

## REVIEW

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## Translational applications of magnetic nanocellulose composites

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Nanocellulose has emerged as a potential 'green' material owing to its inimitable properties. Furthermore, the significant development in technology has facilitated the design of multidimensional nanocellulose structures, including one-dimensional (1D: microparticles and nanofibers), two-dimensional (2D: coatings), and three-dimensional (3D: hydrogels/ferrogels) composites. In this case, nanocellulose composites blended with magnetic nanoparticles represent a new class of hybrid materials with improved biocompatibility and biodegradability. The application field of magnetic nanocellulose composites (MNCs) ranges from biomedicine and the environment to catalysis and sensing. In this review, we present the major applications of MNCs, emphasizing their innovative benefits and how they interconnect with translational applications in clinics and the environment. Additionally, we focus on the synthesis techniques and role of different additives in the fabrication of MNCs for achieving extremely precise and intricate tasks related to real-world applications. Subsequently, we reveal the recent interdisciplinary research on MNCs and discuss their mechanical, tribological, electrochemical, magnetic, and biological phenomena. Finally, this review concludes with a portrayal of computational modelling together with a glimpse of the various translational applications of MNCs. Therefore, it is anticipated that the current review will provide the readers with an extensive opportunity and a more comprehensive depiction related to the types, properties, and applications of MNCs.

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## 1. Introduction

The emerging interest in renewable, green, and sustainable materials in various applications is due to their high production value with minimal health and environmental impacts.<sup>1–3</sup> From the discovery of the terminology of nanocellulose (NC) in 1970 to the introduction of NC in the form of patents and publications in 1980, the interest in this attention-grasping material by researchers has expanded to a wide variety of biomedical and environmental applications.<sup>4</sup> The expansion of NC-based research studies in the last 40 years presents a vibrant depiction of the passion for this material (Fig. 1). The number of articles has tremendously increased from just 100 publications in 1990 to more than 15 000 NC-related articles in 2024.

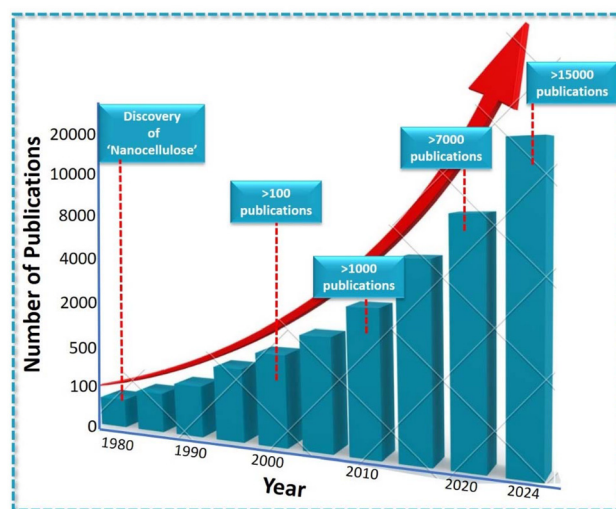
Nanocellulose can be isolated from plant materials or biochemical processes and possesses several desirable features, including high mechanical strength (Young's modulus = 20–50

GPa), ability to self-assemble, non-toxicity, and biodegradability owing to its size, shape, crystallinity, and surface chemistry.<sup>5–7</sup> NC is specifically categorized into bacterial nanocellulose (BNC), cellulose nanofibrils (CNFs), and cellulose nanocrystals (CNCs).<sup>8</sup> Among them, BNC is extracted from bacteria through a 'bottom-up' process, whereas CNCs and CNFs are produced from algae or plants using a 'top-down' method. From the perspective of researchers, using agricultural waste to recover nanocellulose is a very interesting subject. The residual agricultural material is lignocellulosic biomass with a high cellulosic component. In this case, several procedures are involved in the extraction of nanocellulose from lignocellulosic biomass, as shown in Fig. 2. Prior to the extraction of nanocellulose using different extraction procedures, its pretreatment removes non-cellulosic substances such as lignin, hemicellulose, and pectin. Considering the remarkable mechanical qualities of CNFs made from natural fibers, they have attracted significant interest. Without significantly destroying cellulose, mechanical treatments can separate CNFs from the primary and secondary cell walls. The degree of polymerization, shape, and aspect ratio of CNFs vary depending on the source of their raw materials and the defibrillation method. Processes such as microfluidization, high-pressure homogenization, grinding, and refining are required for the mechanical separation of

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CNFs from wood and non-wood fibers. The rotor and stator disks of a disk refiner feature surfaces that are equipped with bars and grooves, which subject the fibers to repeated cyclic stresses. The dilute fiber suspension is driven through this gap and this action irreversibly alters the shape and size of the fibers, hence enhancing their bonding capacity. Diluted slurries of previously refined cellulose fibers are fed through a spring high-pressure-loaded valve assembly while under high pressure during the homogenization process. The shearing and impact forces cause a significant decrease in pressure on the fibers when this valve rapidly opens and closes. This interaction of forces causes the cellulose fibers to fibrillate to a high degree, which causes the gradual release of CNFs.<sup>9</sup> It should be noted that although this is the most common method for the production of CNFs, mechanical treatments require a large amount of energy.<sup>10</sup> In this case, several pretreatments must be carried out before mechanical fibrillation to reduce the energy consumption during the CNF separation process. These pretreatments include mechanical, enzymatic, and chemical methods. The effective chemical pretreatments to reduce the energy consumption during the mechanical fibrillation process include TEMPO-mediated oxidation, carboxymethylation, and enzymatic pretreatment.<sup>11–13</sup> The raw sources must be subjected to sonication and very acidic conditions to isolate CNCs. This causes cellulose to hydrolyze into noncrystalline domains, from which rod-like structures known as CNCs can be formed. Concentrated sulfuric acid is the most



**Fig. 1** Since the discovery of the terminology of nanocellulose in 1990, nanocellulose-based research publications have increased exponentially to date (Web of Science data).

often used mineral acid. The esterification of the surface hydroxyl groups of cellulose occurs during the hydrolysis of sulfuric acid. This introduces a large number of negatively charged sulfate groups on the surface of CNCs, which prevent CNCs from clumping and flocculating in an aqueous solu-



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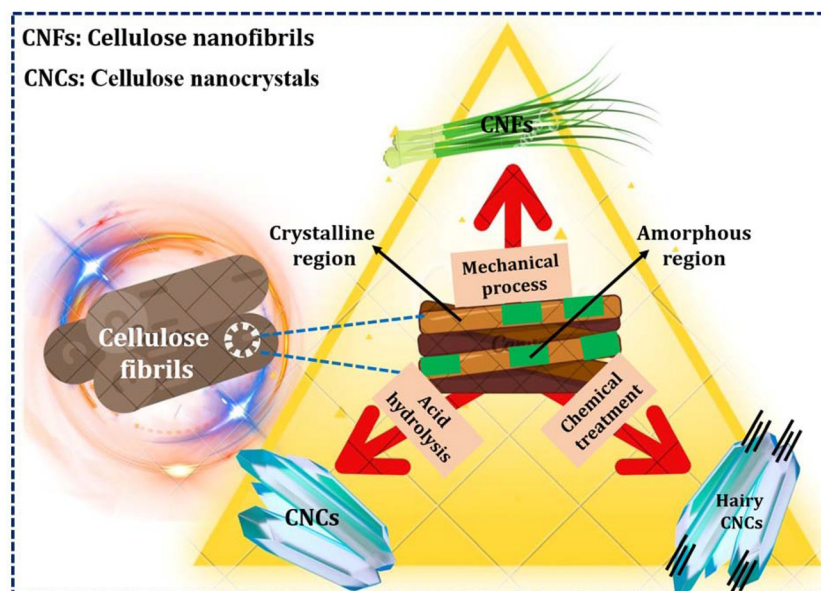
*and carbonaceous additives for cartilage repair applications. She has published extensively with one Indian patent and more than 35 publications in renowned journals. She worked at the Indian Institute of Technology, Kanpur, India, for her doctoral work and obtained her Ph.D. in 2019. She received the Dr D. S. Kothari's Postdoctoral Fellowship and completed her postdoctoral work at the Indian Institute of Science Bangalore. Being a silver medalist upon graduation and postgraduation, Dr Shikha also received the Best Researcher in Surface Engineering Award from the World Research Council, Young Scientist Award, Best Oral, Best Poster Award, etc. She also served as a reviewer for several prominent journals such as Elsevier, RSC, Springer Nature, and IOP.*



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**Fig. 2** Schematic showing the process for the extraction of nanocellulose from lignocellulosic biomass. The concept was adapted from ref. 15 and redrawn by the authors.

tion.<sup>14</sup> Nonetheless, the CNCs made *via* the hydrolysis of sulfuric acid exhibit a reasonable degree of thermostability. This may be resolved by employing sodium hydroxide to neutralize the nanoparticles.



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Presently, although magnetic nanoparticles possess numerous exceptional features for various applications,<sup>16,17</sup> their removal from a suspension is challenging due to their aggregation and oxidizing properties, which cause some shortcomings in any process. Accordingly, researchers have developed several approaches to avoid the accumulation of magnetic nanoparticles such as the use of a polymeric template for the *in situ* synthesis of magnetic nanoparticles, functionalization of magnetic nanoparticles, and designing composites of polymers.<sup>18</sup> Amongst the diverse resources used to diminish the problems associated with magnetic nanoparticles, nanocellulose unveils remarkable prospects due to its noteworthy characteristics. In this case, magnetic nanoparticles condense due to the hydroxyl groups present in nanocellulose, which provides a network of nanofibers through hydrogen bonding. Magnetic nanocellulose composites (MNCs) composed of magnetite (Fe<sub>3</sub>O<sub>4</sub>) nanoparticles display superparamagnetic properties and can be significantly pertinent to biomedical and environmental applications (Fig. 3). The biomedical or bioengineering applications of MNCs are related to diagnostics (magnetic resonance imaging),<sup>19</sup> disease treatment,<sup>20</sup> biosensing,<sup>21</sup> biolabeling,<sup>22</sup> and controlled release of drugs,<sup>23</sup> while their environmental applications include water purification remedies,<sup>24</sup> water filtration, adsorbents, photocatalysts, and fuel cells.<sup>25</sup> Fig. 3 shows a schematic representation of the various translational applications of MNCs associated in the environment and biomedical fields.

