

Review Article

Austine Ofondu Chinomso Iroegbu and Suprakas Sinha Ray*

Recent developments and future perspectives of biorenewable nanocomposites for advanced applications

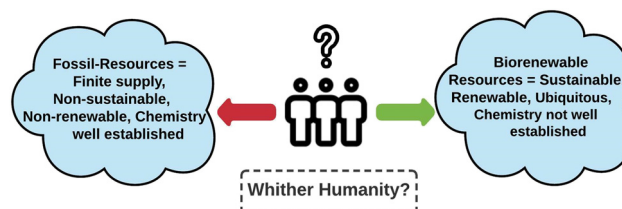
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Abstract: Fossil-based resources are finite; thus, materials derivable from these resources are nonsustainable. As no methods for renewing the depleting fossil reserves have been discovered, humanity must seek renewable, sustainable, and efficient alternatives to satisfy its material needs and preserve civilization. Solar energy remains the “capital income” of the global community and is pivotal to meeting the current needs of humankind by using biorenewable resources. Hence, the scientific community should ultimately shift its focus away from the well-established field of fossil-based chemistry to the less examined area of biorenewable chemistry. Although nature-based materials offer enormous opportunities as eco-friendly and renewable resources toward materials sustainability, their application ranges are beset with limitations compared to conventional materials – hence, enhancing their properties for advanced applications toward sustainable development represents a very challenging task. This review discusses the existing techniques for various synergizing approaches developed in the fields of biology, chemistry, materials science, and nanotechnology to overcome the limitations of biorenewable nanocomposites, expand their application range into previously restricted areas, and establish biorenewables as viable alternatives to conventional materials fabricated from fossil-based resources. In addition,

* **Corresponding author: Suprakas Sinha Ray**, Centre for Nanostructures and Advanced Materials, DSI-CSIR Nanotechnology Innovation Centre, Council for Scientific and Industrial Research, Pretoria 0001, South Africa; Department of Chemical Sciences, University of Johannesburg, Doornfontein 2028, Johannesburg, South Africa, e-mail: rsuprakas@csir.co.za, ssinharay@uj.ac.za, suprakas73@yahoo.com

Austine Ofondu Chinomso Iroegbu: Centre for Nanostructures and Advanced Materials, DSI-CSIR Nanotechnology Innovation Centre, Council for Scientific and Industrial Research, Pretoria 0001, South Africa; Department of Chemical Sciences, University of Johannesburg, Doornfontein 2028, Johannesburg, South Africa



Graphical abstract

we briefly outline various preparation methods for bio-based nanocomposites as their limitations and directions for future studies.

Keywords: fossil-based resources, biorenewable resources, biobased nanocomposites, biobased chemistry, sustainability.

1 Introduction

Composites are combinations of two or more materials (heterogeneous systems) with a multiphase matrix structure produced *via* natural or synthetic processes. Each component of the composite matrix contributes to the bulk properties of the aggregate material [1]. A major attribute of composites is that both their constituents and interfaces between different components are physically distinguishable; in other words, composites represent the combinations of materials dispersed in a continuous matrix [2]. Composites can be found virtually anywhere, from the tiniest bones that support a bird's structure to the towering skyscrapers lining our cities, and the significance of composite materials for the structural integrity of living and nonliving systems cannot be overemphasized. Hence, advances in composite material science and technology are of key importance for the advancement of human civilization. In spite of the synthetic composite industry that has grown extensively over the last seven decades [2], studies on the effective mimicking of the outstanding designs of natural composites with superior properties and multiple functionalities are very scarce. Such materials are purposefully

designed to satisfy specific requirements; at the same time, they are environmentally benign, biodegradable, and biocompatible [3,4]. For example, biological composites such as bones in the human body function not only as support structure but also as agents for synthesizing red blood cells, platelets, and white blood cells; storing fats and minerals (such as calcium and phosphorous); and stabilizing the pH of the human blood by either releasing or absorbing alkaline salts [5], demonstrating the superiority of nature over humanity in terms of material design.

In terms of particle size, composites can be broadly classified into macrocomposites (size range: $\geq 5\ \mu\text{m}$, concrete), microcomposites (size range: $1\text{--}5\ \mu\text{m}$, particle-board), and nanocomposites (size range: $\leq 1\ \mu\text{m}$, natural bone tissues). Generally speaking, the ideal nanocomposite is a material with at least one constituent phase in the nanoscale range (up to 100 nm) or a solid structure with nanometer-scale dimensions and identifiable distances between its similar or dissimilar phases. These structural phases can be zero-dimensional (graphene quantum dots), one-dimensional (nanowires), two-dimensional (layered graphene), and three-dimensional (fullerenes) [4]. Because nanoscale materials possess high surface-to-volume ratios with exceptionally large fractions of surface atoms, they exhibit unique mechanical, electrical, optical, electrochemical, magnetic, catalytic, and structural properties. Hence, their combination into aggregate materials results in multifunctional behavior with characteristics deviating from the properties of the individual constituents [4,6]. Nanocomposites are fabricated to achieve properties and multiple functionalities that are ordinarily not demonstrated by conventional single-phase materials; however, there

are numerous challenges to realizing this goal [4]. The science and technology of nanocomposites is a relatively new field with the term “nanocomposites” coined around 1982–1983 [7,8]; however, reports dated many centuries ago demonstrate that the fabrication of nanocomposite materials is as old as humankind [9]. For example, the Lycurgus cup (Figure 1) [10] dubbed “an exceptional piece of work” was manufactured around AD 400. First reported in 1845, this glass nanocomposite is dichroic and exhibits opaque greenish-yellow coloration under indirect light exposure (Figure 1a); however, under transmitted light, it assumes a translucent ruby-like color (Figure 1b) [10]. Chemical analyses have shown that the cup consists of a colloidal soda–lime–silica mixture with trace amounts of well-distributed silver–gold alloy nanoparticles (NPs) ($\approx 1\%$) and approximately 10% of copper [10]. However, how exactly these metals were dissolved in the glass to produce the characteristics of the Lycurgus cup remains a subject of speculation for modern scientists [10].

In 1924, Zschokke [11] reported the exceptional properties of the “Damask and Damascus blades” from the Middle East dated around AD 300 and AD 1700. These ultrahigh carbon steels possess extraordinary strength, are almost indestructible and shatter-proof, and exhibit extremely high resistance to blunting [11]. In recent years, it has been argued that in the blades, cementite nanowires are encapsulated into carbon nanotubes (CNTs), which may explain their remarkable properties. However, numerous attempts to replicate these blades by modern methods were unsuccessful as the exact manufacturing procedure has not been identified [12,13]. Moreover, archaeological excavations dated around AD 800 revealed the fabrication of a resilient



Figure 1: Portion of the Lycurgus cup with dichroic properties: (a) Opaque greenish yellow color under reflected light. (b) Translucent ruby-like color under transmitted light. Reproduced from ref. [10], Copyright 2008, Springer-Nature.

and robust material from nanomaterials by the Mayans [14]. Referred to as “Maya blue,” it was manufactured by embedding indigo dye into the nanopores of palygorskite clay (magnesium–aluminum nanoclay). This relic nanocomposite exhibits stable pigmentation despite the exposure to harsh environmental conditions, including acidic, alkaline, and chemical solvents [14]. The interfacial chemistry between the organic and inorganic phases endows it with an exceptionally high resistance to discoloration, thereby providing new directions for fabricating stable hybrid nanostructured pigments [14] and furnishing a clear evidence of the existence of advanced art and possibly the science of nanocomposite fabrication long before the development of the modern instrumentation and technologies.

Because we live in the nano-age, there is scarcely any field of human endeavor that does not utilize nanoscale composites (Figure 2) [4,15]. The advancement of human civilization is directly related to the advancements in material science and engineering, hence a future without nanocomposites is unlikely even as humanity constantly pushes the boundaries of science and technology. However, the concept of nanocomposite science and technology involves controlled manipulation of the structures and properties of matter on the nanoscale, resulting in materials and states of matter with exceptional mechanical strength, resilience, toughness, and resistance to harsh environmental conditions [4], it also becomes a source of concern from the viewpoints of ecological integrity, human health, and sustainability as these engineered nanoscale materials can potentially resist to undergo decomposition and mineralization in the natural environment. When such nanomaterials are exposed to the natural environment through leaching and direct or accidental release, they can compromise and upset the Earth’s intricate and inter-related systems and generate complex environmental stresses for both the living and nonliving components [16]. Hence, the consensus in the scientific community is to pursue greener, biodegradable, and biocompatible alternative resources for the fabrication of nanocomposites, which satisfy the existing application needs without compromising their efficiency [17,18]. These materials can be fabricated as either stand-alone nanocomposites or integrated into other

nonbiodegradable or nonbiocompatible materials to increase the potential for their breakdown in the natural environment, thereby promoting the nanocomposite decomposition process [19]. Biorenewable resources, such as cellulose, natural rubber, proteins, starch, and lignin, are considered promising candidates for the fabrication of such eco-friendly nanocomposites. Moreover, with the persistent calls for sustainable development [20], it has become necessary for stakeholders, such as chemical scientists, technologists, and engineers to respond and change the narrative by shifting the research focus toward materials and environmental sustainability for a better earth.

Although the nanocomposites fabricated from biorenewable materials possess many advantages, ranging from eco-sustainability to serving as greener alternatives to many conventional nanocomposites used in food packaging, drug delivery systems, and tissue engineering, they are also limited by certain drawbacks, including chemical instability and inferior mechanical and thermal properties compared with those of derived from fossil resources [7]. Unlike the previously published reviews on biorenewable nanocomposites (Table 1), this study discusses possible methods for synergizing various approaches currently used in biology, chemistry, materials science, and nanotechnology from a broader perspective to overcome the limitations of biorenewable nanocomposites, expand their application range into previously restricted areas, and establish biorenewables as viable alternatives to the conventional materials. In addition, various preparation methods for these biobased nanocomposites as well as their limitations and directions of future research studies are discussed.

2 Importance of biorenewable nanocomposites

Nanoreinforced polymeric systems (such as plastics), also known as polymer nanocomposites (PNCs), remain the bedrock of nanotechnology while ranking top in the fabrication and application ranges of nanoscale materials

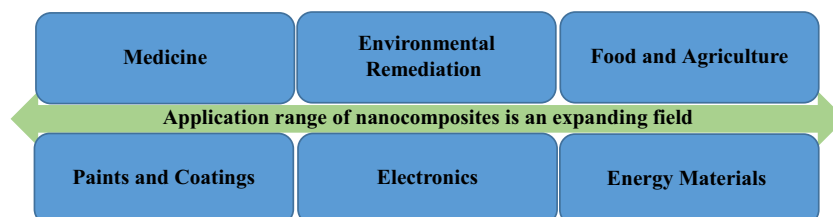


Figure 2: Various application ranges of nanocomposites that are continuously growing and expanding.

