Applications of Nanocellulose/Nanocarbon Composites: Focus on Biotechnology and Medicine

Subjects: Nanoscience & Nanotechnology | Biotechnology & Applied Microbiology | Others

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Nanocellulose/nanocarbon composites are newly-emerging smart hybrid materials containing cellulose nanoparticles, such as nanofibrils and nanocrystals, and carbon nanoparticles, such as “classical” carbon allotropes (fullerenes, graphene, nanotubes and nanodiamonds), or other carbon nanostructures (carbon nanofibers, carbon quantum dots, activated carbon and carbon black). The nanocellulose component acts as a dispersing agent and homogeneously distribute the carbon nanoparticles in aqueous environment. Nanocellulose/nanocarbon composites can be prepared with many advantageous properties, such as high mechanical strength, flexibility, stretchability, tunable thermal and electrical conductivity, tunable optical transparency, photodynamic and photothermal activity, nanoporous character and high adsorption capacity. They are therefore promising for a wide range of industrial applications, such as energy generation, storage and conversion, water purification, food packaging, construction of fire retardants and shape memory devices. They also hold great promise for biomedical applications, such as radical scavenging, photodynamic and photothermal therapy of tumors and microbial infections, drug delivery, biosensorics, isolation of various biomolecules, electrical stimulation of damaged tissues (e.g. cardiac, neural), neural and bone tissue engineering, engineering of blood vessels and advanced wound dressing, e.g. with antimicrobial and antitumor activity. However, the potential cytotoxicity and immunogenicity of the composites and their components must also be taken into account.

nanofibrillated cellulose | cellulose nanocrystals | fullerenes | graphene | carbon nanotubes | diamond nanoparticles | sensors | drug delivery | tissue engineering | wound dressing

1. Introduction

Nanocellulose/nanocarbon composites are hybrid materials containing cellulose and carbon nanoparticles. Integration of nanocarbon materials with nanocellulose provides functionality of nanocarbons, using an eco-friendly, low-cost, strong, dimension-stable, nonmelting, nontoxic and nonmetal matrix or carrier, which alone has versatile applications in industry, biotechnology and biomedicine (for a review, see [1][2]). In addition to its advantageous combination with nanocarbon materials, nanocellulose is an appealing material for biomedical applications due to its tunable chemical properties, nonanimal origin, and resemblance to biological molecules in dimension, chemistry and viscoelastic properties, etc. [3][4][5][6].

2. Applications of Nanocellulose/Nanocarbon Composites: Focus on Biotechnology and Medicine

Cellulose nanomaterials include cellulose nanofibrils (CNFs) and cellulose nanocrystals (CNCs) [3]. CNFs are manufactured using either a bottom-up or a top-down approach. The bottom-up approach involves bacterial (*Gluconacetobacter*) biosynthesis to obtain bacterial cellulose (BC), while, in the top-down method, cellulosic biomass from plant fibers is disintegrated into smaller CNFs [7] that contain amorphous and crystalline regions [3]. The fibrillation of cellulose is achieved using mechanical forces, chemical treatments, enzymes or combinations of these. After fibrillation, the width of CNFs is typically between 3 and 100 nm, and the length can be several micrometers [8]. Separation of the crystalline parts from the amorphous regions of the fibers or fibrils to obtain CNCs typically requires acid hydrolysis, which destroys the amorphous regions [9]. Entangled CNFs are longer, while CNCs possess shorter needle- or rod-like morphology with a similar diameter and a more rigid molecule due to their higher crystallinity [3][9]. In general, the properties of nanocelluloses are variable and depend on their origin, type, processing, pretreatments and functionalization. Integration with other materials, as well as fabrication of the final product, further affects the properties of the resulting composite or hybrid structure.