However, although researchers have probed a diverse range of conventional stimuli-responsive biomaterials thus far for desirable applications, in the pursuit of technical merits and functional versatility, MNCs have marked a significant stride compared to materials. MNCs exhibit suitable attributes such as high Young's modulus, high surface area, high storage





**Fig. 3** Schematic presentation showing the various translational applications of magnetic nanocellulose composites (MNCs) in the biomedical and environmental fields.

stability, prolonged half-life, high pH tolerance, and most notably, high stability in reuse assays and great potential for chemical modification. Furthermore, no added purification procedures are required for their recovery from the reaction mixture.<sup>26</sup> Their magnetic responsiveness combined with the natural properties of cellulose enables remote and non-invasive control. This permits manipulation using an external magnetic field, which is an important characteristic for remote actuation, tissue engineering, and targeted drug delivery. The presence of cellulose components in MNCs provides high biocompatibility, biodegradability, and flexibility in comparison to conventional biomaterials such as poly(*N*-isopropyl acrylamide) (PNIPAM), polyurethane-based shape memory polymers, polycaprolactone, polyaniline, poly(ethylene glycol), and poly(acrylic acid), which often require additional surface modification, special designs or additives to enhance compatibility and achieve degradable properties. The production of nanocellulose from renewable resources/bio-wastes is cost-effective; however, the higher costs associated with other advanced materials can constrain their widespread implementation, specifically in cost-sensitive applications. Besides their huge potential for multifunctional applications in diverse fields including wound dressings, intelligent drug delivery, magnetically targeted drug preparations, thermally responsive optically adjustable devices, electronic skin, and sensors, MNCs are associated with certain limitations, *e.g.* their fabrication involves the synthesis of cellulose nanomaterials and the embedding of magnetic nanoparticles, which requires precise control of the size and distribution of nanoparticles for consistent performance and can be complex and costly. Also, the incorporation of magnetic nanoparticles in higher amounts may affect the mechanical properties, potentially leading to an increase in stiffness. Furthermore, the long-term environmental impact and biodegradability of magnetic nanoparticles need to be assessed, particularly concerning their release and accumulation in the environment. MNCs can be prepared as coatings, microspheres, fibers, hydrogels, and aerogels that

respond distinctly to an external magnetic field due to their different morphological structures and features. Magnetic nanocellulose films possess significant magnetic properties, which increase with an increase in the concentration of magnetic nanoparticles.<sup>27</sup> Magnetic films can be prepared using  $\text{Fe}_3\text{O}_4$  or  $\text{CoFe}_2\text{O}_4$  nanoparticles. Between them, the magnetic nanocellulose film synthesized using  $\text{CoFe}_2\text{O}_4$  nanoparticles show a higher magnetic coercivity than that prepared with  $\text{Fe}_3\text{O}_4$  nanoparticles. The thermal stability of magnetic nanocellulose films depends on the crystallinity of the dispersed particles, which significantly contributes to the strength of magnetic nanocellulose films. Also, superparamagnetic nanocellulose microspheres have gained many advances due to their remote responses to an external magnetic field. Potential magnetic nanocellulose microspheres must possess a small size, high magnetic saturation, chemical stability, biocompatibility, and biodegradability together with a cheap and simple preparation process. The absorption performance of magnetic nanocellulose microspheres is an important aspect for attaining substantial outcomes in various clinical, and environmental applications. The Langmuir theory states that the smaller the size of magnetic nanocellulose microspheres, the higher their surface area, and thus the stronger their absorption capability. After a brief discussion on magnetic nanocellulose films and magnetic nanocellulose microspheres, the focus on magnetic nanocellulose fibers suggests that these fibers can be prepared *via* the strong agitation of nanocellulose pulp with a concentrated suspension of magnetite and maghemite particles, followed by washing of the composite to eradicate unbound magnetic nanoparticles from the surface of the polymer. Moreover, magnetic nanocellulose gels have attracted significant attention from researchers due to their stimuli-responsive feature. A homogenous distribution of magnetic particles in nanocellulose hydrogels was achieved by Chatterjee and co-workers<sup>28</sup> using a two-step method for their synthesis. In the first step, the emulsion method was used to synthesize hydroxypropyl cellulose with functionalized maghe-

mite. In the second step, a cross-linker was used to generate a network structure in the hydrogel. Magnetic nanocellulose aerogels exhibit different characteristics than hydrogels given that aerogels consist of air as a medium (the liquid in hydrogels is removed by the freezing-drying method), whereas hydrogels involve liquid as a medium.<sup>29</sup> Magnetic nanocellulose aerogels are extra flexible porous materials with additional characteristics and advantages such as high porosity, low density, and high surface area. The concise deliberation on nanocellulose to magnetic nanocellulose and their types can give an exciting boost to these materials. Thus, the present review aims to demonstrate the relationship between the theoretical and experimental contributions on magnetic nanocellulose composites. Firstly, the techniques for the synthesis and surface grafting of MNCs are discussed, followed by how these systems work to provide a variety of potential real-world applications and technologies. The main factor in mechanical strengthening is summarized in the following sections. The study also includes an in-depth analysis of MNCs with various reinforcements and the wealth of research on the tribological behavior of these composites. This study expands the conversation on the most exciting and developing area of systems, including therapeutic variations achieved by incorporating magnetic particles into nanocellulose. The overall applications of MNCs considering the current panorama include bone tissue engineering, wound dressing, medical implants, and medication delivery. Thus, the use of magnetic nanoparticles and their shortcomings and advancements are also highlighted, with an emphasis on the toxicity or biocompatibility of these structures. This focus of this review is bridging the gap between theoretical and experimental contributions. The illustration of the essential progressive computational tool-based parameters, such as compliance constant (CC), relaxed force constant (RFC), binding energy (BE)/interaction energy (IE), QTAIM topological indices, hydrogen bond (HB) strength based on interaction coordinate (HBSBIC), and NCI-plot based on bonding/nonbonding interaction strength deliver the essence of *in silico* insights on MNCs. In the following sections, we offer a comprehensive overview of the diverse applications of the combination of magnetic particles and nanocellulose. Overall, this study is structured to provide readers with a comprehensive understanding and prevailing opportunities for MNCs to offer practical benefits to society and to dispense their potential for translational applications. Also, a discussion on the processes for the synthesis and functionalization of MNCs is presented in the subsequent sections.

## 2. Synthesis and functionalization of magnetic nanocellulose composites

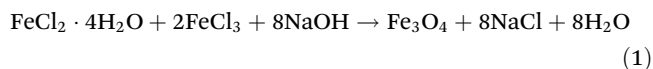
### 2.1. Synthesis of MNCs: lumen loading and *in situ* methods

Magnetic nanocellulose composites can be synthesized *via* lumen loading and *in situ* methods. In the lumen loading method, which is a physical approach, ferrimagnetic nanocel-

lulose composites are produced by introducing commercially available magnetic particles into the nanocellulose pulp.<sup>30,31</sup> The process of lumen loading takes place in three steps. Firstly, there is a brief phase, where the diffusion front advances and penetrates the lumen, leaving a minimal amount of filler particles deposited on the interior surface. The second step, a crucial stage of the lumen loading method, is known as the quasi-steady state regime. In this stage, the content of magnetic particles dispersed in the lumen is approximately constant and the number of magnetic particles entering the lumen matches the amount being added to the wall of the lumen. The diffusion kinetics of this method are primarily restricted by the passage of the filler particles through the pit gaps of the fiber wall.<sup>32</sup> It frequently produces a heterogeneous composite with toxic particle aggregation and dispersion, leading to reduced activity. Alternatively, more uniform and homogenous magnetic nanocellulose composites can be produced by concurrently co-depositing the matrix and magnetic particles using a premixed suspension. One typical process involves the use of a specific retention agent to help integrate the as-synthesized magnetic nanoparticles into the lumens of decomposing nanocellulose.<sup>33</sup> The filler particles are only injected into the fiber lumen after impregnation, which is followed by an agitation and cleaning process to remove any undesired particles. The exterior surfaces remain filler-free. The cell wall prevents the filler from becoming dislodged, and the bonds between the interfiber are not disturbed by particles during the process. Furthermore, the resulting magnetic nanocellulose composite material exhibits significantly higher coercivity and saturation magnetization. Nonetheless, the short-range magnetic dipole effect is most likely the reason for the spatial aggregation of the particles. This exemplifies a phenomenon frequently encountered by particle nanocomposites. Furthermore, in comparison to the host matrix, this shape produces a brittle material and reduces the mechanical qualities such as tensile strength. Surface coating has been used to execute a modified approach to overcome these issues. In this procedure, cellulose fibers are first disassembled and aggressively agitated in a colloidal solution of magnetic nanoparticles. However, particles show persistent bonding with the surface of the fibers even after repeated washing and sonication. One vital discovery is the formation of a novel bonding phase of  $\alpha$ -FeOOH at the interface of cellulose fibers and  $\text{Fe}_3\text{O}_4$  particles. The integrity of heterogenous composite materials throughout processing and real-world applications depends on the creation of this bridge. Meanwhile, it endows the matrix with magnetic characteristics, while preserving the intrinsic qualities of the fiber, such as its tensile strength and flexibility. Furthermore, the nanoparticles are fully and evenly embedded in the surface of the fibers.<sup>34</sup>

Another method for the synthesis of magnetic nanocellulose composites is the *in situ* co-precipitation approach. In this process, cellulose pulp is vigorously agitated in an aqueous solution of iron ions, which changes the iron ions to iron oxide in the nanocellulose matrix by the mixing of an excess

alkaline solution of NaOH. The reaction associated with this procedure is as follows:



Compared to the lumen-loading technique, this approach allows greater control of the magnetic characteristics and diversity of magnetic particles included in the final product.<sup>35</sup> Magnetic cellulose materials are produced using this approach on a large scale. The *in situ* co-precipitation technique was modified by Katepetch *et al.*<sup>36</sup> This procedure involved immersing bacterial cellulose pellicles in a suspension of  $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$ , and then dipping them in a freshly prepared NaOH solution. Subsequently, a hydrogen peroxide solution was added to the mixture after heating the suspension to 65 °C in a water bath. Lastly, distilled water was used to wash the samples. By employing this technique, the specific reaction occurred stepwise inside the bacterial cellulose. However, this dipping process, which occurred stepwise, showed some drawbacks. The fabricated samples exhibited the agglomeration of encapsulated nanoparticles in the cross-sectional regions of the bacterial cellulose. The bulk development of the ferrites at the bacterial cellulose surface caused a darker covering at the surface. In addition, the dipping procedure was carried out under ambient conditions. Consequently, maghemite ( $\gamma\text{-Fe}_2\text{O}_3$ ) and hematite ( $\alpha\text{-Fe}_2\text{O}_3$ ) were formed fluently by the oxygen gas present in the atmosphere. Ammonia gas-enhancing in the co-precipitation technique functioned in a packed system without the presence of oxygen to create a non-accumulated, homogenous dispersion and regulate the crystalline structure of magnetic nanoparticles in the matrix of nanocellulose. The use of ammonia gas (instead of traditional aqueous basic solutions) in this process prevented the agglomeration of the particles, resulting in the uniform distribution of magnetic nanoparticles in the cellulose matrix. Consequently, the cellulose sheet with magnetic nanoparticles showed consistent magnetic characteristics across the cellulose matrix. At ambient temperature, the cellulose sheet integrated with magnetic nanoparticles exhibited a saturation magnetization in the range of 1.92 to 26.20 emu g<sup>-1</sup>, accompanied by a reduction in the magnetic remanence to 0.15–2.67 emu g<sup>-1</sup> and magnetic coercivity to 40–65 G.

## 2.2. Functionalization of magnetic nanocellulose composites

Typically, the magnetic constituents within MNCs possess superparamagnetic behavior due to their small radius, extensive specific surface area, and abundant porosity, leading to a pronounced propensity for agglomeration.<sup>37,38</sup> This agglomeration causes MNCs to exhibit low structural durability, inconsistent physical stability, and inadequate interfacial adhesion. Thus, considering the above-mentioned challenges, developing suitable strategies to augment the mechanical robustness, dispersion capability, and compatibility of MNCs with various matrices has become crucial. Among the different strategies, surface functionalization has emerged as the predominant

methodology to enhance the intrinsic properties of MNCs together with preserving their physicochemical integrity, quality, and environmental sustainability.<sup>39,40</sup> This approach has demonstrated considerable efficacy in customizing the properties of materials for further expanding their range of applications.<sup>41</sup>

Structurally, each monomer unit of cellulose is composed of an anhydro glucose unit (AGU) featuring three hydroxyl groups, providing a foundation for a variety of chemical surface modifications aimed at enhancing its characteristics and selectivity depending on its applications.<sup>42</sup> The intrinsic polarity of MNCs, which is attributed to the hydroxyl groups of cellulose, results in poor compatibility with non-polar entities, confining their suitability to polar solvents and resins.<sup>43</sup> Accordingly, their chemical modification techniques involve substituting these hydroxyl moieties, thereby reducing the fibril entanglement. Studies have indicated that the reactivity of the free hydroxyl groups in cellulose follows the order of C-6 > C-2 > C-3.<sup>44</sup> Although nucleophilic reactions can occur at the secondary hydroxyl groups located at the C-2 and C-3 positions, the primary hydroxyl group at C-6, due to its absence of steric hindrance, presents the most favorable site for nucleophilic substitution reactions following the S<sub>N</sub><sup>2</sup> mechanism. This modification can be executed in both homogeneous and heterogeneous phases; however, reactions in the heterogeneous phase are predominantly surface-oriented due to the limited dispersibility. The key methods for chemical modification include oxidation, esterification, silanization, amination, acetylation, and phosphorylation. A detailed discussion on the different routes for surface functionalization is presented in the following sections.

