

Review of Silver Nanoparticles (AgNPs)-Cellulose Antibacterial Composites

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With the improvement of living standards, the human demand for antibacterial materials has increased. Cellulose, as the most abundant polymer in the world, is natural, biodegradable, and renewable, which makes it a promising raw material for the production of antibacterial materials. Silver nanoparticles (AgNPs)-cellulose antibacterial composites exhibit good biocompatibility and antimicrobial properties. These materials are easily degraded chemically and are environmentally friendly. Therefore, the AgNPs-cellulose antibacterial composites exhibit broad utilization prospects in environmental protection, medicine, chemical catalysis, and other fields. Several methods are used to manufacture such materials. This paper reviews three common techniques: the physical method, the *in situ* chemical reduction method, and the covalent bonding method. The differences and relationships are identified, and the advantages and disadvantages are compared among these three methods. Lastly, the present situation and the development potential of the AgNPs-cellulose antibacterial composites are discussed in this review.

Keywords: Silver nanoparticles; Cellulose; Cellulosic antibacterial materials

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INTRODUCTION

With the improvement of environmental awareness, research for development of biodegradable materials from renewable sources is increasing. Biopolymers, which are less expensive and occur in abundance in nature, have become the focus. A representative of biopolymers presenting these advantages is cellulose (Cherian *et al.* 2011). However, similar to the great majority of biopolymers, the characteristics of cellulosic materials determine that they can undergo bacterial attachment (Pasmore *et al.* 2001). Since cellulosic materials are commonly used in biomedical fields, the spreading of infections within hospitals has motivated scientists to develop new and efficient antibacterial materials for fighting infections, more especially as components of wound dressings and antifouling coatings (Drogat *et al.* 2011; Tsai *et al.* 2017). Fortunately cellulose and its derivatives have been demonstrated to be good materials for functionalization, and it is a favorable base material to address the antibacterial problem (Shin *et al.* 2008; Díez *et al.* 2011).

Cellulosic antibacterial materials generally can be described as the combination of cellulose and antimicrobials. This combination endows new properties and expands the

applications of cellulosic materials (Bakumov *et al.* 2007; Li *et al.* 2009). Generally, there are three types of common antimicrobials: inorganic (Xu *et al.* 2016), organic (Roy *et al.* 2008), and natural (Alonso *et al.* 2009). The main raw materials of organic antimicrobials are lipids, alcohols, and phenols, such as quaternary ammonium salts, ethanol, metformin, formaldehyde, organic halogen compounds, organic metal, pyridine, imidazole, haloalkane, and iodide. Although organic antimicrobials are widely used in all aspects of life, due to their bactericidal, antiseptic, and mildew-proof effects, their applications in the fields of textile, health, medicine, and some other areas are limited due to their intrinsic toxicity. Natural biological antimicrobials, which mainly come from plant extraction, include amino acids, natural peptides, and polysaccharides and are abundant and environmentally friendly. However, natural biological antimicrobials require a complex preparation process and have higher costs, which restricts the research and applications of these antimicrobials. Represented by silver, zinc, and copper, the inorganic antimicrobials, with high antimicrobial activity and low toxicity, have gained extensive attention (Jensen *et al.* 2000; Métraux and Mirkin 2005; Torres *et al.* 2007; Tao *et al.* 2008; Liu and Hurt 2010; Zhang *et al.* 2011; Henzie *et al.* 2012). Among these antimicrobials, silver nanoparticles (AgNPs), which has been demonstrated to possess excellent antibacterial activity through mechanisms involving the release of Ag⁺ ions that affect the replication of DNA (Marini *et al.* 2007) or the collapse of the proton-motive force across the cytoplasmic membrane (Holt and Bard 2005), have become a target of great interest for their relatively nontoxicity to human cells (Vimala *et al.* 2010).

Cellulose, the most abundant renewable resource on earth, mainly comes from plants and bacteria. Multifarious cellulosic materials, such as nanofibrillated cellulose (NFC) or regenerated cellulose (RC), can be obtained *via* physical, chemical, or other methods. Hence, there is an enormous potential for the development of AgNPs-cellulose antibacterial composites.

This paper reviews three methods for the combination of cellulose and AgNPs, *i.e.*, physical methods, the *in situ* chemical reduction method, and covalent bonding methods. The differences and relationships between these methods, as well as their advantages and disadvantages, are compared. Furthermore, the specific forms and applications of AgNPs-cellulose antibacterial composites are discussed. This paper aims to promote the development of AgNPs-cellulose antibacterial composites.