Carbon nanoparticles include fullerenes (usually C_{60}), graphene-based particles (graphene, graphene oxide, reduced graphene oxide, graphene quantum dots), nanotubes (single-walled, double-walled, few-walled or multi-walled) and nanodiamonds (for a review, see [10][11][12][13][14][15][16][17][18][19][20]). The most frequently used nanocellulose/nanocarbon composites contain graphene or carbon nanotubes, while composites of nanocellulose with nanodiamond, and particularly with fullerenes, are less frequently used (Figures 1-4). Other carbon nanostructures, which are less frequently used in nanocellulose/nanocarbon composites, at least for biomedical applications, include carbon nanofibers [21][22][23][24][25], carbon quantum dots [26][27][28], activated carbon [29][30] and carbon black [31][32][33].

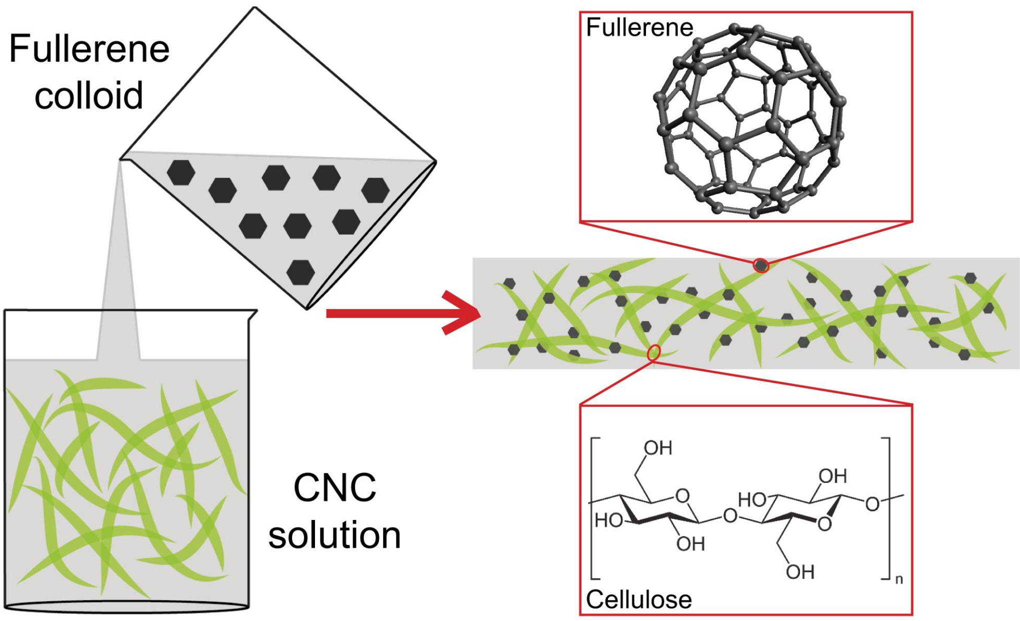

Figure 1. Scheme of the preparation and structure of nanocellulose/fullerene C_{60} composites