**2.2.1. Surface functionalization by oxidation.** Oxidation processes can be applied to MNCs to incorporate functionalities such as carboxylic acid and aldehyde groups.<sup>45</sup> The two predominant approaches for the chemical modification of MNCs involve nitroxyl-mediated oxidation to selectively yield carboxylic acids at the primary alcohol groups and periodate oxidation to generate aldehydes from vicinal diols.<sup>46,47</sup> Oxidation using the 2,2,6,6-tetramethylpiperidinyloxy (TEMPO) radical has gained prominence for both the surface and bulk modification of MNCs.<sup>48</sup> Typically, this process employs a catalytic quantity of TEMPO in conjunction with a secondary oxidant, such as sodium hypochlorite and sodium chlorite, to regenerate the TEMPO radical at pH 10–11. Frequently, sodium bromide is utilized to enhance the oxidation rate by the *in situ* generation of sodium hypobromite. A novel advancement in the modification of nanocellulose through TEMPO-mediated oxidation involves the application of ultrasonic treatment. This technique preferentially eliminates the amorphous regions of cellulose, while conserving its crystalline domains, resulting in an increase in the density and enhancement in the tensile strength of MNCs.<sup>49</sup> Additionally, periodate-based oxidation represents another method, employing aldehydes to cleave the bonds between adjacent carbon atoms. In this reaction, sodium periodate ( $\text{NaIO}_4$ ) and potassium periodate ( $\text{KIO}_4$ ) serve as oxidizing

agents to specifically target the secondary hydroxyl groups, thereby breaking the chemical linkage between the C2 and C3 positions, and consequently generating two aldehyde groups.<sup>50</sup>

**2.2.2. Surface functionalization by esterification.** The hydrophobization of the surface of MNCs is usually accomplished through the well-known esterification process, which uses functionalities such as carboxylic acid, acyl chlorides and acid anhydrides as reacting agents.<sup>51</sup> The esterification of MNCs can be achieved through condensation reactions to link the hydroxyl groups present on MNCs with an ester functional group (O–C=O) through a biphasic process employing either a homogeneous or heterogeneous methodology. In the heterogeneous approach, the insoluble modified NC encapsulates the crystalline cellulose chains, which remain unreacted. Alternatively, in the case of the homogeneous methodology, the modification leads to the immediate dispersion and solubilization of MNCs. Acetylation, a specific form of esterification, introduces acetyl groups (CH<sub>3</sub>–C(=O)–) onto the surface of MNCs. This modification significantly enhances the thermal stability and mechanical properties of the material, additionally rendering it hydrophobic.<sup>52</sup> The acetylation process also enhances the dispersion of MNCs within other matrices, thereby facilitating improved material compounding. This method offers advantages such as cost efficiency, convenience, swift production times, and an environmentally benign profile.<sup>53</sup>

**2.2.3. Surface functionalization by silylation.** Silylation serves as a pivotal strategy for enhancing the interfacial adhesion between the NC matrix and magnetic fillers within composites. Utilizing silanes as coupling agents plays a crucial role in the stabilization of composite materials by minimizing the presence of cellulose hydroxyl groups. The chemical grafting initiates with the hydrolysable silyl groups reacting with moisture to generate silanols, which subsequently react with the hydroxyl groups present on the substrate. Additionally, these silanol groups are prone to self-condensation, leading to the creation of polysiloxanes.<sup>54</sup> The Si–OH and C–OH condensation culminates in durable surface grafting, solidified through a subsequent temperature curing process.

(3-Aminopropyl)trimethoxysilane (APMS) represents one of the most extensively researched aminoalkylsilanes, which is commonly grafted to cellulose derivatives for diverse applications.<sup>55</sup> Accordingly, extensive research has been conducted on the exploration of a variety of factors, such as the type of solvent used, the initial silane concentration, and pH levels, among others, to reduce self-condensation, while enhancing the efficiency of the grafting process.<sup>56,57</sup> These investigations aim to optimize the conditions under which silanol groups preferentially bond to the substrate hydroxyl groups over engaging in self-condensation, thereby improving the overall effectiveness and specificity of the chemical grafting procedure.

**2.2.4. Surface functionalization by amidation.** The amidation process has emerged as the most proficient method for the covalent modification of MNCs, exploiting the high reactivity between carboxyl and amino groups to produce a durable

amide linkage. Generally, amidation modification is preceded by TEMPO/periodate oxidation of the surface of MNCs. A widely used protocol for the amidation of MNCs is the process reported by Bulpitt and Aeschlimann,<sup>58</sup> which employs *N*-(3-dimethylaminopropyl)-*N'*-ethylcarbodiimide hydrochloride (EDC) together with *N*-hydroxysuccinimide (NHS). Here, EDC aids in the activation of the carboxylic acid groups, converting them into an *O*-acylisourea intermediate or a carboxylic ester, which is inherently unstable and susceptible to hydrolysis. The incorporation of NHS results in the formation of a more stable NHS-ester, significantly enhancing the resistance to hydrolysis. Upon introducing the amine, the amidation reaction progresses with the reduced formation of by-products, which is attributed to efficiency of NHS as a leaving group.

Amidation modification is instrumental in widening the potential application scope of MNCs given that this modification allows the introduction of various biochemical and biophysical properties tailored to specific applications, including drug delivery systems,<sup>59</sup> biocompatible materials,<sup>60</sup> and environmental sensors.<sup>61</sup> The stability of the amide bond under physiological conditions makes it particularly suitable for biomedical applications, where controlled release and biocompatibility are crucial. This strategic approach for the modification of MNCs opens avenues for creating highly specialized materials designed for advanced applications.

**2.2.5. Surface modification by phosphorylation.** Phosphorylation stands out as one of the most efficacious pre-treatments developed to produce greener MNCs with enhanced quality, low energy consumption, and new benign properties.<sup>62</sup> Phosphorylation represents a viable pathway that can be applied for the industrial-scale production of MNCs. This modification has been observed to maintain the crystallinity and degree of polymerization of NC. These promising outcomes have successfully transitioned to commercial application, as evidenced by the initial production disclosure in Japan. Phosphorylation expands the application scope of MNCs to orthopedics, biomedicine, environmental decontamination, and biochemical separation.<sup>63–65</sup> The phosphorus functionality can be grafted on MNCs through the homogeneous or heterogeneous methodology. In many reports, phosphorylation has been performed using an organic solvent such as pyridine<sup>66</sup> and dimethylformamide<sup>67</sup> using phosphate salt, phosphorous pentoxide, or phosphoric acid as phosphorylating agents. One of the classical pathways to introduce phosphoric functionalization in MNCs is the phosphorylation of NC within a water-based urea system utilizing phosphate salts.<sup>68,69</sup>

### 3. Recent experimental studies on magnetic nanocellulose composites

Researchers around the globe have devoted their efforts to exploring and tailoring the beneficial properties of magnetic nanocellulose composites. Amiri *et al.*<sup>70</sup> investigated the



potential of magnetic cellulose nanocrystals derived from cotton for the demulsification of water in crude oil emulsions. The authors evaluated the effect of temperature, time, and demulsifier concentration on the efficacy of demulsification. Their findings indicated that the evaluated demulsifier demonstrates high efficacy in facilitating both chemical and magnetic demulsification processes. Specifically, it achieved a demulsification efficiency (DE) of 100% at a temperature of 50 °C without the application of a magnetic field and 90% at 20 °C when a magnet was employed. Moreover, this study explored the DE of Fe<sub>3</sub>O<sub>4</sub> nanoparticles in chemical demulsification, revealing that it is less effective compared to that of magnetic cellulose nanocrystals. The crystals were observed to be reusable up to 4 times. Liu *et al.*<sup>71</sup> synthesized magnetic nanocrystalline cellulose (MNCC) via the combination of Fe<sub>3</sub>O<sub>4</sub> and a nanocellulose support. MNCC was utilized to immobilize esterase from porcine liver (PLE) for the degradation of mycotoxins present in fruit juice. MNCC was obtained through a simple self-assembly methodology involving electrostatic coprecipitation. The properties and structure of PLE@MNCC were thoroughly characterized. Furthermore, the detoxification process of patulin present in apple juice was thoroughly explored and optimized by cytotoxicity tests and quality evaluation.

Hegazy *et al.*<sup>72</sup> developed a cost-effective and efficient methodology for the fabrication of a magnetic Fe<sub>3</sub>O<sub>4</sub>-cellulose-graphene oxide nanocomposite through free radical graft copolymerization. Distinct grafting parameters including reaction time, temperature, and molar ratio were optimized. The authors carried out a comprehensive analysis of the morphological and structural characteristics of the nanocomposite. The adsorption efficiency of methylene blue (MB) dye on the nanocomposite was assessed under varying conditions, with the optimal adsorption observed at pH 7, after 7 h of contact, and using an adsorbent dose of 0.0375 g in a 30 mg L<sup>-1</sup> dye solution. The integration of magnetite and graphene oxide into the cellulose significantly improved the dye removal, achieving a 97% removal rate using the nanocomposite. Shaltout *et al.*<sup>73</sup> proposed a strategy to formulate an effective adsorbent material by combining nanocellulose, nanohydroxyapatite, and ferric chloride using natural sources such as cotton stalks and scallop shells. The magnetic nanocomposite exhibited a remarkable removal efficiency of 239.23 mg g<sup>-1</sup> for Safranin-O dye. The adsorption of Safranin-O has been demonstrated through thermodynamic and kinetic analyses to be advantageous, spontaneous, endothermic in nature, and governed by physisorption mechanisms.

In another study, Chacko *et al.*<sup>74</sup> presented a novel ternary nanocomposite fiber system, integrating poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP), microcrystalline cellulose (MCC), and LiFe<sub>5</sub>O<sub>8</sub> (LFO) nanoparticles, demonstrating magnetoelectric coupling and antibacterial capabilities. This offered a promising approach for developing field-responsive antibacterial coatings. The inclusion of 8 wt% LFO enhanced the electroactive phase to 95%, significantly improving both the dielectric and ferroelectric properties, with the maximum

magnetoelectric coupling coefficient (MECC) of 20.3 mV cm<sup>-1</sup> Oe. The antibacterial efficacy of the nanocomposite, as validated through the minimal inhibitory concentration (MIC) method, underscores its potential for advanced antibacterial surface applications. Top of Form Tang *et al.*<sup>75</sup> prepared a magnetic nanocellulose-based multifunctional aerogel with enhanced mechanical properties and improved oil-water separation capability. The aerogel consisted of an amalgamation of polyethyleneimine, magnetic exfoliated bentonite, and carboxy cellulose nanofibers prepared using chemical vapour deposition and hydrothermal methodologies. The findings indicated that the fabricated aerogel demonstrated outstanding superhydrophobicity (with a contact angle of 151°), superior mechanical resilience (fully recovered at 80% compression), high anti-mold effectiveness (exceeding 90% efficiency), and magnetic responsiveness.

Sun *et al.*<sup>76</sup> fabricated a similar composite from polyethyleneimine (PEI), magnetic bentonite (MB), and TEMPO-oxidized nanocellulose (TOCN) and utilized it as a biosorbent (PNMBC) for the effective removal of Cu(II) from water. The adsorptive removal of Cu(II) reached 757.45 mg g<sup>-1</sup> within 10 min of contact between the adsorbate and adsorbent. The synergistic interactions between the different constituents of adsorbent are depicted in Fig. 4. It was concluded from this study that the adsorptive process was mainly controlled by chemisorption, electrostatic forces, and functional group chelation mechanisms. Recently, Naznin *et al.*<sup>77</sup> developed a magneto-responsive composite using magnetic iron oxide nanoparticles (MIO) and cellulose sourced from sugarcane bagasse (SCB) and waste tissue paper (WTP). The obtained nanocomposite particles (NCPs) were characterized by advanced analytical techniques and investigated for their anti-oxidative properties and applicability in metronidazole drug delivery. Overall, it was concluded that the combination of MIO-NPs in the cellulose matrix enhanced the drug loading capacity, swelling capacity, and drug-releasing time.

