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## Surface modification of Nano cellulose: The path to advanced uses of smart and sustainable bio-material

Shubhangi P Patil <sup>1</sup>, Prajakta Baikar <sup>1</sup> and Rajendra R Tayade <sup>2,\*</sup>

<sup>1</sup> Department of Chemistry, The Institute of Science, 15, Madam Cama Road, Mantralaya, Fort, Mumbai, Maharashtra 400032, India.

<sup>2</sup> Department of Chemistry, The Institute of Science, Nagpur, R. T. Road, Civil Lines, Nagpur – 440008, India.

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### Abstract

In order to develop sophisticated applications for intelligent and sustainable biomaterials, our work focuses on the surface modification of nanocellulose. Nanocellulose has the ability to be modified to improve its surface characteristics, making it appropriate for a variety of uses in industries including biomedicine, packaging, textiles, and water treatment. There have been discussions on a number of physical and chemical surface modification approaches, including mechanical treatment, high-pressure homogenization, and chemical functionalization. The study also highlights the results of different characterization methods that were employed to examine the surface-modified nanocellulose. The review addresses the difficulties in integrating nanocellulose into many applications, despite its potential, including manufacturing scale-up, standardisation, and toxicity issues. The article's conclusion highlights the need for continuing research and development of nanocellulose-based materials in order to meet these obstacles and offer long-term solutions to a range of social issues.

**Keywords:** Nano cellulose; Energy storage; Tissue engineering; Surface modification; Sustainable material

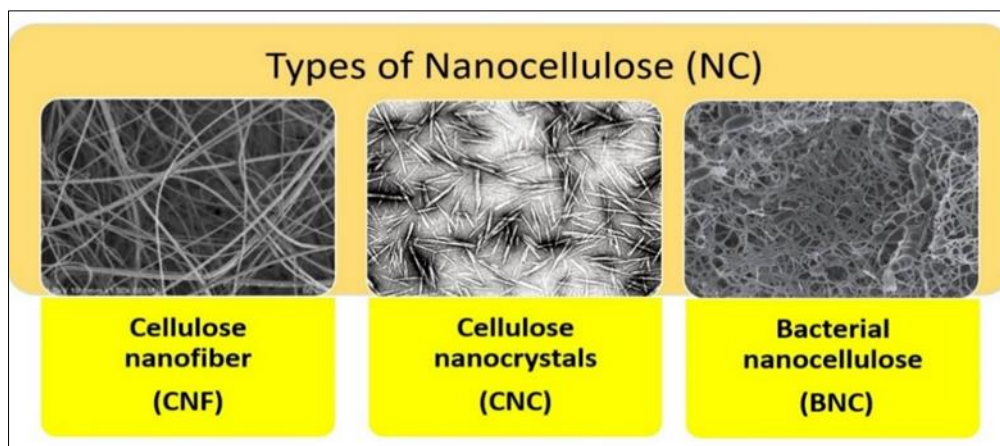
## 1. Introduction

### 1.1. Overview of Nanocellulose

A form of nanomaterial called nanocellulose is produced by bacteria and plants, which are natural sources. One of the tiniest naturally occurring substances, it is made of cellulose fibres with a diameter of less than 100 nanometers. Nanofibrillated cellulose (NFC) and nanocrystalline cellulose (NCC) are the two primary varieties of nanocellulose (NFC) as shown in figure.1. Whereas NFC is made by the mechanical disintegration of cellulose fibres into minute fibrils, NCC is made by converting cellulose fibres into small crystalline particles[1]. A variety of applications find nanocellulose to be appealing due to its numerous distinctive qualities. It has a large surface area to volume ratio and is rigid, robust, and lightweight[2]. A sustainable replacement for many conventional materials, it is also biodegradable, renewable, and non-toxic[3], [4].

Potential uses for nanocellulose include biomedicine, food packaging, building materials, and textiles, among other industries. It can be used to biomedicine to develop medication delivery systems or scaffolds for tissue engineering. It may be used to make barriers in food packaging that stop the transmission of oxygen and moisture, extending the shelf life of food goods. It can be used to increase the strength and durability of concrete in building materials. Overall, research into the characteristics and prospective uses of nanocellulose is still ongoing. It is a promising nanomaterial with a wide range of potential applications[8], [9].