Table 1: Previously published reviews on biorenewable nanocomposites for sustainable development

Title	Scope of review	Date published	Ref.
Cellulose-based bio- and nanocomposites: a review	The progress on cellulose macro- and nanofibers and their production and application in composite materials	2011	[21]
Bio-nanocomposites for food packaging applications	Focuses on bionanocomposites for packaging applications, such as PLA, polycaprolactone, poly (butylene succinate), chitosan, <i>etc.</i>	2013	[22]
A review on bionanocomposites based on chitosan and its derivatives for biomedical applications	Chitosan-based nanocomposites for biomedical applications. Their modifications and properties	2015	[23]
Polysaccharide-based bionanocomposites, properties, and applications: a review	Discusses the preparations, modifications, characterization, and applications of bio-nanocomposites based on various polysaccharides functionalized by different nanofillers	2016	[24]
Hybrid cellulose bionanocomposites from banana and jute fiber: a review of preparation, properties, and applications	Summarizes the importance of employing banana and jute in reinforcing polymer matrix. Further discusses cellulose nanoparticle reinforced composites and their preparation techniques with associated merits in their applications for sustainable development	2017	[25]
Nanocellulose, a versatile green platform: from bioresources to materials and their applications	Reviewed the current knowledge and limitations of nanocellulose-based materials from source, synthesis, and modification methodologies to material applications	2018	[26]
A review on electrically conducting polymer bionanocomposites for biomedical and other applications	Presents the progress made in bio-nanocomposites from renewable resources and their unique structure, functionality, and conductivity in various applications. Further presented are the merits of biocompatibility, biodegradability, and challenges toward the pursuit of sustainable development	2020	[27]
Chemistry, structures, and advanced applications of nanocomposites from biorenewable resources	Reviews the chemistry, structures, and applications of biobased nanocomposites	2020	[7]
Review on cellulose nanocrystal-reinforced polymer nanocomposites: processing, properties, and rheology	Demonstrates the recent progress of cellulose nanocrystals (CNCs) as reinforcing fillers. Further presented are the processing techniques, rheological behaviors, and performances of CNCs and advantages over conventional carbon/metal nanofillers in various applications	2021	[28]
Cellulose biomass-based sustainable hydrogels for wastewater remediation: chemistry and prospective	Focuses on cellulose biomass hydrogels toward water remediation. It also presents the latest technological advancements with current challenges and opportunities	2022	[29]
Biopolymeric nanocomposites for orthopedic applications	Reviews mechanical, biocompatibility, bioactivity, and structural viability of biopolymeric nanocomposites in orthopedic applications	2022	[30]

[1,31]. The PNC application range covers a vast area including automobiles, electronics, medicines, and environmental remediation materials because these categories of nanocomposites demonstrate outstanding thermal, mechanical, chemical, and gas barrier characteristics that are considerably different from those of their bulk counterparts [1,31]. The improved properties of PNCs are attributable to the large interfacial volume between the nano-sized heterogeneities and polymer chains in a multiphase matrix coupled with the physical, chemical, and/or biological interactions at their boundaries, which do not exist in the traditional micro-filled polymer composites [32]. Furthermore, the polymer chain mobility (*i.e.*, glass

transition) and relaxation properties are strongly influenced by the interactions between polymer chains and NPs, thus allowing manipulation of the rheological properties and glass transition temperature of a polymeric system by varying the NPs characteristics. For example, the mobility of polystyrene chains increases after their intercalation between the layers of a smectic clay (nanofillers) [33]. This phenomenon transcends the concept of local immobilization of polymer chains by the NPs and suggests the existence of a strong far-field effect of the nanofiller creating weakly bound structures with physical entrapments (such as bridges) in the matrix phase and temporary interactions between the filler particles [4,34].

PNCs have several advantages, including low cost, ease of fabrication, and high operation efficiency under harsh conditions, which make them the preferred candidates for nanocomposite fabrication [1,35,36]. Because we live in a polymer world [16], the dominance of polymer-based materials in nearly every human endeavor cannot be overstated [16]. Polymers have outperformed many competing materials, including metals, wood, and glass, owing to their resilience, high adaptability, low weight, low fabrication cost, and other advantages [37]. Furthermore, because the processability of nanocomposites critically affects their final characteristics and application range [38,39], PNCs are suitable for the fabrication of nanocomposites with different shapes and sizes through the well-known conventional processing techniques, such as extrusion, thermoforming, and injection molding, making PNCs one of the most versatile classes of nanoreinforced materials in these modern times [38,39]. However, despite the benefits and huge application potential of PNCs, there are growing concerns regarding the negative environmental and health effects of plastic materials on the natural environment [16,40]. Furthermore, because most commercial polymers employed in PNCs are fossil-based materials or synthetic polymers (Table 2), the microbial community in the natural environment has not been able to efficiently biodegrade or depolymerize, assimilate, and mineralize them. Although microbial and enzymatic degradation of synthetic polymers were observed in some studies [41,42], these processes were very limited. For example, certain conditions, such as temperature, pH, moisture content, ultraviolet (UV) radiation, and molecular weight of the polymer (its upper limit, *e.g.*, 2,000 Da for polyethylene, cannot be exceeded), are required for efficient microbial degradation [43,44]. Nevertheless, owing to the chemical inertness, resilience, and robustness of conventional plastics, their incorporation into nature would take a significant amount of time, generating a considerable

stress for the living and nonliving components of the natural environment [16]. In addition, the presence of plastic debris in the oceans results in a lower ocean efficiency (affecting the global heat balance) and revenue loss (estimated to exceed 1 trillion USD per annum) [16]. Furthermore, fossil resources are finite [45] and cannot be replenished by any modern technologies, and this causes a sustainability problem. Considering the negative contribution to the climate change (global warming), rising exploration costs, volatility, and related environmental degradation [46,47], fossil resources have recently come under serious scrutiny while potentially becoming a threat to the environmental and economic sustainability rather than an asset [48–50].

It is unarguable that materials science and engineering play a key role in human civilization (including construction materials, medical tools, cooking utensils, agricultural yield optimization, packaging and energy storage materials) and that human existence without these materials is impossible [56]. Hence, the scientific community is currently focused on the fabrication of benign PNCs from biorenewables due to their environmental friendliness, closed material loop, cost efficiency, biodegradability, nontoxicity, biocompatibility (the ease of mineralization and assimilation in the natural environment), and sustainability (renewability) with related properties that are commensurate with or even exceed those of the materials fabricated from conventional polymers [6,57].

2.1 Biorenewables: a brief background

Biorenewables (the chemicals and materials derived from organic or biobased sources), such as plant matter, have

Table 2: Commonly employed polymers for PNC fabrication with the corresponding application areas

Common polymers used in fossil-based PNCs		
Polymer type	Select application range	Ref.
Poly(vinylidene fluoride) and polydopamine (PDA)	Energy storage systems, photovoltaic (PV) backsheets, <i>etc.</i>	[36]
Polyamide and polysulfone	Membrane technology, fibers, films, <i>etc.</i>	[51]
Polyethersulfone	Membrane technology, environmental remediation	[52]
Poly(tetrafluoroethylene), polypropylene, polyethylene, polyethersulfone, polyetheretherketone, polyacrylonitrile, polyethersulfone, poly(phthalazinone ether sulfone ketone), poly(vinyl butyral), polyamide, polysulfone, polyol, and polyphenol	Membrane technology, food contact materials-packaging, PV backsheets, automobile composites, <i>etc.</i>	[1]
Polyepoxide (<i>i.e.</i> , epoxy resin)	Tire reinforcement, coating technology, <i>etc.</i>	[53]
Polyacrylamide and polyaniline (PANi)	Drug release systems, anticorrosive coatings, <i>etc.</i>	[54]
Polymethylmethacrylate and polyimide	Electromagnetic (EM) interference shielding, sensors, supercapacitors, <i>etc.</i>	[55]

long been proposed as promising alternative resources to fossil-derived chemicals and materials [58–60]. For example, sugarcane and pineapple crops can be optimized for the production of fermented alcohol, an intermediate chemical that may be subsequently used to produce formaldehyde used in the synthesis of various plastics, including urea–formaldehyde and phenol–formaldehyde resins [61,62]. The proliferation of biorenewable resources such as plant biomass for the fabrication of nanocomposites has several environmental benefits, including the possibility of mitigating the negative effects of CO₂ emissions and the global warming. For example, during photosynthesis, plants consume CO₂ while generating oxygen. The consumed CO₂ is spread across the plant leaves, trunks, and roots. At the roots, CO₂ is taken by microorganisms and converted to food sources by a fungal network inside the ground, thereby fueling a vast expanse of microbial communities in the food chain and making CO₂ a source of nourishment in the ecosystem. Hence, plant biomass and the microbial community are extremely important for CO₂ stabilization in the natural environment [63,64]. Despite the efforts spent on creating photosynthetic bioreactors [65,66], to the best of our knowledge, no technology developed by humankind for the capture and storage of CO₂ from the atmosphere is as cost-effective, benign, and efficient as that purposefully designed by the nature [67,68].

The concept of biorenewables includes the purposeful use of organic and natural materials for the fabrication of nanocomposites. By incorporating biorenewables as organic additives or nanofillers, a matrix platform, or their combination, the application range of these materials can be expanded considerably. Biorenewables may also make conventional nanocomposites biodegradable and/or fundamentally alter their characteristics, thereby opening new avenues for reengineering traditional PNCs. For example, CNCs and chitosan nanowhiskers, both biorenewable NPs, were used to reengineer and modify Nylon-6,6, a fossil-based commercial polymer [69]. The utilization of an all-organic reinforcement even at low filler loadings (≤ 0.5 wt%) allowed toggling the mechanical properties of the PNCs between those of a rigid structure and a mechanically robust material with a tensile strength exceeding 100 MPa [69]. It is noteworthy that nanocomposite polyurethane (PU) foams, an important class of engineering materials traditionally prepared from fossil-based polyisocyanate and polyalcohol, can be now synthesized entirely from epoxidized vegetable oils and biobased diisocyanate using glycerin extracted from plant biomass as an additive, water as a blowing agent, and natural fibers or particles as nanofillers [70]. Moreover, due to the increasing

concerns regarding the toxicity/environmental challenges of nonecofriendly and nonbiocompatible NPs (such as silver, zinc oxide [ZnO], and ferrous oxide NPs and CNTs) [71], biorenewables enable the fabrication of biocompatible and green nanofillers (utilized as modifiers and plasticizers) in a wide range of applications to satisfy the current material needs while aiming to achieve environmental sustainability [6,72,73]. Various biorenewable resources, such as polysaccharides (starch), chitin, chitosan, proteins, vegetable oils (including oil waste), lignin, cellulose, eucalyptus tar, cashew nut shell oil, hemicellulose, natural rubber, and wood biomass, have been successfully used in the fabrication of nanoreinforced polymer composites [74,75]. Generally, all biobased PNCs can be grouped into four major classes [76–80]: (i) plant-based polymers and their derivatives, such as starch, natural rubber, terpenes, terpenoids, vegetable oils, seed oils, cellulose, regenerated cellulose, and modified starch; (ii) animal-based polymers and their derivatives, including animal tissues and microbial biosynthesis such as bacterial cellulose, keratin, silk, albumin, chitin, and poly(hydroxy alkanoates); (iii) fungus-based polymers derived from fungi, such as mycelium (mainly composed of chitin, glucans, mannoproteins, and hydrophobins); and (iv) polymers synthesized from biobased resources, such as polylactic acid (PLA), poly(butylene succinate), furans (e.g., poly[furfuryl alcohol]), and poly(ethylene furanoate).

Nature has always provided humanity with templates for prototyping materials to satisfy its needs and advance our civilization. Unlike the well-established field of fossil-based chemistry, the chemistry of biorenewables includes complex hierarchical architectures and chemical constituents whose functionalities are modified at various structural levels to produce self-assembled systems with unique properties. In spite of many attempts to mimic and recreate these biological nanocomposite structures, the obtained results have not always been satisfactory [3,4,81]. For example, spider silk, a biocomposite fibrous material demonstrating a unique combination of strength and toughness typified by a sigmoidal, balanced stress–strain relationship with impressive characteristics such as exceptional shear rigidity (compared with that of industrial fibers) and a toughness of 160 MJ/m³ exceeding that of nylon (80 MJ/m³) by a factor of two [82], is bio-produced by silk-spinning spiders *via* a sophisticated spinning process whose mechanism is not yet fully understood [83]. Although various studies have extensively demonstrated the possibility of biomimicking spider-silk bioproduction [84,85], no successful industrial-scale technology has been developed in this area. Hence, to increase the application potential of biorenewables and overcome their well-known limitations

outlined by Abe *et al.* [86], it is necessary to elucidate their structural design, chemical properties of their interfaces in target environments, and highly evolved structure–function parameters purposefully designed to satisfy specific objectives while remaining ecofriendly, benign, and sustainable in the natural environment.