Janudin *et al.*<sup>78</sup> attempted to combine nanocellulose with nickel ferrite nanoparticles in different amounts (0.1 g–0.5 g) via an *in situ* synthesis technique. The composite could accurately monitor chlorine gas. Various regulating factors such as gas concentration, reusability, repeatability, and nickel ferrite content were investigated to achieve the optimum gas sensing conditions. The responses of the discrete quantities of nickel ferrite loaded on the nanocellulose to 1% chlorine gas are shown in Fig. 5. It was found that 0.3 g nickel ferrite demonstrated a superior response and sensitivity in comparison to other samples with a response time of 40 s and recovery time of 50 s.

T. da Rosa Salles *et al.*<sup>79</sup> prepared magnetic nanocrystalline cellulose using Fe<sup>2+</sup> as the iron source under mild conditions, facilitating the precise regulation of Fe<sub>3</sub>O<sub>4</sub> integration. The magnetic composite possessed remarkable adsorption characteristics for removing azithromycin from water. The adsorption data indicated that the peak adsorption of azithromycin occurred at a pH of 3 when using an NC:Fe<sub>3</sub>O<sub>4</sub> ratio of 1:10. In addition, the *in vitro* biological tests proved that the compo-



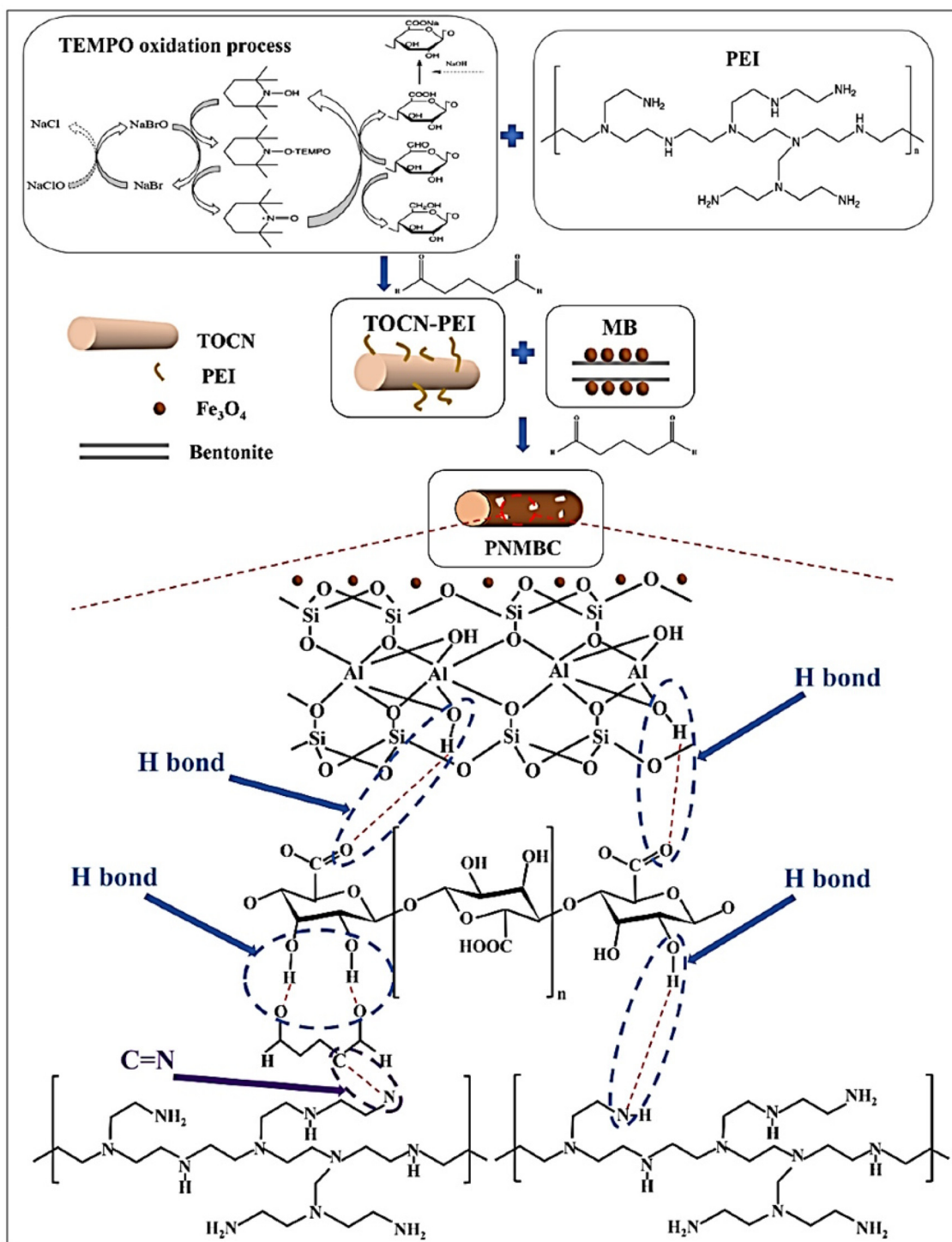
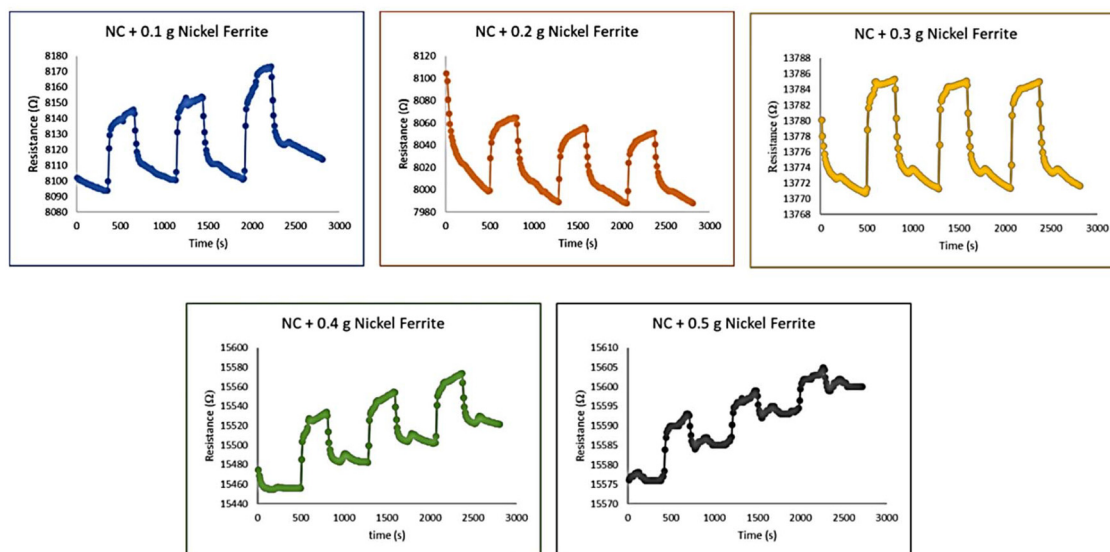


Fig. 4 Schematic of the interactions in PNMBC biosorbent. Reproduced with permission from ref. 76. Copyright 2022, MDPI.

site exhibited excellent biological activity against melanoma cells. Markovic *et al.*<sup>80</sup> fabricated magneto-responsive hybrid hydrogels made from poly(methacrylic acid) and nanocellulose derived from *Eucalyptus* wood. The magnetic and pH sensitivity of the hydrogels enabled precise targeting and regulated release of ibuprofen. It was observed that as the weight per-

centage of nanocellulose (NC) increased, the hybrid hydrogels swelled less and released less ibuprofen over time.

Bakr *et al.*<sup>81</sup> performed a comparative study on the co-delivery of curcumin (CUR) and 5-fluorouracil (FU) using two distinct morphological structures based on nanocellulose (NC), polylactic acid (PLA), and magnetite (M). The two formu-



**Fig. 5** Sensing response of nickel ferrite-loaded nanocellulose composites to 1% chlorine gas. Reproduced with permission from ref. 78. Copyright 2022, MDPI.

**Table 1** Applications of magnetic nanocellulose-based functional materials

| S. no. | Material                                                                                                | Application                                                     | Publication year | Ref. |
|--------|---------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------|------------------|------|
| 1      | Fe <sub>3</sub> O <sub>4</sub> @nanocellulose                                                           | Magnetic transparent composite film                             | 2021             | 82   |
| 2      | Polycitric acid@Fe <sub>3</sub> O <sub>4</sub> @nanocellulose                                           | Contrast agent for magnetic resonance imaging                   | 2019             | 83   |
| 3      | Fe <sub>3</sub> O <sub>4</sub> @grass nanocellulose from <i>Cyperus rotundas</i>                        | Adsorption of rare earth element cerium(III)                    | 2022             | 84   |
| 4      | Magnetic nanocellulose fibers@polydopamine@Ag nanocatalyst                                              | Degradation of organic pollutants and bacterial disinfection    | 2022             | 85   |
| 5      | Thiourea-functionalized magnetic ZnO/nanocellulose composite                                            | Adsorptive elimination of Pb(II) ions                           | 2020             | 86   |
| 6      | Bacterial cellulose@Fe <sub>3</sub> O <sub>4</sub> @hydroxyapatite                                      | Bone tissue engineering                                         | 2019             | 87   |
| 7      | Oleic acid@Fe <sub>3</sub> O <sub>4</sub> @nanocellulose aerogel                                        | Oil adsorption                                                  | 2020             | 88   |
| 8      | Magnetic nanocellulose@polyethersulfone@ <i>Aspergillus oryzae</i> lipase                               | Catalyst for the synthesis of ethyl valerate                    | 2021             | 89   |
| 9      | Palladium nanoparticles@Fe <sub>3</sub> O <sub>4</sub> @nanocellulose                                   | Reduction of Cr(VI) and organic dyes                            | 2020             | 90   |
| 10     | Fe <sub>3</sub> O <sub>4</sub> @2,3-dialdehyde nanocrystalline cellulose                                | Preconcentration of glycoproteins                               | 2020             | 91   |
| 11     | Papain@magnetic nanocrystalline cellulose                                                               | Biocatalyst for the synthesis of dipeptides                     | 2020             | 92   |
| 12     | Fe <sub>3</sub> O <sub>4</sub> @bacterial nanocellulose crystals                                        | Flexible transparent films for fast actuating optical materials | 2022             | 93   |
| 13     | Magnetic cellulose nanobiocomposites obtained from Fe <sub>3</sub> O <sub>4</sub> and sugarcane bagasse | Adsorptive removal of Cd(II), Cu(II) and Pb(II)                 | 2021             | 94   |
| 14     | Nanocellulose Iron Oxide Nanobiocomposites (NIONS)                                                      | Adsorptive remediation of Arsenic from wastewater               | 2020             | 95   |
| 15     | Nano-γ-Fe <sub>2</sub> O <sub>3</sub> @cellulose dialdehyde@urease                                      | Bio-catalytic activity for urea hydrolysis                      | 2020             | 96   |

lations, *i.e.* PLA/M-NC and M-PLA-co-NC, were obtained by the blending technique and grafting copolymer self-assembly, respectively. Both morphological forms were almost equally efficient in loading 5-FU, while in the case of CUR, the M-PLA-co-NC nanocarrier could hold twice that of the PLA/M-NC carrier. This shows that the copolymeric micelle was superior in comparison to the blended formulation. The aforementioned reports clearly highlight the potential of MNCs for advanced applications. A few more recent experimental studies based on utilizing MNCs for distinct applications are presented in Table 1.

