Carbon Nanotubes-Based Hydrogels

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Carbonaceous materials, including carbon nanotubes (CNTs), have been widely explored in wound healing and other applications because of their superior physicochemical and potential biomedical properties to the nanoscale level. CNTs-based hydrogels are widely used for wound-healing and antibacterial applications. CNTs-based materials exhibited improved antimicrobial, antibacterial, adhesive, antioxidants, and mechanical properties, which are beneficial for the wound-healing process.

Keywords: carbon nanotubes (CNTs) ; conductive hydrogels ; antibacterial ; wound healing

1. Introduction

The annual wound care costs are estimated to be several billion dollars, which constitute approximately 2–3% total expenditure on health [1]. Wound healing is one of the most complex and essential treatments in the human body. Skin injury also occurs in daily life, where the skin loses its protective action, leading to the formation of a wound in the skin [2]. Multiple cell types are required within the damaged skin layers to accelerate the healing process, such as hemostasis/inflammation, angiogenesis, proliferation, and remodeling. Several therapies are available for the wound-healing process [3]. These therapies include both conventional and modern treatments. Some examples of modern and currently used treatments are stem cell therapy [4], oxygen therapy [5], nitric oxide therapy [6], artificial dressing [7], and growth factor therapy [8]. Comparatively, conventional treatments include using natural substances such as plant extracts, honey, propolis, larvae, etc. [9]. The treatments are effective when the healing materials are fabricated with excellent wound-healing potential. Different materials are developed for this process; still, few reports are available on more effective therapies [2][10]. The incorporation of nanomaterials with biocompatible polymers has been an emerging area for wound-healing application. Commonly, two types of approaches are used where nanomaterials either act as drug or vehicle to deliver the drug. Materials such as silver, gold, zinc, gold copper, titanium, and terbium are used in the form of nanoparticles to act as a drug. Whereas, in another approach, nanomaterials are used to deliver antibiotics, growth factors, nucleic acids, and antioxidants [11]. The design of effective materials for rapid wound healing is emerging, and nanomaterials-based hydrogels exhibited an improved healing process [12][13]. The utilization of inorganic nanomaterials for biomedical applications is an emerging research field. This research topic received significant attention from the researcher community to develop nanocarrier devices for medical applications. Nanomaterials (NM) are classified into four classes: carbon-based NM, inorganic-based NM, organic-based NM, and composite-based NM [14]. Several studies have investigated the development of inorganic conductive materials in combination with non-conductive polymers to support the proliferation and electro-stimuli responsive cell activities [2]. These materials are mainly carbon-based (CNTs and graphene oxide) and metals, which acquired the potential properties for biomedical applications. Since the last decade, CNTs-based nanomaterials have received significant interest from the scientific community and have been widely studied for broad research topics due to their excellent electrical, electronic, and physicochemical properties [15] for electrode materials, biomedical applications, biosensors, bio imaging, drug delivery, tissue engineering, wound healing, sorption materials, and catalysis [16]. Various CNTs composites and their biological applications are listed in **Table 1**. CNTs-based materials also exhibited antibacterial properties. Antibacterial materials can kill or suppress the growth of bacteria. CNTs are an allotrope of carbon and have high electrical and thermal conductivity and superior mechanical properties [17].

CNTs-based materials can be produced at a large scale that could be an additional advantage in developing industrial-scale biomedical devices [17][18]. Several studies have been conducted to explore the potential of CNTs-based materials in biomedical applications, drug delivery, tissue engineering, cancer therapies, implantable devices such as nanosensors, nanorobotics, antibacterial, and wound dressing [17][19][20]. However, there is broad scope for the utilization of CNTs-based materials in other clinical applications. In the wound-healing process, electro-conductive composites of CNTs and non-conductive polymeric materials have been efficiently utilized. The combination of intelligent hydrogels and CNTs was very effective in wound dressing and antibacterial treatment [21][22].

Table 1. List of CNTs-based composites and their biological applications.