3 Preparation of biorenewable nanocomposites

Among various factors, a processing technique or methodology employed during the preparation of nanocomposites strongly influences their ultimate properties. Hence, special attention must be paid when choosing a fabrication technique for a target application to obtain optimal results [4]. Conventional techniques utilized for nanocomposite fabrication include ball milling, free-radical grafting, and melt compounding; however, not all of these preparation methods are suitable for manufacturing biorenewable nanocomposites [39,87]. The latter are currently produced by chemical solution processes (such as sol–gel and coprecipitation methods), *in situ* polymerization, intercalation, and extrusion (Figure 3) [57]. Moreover, newer techniques such as rotational molding, centrifugal casting, pultrusion, electrospinning, and nanofiber direct dispersion are emerging as promising manufacturing methods, although they are currently at the pilot stage [7,88,89]. As pointed out previously, the concept of biorenewable nanocomposites implies their incorporation as additives or nanofillers, utilization as a matrix platform, or a combination of the first two approaches. Therefore, these

nanocomposites are generally modified (physically or chemically) to enhance their usability and particular characteristics for target applications. In this section, several techniques employed for the fabrication of biorenewables are briefly discussed.

3.1 Sol–gel methods

Template synthesis is one of the oldest bottom-up approaches utilized to fabricate nanomaterials. It was originally introduced as a “gel route” by Roy [90] who used inorganic gels as templates for the fabrication of diphasic materials such as ceramic–ceramic nanocomposites at low temperatures well below their melting points. This conceptualized innovation represented a new direction for the preparation of ceramic nanocomposites that demonstrated nano-heterogeneity or ultra-heterogeneity by exploiting the thermodynamics of metastable materials including the heat generated during the reaction between separate phases and benefits of epitaxial growth [8]. As illustrated in Figure 4, the processing of multicomponent nanomaterials *via* a sol–gel (multicomponent colloidal suspension) technique allowed the densification of different types of sol–gel multicomponent systems such as mullite, alumina, and zircon ceramics with a wide range of applications in high-temperature structural materials, refractory materials, and infrared transmitting materials [8]. Owing to the high flexibility of this method, it has been significantly modified within the last several years and successfully employed for the fabrication of multicomponent nanomaterials such as hybrid organic–inorganic nanocomposites [91,92], thus opening new frontiers for the development of a wide range of advanced materials. The sol–gel process can produce spun fibers, layered densified materials, and lightweight porous aerogels with various mechanical properties from multiple resources [93]. For example, Silva *et al.* [94] reported a green technique for synthesizing porous chitin nanostructures for biomedical applications. This ecofriendly process employed an ionic liquid (1-butyl-3-imidazolium acetate) in conjunction with supercritical fluid technology for chitin dissolution, after which the biopolymer was regenerated in ethanol. The final mesoporous material exhibited low density and high porosity [94]. Nevertheless, although the sol–gel process is suitable for the preparation of various nanocomposites [91–93,95,96], it is capable of fabricating biorenewable nanocomposites only on a laboratory scale [7], owing to the complex solubility and chemical properties of biobased resources that undergo solubilization with subsequent regeneration [97,98].

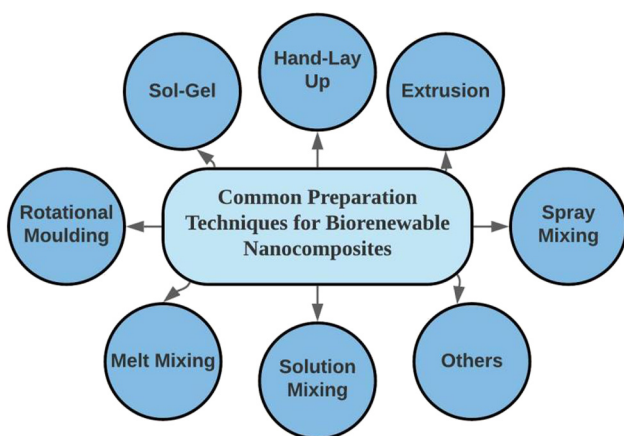


Figure 3: Various preparation methods employed for the fabrication of biorenewable nanocomposites.

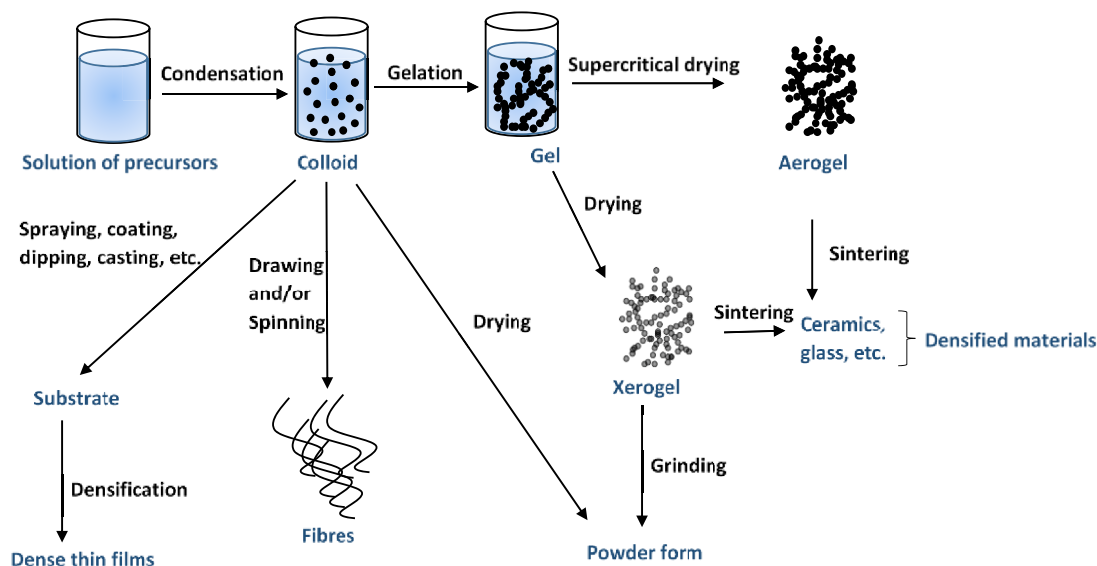


Figure 4: Simplified flow diagram of the sol–gel process used in the fabrication of nanocomposites. Adapted from ref. [99], Copyright 2010, Springer-Nature.

3.2 Solution mixing

The solution method is very similar to the sol–gel method; however, composite materials in the former case are obtained by spraying, coating, dipping, casting, or molding after mixing the dissolved matrix with additives [87]. This method has been argued as a nonenvironmentally friendly process (especially in terms of sustainable development) because it produces toxic chemical footprints due to the use of solvents during the recovery and evaporation processes [100], hence the growing interests toward solvent-free and green(er) solvents in this process [101,102].

3.3 Melt mixing

This straightforward technique is widely employed to produce nanocomposite materials. It generally involves melting the matrix phase, after which additives/fillers are incorporated therein, and the resulting mixture is molded or cast into a desired configuration [100]. Various biorenewable nanocomposites have been fabricated using this approach. For example, thermoplastic PU derived from long-chain polyols produced from fatty acids (*i.e.*, linoleic acids) and incorporated into halloysite nanotubes (HNTs) was used to prepare a biorenewable composite material for biomedical applications [103]. In another work, nanocomposite materials consisting of bioderived polyamide 4, 10, and CNTs were produced in a twin-screw extruder by the melt mixing technique and demonstrated

high application potentials in the electrical, electronic, and automotive industries [104]. Note that the melt mixing technique utilizes a more environmentally friendly method than the solution mixing process as it produces a smaller chemical footprint due to the use of solvents, and therefore, should be employed for the fabrication of biorenewables whenever possible [87].

3.4 *In situ* polymerization

In this technique, a filler is swelled inside a low-molecular-weight monomeric system (such as furfuryl alcohol monomer), which promotes the monomer diffusion into the filler/additive. After this step, an *in situ* polymerization of the low-molecular weight monomer into an oligomer/polymer is initiated inside the filler/additive to form intercalated or exfoliated nanocomposites [87]. Unlike the melt and exfoliation adsorption techniques, *in situ* polymerization results in better exfoliation, which can be attributed to the homogenous penetration of the monomer and distribution of fillers/additives in the resulting polymer matrix [105]. Figure 5 illustrates the fabrication of the nanofibrillated cellulose (NFC) and PANi composite material with good mechanical properties and high electrical conductivity. A variety of biorenewable composites can be produced *via in situ* polymerization. For example, epoxidized soybean castor oil fatty acid-based polyol was successfully employed for the production of thermosetting PU/graphene oxide (GO) nanocomposites [106].

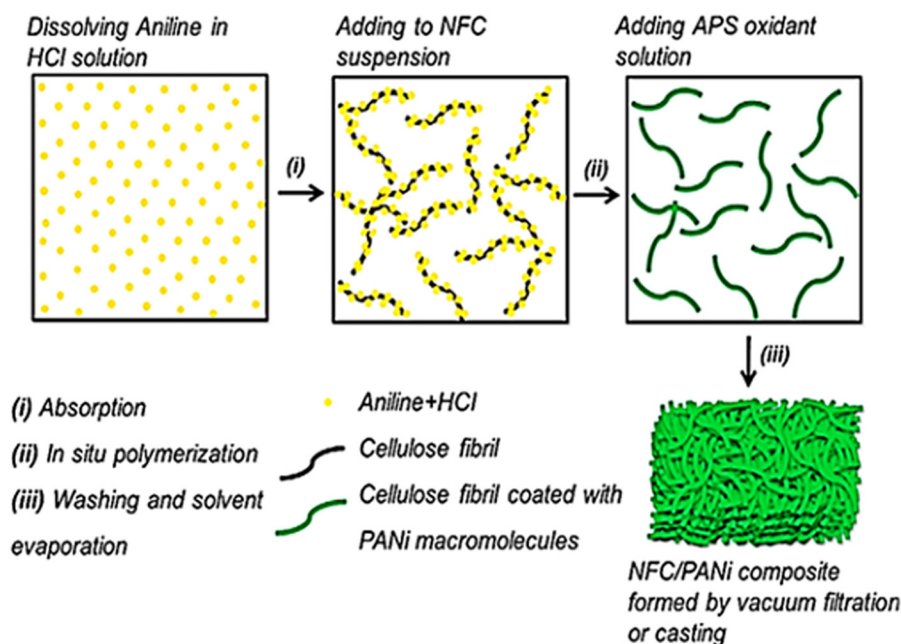


Figure 5: Schematic of the *in-situ* polymerization process used for fabricating NFC/PANi composite paper. Reproduced from ref. [107]. Copyright 2012, Elsevier Science Ltd.

4 Applications of biorenewable nanocomposites

The number of advanced new materials fabricated from biorenewable, and sustainable resources have substantially increased in recent years. Biorenewable materials have many applications ranging from the automotive industry to the built environment. In this section, we highlight several important developments and applications of the nanocomposites produced from biorenewable resources.

4.1 Membrane technology

Basically, membrane technology (*i.e.*, selective barriers) remains one of the most widely employed methods in various separation processes, including purification, remediation, recycling, recovery, and phase separations [1]. In 2020, the global membrane market exceeded 24 billion USD and is projected to grow at a compounded annual growth rate of 7.5% between 2021 and 2028 [108]. These selective barrier systems can be manufactured from organic (polymers), inorganic (ceramics, silica, zeolites), or hybrid (organic–inorganic) materials. However, on a comparative level, polymer-based systems such as organic membranes are extensively employed in a wide range of industrial and domestic applications due to their various

advantages, including low manufacturing costs, amenability, versatility, ease of fabrication, high surface area (surface-to-volume ratio), inherently high reactivity, and high performance efficiency [1,109]. Owing to the increasing water scarcity, environmental degradation, pollution, and global health challenges, the demand for membrane technology continues to increase in various areas from oxygen generation [110] to water purification, desalination, and environmental remediation [1]. Moreover, to achieve high sustainability [20], the scientific community has intensified research efforts aimed at producing biocompatible and biodegradable membranes from biorenewable resources, thus advancing and opening new frontiers for the production of innovative and sustainable materials. This section highlights the recent advances in the utilization of biorenewable nanocomposites in separation technologies.