## 4. Theoretical studies on magnetic nanocellulose composites

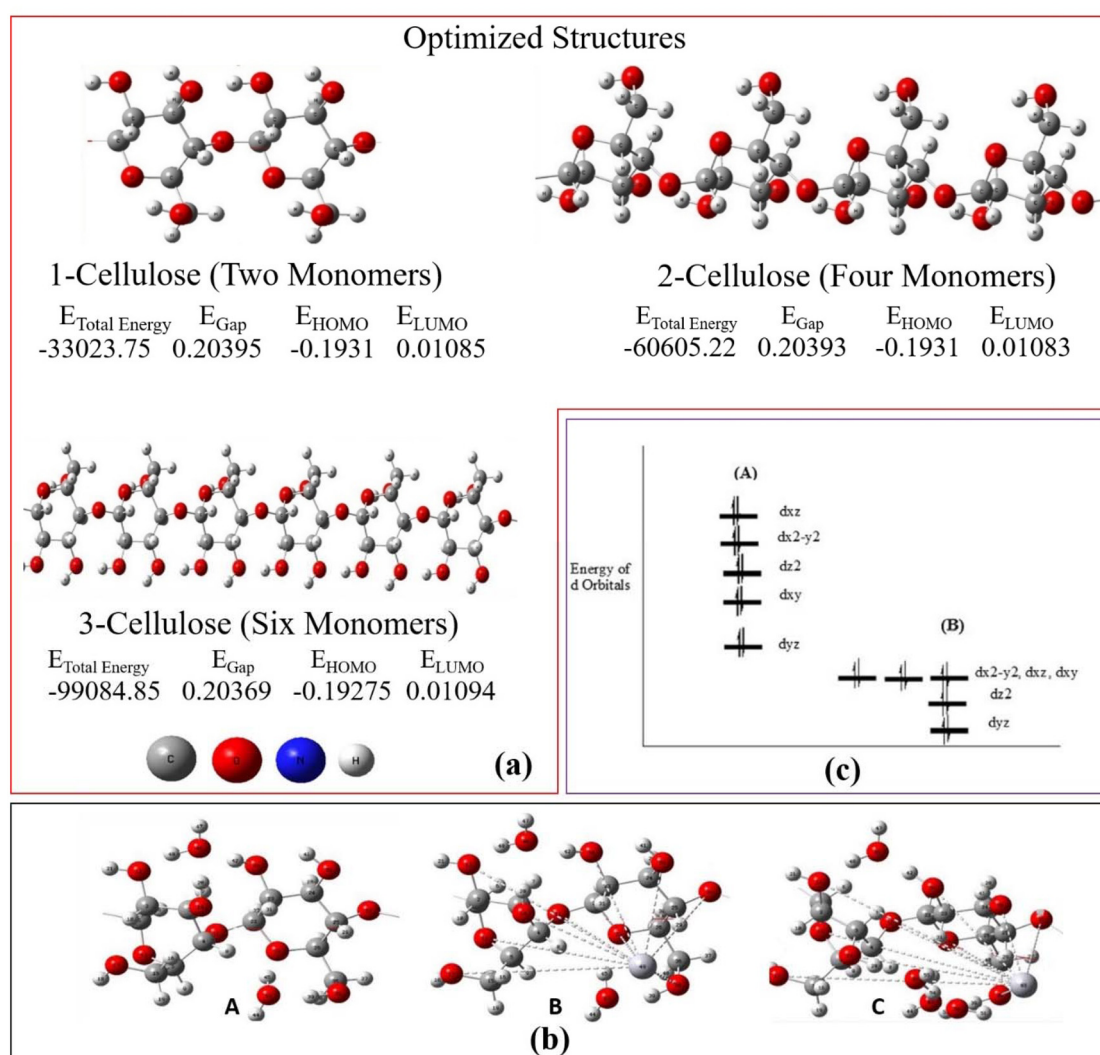
To date, although a range of research works on the synthesis and characterization of magnetic nanocellulose has been reported in the literature, and there is still room for *in silico* studies ranging from molecular mechanics (MM) to MM/quantum mechanical (QM) and molecular dynamics (MD) simulations. This section provides glimpses into the theory-based analyses, portraying the scarce interesting works (rarely

reported in the literature) on the structural characterization or removal of mercury ion pollution based on the metal oxide  $\text{Fe}_3\text{O}_4$ -nanocellulose biocomposite model using the density functional theory (DFT) approach.

Zarei *et al.*<sup>97</sup> reported research on the removal of mercury ion pollution using an  $\text{Fe}_3\text{O}_4$ -nanocellulose including its synthesis, characterization, and DFT studies. In inspecting the essential aspects to biochemists, the application of *in silico* studies has been enabled by the successive advances in computational skills. Moreover, these advancements in theory-based skills have been widely accepted to be truthful by appropriate evidence acquired from diverse experiments. For instance, structural (optimization and frequency calculations, *i.e.* minimum energy or equilibrium structure of any species), stability (total energy, binding energy, interaction energy, *etc.*),

and electronic feature (HOMO, LUMO, HOMO-LUMO gap, HOMO-LUMO three-dimensional isosurface maps, charge transfer phenomena, molecular electrostatic potential plots, dipole moment, *etc.*) analyses are imperative and noteworthy parameters obtained from the DFT approach. In this research, initially the researchers modelled some nanocellulose having three different (2, 4, and 6) numbers of monomer units, and the optimized structures are presented in Fig. 6(a). The authors found that the synthesized nanocellulose having longer chains is more stable ( $6 > 4 > 2$ ) using the total energy (*E*) parameter.

In this study, the researchers concluded that due to the high HOMO energy, high stability with longer chains, and high flexural and mechanical strength of the cellulose and the high abundant matter in the plants than the other polymers



**Fig. 6** (a) Molecular structures and electronic feature-based energies of (1) cellulose (having 2 monomer units), (2) cellulose (having 4 monomer units), and (3) cellulose (consisting of 6 monomer units), inspected using the Gaussian electronic structure calculations package. (b) Equilibrium structure of nanocellulose as an external part of the  $\text{Fe}_3\text{O}_4$ -nanocellulose biocomposite in the presence of water ( $\text{H}_2\text{O}$ ) and mercury ions as  $\text{Hg}(\text{II})$ . (c) Orbital energy levels of the mercury cation,  $\text{Hg}(\text{II})$  d-orbitals in higher (A) and lower (B) concentrations probed using the DFT level of approach. Reproduced with permission from ref. 97. Copyright 2018, Elsevier.



used in this work, nanocellulose has been recognised as a suitable absorbent, attracting wide attention from the research community.

Moreover, the optimized structure of the external part of the nanobiocomposite ( $\text{Fe}_3\text{O}_4$ -nanocellulose) in the presence of water and  $\text{Hg}(\text{II})$  can be seen in Fig. 6(b), where the analysis of the minor energies resulted from nanocellulose having two monomer units and the implementation of the DFT approach. It should be noted that Fig. 6(b) (B) shows that the reaction complex (taking part in the reactions) is related to the lower concentration of water molecules (2 water molecules), whereas the reaction complex (engaged in reactions) corresponding to Fig. 6(b) (C) ensues in the presence of a smaller number of adsorbents, which is because of the existence of large number of water molecules (4 water molecules). A few chosen optimized parameters (bond lengths and bond angles) of the reaction between  $\text{Fe}_3\text{O}_4$  nanocellulose and  $\text{Hg}(\text{II})$  are discussed, where Fig. 6(b) (B and C) portrays the uptake of the  $\text{Hg}(\text{II})$  *via* ion-exchange and adsorption on the surface. The above-mentioned simulation-based results (bond lengths and bond angles) related to the ion exchange and adsorption are consistent with the outcomes of the Langmuir isotherm model.

Importantly, deep insight into the bond formation taking part between the heavy metal ( $\text{Hg}$ ) and adsorbent (ligand) has been gained, where the energies of the d-orbitals for a smaller concentration ( $d_{x^2-y^2} = d_{xy} = d_{xz} > d_{z^2} > d_{yz}$ ) were detected to be lower than that of a larger concentration ( $d_{xz} = d_{x^2-y^2} = d_{z^2} > d_{xy} > d_{yz}$ ) (see Fig. 6(c)). According to this analysis, the complex does not show a definite shape, where the complex system is formed between the  $\text{Hg}(\text{II})$  cation and metal oxide associated nanocellulose with a larger concentration. This indicated that the shape of the resulting disorder complex form can be controlled by the adsorbent concentration.

Pavlović and co-workers<sup>98</sup> reported a thorough study on the structural characterization of nanocellulose/ $\text{Fe}_3\text{O}_4$  hybrid nanomaterials, in which they also studied the magnetic interactions and magnetic couplings between the Fe atoms in the magnetic structure within the probed nanocomposite materials. In this work, they employed a theoretical approach (DFT-broken symmetry) using the ORCA program package in the framework of the B3LYP hybrid functional and TZP basis set.

Moreover, the OPBE function was used to calculate the coupling constants. In analysing the exchange coupling constants between  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  ions, the first coordination sphere located near the Fe atoms was chosen directly from the crystal structures of the  $\text{Fe}_3\text{O}_4$  model system, as can be seen in Fig. 7. It was found that the exchange coupling constants values were somewhat smaller at the B3LYP level of theory compared to the OPBE approach.

The outcomes of the coordination environment near the metal ions and electronic structure are represented by the magnetic couplings between the paramagnetic centers. It can be seen that the tetrahedral coordination having a high-spin  $e(2)t_2(3)$  electronic configuration is occupied by half of the  $\text{Fe}^{3+}$  ions in the case of the inverse spinel, whereas the octa-

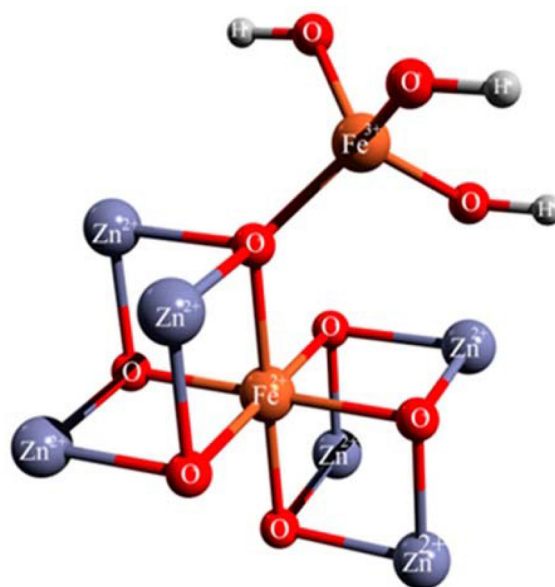


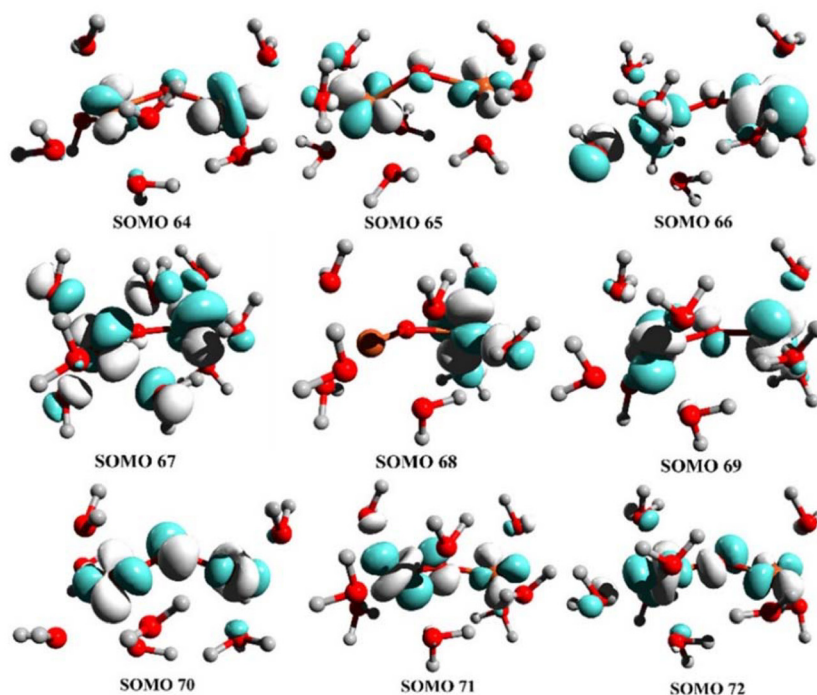
Fig. 7 Structure of  $\text{Fe}_3\text{O}_4$  model system. Reproduced with permission from ref. 98. Copyright 2022, MDPI.

hedral voids consisting of high-spin  $t_{2g}(4)e_g(2)$  electronic configuration are engaged by  $\text{Fe}^{2+}$ . The total interactions between the  $\text{Fe}^{2+}$  ions with octahedral coordination and  $\text{Fe}^{3+}$  with a tetrahedral environment were portrayed by the analysis of molecular orbitals having a predominant metal character, as displayed in Fig. 8 and 9. According to the above-displayed SOMO diagrams, ferromagnetic couplings can be observed, where the probable antiferromagnetic interactions are indicated by the orbital analysis through a super-exchange procedure represented by bridging the oxygen ligand. Additionally, it was shown that the electron density was spread over both metal centers. The magnetic interactions between the paramagnetic centers were explained by the structural or geometrical parameters such as bond distances and bond angles given that they are strongly influenced by such parameters.