\* Corresponding author: Rajendra R Tayade



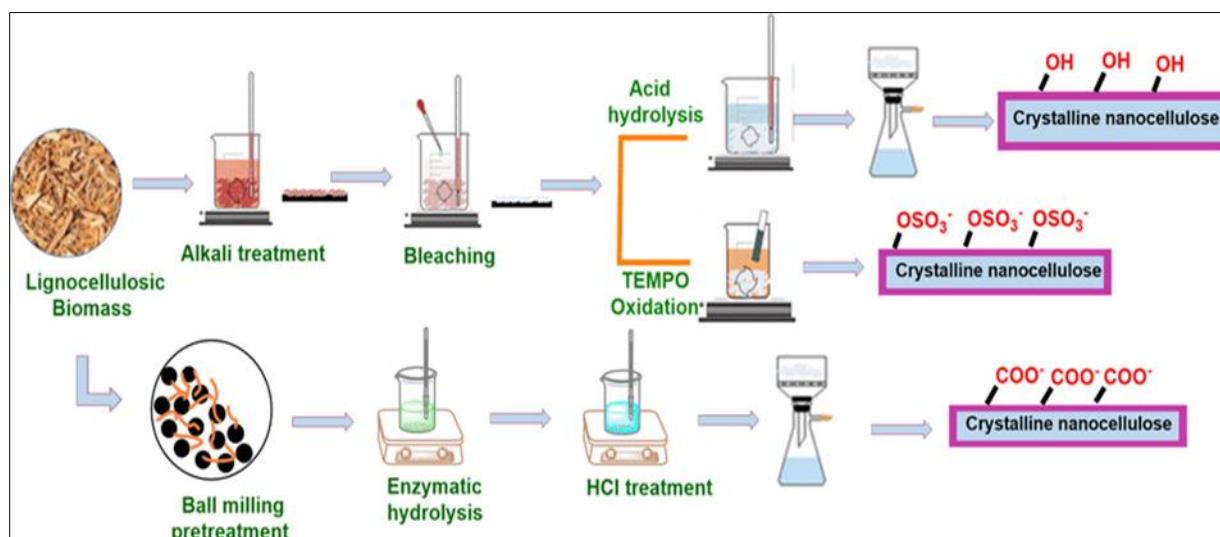
**Figure 1** Classification of nanocellulose. Adapted with permission from [5]–[7] Copyright @2019 Elsevier

## 1.2. Importance of Surface Modification of Nanocellulose

An essential procedure that can improve the characteristics of nanocellulose and increase its suitability for use with different materials and applications is surface modification. The following are some major justifications for the significance of surface modification of nanocellulose:

**Increased dispersibility:** Surface modification can make nanocellulose more dispersible in a variety of solvents and matrices, which will make it easier to process and more compatible with other materials. **Improved mechanical qualities:** Surface modification can help nanocellulose's mechanical qualities, including tensile strength, toughness, and flexibility. This can improve its appeal as a material for a variety of uses, such as coatings and the reinforcement of composites[10].

**Customized functionality:** Surface alterations can provide nanocellulose new or better properties, such as increased heat stability, water resistance, and chemical reactivity. This may increase the breadth of possible uses for nanocellulose. Nanocellulose's biocompatibility may be increased by surface modification, making it a more desirable material for biomedical applications including medication delivery and tissue creation[11], [12].



**Figure 2** Chemical and Physical surface modification of nano cellulose [15]

Chemical functionalization, physical adsorption, and covalent bonding are a few typical techniques for nanocellulose surface modification. By applying different functional groups to the surface of nanocellulose using these techniques, its characteristics and suitability for different applications are enhanced[3], [4]. Overall, surface modification of nanocellulose is a crucial procedure that can improve its characteristics and increase the range of sectors in which it may be used[10], [13]. Here, we review the significance of surface modification as shown in Figure.2 of nanocellulose,

as well as the numerous methods utilised for it, as well as its characterisation and uses in a variety of industries, including biomedicine, food packaging, paper and pulp, construction, and textiles. The review also intends to emphasise the significance of this study for potential future uses and societal advantages, as well as future directions and obstacles in the field[14].