4.1.1 Wastewater treatment

Water is essential for sustaining life, although the access to adequate, safe, and usable water supplies can potentially become a source for conflicts in the future [111,112]. Coupled with the impacts of natural disasters and environmental degradation resulting from human activities, the increasing stress on safe and usable water cannot be overemphasized [113,114]. The considerable volumes of wastewater generated globally per annum can potentially mitigate the problem of water scarcity and increase

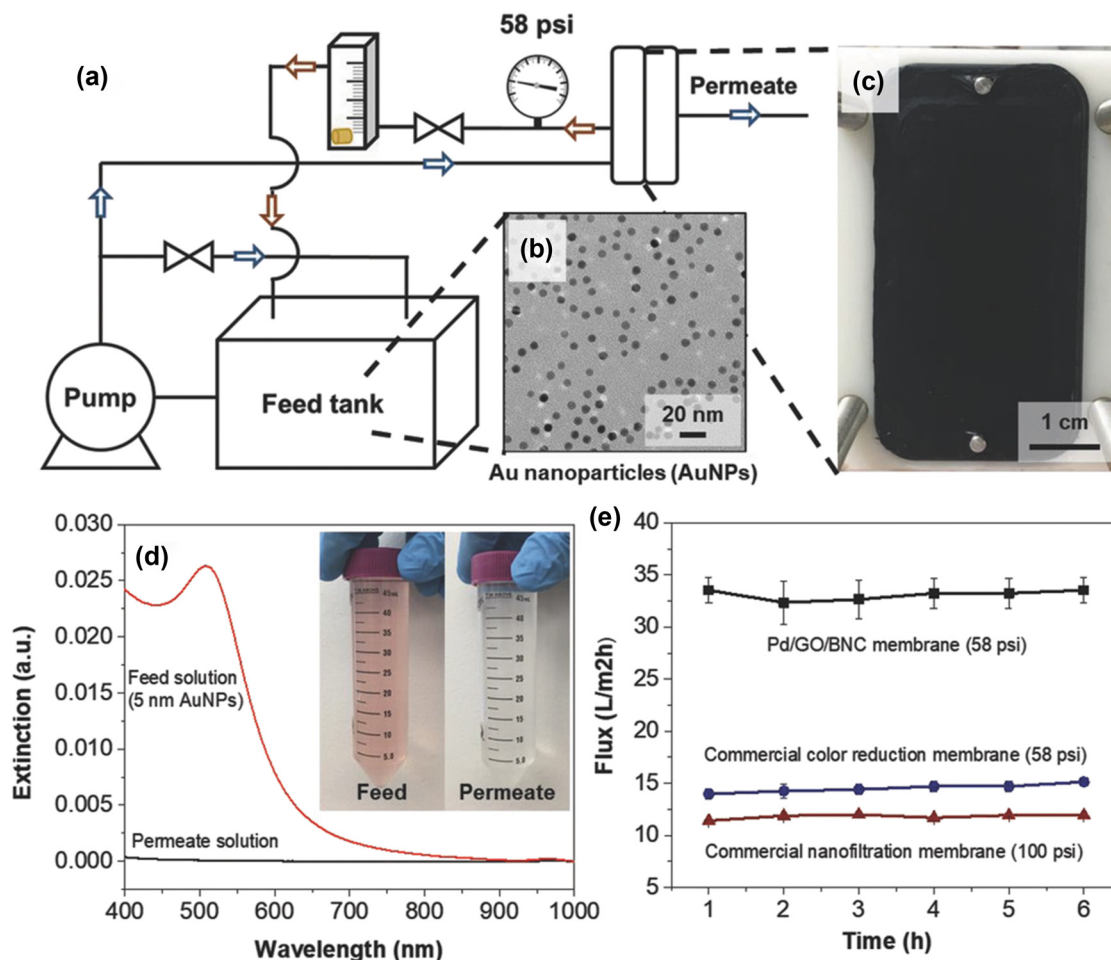


Figure 6: Schematic of the particle rejection and flux tests performed for the Pd/GO/BNC nanocomposite membrane: (a) layout of the crossflow test setup; (b) transmission electron microscopy images of the AuNPs with diameters of 5 nm; (c) cross-sectional photograph of the membrane in a flow cell; (d) UV transmittance spectra showing the rejection of AuNPs filtered through the nanocomposite material. The inset depicts the feed and permeate solutions. (e) Graphical representation of the water flux characteristics of nanocomposite membranes, including those of the commercial color reduction and nanofiltration membranes obtained at applied pressures of 58 and 100 psi, respectively [116], Copyright 2018, John Wiley & Sons.

the water security after proper capture, remediation, and reuse [1,115]. Therefore, effective processes must be developed for the optimal recovery, upgrade, and purification of wastewater to obtain safe and reusable water supplies. In recent years, the scientific community has initiated ambitious research studies on the production of biorenewable materials with high ecosustainability and efficiency for water and wastewater treatment applications. As demonstrated previously [6], biorenewables such as nanocellulose can be potentially used for the efficient treatment of degraded water. In addition to its ubiquity, biodegradability, biocompatibility, wettability, good mechanical properties, and nontoxicity, nanocellulose can efficiently replace many toxic and unfriendly conventional nanomaterials, such as titanium oxide, silver, and ZnO, which are currently employed for the fabrication of membranes

utilized in water treatment processes [6]. For example, bacterial nanocellulose (BNC) loaded with GO and palladium (Pd) NPs for the efficient treatment of wastewater [116]. A Pd/GO/BNC biorenewable nanocomposite was obtained through the *in situ* incorporation of GO flakes into BNC matrix during growth. The resultant material was highly efficient in the removal of methylene orange (MO) (up to 99.3% over a wide range of concentrations) and changing the pH value despite the multiple reuse cycles. This membrane also effectively removed other wastewater contaminants such as rhodamine 6 G (R6G) and even a cocktail of 4-nitrophenol and methylene blue (MB) [116]. As shown in Figure 6, the particle rejection capability of this composite membrane was achieved using gold NPs (AuNPs), and the membrane rejection rate for the particles with diameters exceeding 5 nm was almost

100%, indicating that the pore size of the Pd/GO/BNC composite was below 5 nm [116].

A similar work of Gholami Derami *et al.* [117] used a facile and inexpensive process to produce an innovative PDA/BNC hybrid membrane for the efficient removal of multiple heavy metal ions (such as Pb^{2+} and Cd^{2+} and organic pollutants (such as R6G, MB, and MO) from wastewater. The hybrid nanocomposite was obtained by the direct introduction (*in situ* incorporation) of PDA particles into the BNC matrix during growth. The utilized fabrication technique exhibited high integration flexibility for other adsorbent systems and processes, robustness and high stability during and after the use without decreasing its efficiency, and a possibility for industrial upscaling combined with biocompatibility and biodegradability [117]. The efficient removal of Cr ions (such as Cr^{6+} and Cr^{3+}) was achieved using biodegradable membranes consisting of functionalized cellulosic nanostructures (CNS) based on eucalyptus residues (in the sawdust form) phosphorylated after preparation [118]. The obtained CNS samples were incorporated into a poly(butylene adipate-co-terephthalate) matrix synthesized by an inversion method. The produced material demonstrated the ability to be integrated into personal and public water tap systems for the direct chromium removal from distributed waters, especially in the regions with water scarcity and high levels of chromium contamination. Considering its low fabrication cost, this material is not only affordable but can become a game-changer for low-income households to mitigate chromium poisoning [118]. By achieving optimal removal values of 93% for Cr^{6+} and 88% for Cr^{3+} , these membranes demonstrated 71% retention of chromium in accordance with the guidelines established for drinking water [118].

With the growth of the numbers of electric vehicles and energy storage systems, tellurium (Te), a rare element that is relatively new in the additive industry, is attracting considerable attention as a strategic element in many applications [119,120]. However, some concerns were expressed regarding the potential short-term and long-term environmental impacts of Te (especially as a food and water contaminant) and the related health implications for living and nonliving systems [121,122]. To mitigate potential health challenges associated with the presence of Te in drinking water, multiple studies have been conducted to explore the removal of Te from wastewater during processing [123,124]. Yao *et al.* [125] successfully fabricated an ecofriendly membrane that was acid-resistant, biodegradable, recoverable, and highly efficient for the selective recovery of Te from acidic wastewater using a two-step hydrothermal reaction. The produced sheet-like iron sulfide (FeS)–lignin–carbon magnetic nanocomposite

was composed of irregular lignin block structures with variable dimensions. The lignin nanospheres had diameters of ~300 nm and were fully embedded into FeS nanosheets. The obtained composite possessed a large surface area and abundant adsorption sites for trapping Te ions. The advantages of FeS as a magnetic metastable and nontoxic mineral with good activation properties were added advantages, demonstrating high potential of achieving phase separation in harsh environments (such as acidic wastewater) [125].

4.1.2 Dye removal

Because dyes constitute a group of very hazardous environmental pollutants and remain in wastewater during the treatment processes, developing effective, ecofriendly, low-cost, and sustainable methods for their removal is an extremely important task [126,127]. Chitosan, an inexpensive, abundant, nontoxic, biocompatible, biodegradable, and biorenewable polymer with excellent adsorption properties due to its surface functional groups have been used to manufacture efficient membrane materials for removing dyes from wastewater. For example, by employing the concept of mixed matrix membranes, a facile and green approach was employed for the fabrication of chitosan-wrapped multi-walled CNT (MWCNT) filler, which was incorporated into a polyether block amide thin-film membrane for efficient dye removal [128]. The performance of the fabricated membranes was investigated inside a cross-flow system at a pressure of 2 bar, and the rejection rate achieved for Malachite green dyes with a concentration of 30 mg/L was over 95% at a permeate flux exceeding $5.3 \text{ L/m}^2 \text{ h}$ with good water permeation properties [128]. Although the applied pressure of 2 bar was lower than the values of 5–15 bar required for nanofiltration systems, it was still possible to achieve high permeate fluxes [128]. In another study, a chitosan membrane modified with ZnO/copper oxide (CuO) heterostructured nanocomposite was prepared for the photodegradation of fast green dyes under artificial and solar irradiation, and its catalytic efficiency under UV irradiation exceeded 80% after three cycles [129]. The obtained scanning electron microscopic (SEM) images (Figure 7) revealed that the incorporation of the ZnO/CuO nanomaterials increased the porosity and active surface area of both the neat chitosan film and chitosan loaded with only ZnO nanostructures [129].