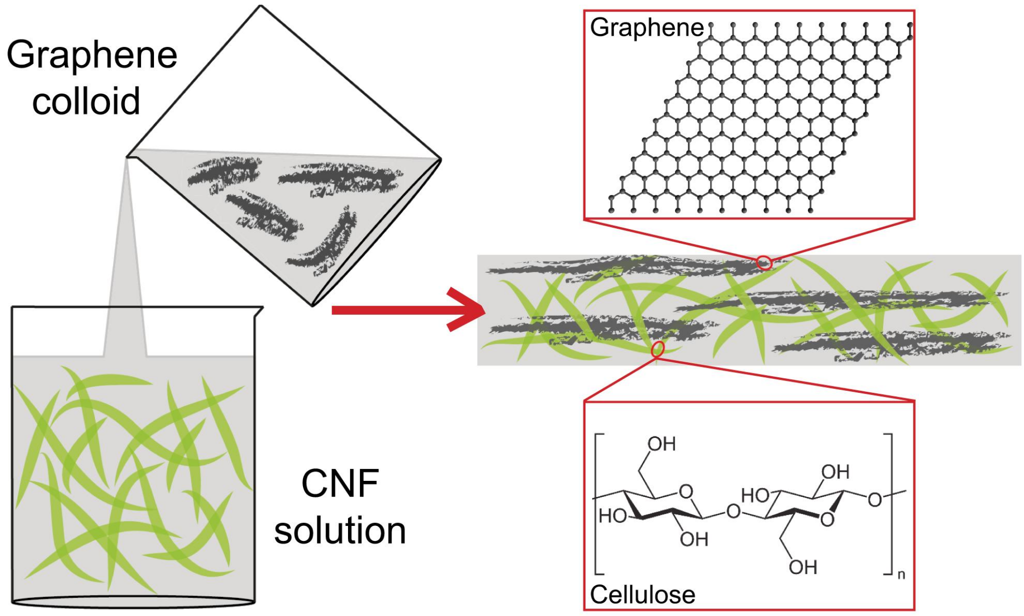

Figure 2. Scheme of the preparation and structure of nanocellulose/graphene composites

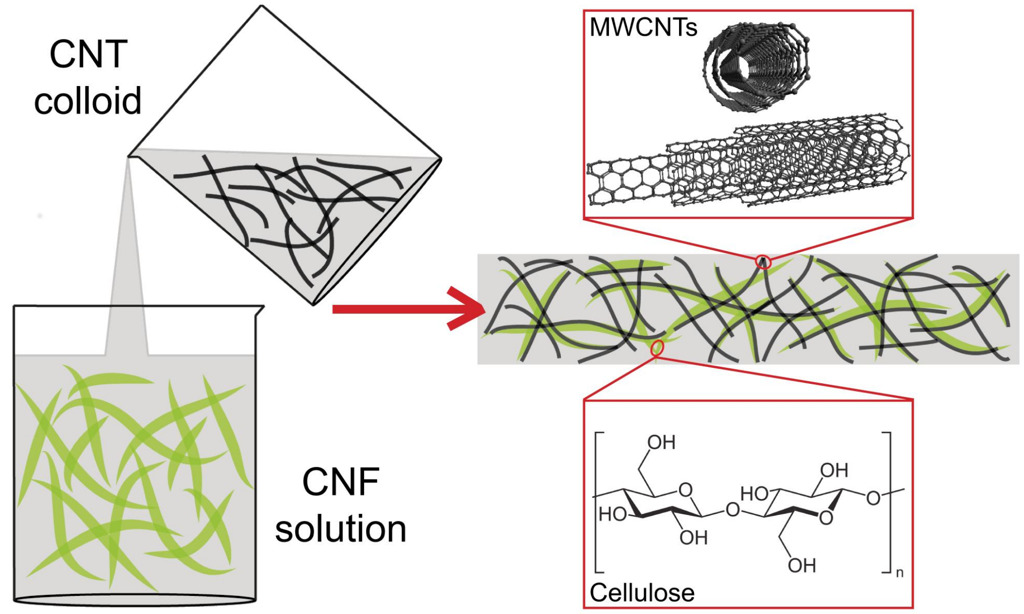

Figure 3. Scheme of the preparation and structure of nanocellulose/carbon nanotube composites