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## 2. Surface Modification Techniques for Nano cellulose

### 2.1. Chemical Modification

By adding functional groups like carboxyl, amino, or hydroxyl groups to the surface of nano cellulose, chemical modification is a technique used to change the surface chemistry of the material. Nanocellulose's characteristics may be enhanced by this method, which will also increase its applicability. Chemical alteration can be done in a number of ways, including as oxidation, esterification, etherification, and grafting. By using oxidising substances like sodium periodate or TEMPO, carboxyl groups are added to the surface of nanocellulose during oxidation[2], [16]. Esterification is the process of adding ester groups to the surface of nanocellulose by reacting it with carboxylic acids or their derivatives. In order to add ether groups to the surface of nanocellulose, it must first react with alkyl or aryl halides. Grafting is the process of combining nanocellulose with monomers or polymers to create a layer that is covalently linked to the surface. Nanocellulose may be chemically altered to increase its compatibility with different solvents and matrices, improve its mechanical characteristics, and provide new or improved functionality. The type of functional group added, the extent of the alteration, and the technique of modification can all affect how successful a chemical modification is. To acquire the desired characteristics and usefulness of the modified nanocellulose for particular applications, careful optimisation of these parameters is necessary[17], [18].

#### 2.1.1. Acetylation

By adding acetyl groups to the hydroxyl groups on the surface of nanocellulose, acetylation is a chemical modification process used to change the surface. It becomes more soluble in nonpolar liquids and less polar as a result of this change, increasing its compatibility with hydrophobic materials. Nanocellulose's mechanical, thermal, and barrier characteristics are all improved by acetylation[4], [17]. By modifying reaction parameters such reagent concentration, reaction time, and temperature, one may regulate the degree of acetylation (DOA). Due to its enhanced characteristics and compatibility with hydrophobic materials, acetylated nanocellulose finds use in a variety of industries, including food packaging, medication delivery, and composites. However, it is important to take into account the possible toxicity of acetic anhydride or acetyl chloride as well as the release of acetic acid during storage of acetylated nanocellulose[19].

#### 2.1.2. Esterification

A renewable and biodegradable substance with special qualities such a high aspect ratio, a large surface area, and mechanical strength, nanocellulose may be esterified by altering its surface chemistry. The procedure comprises stirring the mixture at an appropriate temperature for a certain amount of time after adding a catalyst and carboxylic acids or their anhydrides to a nanocellulose suspension[20]. By washing with solvents to get rid of any unreacted chemicals and catalysts, the resultant esterified nanocellulose may be made purer. Many potential uses for esterified nanocellulose exist, including tissue engineering, medication delivery, and reinforcing in polymer composites. A viable strategy for enhancing the characteristics and broadening the scope of uses for this adaptable material is esterification of nanocellulose[14], [21].