PHYSICAL METHODS FOR THE PREPARATION OF AgNPs-CELLULOSE COMPOSITES

The physical methods can be classified as either wet or dry processes according to whether there is water medium during the production process. There is no morphology requirement for raw material in wet process; however, it is generally a block in the dry process.

Wet Process

The wet process includes the adsorption characteristics of natural cellulosic materials. The cellulosic materials and nano-silver colloid are prepared separately. The nano-silver colloid can be obtained by varied methods, such as chemical reduction (Chen *et al.* 2002; Hao *et al.* 2002; Anderson *et al.* 2014), photoinduction (Jin *et al.* 2001; Callegari *et al.* 2003; Jin *et al.* 2003; Basuny *et al.* 2015), or electrochemistry (Braun *et al.*

1998). Then AgNPs are blended with cellulosic materials, without chemical reactions. The cellulosic materials merely serve as a carrier or matrix. Figure 1 illustrates the basic principle of this method. AgNPs-cellulose antibacterial composites should be prepared according to the expected product types (such as membrane, fiber, or powder) and the categories of raw cellulosic material, as reviewed below.

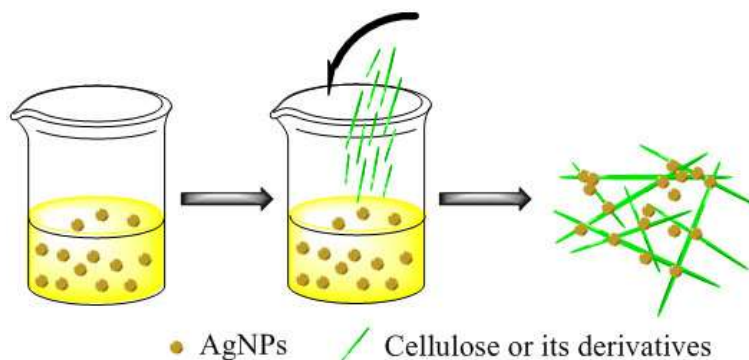


Fig. 1. The wet process

Membrane type products

Cellulosic membranes are widely used in ultrafiltration, microfiltration, reverse osmosis, and forward osmosis processes (Shibata 2004). Due to the rapid development of novel cellulose solvents, regenerated cellulose (RC) membranes have captured extensive attention. Typically, there are two approaches to load AgNPs onto cellulose membrane materials.

RC membranes can be prepared before the AgNPs are loaded (Ahamed *et al.* 2015). Benavente *et al.* (2017) fabricated RC membranes and nano-silver colloids. The AgNPs-containing RC membrane products were obtained by dipping the RC membranes into the nano-silver colloid. However, the disadvantage of this approach is that the AgNPs cannot enter the interior of the RC membrane.

The second method solves the above problem. The AgNPs and the cellulose solution are mixed before the casting of the membrane. For instance, AgNPs can be prepared using the modified Tollens' process and added into the cellulose-alkali/urea solution to obtain an approximately 0.5 mm-thick AgNPs-containing RC membrane (Chook *et al.* 2012). The AgNPs and cellulose are mixed uniformly in the homogenous cellulose solution. Thus, the AgNPs are incorporated into the interior of the RC membrane during the casting process. The final RC has antibacterial ability for a long period.

Due to alternative solvent systems, cellulose derivatives are more favorable for fabricating membrane materials, such as the AgNPs-containing hydroxypropyl methylcellulose membrane (Lloret *et al.* 2012). In addition, Caloca *et al.* (2017) fabricated AgNPs-containing polyethylene glycol/cellulose acetate ultrafiltration membranes by filtering nano-silver colloid through the polymer membrane. Faria *et al.* (2017) proposed the fabrication of antimicrobial membranes through the incorporation of graphene oxide-silver nanocomposites into a cellulose acetate polymeric matrix. The membranes presented strong antibacterial activity, being able to inactivate adhered bacteria at a rate of 90% compared to pristine cellulose acetate membranes.