CNT Composite	Highlight	Biological Application	Reference
CP@CNT	Examine CNT as a nanocarrier of drug CP	Cyclophosphamide, anticancer drug delivery, reduced side effects	[23]
CNT-Alg-Ch-FA	Penetration of functionalized CNT through cell membrane	Doxorubicin hydrochloride delivery for cancer treatment	[24]
CNT-UHMWPE	CNT incorporation showed high mechanical and tribological properties	Sustained release (up to 429 h) of gentamicin	[25]
CNT-3H_2PO_4	Binding energy of drug to CNT increased with more H_2PO_4moeities	Delivery for anti-osteoporosis zolendronate and risedronate drugs	[26]
PLGA-CNT-PDA-lam	PDA modified scaffold can adhere laminin for longer time and promote neurite outgrowth	Enhancement of PC12 cells for nerve tissue engineering	[27]
Rh-CNT	Analysis of gases of lung cancer (C_6H_6 and C_6H_7N)	Biosensor for prediagnosis of lung cancer	[28]

CP-cyclophosphamide; Alg-alginate; Ch-chitosan; FA-folic acid; UHMWPE-ultra-high molecular weight polyethylene; PLGA-polylactide-co-glycolide; PDA-polydopamine; lam-laminin; PC12-adrenal phaeochromocytoma cell line; Rh-rhodium.

Hydrogels belong to the group of biomaterials composed of a cross-linked polymer network to form 3D structures that can hold large proportions of water and retain their primary system even after swelling of the wound [21][29][30]. Nanomaterials-based hydrogels are considered an attractive platform for wound-healing applications [13]. These hydrogels demonstrated superior mechanical strength, antioxidant, antibacterial, electrical, and tissue regeneration potentials [31][32][33]. The physicochemical properties of nanomaterials-based hydrogels are profoundly affected by the surface functionalizations [22]. The appealing properties of multifunctional hydrogel make it a promising candidate for wound healing, antibacterial treatment, and other biomedical applications (**Figure 1**).

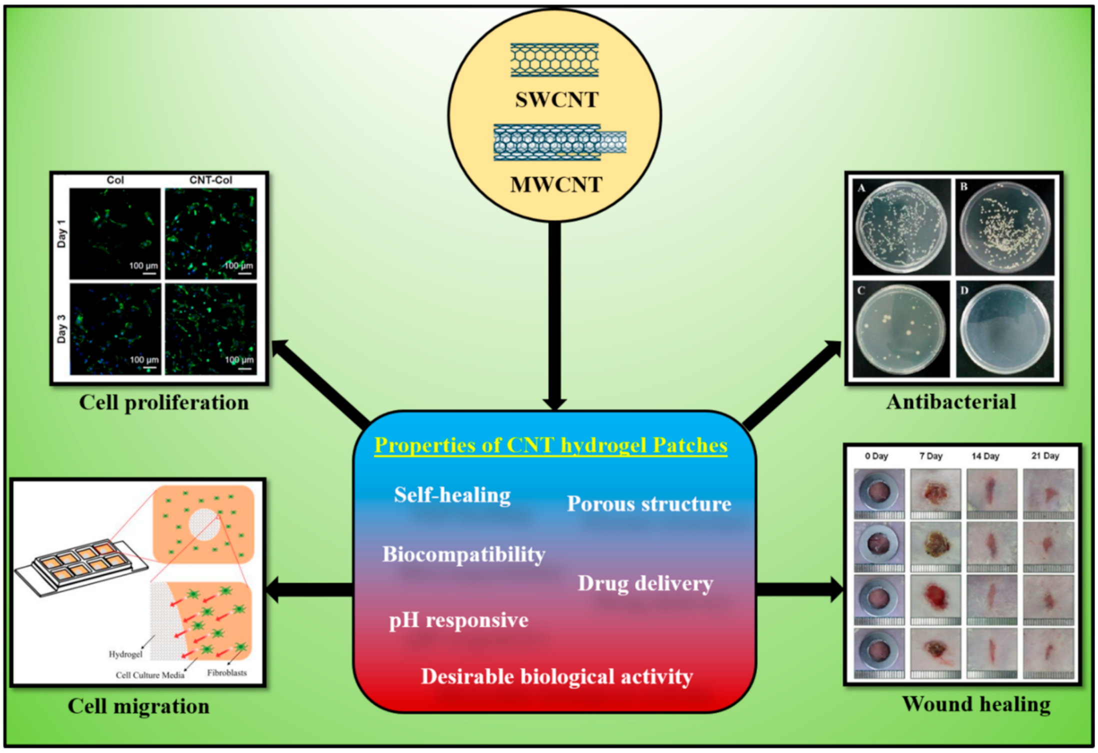

Figure 1. A scheme showing the types of CNT, properties of CNT hydrogel patches, and their application in cell proliferation [34], cell migration [35], antibacterial [36], and wound healing [37].

