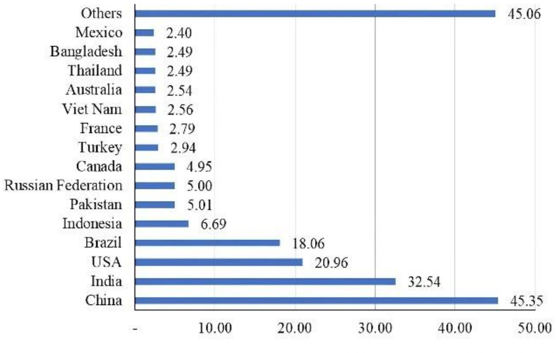


Figure 2: Consumption of fertilizers worldwide in 2020 by country.

However, achieving these agricultural benefits while minimizing environmental impacts requires fundamental shifts in fertilizer technology. Comprehensive analyses have demonstrated that nanobiofertilizers offer improved crop nutrition while minimizing environmental losses through slow release properties and enhanced nutrient absorption at the nanoscale [4]. Similarly, the adoption of biofertilizers as eco-friendly alternatives has gained momentum, particularly for addressing soil degradation and reducing greenhouse gas emissions associated with chemical fertilizer production. These developments underscore the growing recognition that sustainable agricultural productivity requires fundamental shifts in fertilizer technology toward biodegradable, controlled-release systems [5]. In line with this transition toward sustainable nutrient management, Torres and Somera reviewed the utilization of organic fertilizers as nutrient sources in hydroponic systems, emphasizing the importance of efficient nutrient release and reduced environmental impact [6]. Their findings highlight that nutrient delivery mechanisms based on organic and bio-based materials can offer controlled nutrient availability comparable to synthetic fertilizers, supporting the broad shift toward environmentally friendly fertilizer technologies such as S/CRF.

The need for fertilizers containing high N elements is higher than those containing P and K elements because N is the most critical requirement for plant growth. After all, N can help plant growth in terms of vigor, color, and yield [7]. N is a component of the chlorophyll molecule, which allows plants to capture sunlight energy through photosynthesis, promoting plant growth and yield [8]. In other words, N plays an essential role in plants to ensure energy availability and to optimize the yield. The most common fertilizer containing the element N is urea fertilizer, which is commercially used in global agriculture. According to the Statista Research Department, from 2009 to 2021, urea fertilizer production continued to increase yearly (Figure 3) [9].

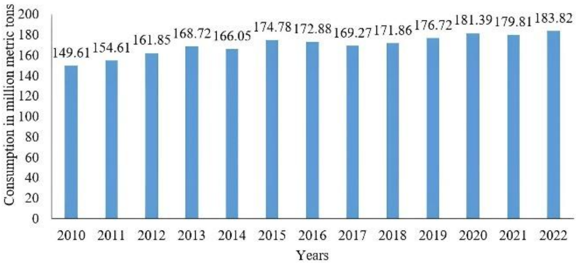


Figure 3: Urea production in the world.

Nitrogen can be found in urea fertilizer at 46% [10]. However, only 20–30% of plant-available nitrogen is used by plants to grow. The remaining nitrogen is released into the environment and can damage the environment and groundwater [11]. On the other hand, periodic utilization of fertilizer doses also promotes concentration fluctuations between ineffectiveness and toxicity levels, which can affect the growth of certain agricultural species [12]. Hydrological loss of N can be caused by the use of excess fertilizer applied to plantations of several types of vegetable crops, where the loss of N by the mechanism of ammonia volatilization (NH_3) and nitrous oxide (N_2O) emissions can occur due to the denitrification reaction. This condition impacts environmental degradation and even global warming from N_2O pollution [13]. In addition, several environmental impacts may occur from non-optimal N application besides global warming [14], [15], which is the occurrence of water pollution and soil acidification [16], [15]. The same thing was stated by the United Nations Environment Programme - UNEP (2022), where the destructive impact of fertilizer is mainly caused by excessive and inefficient use [17]. This condition leads to a decrease in vital nutrients from the ecosystem, along with other harmful effects, including the degradation of safe water supplies and the over-enrichment of freshwater environments and coastal regions. Additionally, certain fertilizers pose risks to human health as a result of improper storage methodologies. The environmental and health impacts tied to the application of pesticides and fertilizers, as well as strategies for their mitigation, warrant thorough examination. This phenomenon has emerged as a significant environmental issue across multiple European nations, the United States, and Asia, including Indonesia [18]. The use of nitrogenous fertilizers in Mexican agriculture affects water quality in the Gulf of Mexico. Nitrogen that is not absorbed by plants results in water pollution. Groundwater pollution increases the risk of colon cancer, undesirable odors and tastes, and increased eutrophication. This problem results in hidden costs estimated at $16.10 per kilogram of nitrogen [19].

Groundwater in Montana, United States, is estimated to contain leached nitrogen worth 1 billion dollars, which causes water pollution. This pollution also has an impact on soil acidification [20]. The risk of eutrophication in Benoa Bay, Bali, Indonesia, is caused by nitrogen content that has exceeded the limit of Ministry Regulation No. 51 in 2004 for marine biota. Benoa Bay, fed by five rivers, has a nitrogen content that fluctuates throughout the year due to differences in water availability in the dry and rainy seasons. The highest nitrogen content was recorded at 16,950 mg/L, while the applicable limit was 0.008 mg/l. The high nitrogen levels are influenced by fertilizer use in agricultural areas upstream of the river [21]. The average loss of nitrogen from food agriculture to the surrounding environment in 2008 was estimated at 141 kg N/ha from using 160 kg N/ha of nitrogenous fertilizer [22].

Therefore, it has been observed that strategies designed to lower fertilizer application could gain valuable perspectives by evaluating the differences in the broad environmental outcomes of fertilizer use across multiple crop systems [23]. Besides that, Jwaideh *et al.*, also reported that it is necessary to make appropriate decisions regarding using N and P fertilizers for certain plant commodities to minimize the impact on water and achieve environmentally friendly plant commodity trade [24].

Therefore, the performance of urea can be enhanced through the application of a slow/controlled release fertilizer (S/CRF) and the alignment of nutrient release with plant absorption. The transformation of CRF across developmental timelines reveals significant insights into how early experiences shape neurodevelopmental trajectories and stress responses. The interplay between environmental factors, caregiver relationships, and biological mechanisms underscores the complexity of development influenced by CRF at multiple levels [25]–[33]. S/CRF can provide nutrients for plants by slowing down their availability for plant absorption from the soil or by decreasing their heightened solubility in both soil and surface water [34]–[36]. However, S/CRF performance can also depend on soil, plant species, and climate [37].

Vejan *et al.*, indicate that slow-release controlled-release fertilizers (S/CRF) serve a critical role within the sustainable agricultural industry, wherein S/CRF has emerged as a transformative solution that fosters environmentally sustainable methodologies aimed at enhancing agricultural productivity by facilitating the optimal development stages of plants via both direct and indirect mechanisms [38]. The S/CRF mechanism mitigates nutrient loss attributed to evaporation and leaching while offering a meticulously designed nutrient release system that aligns with the physiological and biochemical requirements of plant growth. However, increasing the efficiency of slow-release urea requires more complex technology because plant roots absorb nitrogen from the substrate primarily in the form of nitrate or ammonium ions. Unlike nitrate-N_2, ammonium-N provided in the soil or applied externally can persist in soil constituents where plants and soil compete for it. Nitrogen competition when added to soil as mineral fertilizer is a significant concern [39]. This discussion is critical to understanding the challenges in optimizing the performance of S/CRF in practical applications. Addressing these challenges, such as nutrient competition and efficient nitrogen delivery, forms the basis for exploring innovative solutions. One such approach is the use of cationic modified MFC/NC, which has the potential to enhance nutrient retention and release properties, thereby improving the efficiency of S/CRF and contributing to sustainable agricultural practices.

S/CRF is a fertilizer that is considered to have added value because it has low carbon characteristics. The global market for S/CRF is projected to experience a compound annual growth rate of 6.5% from 2014 to 2019. Geographically, the S/CRF market is categorized into several principal areas, namely Europe, North America, Asia-Pacific, Africa, the Middle East, and Latin America. The American S/CRF market is expected to dominate regional revenues due to advanced agricultural technologies. It tends to adopt new cultivation technologies to produce cereals and grains. As the foremost producer of fruits and vegetables, Europe is anticipated to significantly contribute to the robust development of the S/CRF market. The Asia-Pacific region is forecasted to represent the most rapidly expanding market, particularly due to the scarcity of cultivable land in nations within the Asia-Pacific, especially in developing countries such as India and China [40].

The Global Market for S/CRF is segmented based on technology, type, application mode, and end-use. According to Data Bridge (2023), the segmentation of the S/CRF market is based on technological attributes, categorizing it into polymer coatings, sulfur-based coatings, and urea reaction products. The S/CRF market is classified by type into urea formaldehyde, urea isobutyraldehyde, and urea acetaldehyde; by application methodology, where the slow-release fertilizer sector is divided into fertigation, foliar, soil, and other application methods; and based on end use, namely non-agricultural and agricultural. The global slow/controlled release fertilizer (S/CRF) market is projected to experience significant growth, with Data Bridge Market Research forecasting a compound annual growth rate (CAGR) of 6.60% from 2022 to 2029 [41], while Fortune Business Insights reported that the market is expected to reach USD 5.89 billion by 2032, reflecting a CAGR of 6.79% [42].