By employing phosphorylated chitosan modified with GO nanosheets, Song *et al.* [130] developed an efficient foul-resistant separation membrane for the removal of anionic dyes and inorganic salts (such as NaCl and NaSO_4) from

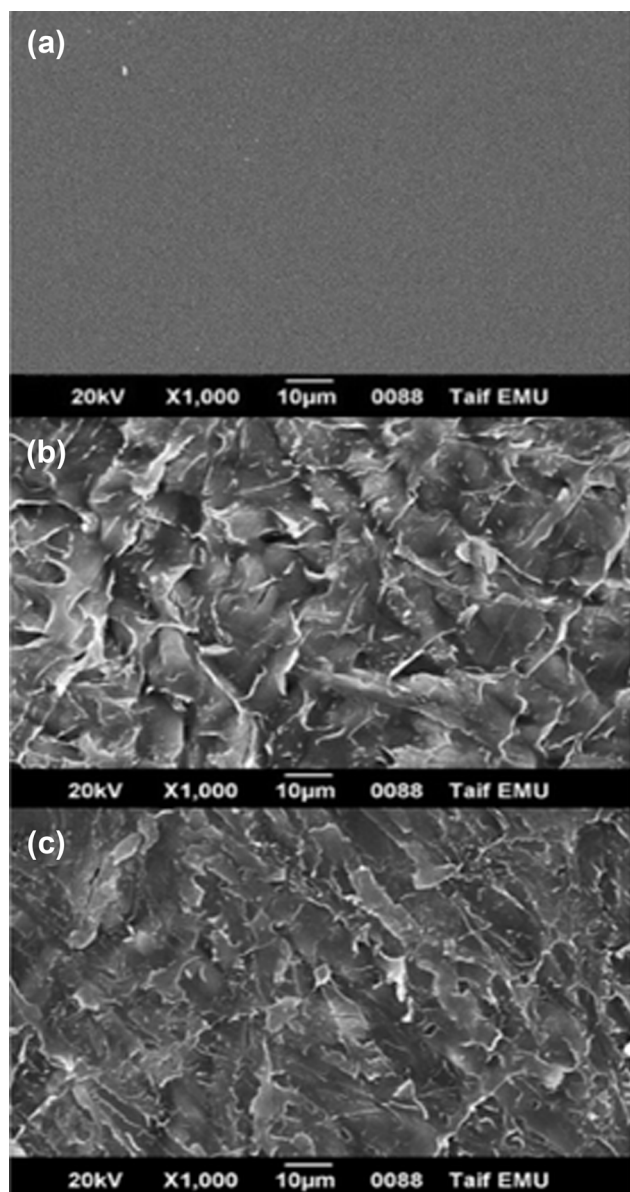


Figure 7: SEM images of (a) neat chitosan membrane with a homogeneous surface, (b) chitosan–ZnO membrane with a rough pore-containing surface, and (c) chitosan–ZnO/CuO membrane with the highest surface roughness and porosity. Reproduced from ref. [129]. Copyright 2018, SAGE Publications.

wastewater. Their innovative concept was based on a hypothesis that phosphorylated chitosan with a high degree of substitution of its hydroxyl group by the phosphate group (generally at the C-6 position) would produce nanofiltration membranes with exceptionally high surface activities. In addition, highly hydrophilic and negatively charged GO rich in surface oxygen groups can promote interactions with chitosan amino groups, which enable the ring-opening polymerization of these groups and the epoxy (oxygenized) groups of GO nanosheets [130]. The separation efficiency of

the chitosan-modified GO nanosheet membrane was evaluated *via* a crossflow permeation test conducted using a nanofiltration membrane evaluation device at a constant inlet flow velocity of 0.068 m/s, feed temperature of $25^{\circ}\text{C} \pm 0.3^{\circ}\text{C}$, feed pH of 6.5 ± 0.1 , and operating pressure of 0.60 ± 0.05 MPa. A single-component permeation test was performed using NaCl or NaSO_4 as a representative monovalent/divalent salt with a concentration of 500 mg/L and anionic dye (direct black, Ponceau S, or xylene orange) with a concentration of 100 mg/L. The salt rejection rates achieved for NaCl and NaSO_4 were 56.8 and 93.6%, respectively. The retention values obtained for the anionic dyes at a permeate flux of $71.6 \text{ L}/(\text{m}^2 \text{ h})$ were 99.7% for direct black, 97.5% for xylene, and 93.4% for Ponceau S. The study revealed that the fabricated biorenewable membrane could efficiently remove salt-containing anionic dyes from wastewater [130].

4.2 Biomedical and pharmaceutical applications

The necessity of improving human health and environmental considerations in the health sector has increased the demand for biorenewable nanomaterials. Because biorenewables exhibit unique surface properties, high biodegradability, and biocompatibility with human tissues and organs, they can be potentially used to advance human health and sustainability. Hence, biorenewable nanomaterials have become promising candidates for biomedical and pharmaceutical applications, including drug delivery/release systems, artificial limbs, tissue regeneration, intraarticular implants, intraocular lenses, and ligament and tendon repair scaffolds [131]. In this section, we describe the recent advances in biorenewable nanomaterials in biomedical and pharmaceutical applications.

4.2.1 Drug delivery

Polysaccharide–protein complexes were successfully utilized in the development of materials for drug delivery systems. For example, by combining bioderived materials, such as sericin (SC), with rice bran albumin (RBA) and embedding it into gellan gum (GG), a smart nanosystem for pH-responsive drug delivery was produced [132]. By dissolving GG in medically benign dimethyl sulfoxide under gentle magnetic stirring at room temperature for 10 min, SC was subsequently added and mixed for 24 h, resulting in the lyophilization of the SC–GG conjugate.

The final SC-GG-RBA product possessed an average particle size of 218 nm and polydispersity index of 0.23, indicating that a nanomaterial has been obtained and that the resulting polymeric system was well distributed [132]. Doxorubicin (DOX), a highly effective anticancer drug with a broad spectrum of activities, was then encapsulated into the SC-GG-RBA nanocomposite to investigate the efficiency of its drug release potential in an acidic tumor environment [132]. The efficiency of DOX release *in vitro* was 84% after 120 h in an acidic environment with pH = 4.0. Only approximately 42% of the MCF-7 cells survived after treatment for a given period; hence, determining the efficiency of green nanocomposites and further demonstrated the capabilities of biorenewables in the biomedical field [132]. In another study, an innovative biocompatible amino-T807-modified human serum albumin nanomaterial consisting of a membrane of erythrocytes (ETm) coating and 1,2-distearoyl-*sn*-glycerol-3-phosphoethanolamine-*N*-[succinimidyl(polyethyleneglycol)] (DSPE-PEG₃₄₀₀-Mal) has been reported [133]. To overcome the challenge of the blood–brain barrier (BBB), which prevents the free movement of blood and transportation of therapeutic medications for the mitigation of brain diseases and increases the efficiency of sustained drug delivery across the BBB, a DSPE-PEG₃₄₀₀-T807 nanomaterial was fabricated. The unique targeting properties of NPs coupled with the brain cell targeting of the T807 ligand resulted in their affixation to brain cells, which subsequently promoted the drug transport across the barrier. *In vivo* live imaging demonstrated that T807/ETm-HSA NPs accumulated in brain cells and produced no cytotoxic effect on these cells at concentrations up to 200 µg/mL; moreover, they did not impair the integrity of the *in vitro* BBB at 125 µg/mL according to the D-lucifer permeability and P-gp efflux activity obtained in an R123 experiment for the cocultured cells. It was concluded that bio-NPs could sustain the continual release of curcumin within 72 h, further suggesting their usefulness and efficiency for drug delivery systems and the transportation of drugs across the BBB for the treatment of chronic neurological diseases [133]. Another work building on the concept of combinatorial chemotherapy for augmenting therapeutic efficiency through multiple mechanisms to effectively mitigate the toxicity and adverse effects have demonstrated the successful production of DOX and gambogic acid (GA) encapsulated in albumin nanocomposites (DOX-loaded albumin NPs [DNPs] and GA-loaded albumin NPs, GNPs, respectively) for synergistic antitumor efficacy [134]. As demonstrated in Figure 8, these drug-loaded albumin NPs exhibited high synergistic efficiency in the ablation of HepG2 tumor cells at a combination index of 0.38. Confirmatory studies employing *ex vivo* fluorescence

imaging showed that the tumor was fully permeated by the drugs due to the enhanced permeation and retention effect of the augmented therapeutic efficiency of the nanoconstruction [134]. It was argued that the synergistic tumor inhibition efficacy was attained at 3-fold lower doses using the combinatory approach than for a single dose *in vivo* [134].

4.2.2 Tissue engineering

The current bone tissue engineering studies are pushing boundaries by employing innovative biocompatible materials with exceptional biocompatibility and efficient mechanical properties. Tissue engineering is extremely promising for increasing sustainability and the quality of human life while lessening the burdens of the societal and economic costs associated with healthcare and life expectancy [135]. To increase the application scope of tissue engineering, the fields of biology, chemistry, and bioengineering were integrated to regenerate or create entirely new tissues or cellular systems for the replacement, repair, or enhancement of biological systems (such as damaged limbs). As a dynamic field, tissue engineering is capable of discovering new approaches to improving health, human longevity, and clinical conditions [135]. For example, to assess the influence of CNCs on the biocompatibility and osteogenic potential of PLA in bone tissue engineering, a PLA/CNC composite scaffold was fabricated by an electrospinning technique [136]. It was found that the incorporation of CNCs enhanced the mechanical properties of the composite and increased its thermal stability compared with those of pure PLA. In addition, the composite scaffold demonstrated impressive biocompatibility and exceptional surface mineralization with good expression of osteogenic gene markers, indicating a high osteogenic potential. It was concluded that the studied material could be used as a functional system for bone tissue engineering [136]. In another work, the biological functionalities of bioactive ternary gum arabic/κ-carrageenan (κ-CG) incorporated hydroxyapatite nanocomposite (n-HA) were leveraged for bone tissue engineering [137]. Because κ-CG is an anionic unbranched sulfated heteropolysaccharide possessing numerous pharmaceutical properties, such as immunomodulation, anticoagulation, and antihyperlipidemic activities, and its chemical structure resembles that of glycosaminoglycans, which are inherently present in human bone tissues and cartilages, it represents a promising platform for the fabrication of bone tissue materials [137]. Ternary n-HA/gum arabic/κ-CG composites were prepared at the following concentrations: 60/30/10 (CHG1),