2. Conductive Properties of CNTs

CNTs are an allotropic form of carbon, as mentioned by SumioIijima in 1991 [38], which have similar properties as graphene. Structurally, CNTs are cylindrical structures composed by the rolling of graphene sheets with sp^2 hybridization. Arc discharge, laser ablation, and chemical vapor deposition (CVD) are commonly utilized to prepare CNTs [39]. In arc discharge and laser ablation methods, the carbon sourced was treated at 3000–4000 °C to generate cylindrical CNTs, whereas the CVD technique involves the pyrolysis of carbon source at a temperature range of 600–1100 °C. The physicochemical properties of the obtained CNTs are widely influenced by synthetic methods [40]. CNTs exhibit remarkable thermal properties due to their structural form and technique of synthesis. The conductivity range of CNTs can vary from 6000 to 0.1 W/mK depending upon the single-walled structure and multi-walled structure, respectively [41][42]. The thermal conductivity is due to the collective vibration of atoms, including phonon and electron transfer [43][44]. The length of CNTs also affects the conductivity [44][45]. Therefore, the optimization of synthetic parameters is required to obtain a certain level of thermal conductivity [46]. Berber et al., studied molecular dynamics simulation to determine the thermal conductivity (k = 6600 W/mK) of CNTs based on Tersoff–Brenner potential, which is similar to a hypothetical isolated graphene monolayer [47]. In comparison, Osman et al., studied the relationship between the physical parameters of CNTs and their thermal conductivity. They examined that the thermal conductivity of single-walled carbon nanotubes (SWCNTs) changes with the temperature. A decrease in the thermal conductivity of armchair (10,10) configured SWCNTs was observed above 400 K, similar to monolayered graphene. The CNTs with similar diameters but different chirality show maximum conductivity at 300 K, and the armchair CNTs have a comparatively sharper peak than zigzag CNTs [48]. **Table 2** shows various properties of single-walled carbon nanotubes (SWCNT) and multi-walled carbon nanotubes (MWCNT).

Table 2. Various properties of SWCNT and MWCNT [49].

Properties	Units	SWCNT	MWCNT
Specific gravity (bulk)	g/cm^3	0.8–1.3	1.8–2.6
Specific area	m^2/g	400–900	200–400
Young’s modulus	Pa	≈1000	≈1000

Properties	Units	SWCNT	MWCNT
Tensile strength	Pa	3.10^{10}–5.10^{11}	1.10^{10}–15.10^{10}
Thermal conductivity	W/m.K	3000–6000	2000–3000
Electrical conductivity	S/cm	10^{2}–10^{6}	10^{3}–10^{5}
Thermal stability temperature in air	°C	550–650	550–650

3. Development of CNT-Based Conductive Hydrogels

Hydrogels comprise a 3D cross-linked polymeric network structure, holding a large amount of water and maintaining their form even after swelling. Hydrogels are similar to the extracellular matrix and can mimic the microstructure of native cellular environments and provide a moist environment [21]. It can adsorb the fluid excreting from the wound through the porous structure and prevent the growth of anaerobic bacteria by the gaseous exchange phenomenon. It can act as a barrier to prevent bacterial infections and improve epithelization and cell migration into the wound [22][50][51][52][53][54]. Hence, hydrogel materials have been broadly used for wound dressing.Conductive hydrogel has received significant attention for various biomedical applications, including wound dressings, drug delivery, biosensors, bio-imaging, and tissue engineering. The hydrogel conductivity is originated due to the presence of conductive ions and electrons. Naturallyderived biopolymers, such as chitosan, peptides, gelatin, and polyamines, are frequently explored to prepare hydrogels due to their excellent biocompatibility [29]. Recently, Zaho et al., developed injectable antibacterial hydrogels for enhanced skin generation. The injectable hydrogel has certain advantages, such as wound site filling with irregular space, wound adherence, and feasibility toward the insitu encapsulation of bioactive molecules and cells [30]. These hydrogels can be designed with advanced features such as bilayer types to control the infiltration of microbes and moistures and additional antibacterial and antimicrobial properties to protect wound sites from infections and inflammations. Different stimuli-responsive properties can be generated in hydrogel to deliver therapeutic molecules, inhibit bacterial infections, and promote cellular proliferation. Multifunctional hydrogels are an emerging approach. It is the composition of hydrogel constituent materials with or without modification and the incorporation of nanostructures in the hydrogels. These construction strategies improve properties (electrical, mechanical, surface, biocompatibility, and biodegradability) for wound-healing applications [55][56][57][58][59].