This information is predicated on the increasing awareness regarding the availability of highly efficient fertilizers, a growing concern for environmental conservation, and heightened levels of crop cultivation, particularly in developing nations, which will emerge as major factors driving market growth. Additionally, escalating apprehensions regarding food safety, an uptick in product innovations, the accelerating pace of industrialization, and augmented financial investment in research and development efforts related to new product introductions constitute other salient factors propelling the market growth [41].

However, the application of S/CRF within extensive commercial agricultural practices remains a source of concern, primarily due to the fact that the coatings are predominantly composed of petroleum-derived synthetic polymers [38], such as polyolefins, polyvinylidene chloride, acrylic resins, and polysulfone. S/CRF products derived from fossil fuel sources often entail intricate manufacturing processes and, unsurprisingly, are composed of toxic chemicals. These S/CRF products originate from non-renewable resources and are generally non-biodegradable, which may exacerbate various environmental and energy-related issues, including the accumulation of non-biodegradable waste in subterranean soils and the progressive depletion of fossil fuel reserves.

To address these limitations, a comprehensive comparison between conventional synthetic coatings and emerging bio-based alternatives is essential for understanding the potential advantages of sustainable coating materials. Table 1 presents a comparison of key performance characteristics and environmental impacts of synthetic versus bio-based coating systems for S/CRF applications.

In addition to nutrient release efficiency, the environmental sustainability of coating materials is strongly influenced by their biodegradability under soil conditions. While synthetic polymer coatings often persist in soil for extended periods with minimal degradation, bio-based coatings exhibit more predictable and shorter degradation timelines. Recent studies have compared the biodegradation of several bio-based coatings evaluated at 30, 60, and 90 days.

Table 1: Comparative analysis of synthetic versus bio-based coating materials for S/CRF-

Aspects	Synthetic Coatings (polymer/ sulfur, etc.)	Bio-based Coatings (chitosan, alginate, lignin, starch, cellulose, etc.)	Ref.
Example materials	Thermoplastic/ resin polymers for S/CRF (e.g., polyolefins, polyurethane), sometimes sulfur/minerals	Polysaccharides (chitosan, alginate), lignin/cellulose derivatives, gelatin/proteins, certain bio-polyesters	[43], [44]
Biodegradability	Generally low	Generally high or moderate	[45]
Environmental risks	Risk of microplastics entering soil/water when coating fragments; may alter metal bioavailability and soil properties	Lower microplastic risk, but rapid degradation may affect nutrient/oxygen dynamics if formulation is not optimized	[46], [47]
Impact on soil microbes	Microplastics can alter microbial communities and soil properties	Some bio-coatings support enzyme activity and beneficial microbial communities	[48], [49]

Shellac-based coatings show relatively rapid degradation, with thinner films releasing nutrients within 11 days, whereas thicker coatings (60–92 μm) extend nutrient release up to 60 days in soil [50]. In contrast, polyvinyl alcohol/starch films reinforced with biochar (U-PVA/ST/BOF) degrade more slowly, with only 18.3% nitrogen released after 28 days, suggesting sustained release beyond 60 days [51]. Pinecone biochar and cellulose acetate composites (PBUA) exhibit moderate degradation, reaching 55.6% release after 30 days and 72.6% after 60 days [52]. Other bio-based hybrids such as lignin–clay nanohybrids (PLC-SRF) and nano-biochar polymer composites (NBC-SRF) demonstrate controlled release behavior, with nitrogen release extended beyond 60 days and moderate biodegradation observed in soil environments [53], [54].

These comparative findings highlight that, unlike synthetic coatings, which may remain in soil for over a year, bio-based coatings can be tailored to achieve predictable degradation within 30–90 days, thereby offering significant environmental benefits. Building on this understanding, cationic nanocellulose emerges as a promising bio-based material that not only provides controlled nutrient release but also retains the intrinsic biodegradability of cellulose, thus aligning with sustainable agricultural practices.

The application of biopolymer-based hydrogels in agricultural systems has demonstrated promising potential beyond traditional S/CRF applications. Recent studies have explored pectin-based hydrogels developed from agricultural residues for environmental remediation [55] and polyvinyl alcohol/chitosan composite systems designed for controlled delivery applications [56], indicating the versatility of biopolymer matrices for encapsulation and controlled release mechanisms. These developments provide valuable insights for designing nanocellulose-based S/CRF systems with enhanced functional properties.

As an alternative, numerous polymers derived from renewable and biodegradable sources are viable candidates for S/CRF coating materials in the form of bio-nanocomposites. Consequently, a distinct disparity exists between the operational mechanisms of S/CRF in fostering plant growth and the significant role of nanocomposites, particularly bio-nanocomposites, as coating materials in the synthesis of S/CRF, underscoring the critical importance of nanotechnology applications in augmenting the efficacy of S/CRF. Therefore, this review focuses on the development, application mechanisms, and future potential of cationic MFC/NC modified as a fertilizer, particularly in the context of S/CRF for enhancing crop sustainability. While extensive research exists on nanocellulose applications [57], [58] and conventional S/CRF technologies [38], this is the first comprehensive review specifically addressing cationically modified MFC/NC as sustainable coating materials for S/CRF systems. Unlike previous works that examine components separately [28], [29], this review systematically integrates cationic modification mechanisms, biodegradable coating performance, and waste biomass utilization [59], [60] for agricultural applications.

2 Slow/Controlled Release fertilizer

The Association of American Plant Food Control Officials (AAPFCO) describes S/CRF as a type of fertilizer that includes plant nutrients in a form that can either delay their availability for absorption by plants after application or extend their availability significantly longer than conventional fertilizers, which release nutrients rapidly [40]. However, the release rate highly depends on soil and climatic conditions [27]. There are three criteria for the slow release of nutrients:

no more than 15% (m/m) should be released within 24 hours, no more than 75% (m/m) within 28 days, and at least 75% (m/m) should be released at the designated release time [25]. Using S/CRF as a substitute for conventional fertilizer provides many benefits for plants and soil. S/CRF helps maintain and enhance environmental safety by lowering toxicity from high ion concentrations in the soil, providing a consistent supply of nutrients throughout the growing season, minimizing nutrient loss from the soil, and reducing eutrophication in water bodies [27]. Apart from the environmental aspect, S/CRF reduces planting costs and labor costs because using S/CRF tends to lead to savings in fertilizer quantity, where fertilizer application rates are reduced by 20–30% compared to conventional fertilizers [25].

To provide a clearer understanding of the position of slow/controlled release fertilizers (S/CRFs) within the broader fertilizer landscape, a comparison with other fertilizer types is necessary. Conventional, organic, synthetic polymer-coated, and biopolymer-based fertilizers differ not only in nutrient release characteristics but also in environmental impact, soil suitability, and crop compatibility. Table 2 summarizes the advantages, disadvantages, and recommended applications of these fertilizer categories across different agricultural contexts.

On the other hand, S/CRF also has a negative aspect: production costs are higher than those of conventional fertilizers. According to the International Fertilizer Industry Association (IFA) report, the price of S/CRF is 4–12 times higher than that of conventional fertilizer. However, with large-scale production, these costs can decrease [27]. Table 3 compares the use of S/CRF with conventional fertilizer.

S/CRF fertilizers include, for example, urea–formaldehyde (UF) and isobutylidenediurea (IBDU), which supply nitrogen. UF is among the most widely used S/CRFs globally. It is created through the synthesis of urea and formaldehyde in a molar ratio of 1.2–1.5:1, resulting in a UF condensate that serves as S/CRF with the general formula $(HOCH_2NHC(O)NH)_nCH_2$. Urea is used as a nitrogen source and converted through mineralisation into NH_4^+, easily available to plants. Despite the slow nutrient release, UF fertilizer increases soil aggregation, increasing permeability and nutrient penetration efficiency. IBDU, on the other hand, is a single compound with the formula $(CH_3)_2CHCH(NHC(O)NH_2)_2$, formed by combining urea and isobutyraldehyde. The rate of nutrient release from IBDU increases as the particle size of the fertilizer decreases and soil temperature rises [27].

Table 2: Comparison of fertilizer types: Advantages, disadvantages, and suitability across soils and crops.

Types of fertilizers	Advantages	Disadvantages	Soil Suitability	Crop Suitability
Conventional Fertilizers (urea, NPK, liquid fertilizers)	Provide nutrients rapidly and are readily available to plants, accelerating growth, increasing crop productivity, and improving quality traits such as color, texture, and size. They are also time–efficient compared to organic fertilizers, which require longer decomposition [43].	Highly susceptible to nutrient losses through leaching; lysimeter studies report the highest nitrogen loads in sandy soils with intensive mineral fertilizer use [44]. Approximately 50% of applied nitrogen may be lost as nitrate to the environment [45].	More effective in fine–textured soils (loam, clay) where leaching losses are lower; less effective in sandy soils where nutrient leaching is severe [46].	Effective for short–cycle crops (e.g., rice, vegetables) because nutrients are immediately available and can be adjusted through split applications, improving yields and fertilizer use efficiency [61].
Organic Fertilizers (compost, manure, biofertilizers)	Environmentally friendly, derived from natural sources, and enhance long–term soil fertility by increasing organic matter and improving soil physical, chemical, and biological properties [43].	Relatively low nutrient content, slow and uncontrolled nutrient release depending on microbial activity, and require higher application volumes than inorganic fertilizers, reducing efficiency when used as the sole nutrient source [47], [48].	Suitable for most soil types; particularly recommended for soils with low SOM, sandy soils, or degraded soils [62].	Effective in perennial and horticultural crops, where they improve yield and quality while enhancing soil health [63].