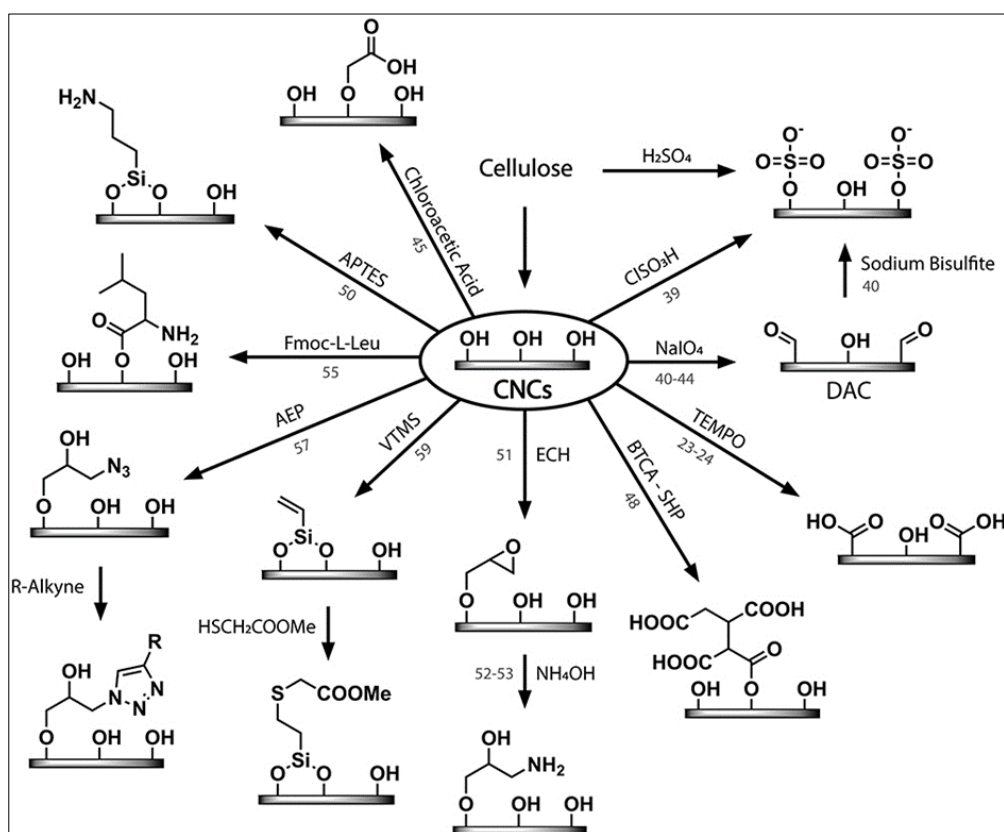
#### 2.1.3. Oxidation

When oxygen-containing functional groups are added to the surface of nanocellulose, a chemical reaction occurs that increases the material's reactivity and compatibility with other substances. Many oxidising substances, including sodium periodate, hydrogen peroxide, and nitric acid, among others, can be used to cause oxidation. By varying the reaction's duration, temperature, and oxidising agent concentration, it is possible to regulate the degree of oxidation[22]. Due to its improved solubility in polar solvents and water, the resultant oxidised nanocellulose offers a wide range of potential uses, including the creation of nanocellulose-based films, coatings, and composites. Moreover, the addition of functional groups like carboxylic acids, aldehydes, and ketones can be employed to further modify the nanocellulose chemically. Nanocellulose's mechanical and thermal characteristics may potentially be impacted by oxidation. For instance, severe oxidation can cause the glycosidic linkages in the cellulose structure to break, which can reduce the crystallinity and mechanical strength of nanocellulose. To preserve the desirable features of the resultant oxidised nanocellulose, careful control of the oxidation conditions is therefore required[8], [10], [23].

### 2.1.4. Etherification

By adding ether linkages to the hydroxyl groups on the cellulose backbone, etherification of nanocellulose is a chemical procedure that modifies the surface chemistry of the material. This can be done by etherifying agents such as alkyl halides, epoxides, or alkylene oxides with nanocellulose in the presence of a catalyst like sodium hydroxide or sulfuric acid. The degree of etherification may be controlled by adjusting the reaction parameters, such as temperature, time, and reagent concentration [14]. The resultant etherified nanocellulose can be utilised as an emulsion stabiliser or dispersion and has increased solubility in nonpolar liquids. Etherification can also improve the mechanical characteristics of nanocellulose-based composites and increase the compatibility of nanocellulose with nonpolar polymers like polyethylene or polystyrene. The reaction conditions must be carefully monitored since excessive etherification can reduce the crystallinity and mechanical strength of nanocellulose. To guarantee the safety of the procedure and the final product, it is also advisable to reduce the use of harmful or dangerous chemicals during the etherification process [19], [24], [25].

### 2.1.5. Silanization



**Figure 3** Chemical surface modification of nanocellulose [26]

Silanization of nanocellulose is a method for altering its surface that entails covering it with an organosilane layer. The procedure is carried out by soaking the nanocellulose in a solution that contains organosilanes that can react with the hydroxyl groups on the cellulose surface, such as aminosilanes or mercaptosilanes. Usually, an acid like hydrochloric acid or a base like sodium hydroxide will catalyse the process. The resultant silanized nanocellulose can be employed as a filler or reinforcement in nonpolar polymer matrices like silicone rubbers or epoxy resins because of its enhanced dispersibility in nonpolar solvents. Furthermore, silanization can increase the bonding strength between nanocellulose and nonpolar surfaces like glass or metals [16], [17]. The mechanical and thermal characteristics of nanocellulose can also be impacted by silanization. For instance, since bulky organosilane groups are introduced onto the cellulose surface as a result of excessive silanization, nanocellulose might lose its crystallinity and mechanical strength. The desirable qualities of the resultant silanized nanocellulose must thus be carefully controlled in the silanization conditions. In conclusion, silanization of nanocellulose is a surface modification technique that can improve nanocellulose's compatibility and adhesion with nonpolar surfaces and polymers. However, the level of silanization needs to be carefully controlled to prevent adverse effects on the properties of the final product [24], [25]. Figure. 3 shows various chemical surface modification of nanocellulose which enhance its applicability in various fields.