Zhang et al., summarized the literature to fabricate different types of hydrogel material with antibacterial agents for wound-healing applications [29]. They included the types of hydrogels and their specific methods of incorporations such as physical combination (incorporation, swelling diffusion, encapsulated with carrier), chemical combinations (chemical bonding, hydrogel with biomedicine activity), and incorporation of photo-assisted antibacterial hydrogels. The physical combination of antimicrobial methods has been considered a straightforward and effective method, in which hydrogel was physically incorporated with antibiotics, biological extracts, antimicrobial peptides, and inorganic nanoparticles. These materials are utilized for wound healing and other biomedical treatments. In chemical combinations, the hydrogel is combined with an antibacterial and antimicrobial agent by chemical treatments. However, these hydrogel derivatives should go through further purification steps before their utilizations. The chemical combination method has been commonly used to synthesize CNT-based hydrogels, and the most common hydrogels are chitosan-based, cationic group-based, polypeptide-based, and halogen-based hydrogels. These hydrogels exhibit strong antibacterial and antimicrobial properties, which are necessary to heal the wound [53] effectively. However, several researchers are developing new hydrogel materials to fulfill their demand in biomedical applications [29][60][61][62][63]. The following sections highlight some recent studies and developments of CNT hydrogels for wound healing and antibacterial property.

As discussed before, CNT functionalization with hydrogel is considered a potential hybrid candidate material for multiple applications in biomedical fields. CNT can be incorporated with hydrogels through their covalent and non-covalent functionalization using different treatment (chemical and mechanical) techniques [64]. Recently, Vashist et al., have nicely summarized the studies on developing CNT-based hybrid hydrogel for wound-healing applications [13]. In addition, they addressed most of the design and synthesis strategies for CNT hydrogel mixed materials and their diverse applications in the biomedical field.

CNT hydrogels are associated with most of the properties of hydrogel polymeric networks. Generally, five polymerization techniques such as (i) covalent cross-linking (insitupolymerization), (ii) exsitu polymerization, (iii) physical cross-linking, (iv) polymer grafting, and (v) smart devices enable techniques that are exploited for the synthesis of CNT hydrogel hybrid. In both insitu and exsitu polymerizations (chemical treatment), CNT hydrogel outfits with excellent mechanical strength, a variety of shapes and surfaces, excellent yields, and it is easy to control the initial and final composition of hybrid gels into the hybrid [65][66]. These techniques involve the introduction of nanofiller during reactions. In physical cross-linking techniques, hydrogels are physically cross-linked with CNT, which showed a high level of biocompatibility but low mechanical strength. These methods have been employed for the synthesis of CNT-based gelatin hydrogels [67]. The schematic representation for the synthesis of CNTs-based hydrogels is shown in **Figure 2**.