## 5. Translational applications of magnetic nanocellulose composites

Because of their special qualities, MNCs are good options for medicinal and environmental applications. Because nanocellulose has natural qualities including low toxicity, biocompatibility, biodegradability, and easy availability, it is a good option for drug delivery, wound healing, biosensing, biolabeling, water purification, fuel cells and adsorbent systems.

MNCs have attracted immense interest for diverse applications pertinent to the biomedical field and material engineering due to their high mechanical strength, biodegradable nature, anisotropic shape, impressive optical properties, excellent biocompatibility, and tunable surface chemistry. However,



**Fig. 8** Singly occupied molecular orbitals (SOMO) in a high-spin state consisting of  $S = 9/2$ . A simpler structure is displayed, in which the oxygen charge (negative) is neutralized by hydrogen atoms (positive). Reproduced with permission from ref. 98. Copyright 2022, MDPI.

interdisciplinary research is fundamental to commercializing MNC-based products for sustainable applications. Economically, the production of MNCs is highly viable due to the low cost and abundance of cellulose sources such as wood pulp, which can be sustainably harvested.<sup>99</sup> The cost of producing nanocellulose is significantly lower than many other nanomaterials such as graphene, graphene oxide, carbon nanotubes, and silicon nanowires. Magnetic nanoparticles can be synthesized and embedded in a nanocellulose matrix using scalable, cost-effective chemical methods. The overall cost competitiveness of MNCs is enhanced by their biocompatibility and functional versatility. The scalability of MNC production is supported by advancements in nanotechnology, allowing the large-scale manufacturing necessary to meet industrial and medical demands. Current pilot-scale production facilities can produce up to several tons of nanocellulose per year, with estimates implying potential scalability to thousands of tons per annum within the next decade. Furthermore, MNCs can be integrated into existing manufacturing infrastructures with minimal modifications, reducing the need for significant capital investment in new equipment and facilities. This ease of integration is due to the compatibility of nanocellulose processing techniques with conventional chemical engineering processes, facilitating their seamless adoption in various industries.

In a specific case study on nanocellulose, a techno-economic analysis was performed by Kargupta *et al.*,<sup>100</sup> wherein the production of nanocellulose was explored through a combination of moderate refining and froth flotation methodologies

with the aim to reduce the traditional production cost. The most sustainable and affordable pathway with the best financial profitability was achieved at the pilot scale (best-case scenario) with 100% yield. The annual production cost of nanocellulose was approximated to be \$7312 AUD per ton and the 25 year NPV turnout was approximately 9.5 million AUD (profit) for the best-case scenario. The major applications of MNCs based on their clinical and environmental utilization are discussed in the subsections of the current section.

### 5.1. Emerging biological applications of MNCs

The hunt for efficient biomaterials that can aid in the renaissance of damaged living tissues is ongoing. A research group devised a novel method for biomedical research that allows for more regulated nanostructured morphology and geometric form. In their investigation,<sup>101</sup> cellulose drops were used to create regenerated cellulose microspheres (RCS) *via* the sol-gel technique. The drops were placed in a solution (precooled) of urea and NaOH. Then, using these porous beads as template microreactors,  $\text{Fe}_3\text{O}_4$  nanoparticles could precipitate *in situ* into the RCS (in the cellulose pores) in a solution containing  $\text{FeCl}_4$  and  $\text{FeCl}_2$ , resulting in the formation of magnetic  $\text{Fe}_3\text{O}_4$ /cellulose microspheres (MRCS). Using these nanocomposite beads as an example, the scientists were able to effectively show magnetic-induced transcription for better-targeted protein delivery and release. An intriguing advancement in cellulose nanofibril template materials may allow for greater control of the characteristics of nanocomposites and lead to the creation of several new multifunctional applications.<sup>102</sup>

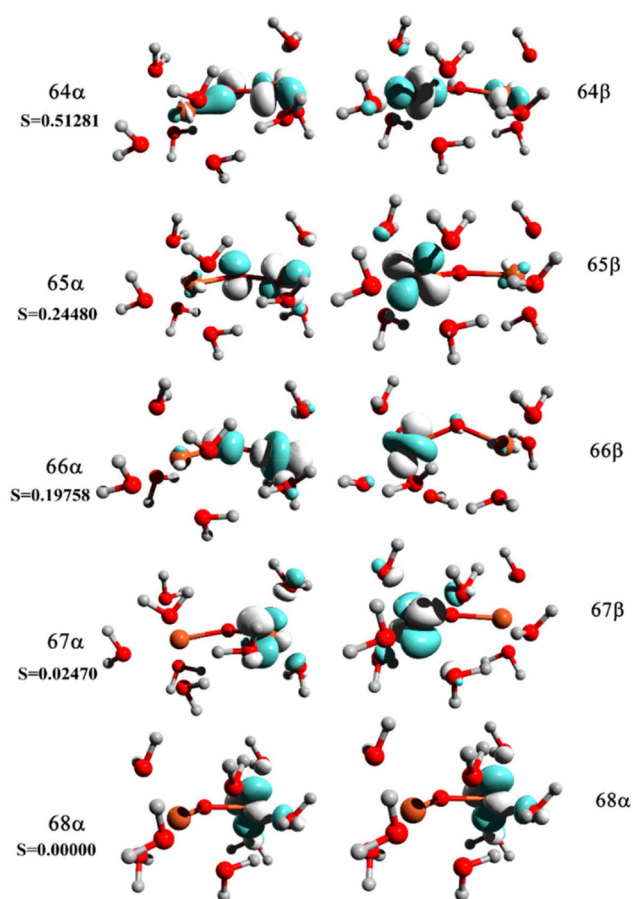


Fig. 9 Corresponding magnetic orbitals and spatial overlap integrals. S. Reproduced with permission from ref. 98. Copyright 2022, MDPI.

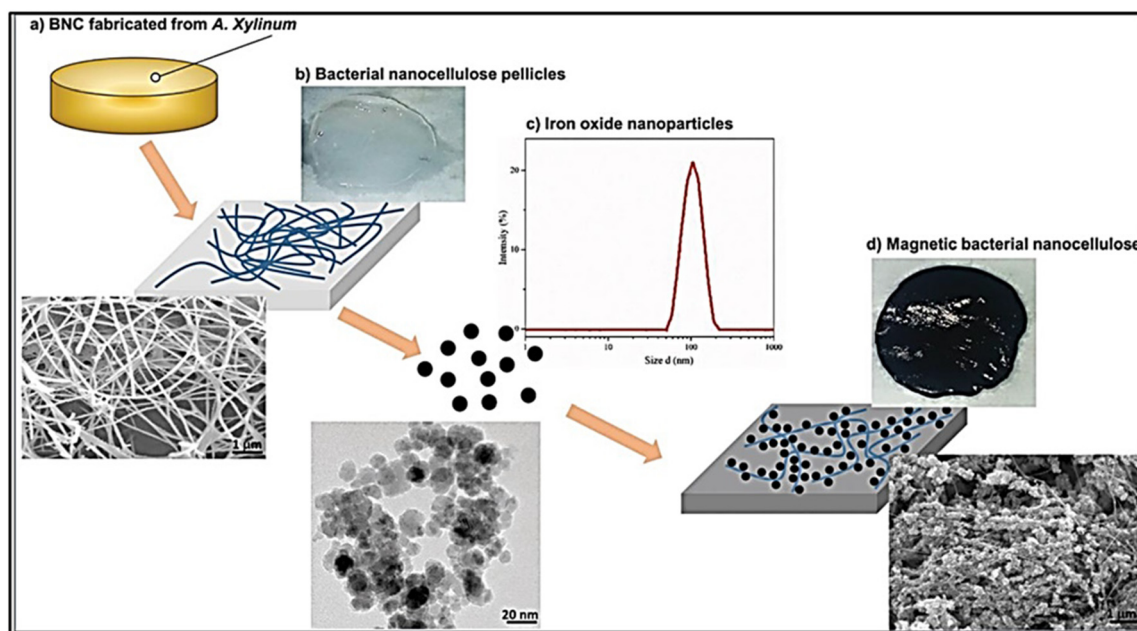
Researchers developed a straightforward but effective method to employ nanofiber scaffolds as a template for *in situ* chemical processes, resulting in the formation of Co nanoparticles. The authors employed a freeze-dried bacterial cellulose nanofiber to produce a multifunctional nanocomposite, which could be adjusted. This method made it possible to create magnetic hydrogels that are very porous, flexible, and magnetic field-responsive. Furthermore, another form of aerogel, which is extremely stiff with a high magnetic nanoparticle content, nanopaper, could be easily generated by pressing it under pressure. These early achievements with adaptable functions indicated a wide range of possible uses, from innovative microfluidics devices and electronic actuators to security documentation.

Pastrana *et al.*<sup>103</sup> studied MNCs for tissue engineering applications and synthesized BNC nanofibers reinforced with magnetite ( $\text{Fe}_3\text{O}_4$ ). The unique nanoporous network of bacterial nanocellulose (BNC), together with its mechanical properties, biocompatible nature, and high-water retention makes it a highly desirable biomaterial. Consequently, BNC has been extensively used in tissue engineering applications as a scaffolding biomaterial. The  $\text{Fe}_3\text{O}_4$ @BNC matrix was designed to attract certain cells and direct them towards targeted areas

to repair damaged tissue. Although the pore areas of the  $\text{Fe}_3\text{O}_4$ @BNC matrix fibres were 25% larger than that of BNC fibres alone (pore size of BNC:  $121 \text{ m}^2$  and  $\text{Fe}_3\text{O}_4$ @BNC:  $161 \text{ m}^2$ ), the diameter of the matrix fibres was 33% smaller. Additionally, the cytotoxicity examination demonstrated 96% survivability of swine aortic smooth muscle cells related to the BNC,  $\text{Fe}_3\text{O}_4$ @BNC, and  $\text{Fe}_3\text{O}_4$ @BNC coated with poly(ethylene glycol) (PEG) and 9% ROS generation for PEG- $\text{Fe}_3\text{O}_4$ @BNC. In contrast, even when cells were metabolically active, 25% of them underwent apoptosis because of the harmful effects of  $\text{Fe}_3\text{O}_4$ @BNC. MNCs have also shown promise in treating diseases involving the brain. Aneurysms are collections of blood pockets at the outer side of the walls of the blood vessel, which make the wall weaker (hematomas are blood pools beneath the skin). In this instance, brain aneurysms are more serious, and the majority of the patients have increased risks, making the present therapies for them invasive, traumatic, and inappropriate. For the *in situ* resetting of the tunica media, scaffold stents need to provide a local and focused attraction force for cells. To achieve this, a biocompatible nanocoating is utilized to provide a non-symmetric domain for the magnetic and bio-mimetic stent scaffold. BNC was utilized as a platform for the attraction of both magnetic and biological cells, leading to their growth. To improve the biocompatibility of hydrogels, Echeverry-Rendon *et al.*<sup>104</sup> synthesized an *in situ* BNC hydrogel membrane soaked with superparamagnetic  $\text{Fe}_3\text{O}_4$  NPs ( $\text{Fe}_3\text{O}_4$ /BNC), and then covered it with PEG (Fig. 10). Using a stent scaffold, this multifunctional hydrogel was tested for neuroendovascular applications, which demonstrated low cytotoxicity and good vitality and movement of the desired cells (porcine aortic smooth muscle cells).