Table 2: (Continued)

Types of fertilizers	Advantages	Disadvantages	Soil Suitability	Crop Suitability
Synthetic polymer–coated fertilizers (polyolefins, resins, etc.)	Provide controlled nutrient release, improve nutrient use efficiency, and reduce leaching losses compared to conventional fertilizers [49], [64], [65].	Poor degradability and risk of leaving microplastic residues in soil, potentially polluting the environment and degrading soil ecosystem quality [49], [66].	Particularly effective in sandy or coarse-textured soils with high leaching risk; shown to reduce nitrate leaching while maintaining crop performance [64].	Well-suited for high-value crops and intensive horticulture (e.g., tomato, strawberry), enabling single pre-plant applications with gradual nutrient release, improving NUE and yield while reducing pollution risks [67].
Biopolymer–based S/CRF	Offer controlled nutrient release, biodegradability, environmental sustainability, and higher nutrient use efficiency compared with conventional fertilizers [68], [69], [70].	High production cost and process complexity; potential risk of incomplete degradation leaving residues [68], [71].	Maintain good performance even in sandy or highly drained soils [71].	Suitable for perennial crops (e.g., rubber, oil palm) due to stable N release profiles, reducing application frequency and increasing efficiency; for annual crops (e.g., rice, corn), improve yield, NUE, and suppress leaching in highly drained soils [72].

Table 3: Comparison of aspects in using conventional fertilizer and S/CRF.

Aspects	Conventional Fertilizer	S/CRF	Ref.
Price	Lower initial cost	Higher initial cost, long-term savings	[73],[74], [75]
Technology	Rapid nutrient release	Controlled nutrient release using advanced materials	[74],[76], [45]
Usage situation	Frequent applications needed	Reduced application frequency, suitable for harsh environments	[29],[77]
Labor	Higher labor costs	Lower labor costs due to fewer applications	[78],[77]
Nutrient supply	Immediate but inefficient	Sustained and efficient nutrient supply	[79],[77]
Environmental effects	High pollution risk	Reduced pollution	[76],[73], [80]

Regarding particle size, applying nanotechnology to S/CRF can promote sustainable agriculture. The creation of nanoparticles or composites can support plant growth by either directly providing nutrients or releasing them gradually. Plants require 16 nutrients in total, with 13 sourced from the soil. Nano-fertilizers have effectively offered these vital nutrients sustainably, thanks to their slow-release properties. This gradual release increases nutrient delivery to the plant, accelerating germination, rapid growth, and high nutrient levels [81].

3 Cationic of Nanocellulose

Cationic nanomaterials are nanoscale structures that measure less than 100 nm and carry a positive charge. For years, these materials have been researched for various applications in the healthcare industry, including clinical diagnosis, gene delivery, drug delivery, and tissue engineering [82]. Recently, a new concept has emerged to utilize cationic nanocellulose as a material that enhances positive charge, as explored in previous studies [83].

Cationic modification of cellulose is the process of attaching positively charged groups (such as quaternary ammonium) onto cellulose. This turns normally neutral/negatively charged cellulose into a positively charged material, which improves its ability to bind anionic substances (like dyes or nutrients) through electrostatic attraction [64]. Nanocellulose is known to have a high degree of polymerization and crystallinity [84] and strong intra and intermolecular hydrogen bonds, making it difficult to increase the surface activity of nanocellulose [83]. Bertsch and Fischer examine the adsorption and interfacial properties of nanocellulose at fluid interfaces, highlighting how its native hydrophilic and hydrophobized surfaces result in distinct interfacial structures and adsorption characteristics [85].

Kumar *et al*., also suggested that nanocellulose in all its forms, including forms like cellulose nanocrystals, cellulose nanofibers, and bacterial cellulose, is attracting significant interest in both research and industry due to its appealing characteristics such as outstanding mechanical properties, a high concentration of hydroxyl groups for modification, large surface area, low weight, and biodegradability [57], [58]. However, its hydrophilic nature restricts its use in many applications [86], [57]. Therefore, the functional groups on its surface offer opportunities for surface modification using various techniques [57].

It is known that nanocellulose is a material with high hydrophilic properties because it contains many hydroxyl groups, so it is very soluble in strong polar solvents [87], [88]. This characteristic makes nanocellulose difficult to apply to hydrophobic media (such as low polar and nonpolar organic solvents) and most polymer matrices [89]. Therefore, several studies have focused on modifying the hydrophilic properties of nanocellulose so that it can be utilized more widely. One way is through physical adsorption modification. The principle is to bind nanocellulose and modified substances with Van der Waals forces and hydrogen bonds so that the shear resistance of the product is weak, easily dissociated by external forces, and unstable [90]. One method of physical adsorption modification is cationic modification, where nanocellulose is reacted with quaternary ammonium groups so that the electronegative surface changes to positive [91]. Thus, cationic nanocellulose is a quaternary ammonium compound that interacts well with negatively charged materials, resulting in good emulsion stability [92]. Figure 4 illustrates the surface modification of cellulose nanocrystals (CNCs) into cationic CNCs via a covalent surface functionalization method adopted from [93]. Table 4 shows several studies of cationic modifications on nanocellulose.

Physical modification can involve the addition of surfactants because surfactants have a hydrophilic side, and the other side is hydrophobic. A bond will be formed between the hydroxyl groups of nanocellulose and the hydrophilic functional groups on one side of the surfactant, and the other side of the surfactant (hydrophobic functional groups) will be exposed to the outside of the nanocellulose and ready to bond with the hydrophobic groups of other media [94]. Numerous previous studies have demonstrated that the properties of nanocellulose can be altered. Nanocellulose is formed into airgel using freeze drying and coated with TiO_2. When placed in a dark room, the properties of nanocellulose will turn hydrophobic. Mariano *et al*., modified cellulose nanofibers (CNFs) by coating them with three cationic surfactants (C_{12}TAB, C_{14}TAB, and C_{16}TAB). Based on the results of this research, it was found that the modified CNF tends to flocculate due to hydrophobic interactions. Another reason for flocculation is decreased electrical repulsion forces between CNFs due to neutralization by cationic surfactants [95].

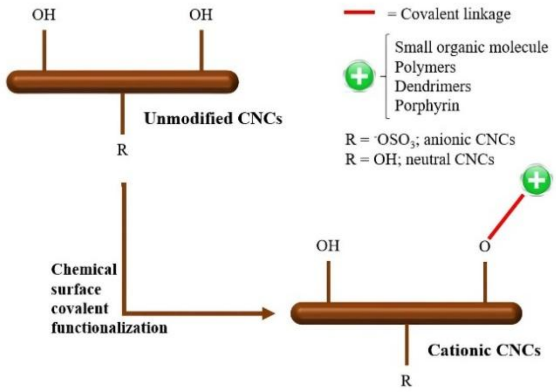


Figure 4: Illustration of surface modification of CNC to cationic CNC (Adapted from [93]).

Table 4: Summary of cationic modification of nanocellulose studies.

Types of Nanocellulose	Cationic Reagent	Process Conditions	Results	Source
CNC	CTMAB	CNC: CTMAB mass ratio (1:1 (w/w)), Reaction temperature: 60 °C, Reaction time: 3 h, Centrifugation speed: 4000 rpm, Duration: 3 min	Cationic modification of CNC can maintain CNC crystallinity	[96]
CNF	EPTMAC	Fiber pulp concentration ratio: NaOH (2.5:5) wt%, Pulp ratio: EPTMAC (1:1.25) w/v, Reaction temperature: 65 °C, Reaction time: 8 h	Cationic modified CNF can increase the biodegradability of composites	[66]
CNC	CTAB	Solution heating: up to 60 °C, Centrifugation: 4000 rpm, 10 min	CTAB surface modification decreases the surface charge of carboxylated CNC and forms hydrophobic regions on the surface of the cellulose nanoparticles.	[97]
CNC	GTMAC	GTMAC: CNC ratio (3:1), NaOH Concentration: 5%	Cationic modification of CNCs can improve their dispersion state and thermal stability. Cationic surface modification gives liquid suspensions of CNC a tendency to gel and gel-like structures that are stronger than unmodified CNC suspensions.	[98]

Beyond the use of surfactants, several other strategies have been developed to modify the behavior of nanocellulose for fertilizer coating applications. According to Castaño *et al*., bacterial nanocellulose (BNC) was modified with CTAB to reduce its hydrophilicity and enhance hydrophobicity, thereby improving the adsorption of nonpolar compounds such as genistein. The contact angle increased from 33.92° (BNC) to 60.66° (BNC-CTAB), confirming significant hydrophobization, while TGA/DTG and FTIR analyses further demonstrated structural modifications associated with CTAB interaction [99]. Similarly, Gong *et al*., reported CTAB-modified cellulose nanocrystals (CNCs) with enhanced hydrophobicity and compatibility with nonpolar media. The modification introduced new FTIR peaks attributable to CTAB, while the zeta potential shifted to more positive values, indicating altered surface charge due to surfactant adsorption [100]. Beyond surfactant-based approaches, Subbotina *et al*., introduced a chemical route in which cellulose nanofibrils (CNFs) were oxidized with $NaIO_4$ to form dialdehyde–CNF (DA-CNF), followed by oxime ligation with O-substituted hydroxylamines. This water–based method avoids costly organic solvents, and the oxidant can be regenerated (e.g., via ozone or electrochemical processes), thereby reducing cost and environmental impact [101].