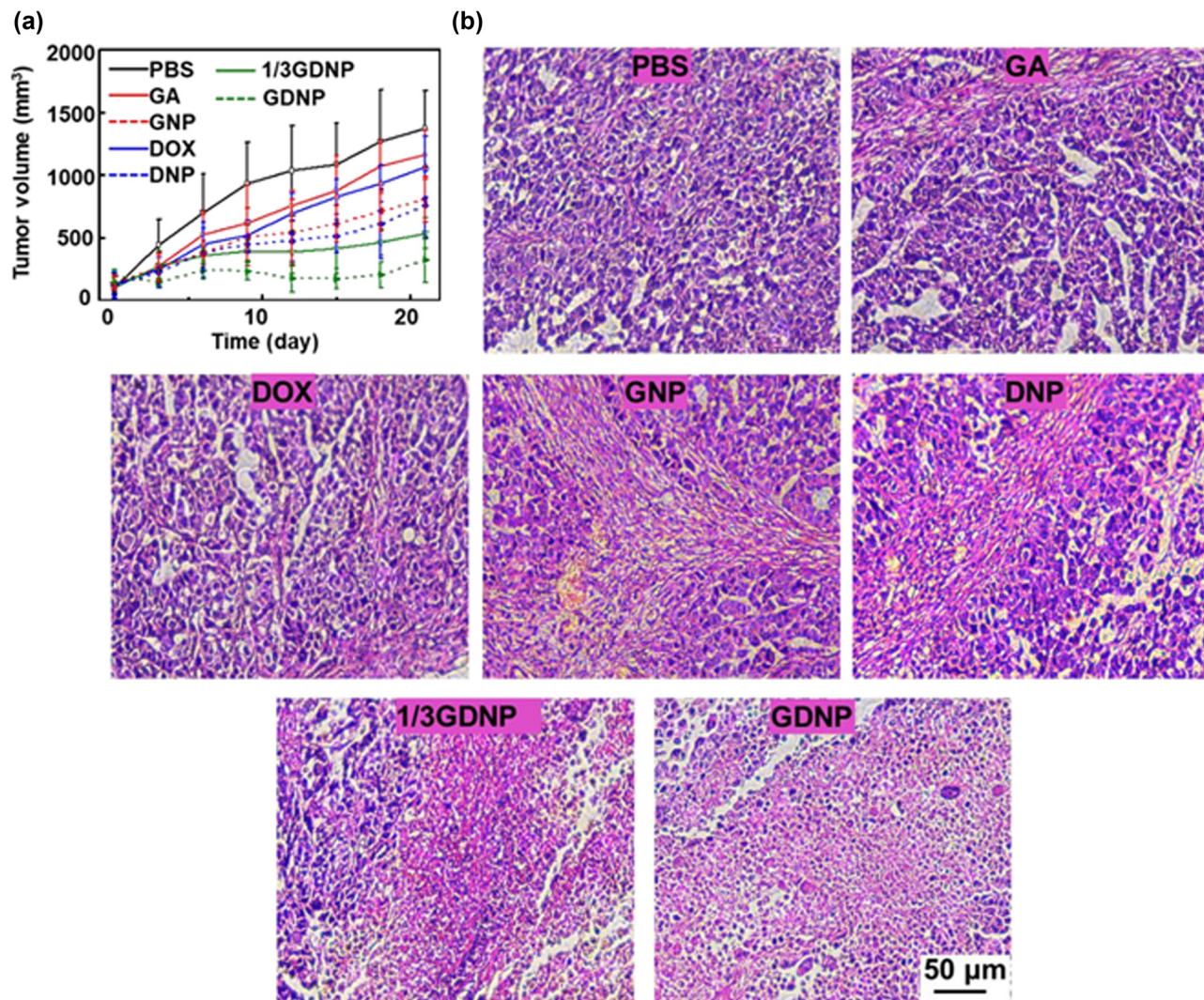


Figure 8: *In vivo* demonstration of the antitumor efficiency of the drug-encapsulated bionanomaterials: (a) Free DOX and GA slightly inhibited the tumor growth as compared with the control treatment; (b) in a comparative assessment, larger necrotic regions were observed for the DNP and GNP groups than those of the free drugs. Meanwhile, the combined treatment with GDNP (combination of DNP and GNP) and 1/3 GDNP produced enhanced necrosis regions, which resulted in $\approx 90\%$ induced apoptosis of cancer cells. Reproduced from ref. [134], Copyright 2020, Elsevier Science Ltd.

60/20/20 (CHG2), and 60/10/30 (CHG3). In addition, an n-HA/gum arabic binary system with the composition 60/40 (HG) was obtained as a control. After 15 days of incubation in a simulated body fluid, rapid mineralization of the apatite layer was observed for the ternary systems, which was confirmed by SEM images [137]. Although CHG2 contained the optimal amount of the deposited apatite layer, the highest mechanical strength was obtained for the scaffold containing 50/50 GA and κ -CG. Moreover, the cytotoxicity of these materials was evaluated by culturing osteoblast-like MG63 cells, and the CHG2 nanocomposite system exhibited the optimal cell viability and proliferation properties. Furthermore, antibacterial activities, protein adsorption

and tolerance, biodegradability, and osteogenic differentiation were observed for all ternary systems. It was concluded that the surface chemical interactions between n-HA, GA, and κ -CG molecules played significant roles in the obtained results [137]. Ferreira *et al.* [138] described the almost endless capabilities of biorenewables in the tissue engineering field. For example, nanocellulose gels and foams can be employed for the fabrication of tissue scaffolds and extracellular matrices from lightweight materials instead of the currently used metals, ceramics, and other polymers with high production costs and unsustainable production steps [138]. Owing to its bioinspired hierarchical construction, pliability, versatility, and biological

origin, nanocellulose exhibits high mechanical strength and biocompatibility, which are required for functional materials in tissue engineering [138].

4.3 Electronic applications

The recent technological advances remarkably decreased the size of electronic devices from phones to television sets by making them slimmer, portable, and lighter. It has gradually become customary to demonstrate how efficiency trumps bulkiness for electronics and energy storage systems. This trend consequently influences the behavior of people and industrial development, which directly impact scientific pursuits, although science and technology thrive on a societal paradigm shift. Hence, the development and fabrication of novel materials (especially those with multifunctional capabilities) to simultaneously fulfill several goals are becoming a growing trend, making smart technologies, the fourth industrial revolution, smart materials, and devices popular buzzwords. Moreover, because of the increasing impact of electronic wastes (e-wastes) [139], the scientific community is spending more research efforts on the development of a new innovative class of materials that undergo biodegradation after use, exhibit biocompatibility during decomposition or mineralization in the natural environment, and are sustainable, creating new frontiers in chemistry and materials science beyond the limitations of conventional materials [140]. As the electronic industry remains pivotal for the advancement of human civilization (including robotics, artificial intelligence, synthetic organs, quantum computing, and smart materials), the fabrication of ecofriendly materials from biorenewables for electronic applications has attracted much attention due to their higher efficiency and lower costs than those of traditional materials.

4.3.1 Sensors

Biobased sensory systems and biosensors are widely employed in various fields, including the medical, environmental remediation, food, and pharmaceutical industries. They are generally categorized according to the mode of physiochemical transduction and/or biorecognition element type. Hence, depending on the utilized transducer, these systems can be classified into electrochemical, optical, and thermal sensors, while according to the biorecognition element type, they are divided into enzymatic, aptamer-

based, and antibody-based ones [141]. The interest in using biorenewables in biosensory applications has recently increased in almost every field of human endeavor. For example, to efficiently monitor and mitigate potentially negative impacts of elemental materials such as Fe, Mn, and Cr in target environments (including both the human body and natural environment), a new class of fluorescent lignin carbon dots (CDs) for reversible responses to high-valence metal ions has been developed [142]. These biosensors exhibit high sensitivity and short response times and are simpler than conventional analytical methods such as differential pulse polarography, electrochemistry, and atomic absorption spectrometry (which are sophisticated, expensive, and time-consuming and utilize complex sample preparation procedures) [142]. Lignin CDs were synthesized by the facile one-pot hydrothermal carbonization of waste lignin biomass obtained from the pulp industry. The produced bright blue photoluminescent CDs demonstrated remarkably high photostability against bleaching in various pH environments and excellent fluorescence properties in the presence of Fe^{3+} , Cr^{+6} , and Mn^{2+} ions [142]. The biosensing of high-valence trace metal ions in fetal bovine serum and ascorbic acid by these CDs was impressive [142]. It was concluded that the valence-dependent response of lignin CDs was related to their sensitive chelation and the formation of stable complexes that inhibited radical recombination and allowed the instantaneous detection of a wide range of metal ions in biological and environmental redox systems and complex composites [142]. Another study described an innovative peptide nucleic acid (PNA) electrochemical biosensor based on reduced GO (rGO) ($\text{NH}_2\text{-rGO}$)/2,2,6,6-tetramethylpiperidin-1-yl)oxyl nanocrystalline cellulose (TEMPO-NCC) for the sensing and detection of *Mycobacterium tuberculosis* [143]. Because PNA recognition layers are widely used for sequence-specific DNA biosensing, such as *M. tuberculosis* DNA, PNA was employed to utilize its sensitivity and specificity detection characteristics for the efficient discrimination toward single-base mismatches. Moreover, PNA significantly reduced the hybridization time and dependence on ionic strength at room temperature [143]. The PNA-modified $\text{NH}_2\text{-rGO/TEMPO-NCC}$ was able to successfully differentiate between complementary, noncomplementary, and one-base mismatch DNA sequences by using MB as an electrochemical indicator. The developed electrochemical biosensor also produced a linear calibration curve in the concentration range from 1×10^{-8} to 1×10^{-13} M with a maximum detection value of 3.14×10^{-14} M. Because of the increasing global challenges of unpredictable terrorist attacks, the detection of trace explosive materials is critical for conducting preventive and security measures aimed at creating a

safer public space and protecting vulnerable targets. Therefore, Wu *et al.* [144] reported a new type of three-dimensional surface-enhanced Raman scattering (SERS) sensors that were obtained by coating Au nanorods (AuNRs) with a shell of Ag nanocubes (AgNCs) (AuNR@AgNCs), which were subsequently loaded onto BNC aerogels *via* electrostatic interaction, leading to the formation of three-dimensional hot spots for the detection of 2,4,6-trinitrotoluene (TNT), a well-known explosive generally used in improvised explosive devices [144]. The key significance of the SERS technique is its ability to enhance the Raman scattering signal of target molecules on a specially primed template fabricated from a noble metal such as Au or Ag. Hence, it was concluded that the intensity of inelastic scattering of photons from a target molecule was enhanced due to the long-range EM field and short-range chemical effects [144]. The EM enhancement effect was attributed to surface plasmon resonance and the chemical enhancement caused by charge transfer. Owing to the unique structure and morphologies of the prepared AuNR@AgNC SERS substrates, traces of TNT with a low limit of 8×10^{-12} g/L were detectable at a SERS enhancement factor of 1.87×10^8 , clearly demonstrating the enormous application capability of the explosive sensors fabricated from nanocellulose aerogels with high detection efficiency for the protection of human lives and property [144].

4.3.2 Energy storage and conversion systems

The access to affordable and clean energy remains a priority for both the developing and developed countries [20]. The

growing interest in mitigating the global warming and pursuing sustainable development have resulted in many scientific breakthroughs ranging from improving the existing energy systems to developing new innovative energy conversion materials and devices. Hence, the utilization of biorenewable materials for energy storage and conversion applications with a potential use in sustainable high-performance storage and conversion systems has been continuously increasing. For example, metal–organic frameworks (MOFs) can be used as sacrificial templates for deriving hierarchical nanostructured materials with high surface areas, porosities, and specific capacitance. Hence, Xiao *et al.* [145] successfully fabricated a layer-by-layer assembly of free-standing and flexible nanocellulose/porous cobalt(II,III) oxide [Co₃O₄] polyhedron hybrid films for supercapacitor electrode applications by employing the MOF concept. By using a cobalt catalyst in the graphitization of carbon, a Co₃O₄-based graphitic material with exceptional nanoporosity, sp² carbon hybridization, and outstanding intrinsic conductivity was obtained. As illustrated in Figure 9, the produced porous Co₃O₄ polyhedron (PCP, active material) was used for the preparation of NFC/porous Co₃O₄ polyhedron (called NFC/PCP or NPC) films supercapacitor electrodes *via* a two-step water-based paper-making technique using NFC as a building block and their substrates with added conductive acetylene black agent [145]. The obtained NPC-60 electrodes exhibited impressive electrochemical performance with a real capacitance of 594.8 mF/cm² at a cyclic voltammetry scan rate of 5 mV/s, which was significantly higher than that of conventional metal oxide electrodes [145].

To develop an inexpensive supercapacitor with high stability, specific capacitance, and energy density while overcoming the challenges of the low electric double-layer

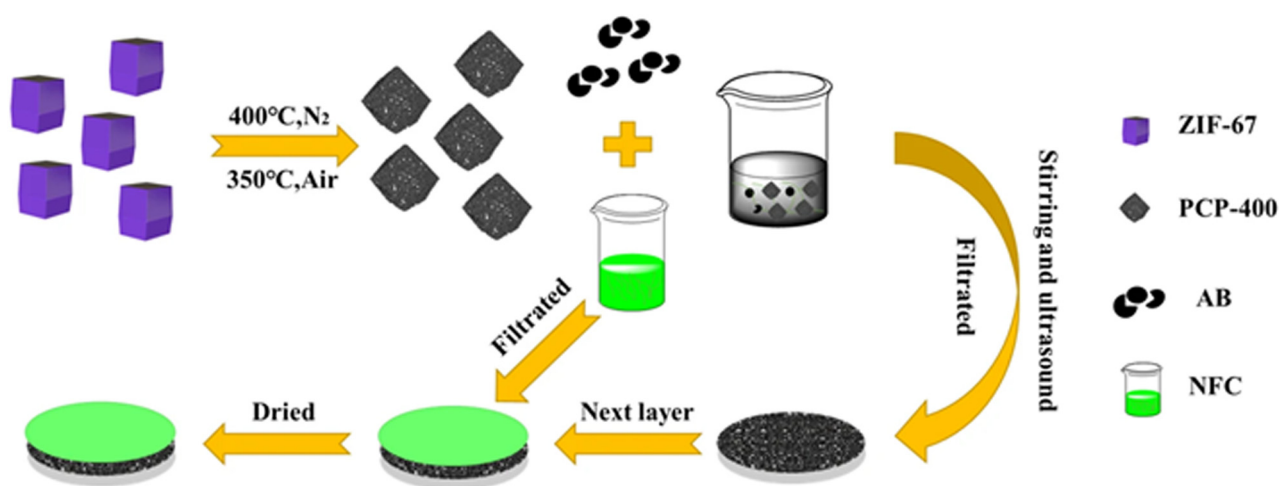


Figure 9: Schematic of the preparation of the NFC/PCP and NPC flexible supercapacitor electrodes. Reproduced from ref. [145], Copyright 2021, Springer Nature.