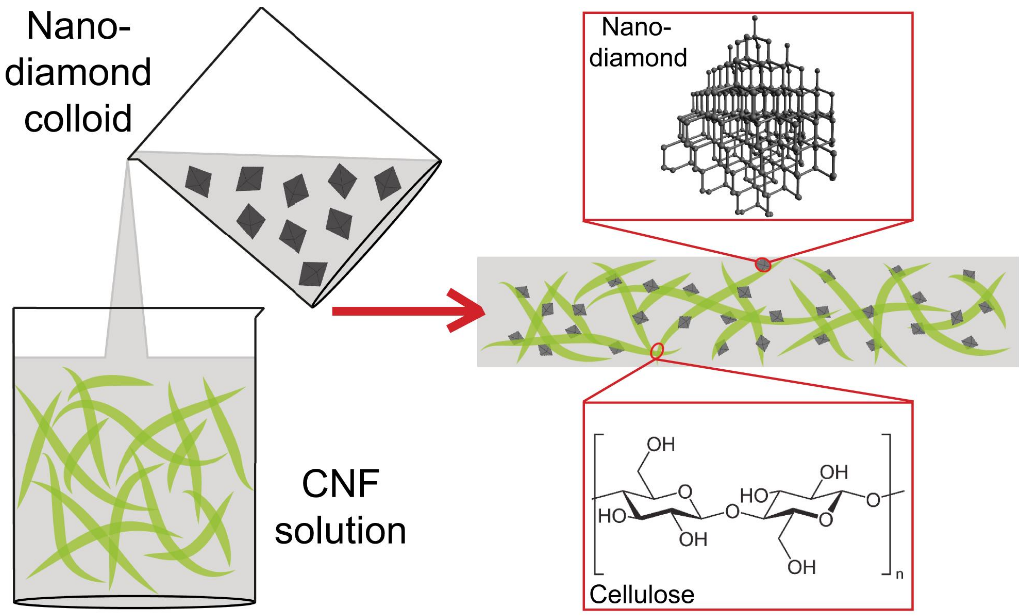

Figure 4. Scheme of the preparation and structure of nanocellulose/nanodiamond composites

Nanocellulose/nanocarbon composites can be prepared in one-dimensional (1D), two-dimensional (2D) or three-dimensional (3D) forms. 1D composites are represented, for example, by C_{60} fullerenes grafted onto cellulose nanocrystals that have undergone amination or oxidation [34][35]. 2D composites are represented by films, which can be self-standing or supported, i.e., in the form of free-standing membranes [29][36][37][38][39][40][41] or in the form of coatings deposited on bulk materials [33][42]. The films can be formed by depositing carbon nanoparticles on a nanocellulose layer [43][44]. More frequently, however, they are fabricated from aqueous dispersions of nanocellulose and carbon nanoparticles [39][42]. It should be pointed out that cellulose nanoparticles are excellent dispersive agents for carbon nanoparticles, as they prevent the aggregation of these nanoparticles and maintain them in long-term stable homogeneous suspensions without the need to subject them to chemical functionalization [45][46]. Suspensions of cellulose and carbon nanoparticles are also starting materials for the creation of 3D nanocellulose/nanocarbon composites in the form of aerogels, foams or sponges [45][47][48][49][50]. In addition, composite 3D scaffolds, especially for tissue engineering and for regenerative medicine, can be fabricated by 3D printing using bioinks based on cellulose and carbon nanoparticles [51][52]. Both 2D composites and 3D composites can also be created by adding carbon nanoparticles to cultures of cellulose-producing bacteria, such as *Gluconacetobacter xylinus*. These nanoparticles are then incorporated into bacterial nanocellulose in situ during its growth [53][54][55][56][57]. Another approach is via the electrospinning or wet spinning of solutions containing cellulose and carbon nanoparticles [58][59][60].

Nanocellulose/nanocarbon composites exhibit several more advantageous properties than materials containing only cellulose nanoparticles or only carbon nanoparticles. Adding carbon nanoparticles to nanocellulose materials can further increase their mechanical strength [59][61]. At the same time, the presence of nanocellulose promotes the flexibility and stretchability of the materials [52][62][63]; for a review, see [64]. Adding graphene, carbon nanotubes or boron-doped diamond nanoparticles endows nanocellulose materials with electrical conductivity [39][50][57][65][66]. Other advantageous properties of nanocellulose/nanocarbon composites include their thermal stability [67][68][69], tunable thermal conductivity and optical transparency [48][57][70], intrinsic fluorescence and luminescence [26][71][72] photothermal activity [56], hydrolytic stability [61], nanoporous character and high adsorption capacity [49][61]. Nanocellulose/nanocarbon composites can therefore be used in a wide range of industrial and technological applications, such as water purification [22][29][43][49][54][56][61][73][74][75][76], the isolation and separation of various molecules [22][74][77][78][79], energy generation, storage and conversion [21][23][44][47][64][80][81][82][83][84][85], biocatalysis [86], food packaging [67][68][69][87], construction of fire retardants [48], heat spreaders [70] and shape memory devices [38][88][89][90]. These composites are also used as fillers for various materials, usually polymers, in order to improve their mechanical, electrical and other physical and chemical properties [67][68][69][87][91].