Cationic nanocellulose serves as an effective biodegradable polymer matrix for S/CRF, playing a key role in improving nutrient delivery efficiency. Thanks to its inherent properties, this material can encapsulate nutrients and release them in a controlled manner that aligns with plant uptake needs. By incorporating cellulose biopolymers into fertilizer formulations, this approach helps tackle the challenges of nutrient loss and environmental pollution typically associated with conventional fertilizers. Recent studies highlight the significance of reversing the surface charge of CNFs or CNCs in developing functional cationic CNFs or CNCs. This is particularly relevant for innovative applications, especially in the biomedical field, such as gene and drug delivery, vaccine adjuvants, and tissue engineering [93]. Therefore, various covalent synthetic methods have recently been discovered to design and manufacture cationic elements in CNFs and CNCs. Table 5 shows the surface covalent functionalization methods for preparing cationic CNFs or CNCs.

Table 5: Surface covalent functionalization method for the preparation of cationic CNFs or CNCs.

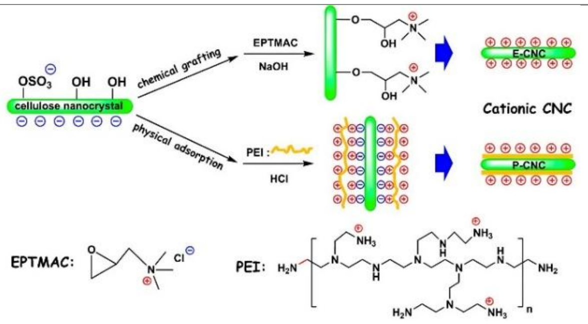
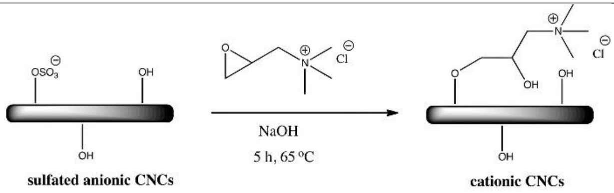
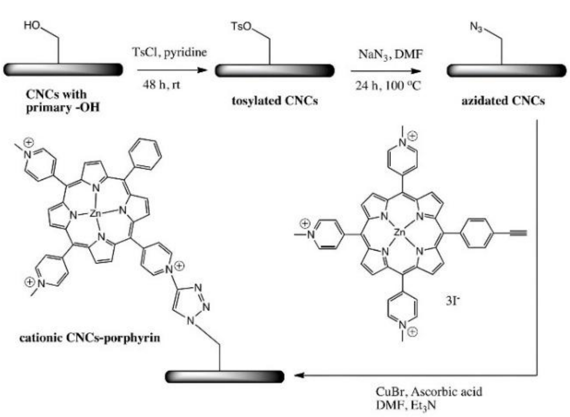
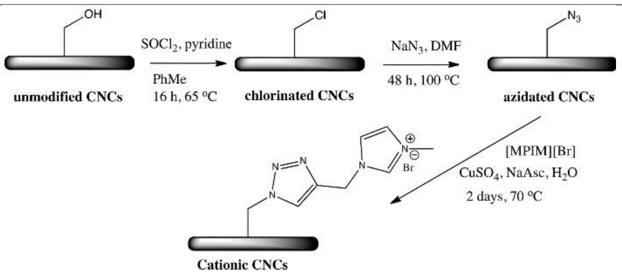
Anticipated Applications	Purpose	Cationization Method	Scheme	Infor-mation
1. Cationization of CNC Surface via Etherification Reaction.				
To develop a biocompatible hydrogel for drug delivery	Developing a hydrogel with a double membrane structure using cationic CNC and anionic alginate	Synthetic route via chemical grafting and physical adsorption		Reproduced from [102] with permission from the American Chemical Society, copyright 2016
To create cationic CNCs for use as a functional component in coatings	Rendering CNCs cationic through epoxypropyltrimethylammonium chloride reaction	Surface cationization of CNCs via a nucleophilic ring-opening reaction		Adapted from [103] with permission from the Royal Society of Chemistry, copyright 2008
2. Cationization of CNC Surface via Photobactericidal Activity.				
To enhance antimicrobial properties for biomedical applications	Synthesizing and characterizing cellulose nanocrystals modified with a cationic porphyrin	Preparation of cationic porphyrin-based CNCs via Click chemistry		Adapted from [104] with permission from the American Chemical Society, copyright 2011
3. Cationization of CNC Surface via Imidazolium and Pyridinium Grafted CNCs.				
To develop versatile ion exchange systems or catalysts	Grafting imidazolium salts onto CNCs	Synthesis of imidazolium grafted CNCs via Click chemistry		Adapted from [105] with permission from the Royal Society of Chemistry, copyright 2011

Table 5: (Continued)

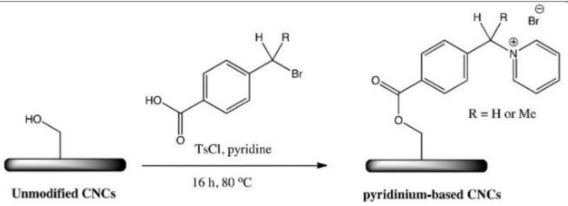
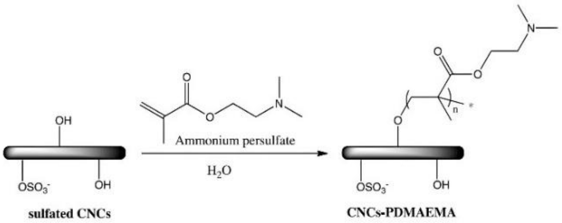
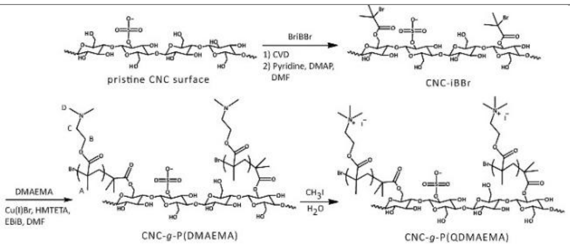
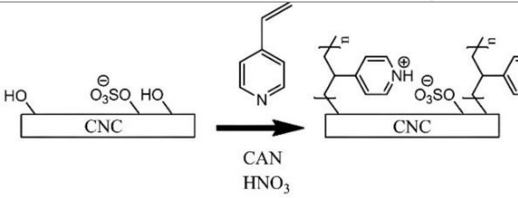
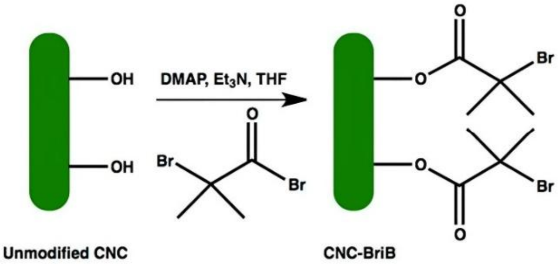
Anticipated Applications	Purpose	Cationization Method	Scheme	Infor-mation
To produce stable positively charged CNCs	Using a one-pot reaction system to graft pyridinium moieties onto CNCs	One-pot synthetic route for the design of pyridinium-grafted CNCs		Adapted from [106] with permission from the Royal Society of Chemistry, copyright 2013
4. Cationic CNC via Polymerization Technique.				
To prepare CNCs with weak polyelectrolytes for advanced materials	Grafting PDMAEMA onto CNCs	Preparation of CNCs grafted PDMAEMA via free radical polymerization		Adapted from [107] with permission from the American Chemical Society, copyright 2014
To modify CNCs for use as colloidal macroinitiators	Modifying pristine CNCs with initiator molecules for subsequent polymer grafting	Preparation of cationic polymer brush-modified CNCs		Adapted from [108] with permission from the Royal Society of Chemistry, copyright 2014
To develop pH-responsive CNCs	Surface-initiated polymerization of 4-vinylpyridine using ceric(IV) ammonium nitrate	One-pot aqueous ceric ion surface-initiated graft polymerization		Adapted from [109] with permission from the American Chemical Society, copyright 2013
To develop biomedical applications such as drug delivery and bioimaging using cationic CNC-based nanoplatforms	To synthesize cationic CNCs using SI-SET-LRP, assess their chemical, morphological, and cytotoxic properties, and explore their potential in gene delivery and nanomedicine	SI-SET-LRP using cationic monomers (AEM and AEMA), involving esterification and subsequent polymerization		Reproduced from [110] with permission from the American Chemical Society, copyright 2015

Table 5: (Continued)

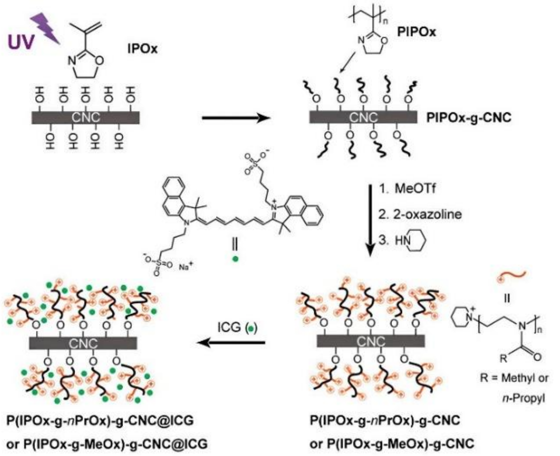
Anticipated Applications	Purpose	Cationization Method	Scheme	Information
To create biocompatible poly(2-oxazoline)s for photothermal cancer therapy	Synthesizing poly(2-oxazoline)-modified CNCs via UV-induced polymerization	Preparation of poly(2-oxazoline)-modified CNCs		Reproduced from [111] with permission from the Royal Society of Chemistry, copyright 2017

Table 6: Application of various nanocellulose in slow and controlled release fertilizers using various methods.