## 2.2. Physical Modification

Other methods for improving the surface characteristics of nanocellulose without introducing chemical modifications include physical surface modification approaches. These methods include surface adsorption, mechanical treatment, and heat treatment[27], [28]. The dispersibility and reactivity of the material can be improved by mechanical processes like homogenization and high-pressure homogenization that result in a reduction in particle size and an increase in surface area. A thin coating of carbon may be formed on the surface of nanocellulose during thermal processing at high temperatures in the absence of oxygen, improving the material's mechanical and thermal durability. To increase compatibility with other materials, surface adsorption might deposit a coating of surfactants or useful molecules on the nanocellulose surface[16], [29].

### 2.2.1. Ultrasonication

A physical surface modification method that may be utilised to alter nanocellulose materials is ultrasonication. Due to its distinct mechanical, optical, and chemical characteristics, nanocellulose is a regenerative and sustainable material that has attracted attention recently. Nanocellulose is frequently subjected to an ultrasonication process, which includes delivering high-frequency sound waves to the substance. In the process of ultrasonication, high-frequency sound waves in the liquid medium produce high-intensity cavitation bubbles that collide with one another and produce significant localised pressure and temperature changes. The nanocellulose surface may become mechanically deformed, fragmented, or eroded as a result of these pressure variations, changing the surface[20], [24]. The ultrasonication's amplitude, frequency, and duration as well as the characteristics of the nanocellulose material all affect how much change occurs. The surface of nanocellulose can be changed by ultrasonication in a number of ways, including surface area, dispersibility, reactivity, and the addition of functional groups. Nanocellulose, for instance, may have carboxylic and hydroxyl groups added to its surface by ultrasonication, which enhances the material's compatibility with other substances and makes it possible to employ it in a variety of applications, including composites, coatings, and biomedical materials. Overall, ultrasonication is a potential approach for altering the surface of nanocellulose. It has numerous benefits, including being easy, inexpensive, and ecologically benign[27].

### 2.2.2. Mechanical Treatment

Materials made of nanocellulose can undergo mechanical treatment, a kind of physical surface modification. Due to its distinct mechanical, optical, and chemical characteristics, nanocellulose is a regenerative and sustainable material that has attracted attention recently. By using mechanical forces on the material, it is possible to change the surface of nanocellulose[27]. Nanocellulose is exposed to mechanical stresses during mechanical treatment, which can result in surface erosion, fragmentation, and mechanical deformation. The degree of alteration is influenced by a number of factors, including the duration and intensity of the mechanical treatment and the characteristics of the nanocellulose material. The surface of nanocellulose may be altered mechanically in a number of ways, including surface area, dispersibility, reactivity, and the introduction of functional groups. Nanocellulose, for instance, can have carboxylic and hydroxyl groups added to its surface through mechanical processing, which enhances the material's compatibility with other substances and makes it possible to use it in a variety of applications, including composites, coatings, and biomedical materials[24], [30].

Nanocellulose's surface may be altered mechanically using a variety of techniques, including grinding, milling, and homogenization. These procedures entail exerting mechanical pressures on the substance using various pieces of machinery, including ball mills, attrition mills, and high-pressure homogenizers. Overall, mechanical treatment is a potential way for altering the surface of nanocellulose[27]. It has numerous benefits, including being easy, inexpensive, and ecologically benign. To accomplish the required surface modification of the nanocellulose material, however, attention should be taken in selecting the precise mechanical treatment method and the treatment parameters[27].

### 2.2.3. High-Pressure Homogenization

A physical surface modification method that may be utilised to alter nanocellulose materials is high-pressure homogenization. Due to its distinct mechanical, optical, and chemical characteristics, nanocellulose is a regenerative and sustainable material that has attracted attention recently. The surface of nanocellulose can be altered via high-pressure homogenization by applying high-pressure pressures to the substance. High-pressure forces are applied to nanocellulose during high-pressure homogenization, which can result in mechanical deformation, fragmentation, and surface erosion. The pressure, flow rate, number of passes, and characteristics of the nanocellulose material are only a few of the variables that affect how much alteration occurs[24], [31]. The surface of nanocellulose may be altered by high-pressure homogenization in a number of ways, including by expanding its surface area, strengthening its dispersibility, boosting its reactivity, and adding functional groups. Nanocellulose, for instance, can have carboxylic and hydroxyl groups added to its surface through high-pressure homogenization, which enhances the material's

compatibility with other substances and makes it possible to use it in a variety of applications, including composites, coatings, and biomedical materials[32], [33].