In addition, nanocellulose/nanocarbon composites are promising for biomedical applications, though these applications are less frequent than industrial applications. Biomedical applications include radical scavenging [34][92], photothermal ablation of pathogenic bacteria [93], photodynamic and combined chemophotothermal therapy against cancer [35][94], drug delivery [16][28][65][72][95][96][97], biosensorics [31][32][33][63][66][71][91][98][99][100][101][102][103][104], and particularly tissue engineering and wound dressings. Hybrid materials containing nanocellulose and nanocarbons stimulated the growth and osteogenic differentiation of human bone marrow mesenchymal stem cells [37][59]. They provided good substrates for the attachment, growth and differentiation of SH-SHY5Y human neuroblastoma cells [51] and PC12 neural cells, particularly under electrical stimulation [105]. They enhanced the outgrowth of neurites from rat dorsal root ganglions in vitro and stimulated nerve regeneration in rats in vivo [106]. They also promoted the growth of vascular endothelial cells, enhanced angiogenesis and arteriogenesis in a chick chorioallantoic membrane model [107], and improved cardiac conduction when applied to surgically disrupted myocardium in dogs [52]. In addition, these materials supported the growth of human dermal fibroblasts [108] and mouse subcutaneous L929 fibroblasts [58][62], promoted wound healing in vivo in mice [109] and showed an antibacterial effect [30]. These materials are therefore promising for bone, neural and vascular tissue engineering, for creating cardiac patches and for advanced wound dressings. The biomedical applications of nanocellulose/nanocarbon composites are summarized in Table 1.

Table 1. Biomedical applications of nanocellulose/nanocarbon composites.

Application	Nanocellulose/Nanocarbon Composites Containing:				
	Fullerenes	**Graphene**	**CNTs**	**Nanodiamonds**	**Others**
Radical scavenging	NH_2-CNC/C_{60} [34];				

	CNC/$C_{60}(OH)_{30}$ [92]				
Photodynamic cancer therapy	TEMPO-oxidized CNC/C_{60}-NH_2 [35]				
Photothermal, chemo-photothermal therapy		Bacteria: [93]			
		Cancer: [94]			
Drug delivery		Anticancer drugs (doxorubicin) [72][95][96]	Anticancer and other drugs [16]	Anticancer drugs (doxorubicin) [97]	Carbon quantum dots: Anticancer drugs (temozolomide) [28]
(Bio)sensors		Electrochemical: cholesterol [98]; glucose and bacteria [110]; avian leucosis virus [111]; organic liquids [112]	Electrochemical: ATP metabolites [102]; oxygen [84]	Electrochemical: Biotin [66]	Carbon black: Electrochemical aptasensor for *S. aureus* [32]; electrochemical sensor for H_2O_2 [33]
		Piezoelectric: strain, human motion [63][99][113]	Piezoresistance and thermoelectric-based: pressure and temperature [103]; pressure [17]; strain, human motion [90][91]		Carbon black: Strain, human motion [31][33]

			[114]; humidity, human breath [104]		
	Optical: oxygen and temperature [115]; oxygen [116]	Optical: *SERS*: bilirubin [100]; *Fluorescence*: laccase [71]			Carbon quantum dots: optical sensor for biothiols [26]
		Acoustic: ammonia [101]			
Isolation of biomolecules		Histidine-rich proteins, hemoglobin [77]; bovine serum albumin [79]			
Electrical stimulation of tissues			Cardiac tissue [52]; neural tissue [106]		
Tissue engineering (TE)		General cell biocompatibility [68][69][87]; bone TE [37][59]; neural TE [105]; vascular TE [107]	Neural tissue engineering [51]; TE in general [117]		
Wound dressing/healing	Polysaccharides/ fullerene C_{60} derivatives [118][	Human dermal fibroblasts in vitro [108]; mouse model in vivo [109]		L929 fibroblasts in vitro [58][62]; HeLa cells in vitro, wound dressings	Activated carbon: antibacterial wound dressing [30]

	delivering doxorubicin [97]

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