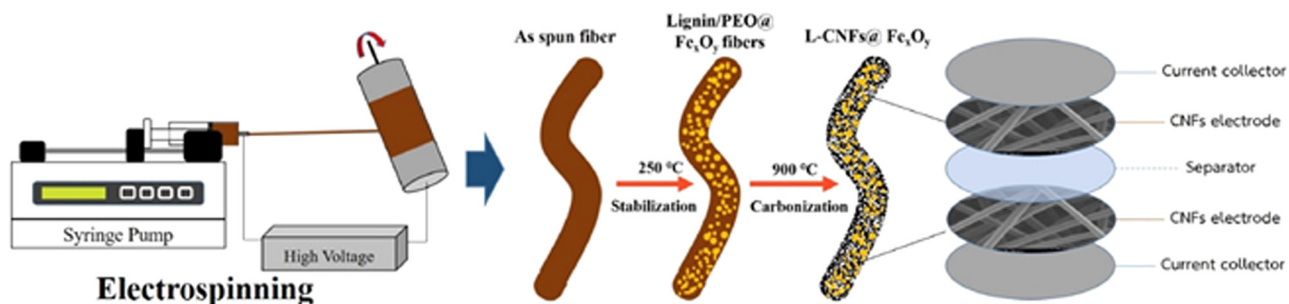


Figure 10: Schematic of the L-CNFs@Fe_xO_y fabrication process. Reproduced from ref. [146], Copyright 2021, Elsevier Science Ltd.

capacitance, Butnoi *et al.* [146] fabricated low-cost flexible lignin-based carbon nanofibers (L-CNFs) incorporated into iron oxide NPs (Fe_xO_y) [L-CNFs@Fe_xO_y] for supercapacitors with high specific capacitance by a one-step electrospinning technique [146]. They hypothesized that because compositional and morphological modifications of carbon nanofibrous materials critically affected their performance in supercapacitors, iron oxides with various valence states (Fe²⁺, Fe³⁺, and Fe⁴⁺) could be incorporated into lignin-based carbon fibers to mitigate the high electrical resistivity and low charge density resulting from the inherent

interspacing of electrospun CNFs [146]. This was achieved by electrospinning a mixture of organosolv lignin and poly (ethylene oxide) with a mass ratio of 99:1 doped with various iron (III) nitrate nonahydrates followed by stabilization and the subsequent carbonization (Figure 10) [146]. The produced L-CNFs@Fe₃O₄ electrodes exhibited an impressive optimal specific capacitance of 216 F/g at a specific current of 0.1 A g⁻¹ and ultra-high energy density of 43 Wh/kg, demonstrating a new manufacturing approach for innovative, cost-effective, and sustainable electrodes [146].

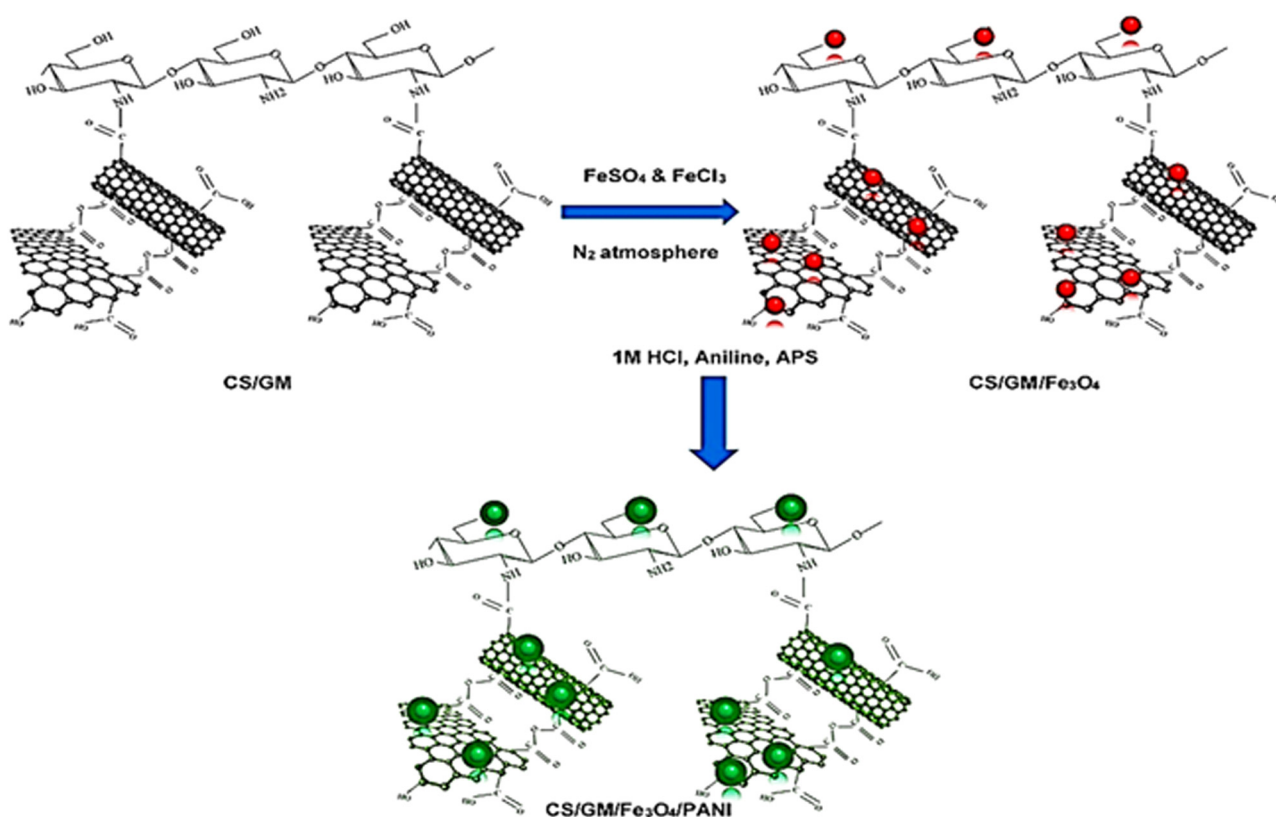


Figure 11: Simplified schematics of the synthesis of CS/GM/Fe₃O₄/PANI nanocomposite material. Reproduced from ref. [147], Copyright 2021, John Wiley & Son.

By incorporating Fe_3O_4 NPs into chitosan/GO MWCNTs (chitosan (CS)/graphene oxide-multiwall carbon nanotubes (GM)), Hosseini *et al.* [147] obtained CS/GM/ Fe_3O_4 nanocomposites grafted with PANi through *in situ* polymerization to produce CS/GM/ Fe_3O_4 /PANi nanocomposite materials for supercapacitor applications (Figure 11). The specific capacitance of the manufactured electrode was 1513.4 F/g at a specific current of 4 A g^{-1} , which was 1.9 times higher than that of a control electrode CS/GM/ Fe_3O_4 without PANi (approximately 800 F/g at 4 A/g). Moreover, both the CS/GM/ Fe_3O_4 /PANi and CS/GM/ Fe_3O_4 electrodes demonstrated remarkable cycle lives of 99.8 and 93.5%, respectively, at a uniform specific capacitance of 100 A/g during charge–discharge cycles [147]. This synergistic effect of chitosan/GM and iron oxide in conjunction with the exceptional properties of PANi indicated a very high potential for the use of biobased nanocomposites in advanced supercapacitor applications.

4.4 Food and agricultural applications

The food and agricultural sector are fundamental for the advancement and sustenance of the human race; hence, through the mandates of the sustainable development goals (SDGs), the global community is resolute on ending hunger across the countries of the earth, attaining food security, improving nutrition (e.g., through food science and processing technology) and the advancement of

agricultural best practices [20]. Recent years have seen the increasing role of nanotechnology in improving crop yield, maintain ecosystem balance (e.g., nitrogen fixation and organic matter cycle), water purification, mitigate the impacts of climate change, improve food security, enhance safe food processing and packaging techniques for better shelf-life and limiting post packaging contamination, plant protection, pest control, *etc.* [148,149]. It has become necessary to eliminate or mitigate the entrainment of biological (e.g., pathogens) and environmental contaminants (e.g., plastic matter) into the food web; to ensure that the loop between the food–water–agriculture–health nexus is efficiently protected, monitored, and continually assessed to maintain a healthy population, advance food security, sustain agricultural best practices, and enhance environmental, social, and economic sustainability [16,150]. Biorenewables have demonstrated efficiency as green(er), ecological, and sustainable materials toward eliminating the current challenges presented by traditional materials and methods employed in the food and agricultural sector toward sustainable development.

4.4.1 Green(er) and smart fertilizers

With a rapidly changing world, farmers across the globe are engaging science and technology to improve food production and related products (e.g., fibers, raw materials, fodders). The annual growth of populations coupled with

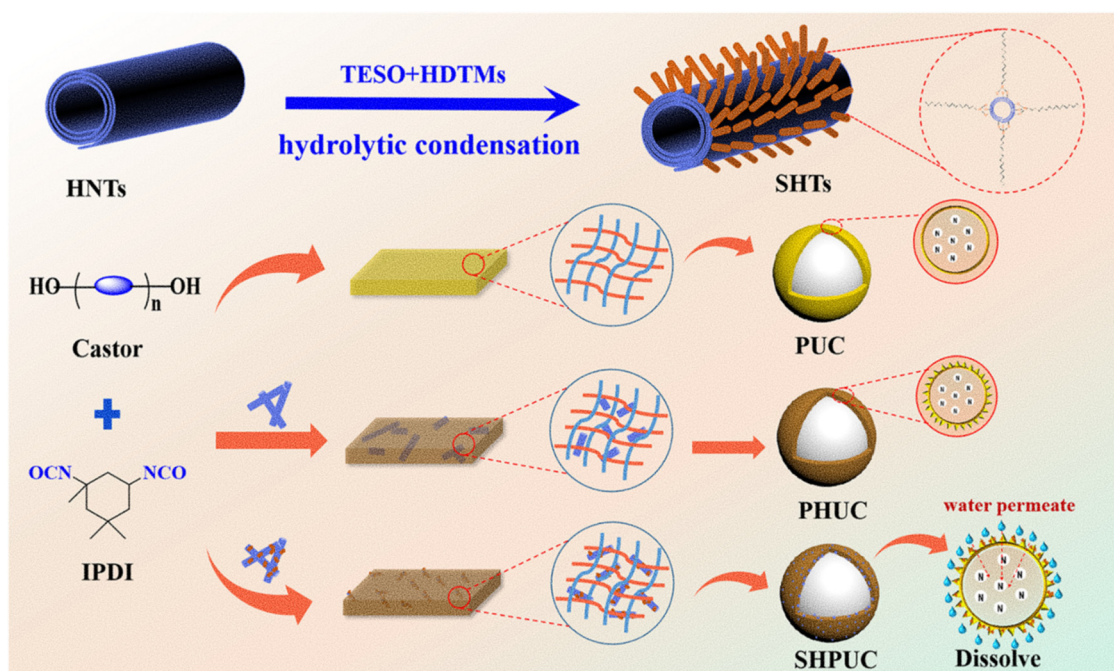


Figure 12: Diagrammatic representation of the various coating modifications employed in the biobased coated CRF [160]. Copyright 2022, Elsevier Science Ltd.

limited resources available and demand for food has necessitated the critical improvement of harvesting systems, crop yields, water recycling and conservation, and optimal land usage, while maintaining good soil structure and fertility all year round [151,152]. In 2015, a conservative population of over 3.5 billion people (more than 50% global population) fed on crops that were produced with synthetic or inorganic fertilizers (*e.g.*, nitrogen-based fertilizers). And with the global population projected to exceed the 9 billion mark by 2050, it is therefore unarguable that necessity is laid upon humanity to do more toward achieving food security and improve on sustainable agriculture despite other limitations such as water and land [152,153].