A flexible method that may be used to change the surface of nanocellulose in a controlled way is high-pressure homogenization. Moreover, it may be utilised to create very homogenous and stable nanocellulose dispersions, which are necessary for a number of applications. In order to accomplish the appropriate surface modification, high-pressure homogenizers can be operated at a variety of pressures and flow rates. Overall, high-pressure homogenization is a promising approach for changing the surface of nanocellulose. It has numerous benefits, including being easy, inexpensive, [2], [29]and ecologically benign. To accomplish the necessary surface modification, however, attention should be taken in selecting the precise high-pressure homogenization conditions and the characteristics of the nanocellulose material.

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### 3. Characterization Techniques of Surface-Modified Nanocellulose

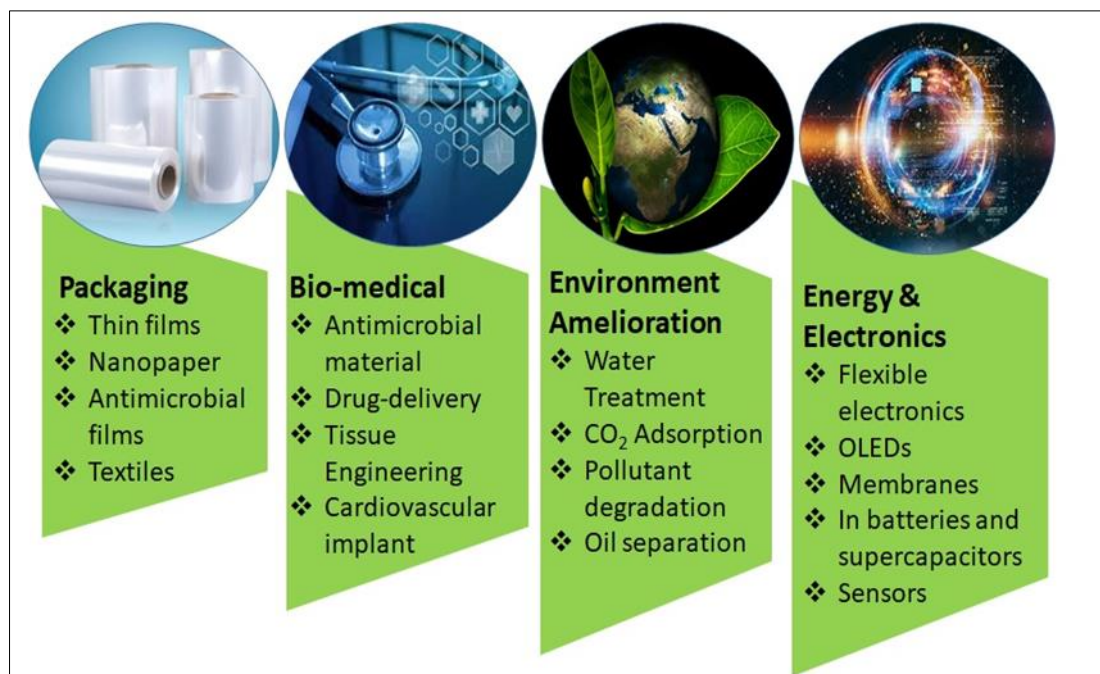
Surface-modified nanocellulose (NC) is a promising nanomaterial with potential applications in a wide range of fields, including biomedical engineering, environmental science, and material science[34]. Several characterization techniques are used to evaluate the properties of surface-modified nanocellulose, including:

- Scanning Electron Microscopy (SEM): SEM is a high-resolution imaging technique that can provide detailed images of the surface morphology and topography of nanocellulose. It can be used to visualize the changes in the surface structure and morphology of nanocellulose after surface modification.
- Transmission Electron Microscopy (TEM): TEM is a powerful imaging technique that can provide high-resolution images of the internal structure of nanocellulose. It can be used to observe the changes in the internal structure and morphology of nanocellulose after surface modification.
- X-ray Diffraction (XRD): XRD is a technique that can provide information about the crystal structure and orientation of nanocellulose. It can be used to analyze the changes in crystal structure and orientation of nanocellulose after surface modification.
- X-ray Photoelectron Spectroscopy (XPS): XPS is a technique that can provide information about the chemical composition and oxidation state of the surface of nanocellulose. It can be used to analyse the changes in the surface chemistry of nanocellulose after surface modification.
- Nuclear Magnetic Resonance (NMR): NMR is a technique that can provide information about the chemical composition and structure of nanocellulose. It can be used to analyze the changes in the chemical composition and structure of nanocellulose after surface modification.
- Thermogravimetric Analysis (TGA): TGA is a technique that can provide information about the thermal stability and degradation behaviour of nanocellulose. It can be used to analyze the changes in thermal stability and degradation behavior of nanocellulose after surface modification.
- Dynamic Light Scattering (DLS): DLS is a technique that can provide information about the size distribution and stability of nanoparticles in solution. It can be used to analyze the changes in the size distribution and stability of nanocellulose after surface modification.
- Fourier Transform Infrared Spectroscopy (FTIR): FTIR is a technique that can provide information about the functional groups and chemical bonding of nanocellulose. It can be used to analyze the changes in the functional groups and chemical bonding of nanocellulose after surface modification.

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### 4. Applications of Surface-Modified Nanocellulose in Various Fields

Nanocellulose, derived from plant-based sources, has found versatile applications across various industries due to its remarkable properties. In the field of materials science, its exceptional strength, lightweight nature, and transparent qualities make it a sought-after component in creating advanced nanocomposites for packaging materials, reinforcing polymers, and enhancing the structural integrity of various products. In biomedicine, nanocellulose's biocompatibility makes it valuable for drug delivery systems, wound healing, and tissue engineering. Additionally, its environmentally friendly nature aligns with the growing demand for sustainable alternatives, positioning nanocellulose as a key player in the development of eco-friendly solutions across multiple sectors[35], [36]. Nanocellulose is used in a variety of disciplines, as seen in **Figure 4**.



**Figure 4** Applications of nanocellulose in various field

#### 4.1. Biomedical Applications

Surface-modified nanocellulose can be used as a carrier for drug delivery systems due to its high surface area and biocompatibility. Surface modification can improve the drug loading capacity, release kinetics, and targeting efficiency of the nanocellulose. Surface-modified nanocellulose can be used as a scaffold for tissue engineering due to its biocompatibility and ability to mimic the extracellular matrix. Surface modification can improve the mechanical properties, cell adhesion, and proliferation of the nanocellulose scaffold. They can be used as a wound dressing material due to its biocompatibility, high water-holding capacity, and ability to promote cell proliferation and wound healing[2], [34].

#### 4.2. Packaging Materials

Surface-modified nanocellulose can be used as a barrier material in packaging due to its ability to reduce the permeability of gases, such as oxygen and carbon dioxide. Surface modification can improve the barrier properties of the nanocellulose and reduce the risk of food spoilage. Surface-modified nanocellulose can be used as a biodegradable packaging material due to its renewable and sustainable nature. Surface modification can enhance the biodegradability of the nanocellulose and reduce environmental pollution. They can be used as a reinforcement material in packaging due to its high tensile strength and stiffness. Surface modification can improve the mechanical properties of the nanocellulose and reduce the risk of package deformation[20], [30], [37].

#### 4.3. Textile Industry

Surface-modified nanocellulose can be used as a dye adsorbent in the textile industry due to its high surface area and hydrophilicity. Surface modification can improve the dye uptake capacity of the nanocellulose and reduce the risk of dye effluent. Surface-modified nanocellulose can be used as a reinforcement material in the textile industry due to its high tensile strength and stiffness. Surface modification can improve the mechanical properties of the nanocellulose and increase the durability of the textile. They can be used as an antibacterial material in the textile industry due to its ability to inhibit bacterial growth. Surface modification can introduce antibacterial agents onto the nanocellulose and improve its antibacterial properties[25], [37], [38].