Nano Cellulose Type	S/CRF Forming Materials	Fertilizer Type	S/CRF Manufacturing Method	Drying Method	Drying Temperature and Time	Discharge Ratio	Ref.
CNF, CNF^+	Starch, PHB, TPS, glycerol, water	KNO_3	Spray Drying and Matrix (in pellet form)	-	-	**Release in Water**: • NO_3^- is released more quickly from fertilizers containing CNF^- than from fertilizers containing CNF^+. • Fertilizer with PHB and starch released NO_3^- as much as 150–200 mg/L and fertilizer with PHB and TPS released <50 mg/L for 240 minutes. **Discharge in soil:** Fertilizers that use starch and CNF^- release NO_3^- faster than K^+ and vice versa for fertilizers that contain CNF^+.	[66]
CNF	Aminated-cellulose nanofibers (A-CNF), Acrylamide, 2-aminoethyl methacrylate hydrochloride, TEMED, V50	N (ammonium nitrate)	In situ radical copolymerization	Freeze Dryer	-50 °C for 5 days	A-CNFs 3 wt% /cationic poly (AM-co-AEM) as much as 4%-based hydrogel is a fertilizer with a late release at neutral pH compared to other combinations. Fertilizer is released continuously for 90 days in irrigated rice soil.	[113]
CNF	Sodium alginate, $FeCl_3 \cdot 6H_2O$, 1,3,5-benzenetricarboxylic acid (H_3BTC)	Urea	Matrix Hydrogel	Freeze Drying	12 h	Fertilizer was released as much as 50% on the 20th day of testing in the soil	[114]
CNC	PVA, CNC, NPK	NPK	Coating	Fluidized Bed Machine	25 min	Release fertilizer in water for 5 hours • PVA 100% Nitrogen: 99% Phosphorus: 99% Potassium: 86% • PVA and CNC 6% Nitrogen: 81% Phosphorus: 85% Potassium: 65% • PVA and CNC 10%	[115]

4 Microfibrillated/Nanocellulose–Based Slow/Controlled Release Fertilizer

S/CRF refers to fertilizers modified by physical, chemical, or physicochemical methods to slow or control the release of nutrients, and have been proven to meet plant nutritional needs better, increase nutrient utilization efficiency, and reduce environmental risks, so its application is a crucial way to achieve green agricultural development [29]. According to Pang *et al.*, nanomaterials have characteristics of small size, large surface area, and significant interfacial effects so that they can be used for S/CRF [29]. Therefore, S/CRF based on nanomaterials has garnered significant interest from researchers in the agricultural field. Previous reports have highlighted the use of environmentally friendly carboxymethyl cellulose hydrogels, incorporated with nanocellulose for agricultural purposes, particularly as soil conditioners and nutrient carriers, and their effects on cucumber growth [112].

Additionally, Pang *et al.*, also revealed that developing natural organic nanomaterials requires low costs, exploring simple and efficient modification methods, and clarifying the performance regulation mechanisms of nanomaterials are still necessary in the future [29]. While the term “low costs” may raise eyebrows among readers familiar with the current market for CNF. Indeed, CNF has traditionally been viewed as an expensive material due to its complex production processes and specialized applications. However, advancements in manufacturing techniques and increased scalability are gradually making CNF more accessible and affordable. The essential functions of nanomaterials are hydrophobicity, adsorption, water retention, environmental responsiveness, and self-repair in S/CRF. Abundant functional groups, physical cross-linking points and micro/nano protrusions of nanomaterials are vital factors in enhancing the slow-release effect. Table 6 shows the application of various nanocelluloses in S/CRF by various methods.

Some coating methods have been reported for the development of cellulose-based slow/controlled release fertilizers. The fluidized-bed coating method was applied, for example, to a combination of PVA and CNC coated onto NPK granules using a Wurster machine for approximately 25 min. This technique has been shown to produce a more controlled release of NPK. PVA-CNC coatings via fluidized-bed were able to extend the release of N, P, and K for up to several weeks in the soil. Furthermore, the spray drying method was used on positively and negatively charged CNF formulations (CNF−/CNF+) carrying KNO_3. The spray drying results were then formed into pellets or matrices, and the NO_3^- release profiles were shown to differ depending on the type of CNF charge [62].

Another study showed that spray-dried fertilizer microparticles encapsulated with CNF and incorporated into a biodegradable matrix (PHB/starch or PHB/TPS) were able to slow the release of nutrients in the soil in a more sustainable manner. In addition, the matrix/extrusion-type method was also reported on CNF-alginate and aminated-CNF/copolymer hydrogels produced through crosslinking and freeze-drying techniques, so that they were able to withstand the release of nutrients in the soil. nutrients for up to 90 days in paddy fields [2]. Another study also confirmed that biodegradable fertilizer-polymer composites can also be fabricated through melt extrusion, a common technology on an industrial scale [3].

The effectiveness of nanocellulose-based slow and controlled release fertilizers relies on three primary physicochemical interactions that govern nutrient retention and controlled release behavior: hydrogen bonding, electrostatic attraction, and physical encapsulation.

First, hydrogen bonding networks. Nanocellulose matrices exhibit extensive hydrogen bonding interactions through abundant hydroxyl groups (–OH) on cellulose chains, forming both intra- and intermolecular bonding networks that enhance structural stability. These hydrogen bonds facilitate the entrapment of water and nutrient molecules within the matrix, thereby improving water-holding capacity and supporting gradual nutrient diffusion. The hydrogen bonding network also enables additional interactions with polymer chains and biomolecules, further reinforcing the hydrogel structure and modulating permeability and swelling behavior [65].

Second, electrostatic interactions. Cationic modification of MFC/NC with quaternary ammonium groups creates positively charged surfaces that interact electrostatically with negatively charged nutrient ions such as nitrate (NO_3^-), phosphate (PO_4^{3-}), and sulfate (SO_4^{2-}). This charge-based attraction mechanism significantly enhances nutrient retention within the coating matrix and prevents rapid loss through leaching, enabling more controlled and sustained nutrient release compared to unmodified cellulose systems [66].

Third, physical encapsulation mechanisms. The three-dimensional porous network structure of nanocellulose-based hydrogels provides physical entrapment of nutrients within interconnected pore spaces. The high surface area of cellulose nanofibrils enhances adsorption capacity while creating tortuous diffusion pathways that nutrients must navigate before release. This physical barrier effect is modulated by swelling-deswelling dynamics, pore accessibility, and gradual biodegradation of the matrix. Such encapsulation mechanisms have been demonstrated in nanocellulose-reinforced starch-chitosan hydrogels for NPK delivery [68] and nanocellulose-based composites for controlled urea release [69]. These synergistic physicochemical interactions enable precise control over nutrient release kinetics, allowing for optimization of release profiles to match plant uptake requirements while minimizing environmental losses.

Nanocellulose presents a revolutionary opportunity for developing sustainable controlled–release fertilizers that align with modern agricultural practices. By leveraging its unique properties, researchers can create efficient nutrient delivery systems that not only enhance crop productivity but also contribute to environmental sustainability. Research indicates that nanocellulose can be utilized to create hydrogels that effectively encapsulate fertilizers. These hydrogels can control the release of nutrients through various mechanisms. There is concern that nanocellulose is impractical and unsuitable for agricultural applications due to the huge time and expense involved in its production. However, microfibrillated cellulose (MFC), which is easier to process than nanocellulose (NC), has many of the properties of nanocellulose, with microbundles of cellulose unravelling to nanosize in places.

The use of cellulose as a hydrogel was carried out by Winarti *et al.*, [116] using the crosslinking method. The use of corn cob cellulose provides a large swelling ratio, so that it has a large water absorption capacity. This is suitable for dry land. NC gives good results as a fertilizer carrier, as in the research conducted by Winarti *et al.*, [117]. NC hydrogels provide a large NPK fertilizer carrying capacity, which is 5%. This carrier provides a slow release of NPK nutrients within 3 weeks and has a positive effect on the development of chili daughters. Research by Priya *et al.*, [118] showed that the utilization of cellulose nanofibers (CNF) combined with carboxyl methyl cellulose (CMC) gave positive results for the release of urea, namely for 30 days. In addition, the hydrogel was also noted to be easily degraded by resistant bacteria within 3 months. In another study conducted by Sharma *et al.*, [119], showed that SRF, by utilizing CNF, provides a fairly long release of N nutrients, namely up to 36 days. In addition, it also supports the growth of microflora so as to maximize the absorption of N to plants.