Fiber type products

In the field of health care, fibers characteristics play an important role in the final product performance. Medical cloth items, such as surgical gowns, will have antibacterial ability if the fibers are loaded with AgNPs. Several reports have focused on the AgNPs-loading modification of cellulosic fibers. Raghavendra *et al.* (2013) fabricated a AgNPs-containing mixed solution with arabic gum and guar gum as reducing agents. The cellulose-Ag compound fibers were obtained by immersing cotton fibers in the mixed solution. Csóka *et al.* (2012) prepared a AgNPs colloid by a modified Tollens method using D-glucose as the reduction agent. The silver colloid solution was added to a water suspension of cellulose fibers to prepare fiber sheets.

Cellulose acetate (CA) is a thermoplastic resin derived from natural fibers. Based on the wide utilization of CA in varied areas such as the pharmaceutical and textile industries, it seems necessary to prepare AgNPs-containing CA fibers. Kendouli *et al.* (2014) prepared a AgNPs colloid *via* polyhydric alcohols reduction, and the AgNPs colloid was further blended with a CA solution to obtain AgNPs-containing CA fibers by electrospinning. They also proposed another approach where prepared CA fibers are immersed into a AgNPs colloid.

Powdery type products

Powdery material is usually employed to fill the other main component, *i.e.* a mobile matrix. Accordingly, the antibacterial properties of compound materials can be achieved by loading AgNPs into the powdery material. Martins *et al.* (2012) prepared a nano-silver colloid *via* glucose reduction. The surface modified NFC was mixed with the AgNPs colloid, and the obtained powdery compounds were used as fillers in starch-based coating formulations to produce antimicrobial paper products.

Dry Process

Wet treatments can cause environmental concerns and high water and energy consumption. Dry routes including sputtering are considered ecofriendly processes and offer the advantage of modifying only the material surface.

Recently, a novel technique for incorporating AgNPs into paper surface using a flame pyrolysis procedure has been proposed by Brobbey *et al.* (2017). The technique is known as Liquid Flame Spray. This method demonstrates a dry synthesis approach for depositing AgNPs directly onto paper surface in a process which produces no effluents. The production technique is scalable for industrial production of antibacterial paper. Irfan *et al.* (2017) also employed the dry process but to prepare antimicrobial functionalized cotton fabric. They deposited antimicrobial silver nanoclusters/silica composite on cotton fabric by radio frequency co-sputtering method. The study is expected to be applied to surgeon gowns in the future.

Summary

In general, the advantages of the physical method, *i.e.*, its convenient operation process and relatively high silver loading ratio, are remarkable. However, due to the relatively weak bonding of adsorption, the AgNPs adsorbed on the surface of cellulosic materials may be partially lost during the utilization process. In other words, the products prepared by this method have poor laundering durability (Ilić *et al.* 2009). Thus, the antibacterial potential may be restricted to a certain degree. Separately, in wet process an obvious disadvantage of this method is that the use of reducing agents can be toxic and

undesirable. In addition, there is heavy water consumption and waste water production with hazardous chemicals. On the contrary, dry routes avoid these problems perfectly. It is an environmentally friendly method and conforms to the needs of industrial production.

IN SITU CHEMICAL REDUCTION METHOD FOR THE PREPARATION OF AgNPs-CELLULOSE COMPOSITES

In situ chemical reduction has been widely used to overcome the inefficiency of physical adsorption. Compared with physical adsorption, *in situ* chemical reduction involves the slow growth of AgNPs in cellulosic materials. In other words, having an ionic radius of about 0.1 nm, the silver ions can pass into the interior of cellulosic materials. After the AgNPs have increased in size, they were wrapped by the skeleton of cellulosic materials and could not drop out. Thus the AgNPs can grow *in situ* in the interior of materials.

Based on whether the cellulosic materials participate in a redox reaction, the *in situ* chemical reduction method can be divided into two branches. First, cellulosic materials can serve as a matrix for the growth of AgNPs. In this case, there is no chemical reaction between the cellulosic materials and the silver precursor. Second, chemical reactions occur between cellulose (or its derivatives) and a silver precursor. In that case, the cellulosic materials play a dual role of reducing agent and matrix.

Cellulosic Materials as a Matrix

Because the cellulosic materials do not participate in a chemical reaction in this approach, an additional reducing agent must be added to reduce the silver ions.