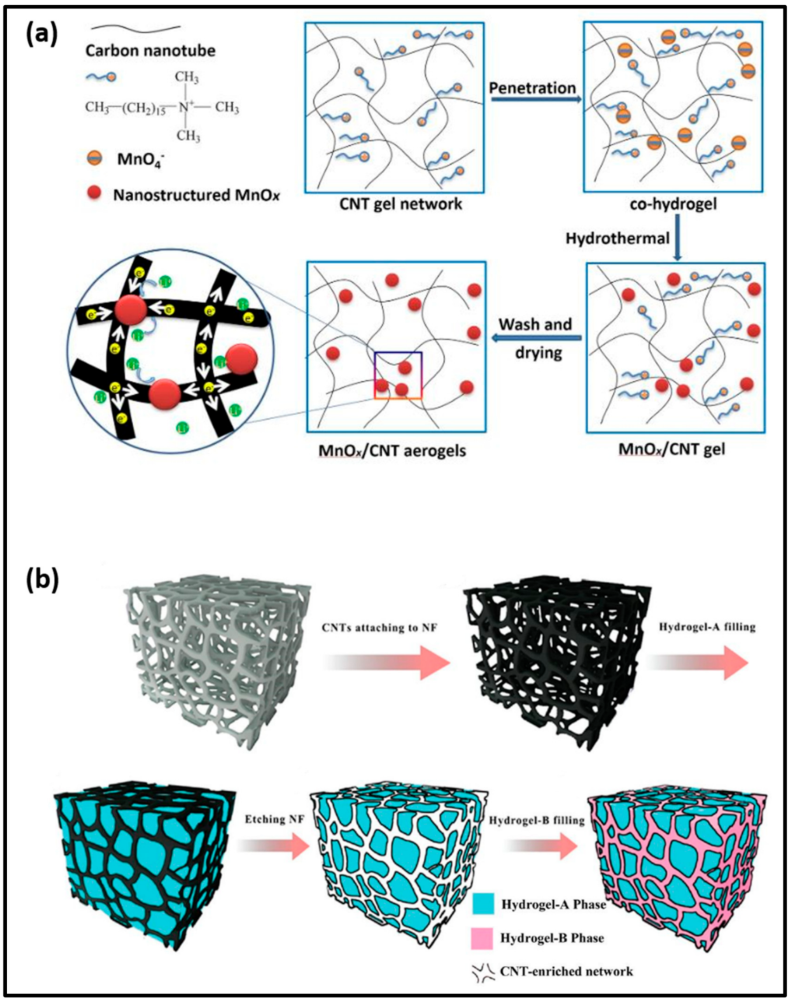

Figure 2. Scheme showing the synthesis of CNT-based hydrogels: (**a**) MnO_x/CNT aerogels [68], (**b**) general process of the double network hydrogel [69].

Polymer grafting is employed for the synthesis of CNTs grafted hydrogels. This technique provides the functionalization of the CNTs surface with a polymeric chain, which results in the shielding of the CNT surface [13][70]. This method offers a good dispersion of nanofillers along with their stronger interaction with polymers. Generally, protein and polypeptide-based CNT hydrogels have been synthesized through grafting techniques. Grafting-to and grafting-from polymerization for CNT polymers are two different grafting strategies involved in fabricating CNT hydrogels. In both approaches, the functional groups of the polymer chains are reacted with CNTs, leading to the formation of bioactive materials for biomedical applications. In addition, the intelligent device enable technique is an exciting technique that was applied to develop responsive hydrogel nanocomposites-based smart devices [71]. This method has been widely applied for the synthesis of hydrogel materials that are pH and ionic responsive. Significant research on the development of CNTs-based hydrogels has been done to obtain the desired functionality, shape, and size for specific applications.

4. CNT-Based Antibacterial Applications

It is well known that the wounded areas are more susceptible to bacterial infections. Depending upon the etiology and severity of the microbial invasion, the infections can cause minor to major damage to human life [72]. In the early stage of infection, the Gram-positive bacteria such as *Staphylococcus aureus* and *Streptococcus pyogenes* are more populated, whereas the Gram-negative bacteria, such as *Escherichia coli* (*E. coli*) and *Pseudomonas aeruginosa* (*P. aeruginosa*), are populated later nearby wounded areas [73]. Usually, infection is avoided by activating the immune system for abolishing the invading pathogens. In this process, macrophages initiate the migration to the wound site and subsequently perform phagocytosis of the pathogens (destroyed in a phagolysosome or by nitric acid production). In a later stage of infection, the immune response is performed by activating T-helper lymphocytes, which secrete interferon-γ and CD40 ligand to coordinate the immune adaptive and humoral response to kill and remove the invading bacteria [74]. In the passive stage of the immune system, infection occurs and causes the deterioration of granulation tissue, growth factors, and extracellular matrix components (collagen, elastin, and fibrin) and alters the normal wound-healing process [75][76]. Therefore, it is necessary to develop wound dressing materials to prevent bacterial penetration into the wound or reduce the microorganism's growth [77].

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