#### 4.4. Water Treatment

Surface-modified nanocellulose can be used as a heavy metal adsorbent in water treatment due to its high surface area and ability to chelate metal ions. Surface modification can enhance the metal adsorption capacity of the nanocellulose and reduce the risk of water pollution. Surface-modified nanocellulose can be used as an adsorbent for organic pollutants in water treatment due to its high surface area and hydrophilicity. Surface modification can improve the

adsorption capacity of the nanocellulose and reduce the risk of water pollution. They can be used as an antimicrobial material in water treatment due to its ability to inhibit microbial growth. Surface modification can introduce antimicrobial agents onto the nanocellulose and improve its antimicrobial properties[4], [33], [39].

#### 4.5. In the world of electronics

Nanocellulose has shown great potential in the field of electronics due to its unique combination of properties, including high mechanical strength, excellent flexibility, a large surface area, and low toxicity. Here are some potential applications of nanocellulose in the field of electronics. Nanocellulose can be used as a substrate material for flexible electronics due to its excellent mechanical flexibility and low cost. It can also be used as a reinforcement material in composite substrates to improve their mechanical properties[40], [41]. Nanocellulose-based sensors can be used in various applications, including environmental monitoring, healthcare, and food safety. For example, nanocellulose-based sensors have been developed for the detection of heavy metals and volatile organic compounds[35], [42]. Nanocellulose can be used as a component in energy storage devices, such as supercapacitors and batteries. Nanocellulose-based electrodes have shown promising performance due to their high surface area and high electrical conductivity[35], [43]. Nanocellulose can be used as a component in optoelectronic devices, such as displays, solar cells, and light-emitting diodes (LEDs). For example, nanocellulose-based transparent electrodes have been developed as a replacement for indium tin oxide (ITO) electrodes in displays and solar cells. Nanocellulose is a biodegradable material, making it an ideal choice for the development of biodegradable electronics. Biodegradable electronics have potential applications in implantable medical devices and environmental monitoring systems. In conclusion, nanocellulose has shown great potential in the field of electronics due to its unique properties and biodegradability. The development of nanocellulose-based electronics can lead to innovative and sustainable solutions in various applications[44]–[46].

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## 5. Challenges and Future Directions

Despite the promising applications of nanocellulose in various fields, there are several challenges that need to be addressed to fully realize its potential. Here are some challenges and future directions for the use of nanocellulose:

- **Production Scale-Up:** The production of nanocellulose is still in the developmental phase and is currently limited to small-scale production. Developing large-scale production methods is necessary to meet the increasing demand for nanocellulose in various applications.
- **Standardization:** The lack of standardisation in nanocellulose production and characterization makes it difficult to compare the properties of different types of nanocellulose. Developing standardised methods for nanocellulose production and characterization is crucial to ensuring the reproducibility and reliability of the results[47], [48].
- **Cost:** The production of nanocellulose is still expensive due to the high cost of raw materials and energy consumption. Developing cost-effective production methods and finding alternative raw materials is necessary to reduce the cost of nanocellulose production.
- **Toxicity:** The potential toxicity of nanocellulose to human health and the environment is still unknown. Further studies are needed to assess the safety of nanocellulose and its potential environmental impacts.
- **Integration with Existing Technologies:** Integrating nanocellulose with existing technologies and materials is crucial to fully exploiting its potential in various applications. Developing compatible technologies and materials for nanocellulose is necessary to enhance its performance and functionality[49], [50].
- In the future, nanocellulose has the potential to revolutionize various industries, including biomedical, packaging, textile, and water treatment. Addressing the challenges mentioned above and developing new applications for nanocellulose can lead to innovative and sustainable solutions for various societal problems.

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## 6. Conclusion

Nanocellulose's surface alterations have created a wide range of possibilities for its use in diverse applications. Its surface qualities, including as stability, dispersibility, and functionality, have been enhanced by the physical and chemical alterations, making it appropriate for a variety of applications. In the biomedical area, nanocellulose has demonstrated considerable promise for use in tissue engineering, medication delivery, and wound dressing. It also has prospective uses in the textile sector, water treatment, and packaging materials, among other things. For the effective incorporation of nanocellulose in diverse applications, there are still a few issues that need to be resolved, such as production scale-up, standardisation, cost, toxicity, and integration with existing technology. Addressing these difficulties and creating new uses for nanocellulose can result in creative and long-lasting answers to a variety of social issues. Overall, nanocellulose's surface alterations have improved its qualities and broadened its possible uses, making it a promising substance for further study and development.



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## Compliance with ethical standards

### Disclosure of conflict of interest

No conflict of interest to be disclosed.

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