Numerous effective methods have been developed to treat and enhance the healing of chronic wounds, consequently having societal effects. This development uses a new tissue engineering technique called 3D cell cultured implants, which are made using undifferentiated cells embedded in pre-seeding drug-loaded scaffolds to produce *in situ* functional neotissue. Consequently, during the biosynthesis process, Galateanu *et al.*<sup>105</sup> directly dispersed different concentrations of  $\text{Fe}_3\text{O}_4$  NPs into a cellulose bacterial growth medium to create  $\text{Fe}_3\text{O}_4$ /BNC bio-nanocomposite membranes. The developed composite membranes demonstrated favourable cytocompatibility characteristics, including viability, proliferation, and cytotoxicity. Molecular biology has enabled a better understanding of gene interactions, the healing process, and wound healing. In this case,  $\text{Fe}_3\text{O}_4$ /BNC nanocomposite films were created by Moniri *et al.*<sup>106</sup> as a wound dressing, and following film treatment, they examined the physicochemical, cytotoxic, antimicrobial, and gene expression aspects of wound healing. In this work, *Aloe vera* extract was used to biosynthesize  $\text{Fe}_3\text{O}_4$  (15–30 nm) in freshly isolated BNC. The  $\text{Fe}_3\text{O}_4$ /BNC films demonstrated non-toxicity ( $\text{IC}$  of  $50.500 \mu\text{m mL}^{-1}$ ) and excellent antibacterial properties (range of  $6 \pm 0.2$  to  $13.40 \pm 0.10 \text{ mm}$ ) against *Pseudomonas aeruginosa*, *Staphylococcus aureus*, and *Staphylococcus epidermidis*, as well as exceptional efficiency for wound healing (after 48 h). Additionally, the





**Fig. 10** (a) BNC produced by the bacterium *A. xylinum*; (b) digital and SEM pictures of the BNC hydrogel membrane; (c) transmission electron microscopy and dynamic light scattering analysis of the Fe<sub>3</sub>O<sub>4</sub> NPs; and (d) comprehensive SEM evaluation of the Fe<sub>3</sub>O<sub>4</sub>/BNC digital image, which shows the strong bonding of the Fe<sub>3</sub>O<sub>4</sub> NPs to the BNC fibres. Reproduced with permission from ref. 104. Copyright 2017, Wiley.

Fe<sub>3</sub>O<sub>4</sub>/BNC films demonstrated an impact on microRNA by reducing its expression, which led to the augmentation of the expression of other genes, ultimately resulting in wound healing. The aim of disease therapy and early diagnosis has led to the exploration of a wide range of materials. Regarding this, Torkashvand *et al.*<sup>107</sup> created a tiny superparamagnetic Fe<sub>3</sub>O<sub>4</sub> nanoparticle-based composite model for magnetic resonance imaging (MRI), which served as a T1–T2 twin positive and negative contrast agent. According to this study, Fe<sub>3</sub>O<sub>4</sub> NPs were created by a thermal breakdown procedure, and subsequently loaded into a CNC–poly(citric acid) (Fe<sub>3</sub>O<sub>4</sub>/CNCs–PCA) platform. The resulting composite showed an ultra-small hydrodynamic size, water dispersibility, hydrophilicity, colloidal stability, and high saturation magnetization (52.2 emu g<sup>−1</sup>). Fe<sub>3</sub>O<sub>4</sub>/CNCs–PCA also showed notable biocompatibility, low cytotoxicity *in vitro* (up to 200 μg mL<sup>−1</sup> Fe), and considerable cellular absorption in HeLa cells. Additionally, compared to various clinical and commercial agents, Fe<sub>3</sub>O<sub>4</sub>/CNCs–PCA demonstrated remarkable advancement in contrast imaging for both T1 and T2-weighted MR images (high values of r1: 13.8 mM<sup>−1</sup> s<sup>−1</sup> and r2: 96.2 mM<sup>−1</sup> s<sup>−1</sup>) relaxivity. To achieve the goal of using MNCs in biomedical applications, it is essential to assess their *in vivo* toxicity and biocompatibility with cells. Toxicology assessments indicate that MNCs generally exhibit low acute toxicity, with minimal cytotoxicity in various cell lines and negligible acute inflammatory responses *in vivo*.<sup>108</sup> The biocompatibility of MNCs is influenced by their particle size, surface charge, and surface functionalization. Long-term evaluations revealed that MNCs are largely biodegradable and do not significantly accumulate in major

organs, suggesting their minimal chronic toxicity. However, slight inflammatory responses have been observed, emphasizing the need for standardized long-term studies to fully elucidate biocompatibility and safety profiles of MNCs for clinical use.<sup>109</sup>

## 5.2. Potential environmental applications of MNCs

Cellulose in combination with magnetic nanoparticles can be utilized as an oil adsorbent.<sup>110</sup> Recently, magnetic hybrid materials were created using a mixture of cobalt–iron oxide particles and cellulose nanocrystals (CNCs).<sup>111</sup> According to Benmassaoud *et al.*,<sup>112</sup> iron-based nanoparticles have recently been used to remove heavy metals and organic dyes from wastewater. Iron-based nanoparticles have a large surface area, interfacial reactivity, and magnetic qualities, making them good adsorbents for hazardous substances found in wastewater. Jodeh *et al.*<sup>113</sup> discovered an innovative method for creating magnetic cellulose nanocrystals (MNCs) from olive industry solid waste (OISW), which could be used as methylene blue-selective magnetic sorbents. About 40% of the solid waste from the olive industry is cellulose. The solid waste from the olive industry was pulped and bleached in many steps to extract cellulose in powder form. After being removed, the cellulose powder was hydrolyzed with acid to create nanocrystalline cellulose (NCs). Following treatment with a colloidal suspension solution containing FeCl<sub>3</sub>·6H<sub>2</sub>O, FeSO<sub>4</sub>, and H<sub>2</sub>O, the NCs formed free-flowing porous MNCs. NCs and cellulose powder had a respectable inclination toward methylene blue. However, MNCs displayed exceptional extraction efficiency for



MB. Thermodynamic investigations showed that MNCs at different temperatures spontaneously adsorbed MB.

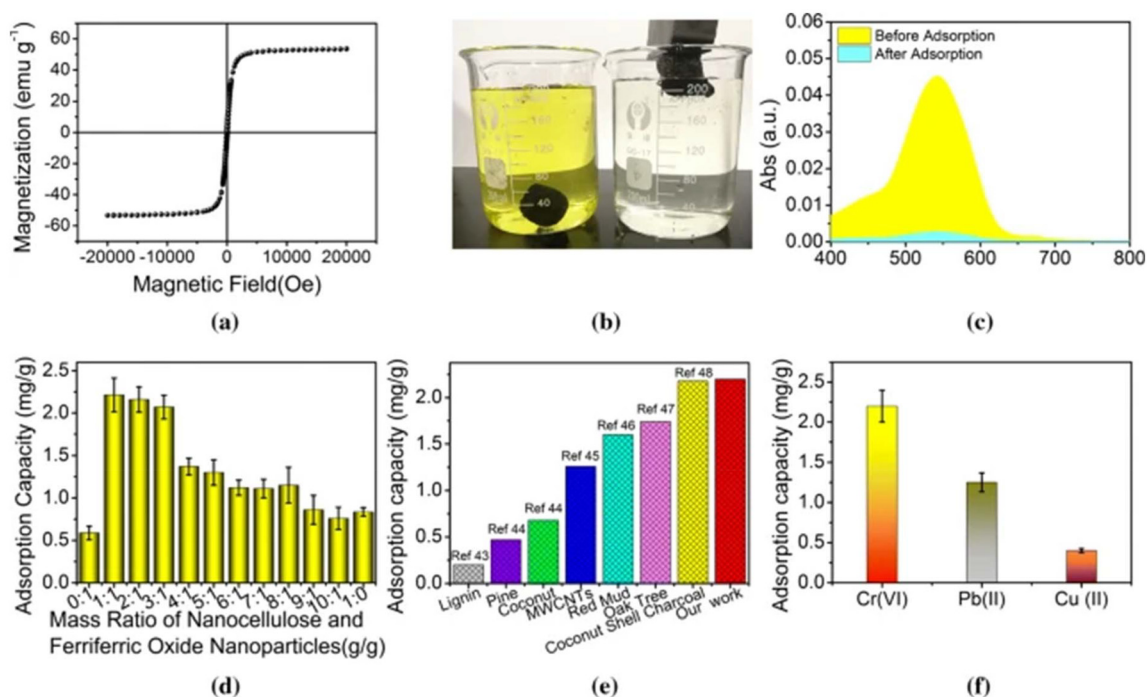
Diverse methods have been documented to eliminate organic contaminants from wastewater, such as the membrane separation technique,<sup>114</sup> absorption technique,<sup>115</sup> and advanced oxidation processes (AOPs). AOPs fully decompose a variety of organic pollutants using active oxygen<sup>116,117</sup> and/or sulphate radicals.<sup>118,119</sup> Active sulphate and oxygen groups have been produced by metal ions ( $\text{Co}^{2+}$ ,  $\text{Cu}^{2+}$ ,  $\text{Ce}^{3+}$ , and  $\text{Fe}^{2+}$ ), their metal oxides, and metal-organic frameworks.<sup>120</sup> In comparison to other traditional methods, this procedure has been proven to be a promising way to remove dyes from water. The substances that produce the radicals in AOPs are difficult to remove from solution, and their removal results in secondary contamination. Thus, to overcome this problem, Amiralian *et al.*<sup>121</sup> used cellulose nanofibers as a template for the synthesis of magnetic nanoparticles. The process of grafting magnetic nanoparticles to nanofibers involves the hydrolysis (*in situ*) of the metal precursors at ambient temperature. The effects of varying contents of nanofiber on the crystallite size, morphology, and thermal and magnetic characteristics of a membrane made from cellulose nanofibers coated with magnetic nanoparticles were examined in this work. The magnetic nanoparticles generated in this investigation possessed diameters of less than 20 nm and their crystallite sizes were observed to be between 96 and 130 Å. With an increase in the content (83–60 wt%) of magnetic nanoparticles, the flexible magnetic membranes exhibited superparamagnetic behaviour and very high magnetic characteristics (67.4–38.5 emu g<sup>-1</sup>). After that, the magnetic membrane was employed in an enhanced oxidation process based on sulphate radicals as an inexpensive, eco-friendly catalyst. The membranes were effective in eliminating Rhodamine B (RhB), which is an organic dye with hydrophilic nature utilized in industry, by activating peroxymonosulphate (PMS). In 300 min at room temperature, most of the Rhodamine B was decomposed (94.9%), showing the powerful ability of the magnetic nanocellulose membrane to catalyze PMS and extract RhB.