In general, modifications such as hydrophobization can significantly slow down nutrient release rates. For example, gas-phase modification of nanocellulose has been shown to reduce the release of phosphorus and potassium by more than an order of magnitude compared to unmodified versions [120]. By functionalizing nanocellulose with positive and negative charges, researchers have successfully enhanced the interaction between the carrier and nutrient ions. This interaction helps modulate the solubility and subsequent release of nutrients like nitrogen, phosphorus, and potassium.

The advantages of using nanocellulose-based S/CRFs consist of the use of biodegradable materials like nanocellulose reduces environmental pollution associated with non-degradable synthetic fertilizers [28], controlled-release properties ensure that nutrients are available to plants over an extended period, reducing leaching losses and improving overall fertilizer efficiency [120], [121], and the ability to modify nanocellulose allows for the creation of S/CRFs tailored to specific crops or soil conditions, optimizing nutrient delivery based on plant needs [68]. Future research should focus on optimizing production methods, exploring various modification techniques, and conducting field trials to validate the effectiveness of MFC/NC-based S/CRFs in diverse agricultural settings.

MFC/NC nanoscale size is pivotal in enhancing its effectiveness in S/CRFs, offering several advantages contributing to improved agricultural practices. One of the most significant benefits of its nano size is the increased surface area it provides. This larger surface area facilitates greater interaction with nutrients, enabling more efficient encapsulation within the nanocellulose matrix [122]. As in the research of Moser *et al.*, cellulose nanofiber provides an increase in surface area over the original structure, from 128 m^2/g to 199 m^2/g. As a result, fertilizers can be loaded more effectively, ensuring that essential nutrients are available to plants in a more uniform and accessible manner [123]. This characteristic is crucial for optimizing nutrient delivery and improving overall fertilizer performance.

Additionally, the smaller particle size of MFC and NC enhances their ability to penetrate soil and interact with plant roots more effectively. The fine dimensions allow for better dispersion in both water and soil, leading to quicker diffusion rates of nutrients. This dimension ensures that plants can absorb nutrients more readily, which is vital for their growth and development. Moreover, the nanoscale structure enables nanocellulose to create a more extensive network within the soil, improving moisture retention and nutrient availability, thus supporting healthier plant growth [124]. In the research of Priya *et al.*, water retention of hydrogels is influenced by OH groups in the polymer chain. In addition, the level of water retention is also influenced by the nature of gelatinization and porosity. At room conditions (25 °C with RH 60%), water retention can last up to 16 days [118].

Furthermore, the controlled release capabilities afforded by MFC's/NC's nanoscale size are critical in reducing nutrient loss and minimizing environmental impact. By encapsulating nutrients within its structure, MFC/NC can regulate its release over time, preventing excessive leaching into water bodies and mitigating issues like eutrophication [125]. This controlled release mechanism not only enhances fertilizer efficiency but also contributes to sustainable agricultural practices by ensuring that nutrients are utilized effectively while reducing the ecological footprint of fertilizer application. Research conducted by França *et al.*, explained that the structure of cellulose nanofiber with negative and positive charges (CNF^- and CNF^+) provides the interaction of the release of NO^{3-} and K^+ nutrients in the soil, up to 80 days [66].

5 Cationic Cellulose for Slow/Controlled Release Fertilizer

Cationic modified cellulose has been used as a raw material for making slow-release media, including in the manufacture of fertilizers and slow-release drugs (drug delivery) [126]–[128], [66]. Its primary function is to prolong the release of nutrients encapsulated in slow-release media. Table 7 summarises several applications of cationic modified cellulose in fertilizers and slow-release drugs. Although cationic modified cellulose has not yet been applied to S/CRF, applying cationized MFC/NC in S/CRF can function as a binder for negatively charged metals in soil, such as waste absorption [83].

Cationic-modified MFC/NC has not yet been applied to S/CRF, which presents a gap in its potential utilization. Current research highlights the advancements in various biomass-derived functional materials, including cationic-modified cellulose, for applications such as enzyme immobilization and biocatalysis, but specific applications in S/CRF remain unexplored. While cationic MFC/NC exhibits properties that could enhance nutrient delivery and retention in agricultural practices, its integration into S/CRF formulations has not been documented. This suggests an opportunity for future research to investigate the efficacy and benefits of cationic-modified MFC/NC in enhancing the performance of slow-release fertilizer systems.

Cationic modification significantly enhances the performance of nanocellulose-based coatings for S/CRF applications through three primary mechanisms that improve coating functionality and nutrient delivery efficiency. First, enhanced coating adherence. Cationic modification improves coating adhesion to fertilizer granules through multiple bonding mechanisms. The positively charged quaternary ammonium groups on modified cellulose create strong electrostatic interactions with negatively charged surfaces, while maintaining hydrogen bonding capabilities through remaining hydroxyl groups. These dual interactions result in superior coating-substrate adhesion compared to unmodified cellulose, reducing coating delamination during handling and storage, and ensuring consistent coating integrity throughout the release period [70].

Second, improved moisture resistance. The introduction of cationic groups alters the hydrophilic-hydrophobic balance of nanocellulose, reducing excessive water uptake while maintaining controlled permeability. Cationic modification partially replaces hydroxyl groups with quaternary ammonium moieties, decreasing the material's affinity for water molecules and creating more stable film structures. This modification results in improved barrier properties against water vapor while allowing controlled diffusion necessary for sustained nutrient release, preventing coating failure due to over-swelling in high-moisture environments [71].

Third, superior nutrient binding capacity. Cationic modification dramatically enhances nutrient retention through electrostatic interactions between positively charged cellulose surfaces and anionic nutrients. Studies demonstrate that cationic modification can increase nutrient adsorption capacity by up to threefold compared to unmodified nanocellulose, particularly for phosphate and nitrate ions. This enhanced binding capacity enables more efficient nutrient loading and provides stronger retention forces that resist rapid leaching, resulting in extended release periods and improved fertilizer utilization efficiency [72].

Table 7: Some applications of cationic modified cellulose in fertilizers and slow-release drugs.

Product	Cellulose Type	Reagents	Role in Product	Ref.
Fertilizer	CNF	EPTMAC	Increases fertilizer encapsulation efficiency and prolongs the process of releasing fertilizer into the environment	[66]
Slow release fertilizer	MC	sodium periodate	Grafting agent and copolymer	[128]
Drug Delivery	CNC	CHPTAC	Increases enzyme stability against RNase degradation	[127]
Drug Delivery	CNC	CTAB	Prolongs the release of curcumin	[126]

6 Challenges of Application of Cationic Microfibrillated cellulose/Nanocellulose in Slow Release Fertilizer

Exploration of renewable resources is crucial to facilitating the shift to a materials economy that is more sustainable. Even though for the last 100 years, fertilizers used on plants have come from renewable resources with environmentally friendly production technology, most manufactured products for fertilization are still based on non-renewable minerals and fossil fuels. This fertilizer impacts surface water eutrophication due to the presence of phosphorus and nitrogen, which come from using chemical fertilizers that are not optimal or controlled. Another harmful impact of chemical fertilizers is that they can kill microorganisms important in plant growth and inhibit the decay of organic compounds that plants need. Fertilizer modification technology, by forming or changing the characteristics of current commercial fertilizers into S/CRF, is an alternative for optimizing the use of chemical fertilizers on plants. S/CRF is considered an environmentally friendly fertilizer widely used in crop cultivation to replace traditional nitrogen fertilizers. This was stated by Guo *et al.* [114], where S/CRF is a useful method for attaining sustainable productivity that concurrently addresses issues related to agricultural sustainability and excessive fertilizer use. However, the challenge is the optimal S/CRF application time and filler materials that do not provide optimal performance when using S/CRF.

According to a number of researchers, cellulose-based materials provide good candidates for creating sustainable and eco-friendly S/CRF. Lignocellulosic biomass consists of three main components: cellulose, hemicellulose, and lignin. Cellulose possesses a strong crystalline structure that requires specific pretreatment conditions to break down into simple sugars such as glucose. Ultrasonic-assisted pretreatment has been shown to significantly improve the efficiency of cellulose hydrolysis by reducing energy input while maximizing sugar yield. Hemicellulose, being more amorphous and less structurally organized than cellulose, is easier to hydrolyze but still benefits from targeted pretreatment. Methods such as microwave-assisted hydrolysis and steam explosion have proven effective in enhancing hemicellulose conversion, resulting in higher concentrations of fermentable sugars. Lignin, which provides structural rigidity to plant cell walls, presents an additional challenge. Ionic liquid-based delignification methods can selectively remove lignin without degrading cellulose or hemicellulose, while advanced oxidative pretreatments have shown promise in breaking down lignin structure and facilitating enzymatic access to cellulose [129].

The production of nanocellulose from lignocellulose biomass has been extensively reviewed, with chemical pretreatment methods proving essential for efficient extraction. Norrahim *et al.*, provided a comprehensive analysis of various pretreatment approaches, including acid, alkali, and steam explosion methods, that enable the conversion of agricultural residues into value-added nanomaterials such as nanocellulose, demonstrating the feasibility of sustainable production pathways [130].