Over the years, the direct application of synthetic agrochemicals such as fertilizers to overcome some of the challenges of crop productivity and enhance agro-economics has been the way to go for conventional agricultural practices; however, the associated negative environmental impacts of this practice such as eutrophication, loss of soil integrity, increasing soil acidification which have consequent impacts on ocean acidity (a significant contributor to global warming and loss in ocean productivity), ground water contamination, and greenhouse gas (GHG) emissions with consequent impacts on climate change [154–156], are significant environmental challenges. In addition, nitrous oxide, a byproduct of nitrogen-based fertilizers, is the third most significant GHGs, after carbon dioxide and methane [157–159]. It is therefore evident that the continual use and direct application of inorganic fertilizers hold more devastation to the living and nonliving systems of the environment than the promise of better crop yield, sustainable agricultural best practices and food security. Hence, research has intensified toward benign and efficient fertilizers from nano systems from biobased resources (*i.e.*, organic or green(er) fertilizers) that offer commensurate or better crop yield than inorganic fertilizers, enhance the mitigation of environmental stressors, and improve ethical agricultural best practices and sustainable development. Because runoff of unused or waste fertilizers in the soil into the surrounding environment is the significant culprit of eutrophication, it has been postulated that smart agrochemical systems such as “slow” and controlled release (*i.e.*, deliberate and regulatory distribution) fertilizer systems (CRFs) hold enormous potentials in ameliorating this challenge [160].

Intelligent and green(er) agrochemical systems such as fertilizers have made notable progress in recent years. For example, inspired by nature, whereby butterfly wings and the legs of water striders are biodesigned with unique micronanoscale structures with reduced surface energies and enhanced superhydrophobic properties to prevent

water penetration. Xie *et al.* [161] fabricated a biomimetic superhydrophobic biobased polyurethane-coated fertilizer (SBPF) for the efficient and controlled release of needed nutrients to crops. The green PU was synthesized from cottonseed oil polyol and isocyanate (as raw materials). At the same time, the outermost coating for the PU, containing terminated OH groups (hydrophobic characteristic), was constructed by increasing the ratio of the cottonseed oil polyol. Subsequently, a two-step technique was then employed to enhance the surface roughness through the construction of micrometer–nanoscale diatomite–SiO₂ configuration, thereby decreasing the surface energy and afterward embedding fluoroalkylsilanes into the fabricated structure, which conferred the coating with overall superhydrophobicity. They determined that the SBPF demonstrated a 2-fold enhanced efficiency compared to the normal biobased PU-coated fertilizer in terms of significantly enhancing nutrient release characteristics, particularly with crops requiring a relatively long growing period [161]. In a similar study by Wang *et al.* [160] encapsulated prilled urea (*i.e.*, solid nitrogen-based fertilizer) into a superhydrophobic bionanocomposite system. As demonstrated in Figure 12, biobased PU films were fabricated from the reaction of castor oil and isophorone diisocyanate at predetermined molar ratios. These films were then modified with cheap and eco-friendly HNTs. Two parts of HNTs were employed in this modification; first, to confer superhydrophobic properties on the HNTs, the surfaces were modified employing hydrolytic condensation of hexadecyltrimethoxysilane and tetraethoxysilane which resulted in superhydrophobic HNTs, and subsequently incorporated into the PU films to produce superhydrophobic polyurethane coating (SHPUC) nanomaterials. Second, unmodified HNTs were incorporated into the PU films to produce unmodified HNTs PU coating (PHUC). Afterward, prilled urea pellets were repeatedly coated and cured under heat for about 8–10 min using SHPUC, PHUC, and normal PU films (PUC) at varying coating percentages, for comparative investigation [160]. It was shown that the urea pellets coated with SHPUC demonstrated the most efficient longevity (2 months longer) and controlled release of nitrogen. Further established was that coating rates directly influenced the release characteristics of the CRFs significantly, with the cumulative percent nitrogen release decreasing with an increasing percentage of the coating material. In addition, the percentage nitrogen release of SHPUC and PHUC compared to that of PUC was considerable slower even at the same coating percentages [160]. The technology of superhydrophobic surfaces and the nonwetting characteristics has enormous application possibilities in various research areas [162,163].

Elsewhere, an innovative CuO NPs encapsulated in a biobased polymeric shell derived from chitosan and sodium alginate is reported [164]. Drawing inspiration from the fact that metal oxide NPs can interact with amino and hydroxyl groups in biopolymeric systems, it therefore holds the possibility of preventing the leaching of the elements into the soil. Fabricated from environmentally friendly polyelectrolyte complexation, the chitosan and sodium alginate complex acted as the biodegradable shell for the controlled release of the encapsulated nutrients (*i.e.*, CuO NPs) [164]. The study demonstrated that optimizing the synthesis conditions, *e.g.*, pH and polymer/crosslinker ratio, good stability, and accurate size particle control was achieved. Furthermore, analytical investigation evidenced that the efficacy of the developed smart fertilizer on seeding and germination enhanced the advantages of seedling growth with a mutual positive impact in developing the epigeal and hypogeal parts [164]. Further demonstrating the endless possibilities in improving crop productivity and sustainable agriculture.

4.4.2 Food packaging and bioactive systems

Conventional materials employed in food and medicinal contact packaging have become a source of environmental and health concerns; thus, research and industrial efforts have intensified toward safer (greener), functional, biocompatible, biodegradable, sustainable, and efficient packaging systems [16,150]. Although challenges such as barrier and mechanical deficiencies have been noted as limitations for exploring biomaterials as effective packaging alternatives, the employment of nanocomposite concepts reverses the trend, expanding the possibilities and advancing the application potentials of biorenewable nanocomposites. For example, biorenewables such as collagen, gelatin, starch, cellulose, chitosan, and soy protein have been used to fabricate films and coatings as selective barriers to moisture, gas, and solvent toward enhancing food safety and antimicrobial activities [7]. In addition, water-soluble gums have found applications as edible coating for fruits and vegetables to limit postharvest losses, improve shelf-life of these perishables, and save the cost of electricity by minimizing or eliminating the need for cold storage [165]. Moreover, it has been shown that postharvest application of gum arabic, as edible coating, on fruits such as persimmon fruits during storage markedly reduced weight losses of the fruits, maintained plasma membrane integrity of the fruit tissues, and so on [166]. Elsewhere, it was reported that aloe vera gel (AVG), a biobased nanocomposite system, is an efficient and functional edible

coating for fruits and vegetable preservation as a result of the good antioxidative and antimicrobial properties [167]. It was further evidenced that overall that AVG has the capacity to retain the color of the coated food for any appreciable length of time, maintain the fruits and vegetable firmness and preserve much-needed vitamins, and improve on the ripening delay [167].

A freshwater durable and ocean water degradable cellulose nanofiber (CNF)-reinforced starch film for food packaging is reported in a related development [168]. By employing the wet pulverization process on cellulose powder and mechanical fibrillation, cellulose nanofibers (MCNFs) were obtained. These MCNFs were afterward mixed with a variation of modified tapioca starch powder and subsequently cast into films. These films demonstrated high efficiency as green(er) packaging films for a wide range of food types, hence opening opportunities for pursuing the blue seas initiative toward zero plastic in our oceans [168]. Another report showed that CNF isolated from pineapple leaves blended with potato starch as nanofillers was used to produce UV-resistant transparent nanocomposite films toward sustainable packaging [169]. It was determined that at 3 wt% loading of CNF, lower water sorption resulted in the films attributable to the crystallinity of the CNF reinforcement acting as a barrier to the starch molecules from swelling in the presence of water [169]. Notwithstanding, at higher CNF loading (≥ 3 wt%), the films began to lose their properties majorly due to possible agglomeration of the CNFs particles in the nanocomposite.

5 Conclusions

It is well known that nature solves material problems through the efficient and unique material designs, which can inspire humans to synthesize new materials for target applications. With the advancement of human civilization caused by the development of smart materials, portable devices, supercapacitors, wearable electronics, electric vehicles, and Internet of Things, biorenewables may be utilized in the fabrication of environmentally friendly, biocompatible, biodegradable, and renewable materials. As a result, it is necessary to expand our knowledge regarding biorenewables and their derivatives to achieve environmental sustainability. The application of green chemistry principles in the processing of bioderived materials for advanced technologies must be prioritized. The nonco-friendly and volatile chemicals used for the derivation, preparation, dissolution, and regeneration of biobased

materials should be urgently replaced with greener alternatives such as ionic liquids, which represent organic salts with unique physicochemical characteristics, high thermal and chemical stabilities, and good recyclability properties that allow mitigating several environmental and safety challenges. Furthermore, achieving a better fundamental understanding of the surface chemistry and interactions of bioderived materials as matrix systems containing fillers/additives is critical for expanding their application range in various fields. Therefore, additional studies must be performed to enhance the controlled properties of these materials for targeted applications.

6 Challenges and future prospects

The role of biorenewable materials in SDGs cannot be overemphasized, especially considering the current environmental challenges, energy security, and local growth of economies related to job creation and foreign exchange conservation. Nevertheless, multiple factors limit the use of biorenewables in domestic and industrial applications. One such challenge is the rapid biodegradation of biorenewables in target environments, which can be mitigated by modifying their chemical and surface properties to increase the biopersistence of these materials. Another problem is the inferior physiochemical and mechanical properties of biorenewables compared with those of the traditional materials obtained from fossil hydrocarbons. However, this drawback can be overcome by achieving a fundamental understanding of their biosynthesis and inherent chemical properties that could be adjusted to improve the physiochemical and mechanical characteristics of these materials. Furthermore, the surface properties and solubility of biorenewable materials as well as their interactions with solvents, additives, and/or fillers are another important factor that requires a proper understanding of fundamental theories, new insights, and more advanced processing technologies for ensuring optimal interactions between the matrix and fillers as well as material dissolution and regeneration/reconstitution. Owing to their unique chemical properties, which are not identical to those of fossil-based materials, biorenewable nanomaterials require better characterization techniques to determine their kinetic parameters and reaction mechanisms in various environments. In addition, the theory of chain dynamics and mobility is not fully understood yet and remains speculative despite the significance it holds for enhancing the mechanical properties of biobased nanocomposite materials.

In summary, our main question is whether a combination of two or more nanoscale materials with different structures, chemical properties, and other parameters can eliminate the constraints of the currently used materials. For example, what material can be obtained by embedding copper NPs into cellulose NPs and then into lignin nanospheres followed by a dispersion in a Kevlar matrix? What material would be produced by combining nanoscale cellulose with nanoscale lignin and nanoscale copper and dispersing the resultant NPs in PLA or furfuryl alcohol under microwave irradiation? In addition to elucidating the chemical and other properties of biorenewable nanomaterials, it is also necessary to determine how these properties can be optimized to fabricate superior materials for advanced applications in extreme environments.

Finally, the fabrication of multifunctional nanocomposites remains a very challenging task. Hence, further research is required to develop materials and systems with multifunctional capabilities in a single use based on a hierarchical structure, in which modified functionalities are integrated at different structural levels.

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