One of the most dangerous environmental issues endangering human health is heavy metal contamination. Wei and co-workers<sup>122</sup> successfully adsorbed heavy metal ions from water and achieved controlled recovery under magnetic conditions by combining nanocellulose and ferroferric oxide ( $\text{Fe}_3\text{O}_4$ ) nanoparticles to create a magnetic hybrid aerogel. The magnetic properties of the hybrid aerogel and its ability to absorb chromium ( $\text{Cr}(\text{vi})$ ) ions, a heavy metal, were investigated in their study. The findings demonstrated that the nanocellulose and ferroferric oxide nanoparticles physically adsorbed each other, while maintaining their original structural and compositional features. Due to its saturation magnetization value of 53.69 emu g<sup>-1</sup>, the magnetic hybrid aerogel showed strong ferromagnetic properties, which enabled its efficient and controlled recovery under magnetic conditions. The maximum value of 2.2 mg g<sup>-1</sup> was achieved by the adsorption effectiveness of the hybrid aerogel for the  $\text{Cr}(\text{vi})$  ion when the mass ratio of ferroferric oxide nanoparticle to nanocellulose was

1 : 1. The hybrid aerogel also exhibited comparable adsorption behaviour for copper ( $\text{Cu}(\text{ii})$ ) and lead ( $\text{Pb}(\text{ii})$ ) ions, indicating its potential use for the removal of heavy metal ions. Because of the presence of nano- $\text{Fe}_3\text{O}_4$  particles, this hybrid aerogel exhibited magnetic properties. As the external magnetic strength increased, Fig. 11a demonstrates how the magnetization of the hybrid aerogel increased, reaching a magnetic saturation of 53.69 emu g<sup>-1</sup>. The ferromagnetism of the hybrid aerogel was evident given that the magnetization direction was aligned with the direction of the applied magnetic field. Under magnetic conditions, this magnetic property helps the efficient recover of the hybrid aerogel. Following the introduction of the hybrid aerogel into a  $\text{Cr}(\text{vi})$  ion-containing aqueous solution, the colour of the solution progressively shifted from yellow to colourless (Fig. 11b). A UV-Vis spectrophotometer provides additional qualitative information on the concentration of the solution before and after adsorption, respectively. The maximum absorption of  $\text{Cr}(\text{vi})$  ions was shown by the absorption peak at 540 nm, which decreased significantly from pre- to post-adsorption, indicating that the aerogel adsorption action effectively removed this heavy metal ion from water (Fig. 11c). The aerogel exhibited varying adsorption capacities for the removal of  $\text{Cr}(\text{vi})$  ions at varying mass ratios. The hybrid aerogel achieved the maximum adsorption capacity of 2.2 mg g<sup>-1</sup> (Fig. 11d), which is superior to that of previously reported adsorbents (Fig. 11e) when the mass ratio of nanocellulose to nano- $\text{Fe}_3\text{O}_4$  was 1 : 1. The high specific surface area of the nanocellulose and nano- $\text{Fe}_3\text{O}_4$  particles and the hierarchical micro-/nanoscale structure are responsible for the high porosity of the aerogel of over 99%. Furthermore,  $\text{Pb}(\text{ii})$  and  $\text{Cu}(\text{ii})$  ions could also be adsorbed from water by the hybrid aerogel with a mass ratio of 1 : 1, with maximum values of 1.25 mg g<sup>-1</sup> and 0.4 mg g<sup>-1</sup>, respectively. This suggests that various types of metal ions can be removed from water by the hybrid aerogel. These findings imply that the design may be expanded to include functional aerogels made of nanocellulose, which can efficiently filter water and achieve regulated recovery in the presence of a magnetic field.

### 5.3. Diverse applications of customized MNCs using 3D/4D printing methods

Using layer-by-layer slicing, additive manufacturing (AM) is a quickly evolving material production technique that helps create sustainable structures.<sup>123</sup> Complex structures can be fabricated using AM technology in contrast to traditional manufacturing methods. Therefore, it has developed into a successful method for achieving great design flexibility and freedom during prototyping at a high resolution. The potential for 3D printing cellulose-based composites has been demonstrated for a range of applications, including dentistry, automotive applications, tissue engineering, complex structures, target drug delivery, and personalized food design.<sup>124–126</sup> The integration of CNCs into various synthetic and natural polymers has been investigated through the use of 3D printing technologies. These printing methods include inkjet printing,



**Fig. 11** Magnetic characteristics and adsorption effectiveness of the hybrid aerogel of nanocellulose and  $\text{Fe}_3\text{O}_4$ . (a) Hysteresis curve of hybrid aerogel, (b) digital image demonstrating the color change of the solution before and after the hybrid aerogel removed  $\text{Cr(VI)}$  ions, (c) UV-vis absorbance spectra of the  $\text{Cr(VI)}$  ion solution before and after absorption, (d) adsorption capacity of the hybrid aerogel at different mass ratios for the  $\text{Cr(VI)}$  ion, (e) comparison of the maximum adsorption capacities of different adsorbents for the removal of the  $\text{Cr(VI)}$  ion, and (f) comparison of the maximum adsorption capacity of the hybrid aerogel for the removal of  $\text{Cr(VI)}$ ,  $\text{Pb(II)}$ , and  $\text{Cu(II)}$  and  $\text{Cr(VI)}$  ions. Reproduced with permission from ref. 122. Copyright 2019, Springer Nature.

vat photopolymerization, and extrusion-based printing. Inkjet printing is a quick, inexpensive, and non-contact 3D printing method that replicates pictures onto a substrate by precisely ejecting ink along a predefined route utilizing acoustic or thermal pressure. Vat photopolymerization techniques such as digital light projection and stereolithography are frequently used to print MNC composites that incorporate photopolymer resins. Using MNC fillers and photoreactive resins as the main components, these approaches can be used to design and build bone scaffolds, robots, and pharmaceutical devices.<sup>127</sup> The mechanical and thermal stability of the printed goods are enhanced by the use of these nanoparticles. Vat photopolymerization methods employ light sources including UV, X-rays, and electron beams to polymerize liquid resins.<sup>128</sup> In addition to printing by inkjet and vat photopolymerization, extrusion printing (fused deposition modelling, direct ink writing, and micro-extrusion bioprinting processes) is a widely used method that allows 3D periodic structures to be assembled programmable. This method selectively distributes the extruded material layer by layer. Furthermore, by aligning every MNC in a single direction, extrusion-based printing is an excellent method for creating MNC-based structures with favourable anisotropic features. These MNC-based composites that are 3D printed and uniaxially orientated have several applications in the development of actuators and optical devices.<sup>129</sup> A notable disadvantage of 3D printing is the

creation of immobile constructions devoid of shape-shifting capabilities. There is a growing emphasis on the creation of complex, multipurpose smart structures that can adjust to their environment. Researchers have developed a new manufacturing technique called 4D printing, which can solve the problems encountered with 3D printing.<sup>130,131</sup> To provide the essential functionality of 3D-printed structures, such as a change in shape, colour, and characteristics upon exposure to external stimuli, 4D printing, which employs time as the fourth dimension, is an improved version of 3D printing. Similar to 3D printing, 4D printing depends on extremely intricate and potent mathematical modelling of the intended structure.<sup>132</sup> Due to alignment during printing and the hygroscopic nature of NPs, which causes the significant predefined bending, twisting, and curling properties for 4D printing, 3D-printed MNC-based composites also exhibit shape-changing behaviour in the presence of external stimuli. These 4D-printed MNC materials are useful for creating flexible sensors for wearable electronics applications as well as lightweight, incredibly fascinating constructions.<sup>133–135</sup> Because of their excellent ion deposition, printability, self-assembly behaviour, unique geometry, and capacity to develop bone scaffold interfaces, as well as their biodegradability, non-toxicity, and biocompatibility, 3D/4D printing technologies have recently attracted significant attention in the fields of wound healing, bone tissue engineering, and regenerative medicine. This is

especially significant for biomineralization agents and mechanical reinforcement.<sup>136,137</sup>

## 6. Conclusions and future aspects

Although diverse topics were covered in the present report, an effort was made to deliver a distinct flavour with the blending of theoretical and experimental research. Magnetic nanocellulose composites have marked a significant milestone in the field of materials science, which are characterized by unique magnetic properties intertwined with the biodegradability and environment-friendly nature of nanocellulose. This review concisely summarized the most significant strides in the realm of magnetic nanocellulosic materials and their applications. The amalgamation of magnetic nanoparticles with nanocellulose has not only introduced multifunctionality, ranging from bio-imaging and targeted drug delivery but has also enhanced its photophysical properties, which are particularly valuable for wastewater treatment and electronic-based applications. The advancements in distinct synthetic methodologies have resulted in the improved dispersion of magnetic entities into the nanocellulose matrix, resulting in composites with enhanced mechanical properties, thermal stability, and superior magnetic responsiveness. Furthermore, this review highlighted the importance of distinct surface functionalization techniques such as silanization, oxidation, esterification, amination, and phosphorylation for enhancing the physicochemical properties of MNCs. Furthermore, a meticulous compilation of recent research reports based on the translational applications of MNCs was presented.

However, despite the remarkable progress in the era of MNCs, challenges such as optimizing the loading of magnetic particles to ensure the maximum performance without compromising the inherent properties of nanocellulose still need to be addressed. Furthermore, interfacial contact, charge transfer, bonding, binding energy, and electronic characteristics studies on hydrogels and carbon nanoallotrope hybrids still need to be narrowly targeted. Clear concepts of adsorption, adhesion, stability, and structural analysis depend on how these factors correlate with the binding energy and electronic structural properties for  $sp^2$ - and  $sp^3$ -configured magnetic cellulose nanoallotropes. Furthermore, a thorough investigation of theoretical and experimental combinations can provide researchers with better results, hence revitalizing future developments in the context of MNCs.

Looking ahead, it is envisaged that MNCs are poised for cutting-edge innovations that can revolutionize multiple domains. Researcher will likely focus on increasing the control of the morphology and nanostructural features of these composites to tailor the properties of MNCs for specific applications. This includes developing novel synthesis techniques that allow the precise manipulation of particle size, distribution, and magnetic properties. Additionally, it is anticipated that sustainability will remain the major driving force for future developments, with emphasis on green synthetic meth-

odologies, enhanced biodegradability, and recyclability. The integration of MNCs with other emerging smart materials or IoT devices may enhance their applicability in extended domains. The outlook for MNCs in practical applications is promising; however, some potential strategies need to be designed to overcome several challenges that need to be addressed. Firstly, a detailed *in vivo* toxicity and comprehensive long-term biocompatibility analysis has not been conducted, which requires thorough investigation. Although the preliminary analysis indicates the good biocompatibility and low toxicity of MNCs, comprehensive and standardized long-term studies are needed to fully comprehend their safety profiles in clinical applications. Secondly, many of the applications of MNCs are still in the laboratory testing stage, and thus one of the key challenges is still the lack of large-scale industrial production testing. Additionally, the interaction between MNCs and biological tissues needs to be better explored and complemented with further analyses. One challenge is the potential environmental impact of the large-scale production of MNCs, particularly concerning the disposal and degradation of magnetic nanoparticles. Strategies to mitigate these impacts include the development of biodegradable magnetic nanoparticles and the implementation of green chemistry principles in the synthesis of MNCs.

Another major challenge is the possibility of batch-to-batch variability, which can affect the reliability and consistency of the properties of MNCs. Finally, looking to the future, the achievable performance and application range of MNCs need to be further expanded by designing standardized production protocols and rigorous quality control measures to produce high-valued specialty materials in large quantities for commercial use. To realize the effective and safe translation of MNCs into clinical practice and industrial applications, collaborative efforts among researchers, regulatory bodies, and industry stakeholders are vital. Although significant effort has been devoted to creating novel cellulose-based 3D printing solutions, some challenges still need to be addressed. Considering the inherent mechanical qualities of cellulose, its full potential as a reinforcement still has not been achieved. Increasing the interfacial bonding and dispersion of cellulose fibres in polymeric matrices is crucial to improving the reinforcing efficacy of cellulose. In this case, cellulose-based formulation 3D printing technology is continuously evolving, with new and remarkable features and uses being described. The research on hygromorphic cellulose composites for 4D printing is still in its infancy, and thus more work is required to fully comprehend how the orientation and dispersion of fibres affect the responsiveness of the resulting materials. One of the issues that has to be resolved is enhancing the mechanical performance of cellulose-based materials that are 4D printed, while also attaining responsiveness. However, studies related to magnetic nanoparticle-incorporated 3D/4D printing of nanocellulose for a broad range of applications are scarce in the literature. Thus, a comprehensive consideration in this direction may expose new insights for future research on MNCs.

Conclusively, MNCs stand at the forefront of advanced materials owing to their exceptional characteristics and eco-friendly nature, having the ability to address some of the most pressing challenges of society. To explore the new era of sustainable innovations, continued collaboration between materials scientists is required to unlock the full potential of MNCs.

## Author contributions

Shikha Awasthi: project administration, conceptualization, investigation, visualization, writing – original draft, validation. Komal: conceptualization, visualization, writing – original draft, validation, and funding acquisition. Sarvesh Kumar Pandey: investigation, visualization, writing – original draft, and validation.

## Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this review.

## Conflicts of interest

The authors declare no conflict of interest.

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