In addition, cellulose can be the main framework for creating advanced agricultural products because of its modifiable structure, which allows for a wider range of applications, such as fillers in S/CRF production [28], [131], [132]. However, it is even more interesting if the cellulose used is nano-sized, as has been done by Rashidzadeh *et. al.*, where the cellulose-based nanocomposite coating induces nutrient release in a more controlled fashion and has good water retention properties [133].

The same concept was conveyed by Kassem *et. al.*, that the nanocomposites showed excellent water retention capacity compared to uncoated fertilizer. This condition is influenced by the hydrophobicity of the polymer material in holding water [134], [135]. However, the obstacle can be seen from the hydrophilic nature of nanocellulose, which can affect the characteristics of the S/CRF that is produced. Arnata *et al.*, [83] revealed that increasing the hydrophobic properties of nanocellulose certainly impacts its increasingly widespread use, where one technique to increase the hydrophobic properties of nanocellulose is by cationic modification on the nanocellulose surface.

It can be difficult to pinpoint the specific methods and functionalization strategies utilized to alter the surface properties of MFC/NC by functionalizing surface hydroxyl groups to produce the appropriate hydrophilic-hydrophobic balance [136], [58]. Therefore, the focus is on creating a balanced proportion of hydrophilic-hydrophobic groups in MFC/NC. Of course, to obtain optimal use of cationic modified MFC/NC in its application to S/CRF.

Research indicates that the methods utilized to obtain MFC/NC from lignocellulosic biomass have a significant impact on the material's form, characteristics, and surface chemistry. Chemical pre-treatment of biomass using alkalis, acids, bleaching agents, ionic liquids, and deep eutectic solvents requires careful consideration of their effectiveness. Therefore, several other parameters are considered when using cationic modified MFC/NC in S/CRF: process, environmental, and economic feasibility. Regarding the availability of commercial MFC/NC products worldwide, it is predicted that they will continue to increase due to their increasing and widespread application [135], such as in various applications in biomedical engineering and materials science [58]. This is not a severe challenge for business or industrial players developing MFC/NC-based products, especially for additional ingredients in S/CRF products.

Another challenge of applying cationic MFC/NC in S/CRF is related to the S/CRF production method. The coating technique or layering on the surface of the fertilizer that will be used is a challenge in itself. Revealed that the S/CRF production method consists of 1) material pretreatment, 2) conditioning, 3) fertilizer dissolution, 4) slow and controlled release fertilizer production, 5) composite connecting material production, 6) composite fertilizer production, and 7) coating [137]. In commercial, S/CRF production generally uses hydrophobic and encapsulation methods. In order to manage the solubility of the nutrients, the first approach involves applying a low solubility layer made of both organic and inorganic compounds to the surface of the fertilizer granules. The encapsulating process is mostly used in spray columns or coated drum batch systems. The second preparation method is more straightforward, using a hydrophobic solid as a matrix mixed and formed with powdered fertilizer. It is also applied commercially in developing countries. Thus, this review will be essential in explaining the advantages and restrictions of both polymeric and nonpolymeric coating materials, as well as the release mechanisms and characterization techniques of conventional and MFC/NC e-coated urea, the slow release properties that the coating imparts to MFC/NC -coated urea, and the release patterns of both.

Oil palm empty fruit bunches (OPEFB) are a potential biomass to produce nanocellulose. OPEFB, as biomass waste, has a high amount of cellulose, making it a suitable raw material for nanocellulose extraction. Researchers have recently focused a lot of attention on nanocellulose extracted from OPEFB because of its qualities, which include low cost and density, superior specific strength, thermal stability, and biodegradability. Previous research results showed that OPEFB has the potential to be used as a filler in the production of S/CRF. The potency can be seen from the high cellulose content of around 42%, with a crystallinity index that is quite similar to the original OPEFB fiber. The morphology of OPEFB shows a porous structure with uneven pore distribution, pore channel structure, and a pore surface that is not too smooth. However, it is necessary to make a slight modification to the cellulose from OPEFB, such as purifying it into single nano–sized cellulose [59].

The application of MFC/NC extracted from OPEFB has not been widely explored, especially in hydrogels. However, the prospects of MFC/NC are growing. OPEFB is now considered to meet the market requirements and product value chain as an MFC/NC raw material for hydrogel applications. Several previous researchers have also identified the challenges and potential future directions of MFC/NC hydrogels, as stated by Padzil *et al*., [60].

OPEFB, based on palm oil mill waste as the primary material for MFC/NC production and applied as a hydrogel filler, is undoubtedly attractive to research. Integrating MFC/NC with other hydrogel materials creates a regeneration product that displays extraordinary properties due to the extraordinary features of MFC/NC. In addition, there is a cationic modification treatment on MFC/NC before it is used as a filler in hydrogel production, which makes the surface interaction properties of MFC/NC even more attractive in various materials for making hydrogels.

Despite their advantages, MFC/NC hydrogels still face numerous challenges in terms of commercialization. One challenge is the long-term biosafety of MFC/NC hydrogels, especially in S/CRF applications. Despite being often regarded as environmentally benign, little is known about the biological effects and life cycle of MFC/NC hydrogels. Studies on hydrogels' life cycle assessment (LCA) are minimal, especially in nanocellulose hydrogels.

Current market prices for nanocellulose vary significantly depending on production method, purity, and supplier. Based on available commercial data, the cost benchmarks are as follows: CNC: $4,792/kg [138] to $25,000/kg ($25/gram) [139], CNF: $2,000/kg ($2/gram) [139]. CNF is approximately 10–12 times cheaper than CNC. These high material costs represent a significant economic challenge for S/CRF applications. However, large-scale production has the potential to substantially reduce costs through improved manufacturing efficiency and economies of scale [27]. The wide price range between different suppliers ($4,792–25,000/kg for CNC) indicates significant variations in quality specifications, processing methods, and market positioning. This price variability highlights the need for standardized benchmarks and quality specifications for industrial S/CRF applications.

In addition, the production cost per kilogram of nanocellulose is more expensive than the production cost of carbon fiber, which impacts the trend of hindering the market growth. However, compared to the chemical synthetic approach used for S/CRF needs, the cost of CNC can be significantly reduced in the near future by considering cellulose resources, which are the most abundant organic polymers in nature, and supported by the development of processing technologies to extract cellulose from various sources into nanocellulose. The same thing was expressed by Lizundia *et al*., [140]. In order to increase the competitiveness of nanocellulose hydrogels in the market, it is advised that future research focus on the hunt for more economical production techniques. Exposure of researchers to market requirements and product value chains is an essential future topic so that laboratory research can be adapted to industrial scale processes [60].

Despite the lack of data on the use of nanocellulose as a hydrogel filler and S/CRF production, it is hoped that in future applications, estimates of cost and energy use in each supply chain for nitrocellulose-based S/CRF production, both cationic modified and non-cationic modified, are deemed necessary to be considered and measured. Estimating the use of materials and energy in input and output in each process unit, which tends to impact the environment, is also a parameter of success in achieving environmentally friendly S/CRF production. Material, energy input, and output certainly impact environmental emissions and the costs incurred. These problems certainly need to be disclosed as an indicator in assessing the level of feasibility and sustainability of the practice of using nanocellulose as a filler in hydrogel production. It is hoped that the various challenges that arise can be overcome shortly.

The extraordinary properties of MFC/NC-based S/CRF will produce cost-effective "green" products that will significantly advance and improve the quality of life of the community. In addition, the need for nanocellulose materials tends to increase, with Inkwood Research anticipating that the global market for nanocellulose will reach USD 2,006.21 million in 2028, growing at a compounded annual growth rate (CAGR) of 18.05% during the forecast period from 2021 to 2028. This is driven by technological advances in the food packaging industry and increased volatile organic compounds (VOC) emissions in paints and coatings [139]. Figure 5 shows the global nanocellulose market forecast for 2021–2028 by end user.

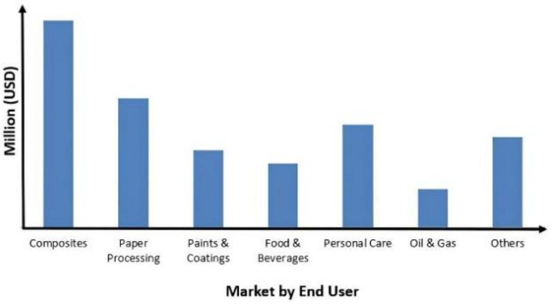


Figure 5: Global nanocellulose market forecast 2021–2028 by end user (Copyright: Inkwood Research ©2023).

If nanocellulose does not reach a level that satisfies the agricultural market in terms of price and quantity, MFC with a more lax level of processing than nanocellulose may still be able to bring the specifications expected of nanocellulose to S/CRF in terms of fertilizer function. Even if the size of MFC sometimes varies, MFC, in which cellulose fibers are subdivided by mechanical or combined with chemical [141], [142] and enzymatic [143] methods in pretreatment to reduce energy consumption and minimal mechanical processing, can still be sufficient for the purpose of S/CRF. Typically, MFC consists of fibrils with diameters ranging from 5 to 60 nm and lengths extending up to several micrometers, depending on the processing method used [144]–[146]. It is necessary to determine the right strategy to achieve all of this. The process stages must be passed, and their feasibility and sustainability status must be measured. These strategies include efficient use of resources and costs, environmental friendliness, the implementation of clean production, and improving community welfare. Figure 6 shows the strategy for applying OPEFB-based nanocellulose as a hydrogel filler. The strategic application of OPEFB-based nanocellulose as a hydrogel filler holds promise across multiple sectors. By optimizing production methods, enhancing hydrogel properties, and addressing existing challenges, this approach can contribute significantly to sustainable material development.

Although nanocellulose and its cationic derivatives are generally regarded as environmentally friendly and biodegradable, concerns have been raised about the potential toxicity of nanomaterials in soil ecosystems. At low concentrations, nanocellulose typically exhibits minimal impact on soil microbial activity and plant growth; however, higher doses or chemically modified forms may alter soil enzyme activity, inhibit microbial populations, or affect nutrient cycling. For example, cationic surfactants used in functionalization, such as CTAB, have been reported to inhibit microbial biodegradation processes at concentrations exceeding 0.3 mg L^{-1} [61]. These findings suggest that while cationic nanocellulose coatings are promising for sustainable fertilizer applications, further ecotoxicological studies are needed to assess their long-term impacts on soil health and biodiversity.

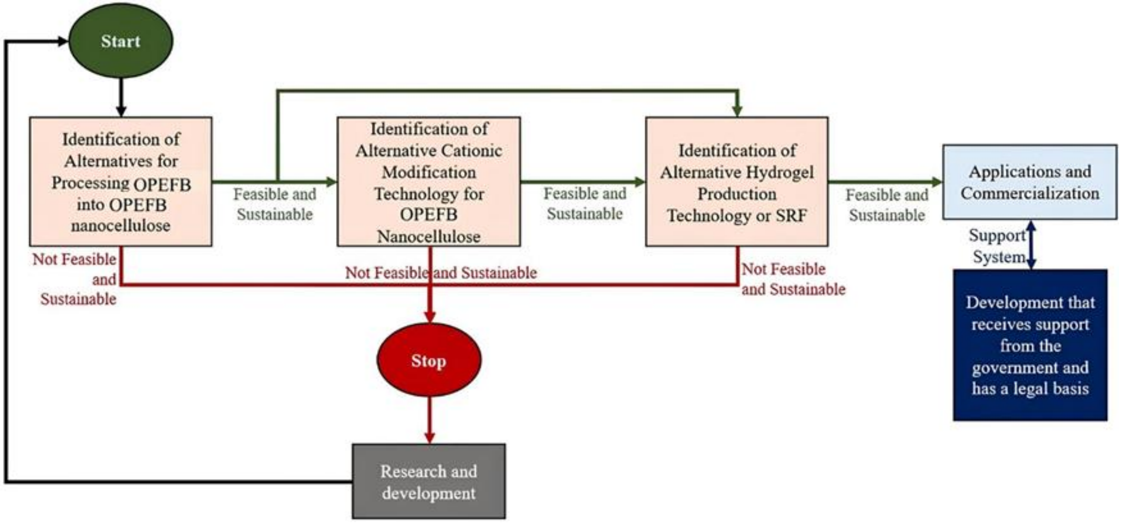


Figure 6: Strategy for applying OPEFB–based nanocellulose as a hydrogel filler.

Despite extensive research progress, industrial-scale commercialization of cationic MFC/NC-based fertilizer coatings remains limited. Current applications of nanocellulose-based S/CRF are predominantly confined to laboratory studies and pilot-scale field trials [66], [113], [119] with no major commercial fertilizer manufacturers openly marketing NC/MFC-coated fertilizers as primary products.

The absence of commercial implementation highlights the substantial cost challenges identified in our analysis, where nanocellulose materials ($2,000–25,000/kg) [138], [139] represent significantly higher costs compared to established fertilizer production systems. However, the projected growth of the global S/CRF market is expected to reach USD 5.89 billion by 2032 [42] suggests considerable commercial opportunities for breakthrough sustainable coating technologies.

Comprehensive analysis of available literature reveals critical quantitative benchmarks that define the performance advantages and limitations of cationic MFC/NC-based S/CRF systems. Cost analysis indicates significant economic barriers, with nanocellulose materials ranging from $2,000/kg for CNF to $25,000/kg for CNC [138]; [139], representing 100–1000 fold increases compared to conventional fertilizer systems. However, performance data demonstrate substantial functional improvements: cationic modification increases nutrient adsorption capacity by up to three-fold compared to unmodified systems, while extending controlled release duration from conventional immediate release to 28–90 days [113], [119].

Surface area enhancement through nanofibrillation increases from 128 to 199 m²/g [123], directly correlating with improved nutrient encapsulation efficiency. Water retention capabilities extend up to 16 days under ambient conditions [118], while complete biodegradation occurs within 3 months [118], contrasting favorably with non-biodegradable synthetic polymer alternatives. These quantitative benchmarks identify three key factors governing successful implementation: 1) cost optimization: requiring 50–80% price reduction through scaled production, 2) surface modification efficiency: maximizing cationic functionality while maintaining biodegradability, and 3) release kinetics control: achieving 60–90 day nutrient delivery aligned with crop uptake patterns.

The projected $5.89 billion S/CRF market by 2032 [42] provides economic incentive for addressing these technical challenges, with comparative performance data indicating that achieving cost parity with synthetic polymers while maintaining biodegradable advantages represents the critical threshold for commercial viability.

7 Conclusions and Recommendations

7.1 *Conclusions*

Cationic modified MFC/NC offers biodegradable alternatives for S/CRF, addressing environmental concerns associated with synthetic polymer coatings. Cationic modification transforms MFC/NC's hydrophilicity into functional advantages by introducing quaternary ammonium groups that enable superior nutrient retention through electrostatic interactions. Performance improvements include threefold increases in nutrient adsorption capacity, extended release duration to multi-month periods, and complete biodegradation within three months. These functional characteristics position cationic nanocellulose as a viable and sustainable coating material for next-generation fertilizer technologies.

However, successful commercial implementation requires addressing critical technical and economic barriers. Substantial cost differentials, limited understanding of long-term environmental impacts, and the absence of standardized production protocols represent key challenges that must be systematically resolved. Bridging these gaps through targeted research and development will enable sustainable fertilizer systems that balance agronomic performance, economic feasibility, and environmental responsibility, contributing to the advancement of precision agriculture and circular economy principles in fertilizer production.

7.2 *Recommendations*

It is recommended that future work should emphasize the search for S/CRF production processes with appropriate, cost-effective methods to increase the competitiveness of S/CRF in national and global markets. Waste-based nanocellulose materials are a top priority in developing nanocellulose-based S/CRF. This is, of course, related to increasing the added value of the waste itself and implementing an adequate circular economy in the S/CRF-based business sphere.

Abbreviations

AAPFCO	The Association of American Plant Food Control Officials
AEM	2-aminoethyl methacrylate
AEMA	2-aminoethyl methacrylamide
CAGR	Compounded Annual Growth Rate
CHPTAC	3-chloro-2-hydroxypropyl trimethyl ammonium chloride
CNC	cellulose nanocrystals
CNF	cellulose nanofibers
CNF$^+$	cationic cellulose nanofibrils
CNF$^-$	anionic cellulose nanofibrils
CTAB	cetyltrimethylammonium chloride
CTMAB	cetyltrimethylammonium bromide
EFB	empty fruit bunch
EPTMAC	2,3-epoxypropyl)trimethylammonium-chloride
FAO	Food and Agriculture Organization
GTMAC	glycidyltrimethylammonium chloride
IBDU	isobutylidenediurea
K	potassium
LCA	life cycle assessment
MC	microcellulose
MFC	microfibrillated cellulose
N	nitrogen
NC	nanocellulose
NPK	nitrogen, phosphorus, potassium fertilizer
OPEFB	oil palm empty fruit bunches
P	phosphorus
PDMAEMA	poly[2-(dimethylamino)ethyl methacrylate]
PHB	poly(3-hydroxybutyrate)
PVA	polyvinyl alcohol
S/CRF	slow/controlled release fertilizer
SI-SET-LRP	surface-initiated single-electron-transfer living radical polymerization
TPS	thermoplastic starch
UF	urea-formaldehyde
VOC	volatile organic compounds

Acknowledgments

These authors would like to thank to the Directorate General of Higher Education, Research, and Technology, Ministry of Education, Culture, Research, and Technology of the Republic of Indonesia for the financial support provided based on Decree No. 0459/E5/PG.02.00/2024 and Contract Numbers 027/E5/PG.02.00.PL/2024 and 22127/IT3.D10/PT.01.03/P/B/2024. Besides, the authors would like to thank IPB University, Indonesia; National Research and Innovation Agency (BRIN), Indonesia; The University of Tokyo, Japan; and Universitas Brawijaya, Indonesia, for the necessary support for the submitted review article.

Author Contributions

I.A.D.: conceptualization, investigation, editing, and writing an original draft; F.F.: investigation, reviewing, editing, funding acquisition, project administration; K.S.: reviewing, editing, funding acquisition, project administration; L.S.: reviewing, editing, and supervision; Y.S.: reviewing, editing, and supervision. All authors have read and agreed to the published version of the manuscript.

Conflicts of Interest

The authors declare no conflict of interest.

Declaration of generative AI and AI-assisted technologies in the writing process

The authors utilized the ChatGPT tool to enhance the language and readability of the manuscript.

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