A one-step method can be used to prepare powder/fibers from cellulosic materials, such as microcrystalline cellulose (MCC) powder (Vivekanandhan *et al.* 2012). Nanocrystalline cellulose (NCC) can serve as the matrix, allowing glucose to reduce Tollen's reagent to produce AgNPs at room temperature (Wang *et al.* 2016). Pinto *et al.* (2009) prepared cellulose nanocomposites by adding a solution of AgNO₃ drop-wise into an ice-cold NaBH₄ solution containing the cellulose and stirring vigorously over 2 h. Gaminian and Montazer (2017) decorated AgNPs on electrospun cellulose nanofibers (CNFs) through a facile method. The CNFs were treated with silver nitrate, ammonia, and sodium hydroxide and subsequently with dopamine as reducing and adhesive agent. Ag ions on the CNF surface were photo-reduced to AgNPs using UVA irradiation to produce a dense layer of AgNPs on the nanofibers. This approach has been employed widely due to its convenient operation (Son *et al.* 2006; Li *et al.* 2011a; Li *et al.* 2011b; Liu *et al.* 2011; Jang *et al.* 2014; Li *et al.* 2016; Prema *et al.* 2017). However, the silver ions and reducing agent are added in a system, which partially hinders the *in situ* synthesis of AgNPs in cellulosic materials. This potential hindrance is the main disadvantage of this method.

Some bulk form AgNPs-containing cellulosic materials, such as membrane, paper, or gel, and can be prepared by a relatively complex two-step method. Figure 2 shows the general operation, which includes the following two steps: i) cellulosic material is immersed in a silver precursor solution to adsorb adequate Ag ions, and the loose Ag ions on the surface of the cellulosic material are washed away by deionized water; and ii) the cellulosic material containing Ag ions is added into a reducing agent solution to form AgNPs. In most cases, silver nitrate is selected as a silver precursor, whereas the reducing agent can be chosen from varied substances, such as sodium borohydride (He *et al.* 2003;

He *et al.* 2005; Luong *et al.* 2008; Maneerung *et al.* 2008; Zhu *et al.* 2009; Ahmad *et al.* 2016; Yan *et al.* 2016), sodium citrate (Tankhiwale and Bajpai 2009; Hebeish *et al.* 2013), triethanolamine (Barud *et al.* 2008; Barud *et al.* 2011), glucose (Chook *et al.* 2017), chitosan sulfate (Breitwieser *et al.* 2013), *Cassia alata* leaf extract (Sivaranjana *et al.* 2017), bioflocculant (Muthulakshmi *et al.* 2017), UV-irradiation (Rehan *et al.* 2017), hydrazine, hydroxylamine, or ascorbic acid (Maria *et al.* 2009). In this method, silver ions are adsorbed inside the cellulosic materials when the materials are soaked in silver nitrate solution. The silver ions are further *in situ* reduced to AgNPs, and the AgNPs grow up in the reductive solution. During *in situ* reduction, the particle size of the AgNPs is controlled by adjusting the soak time in the reducing agent solution.

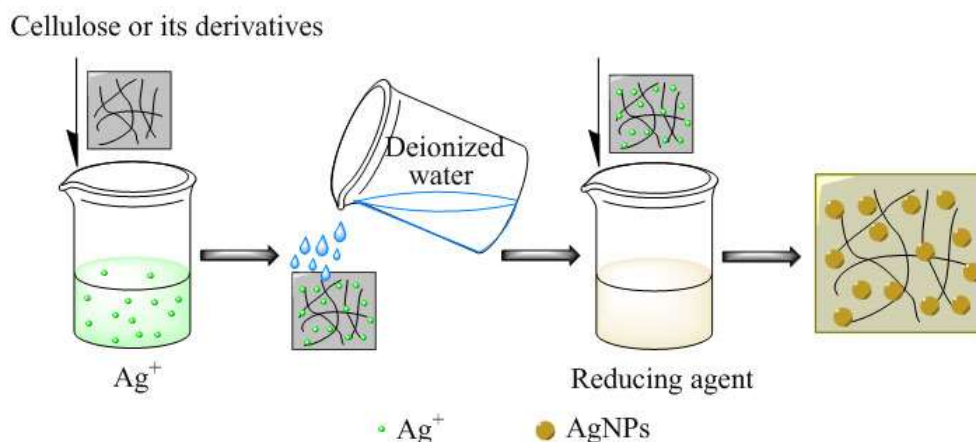


Fig. 2. The two-step method

Cellulosic Materials Serve as Both a Matrix and a Reducing Agent

A remarkable characteristic of this method is that it has no reducing agents other than the cellulosic material. Thus, an advantage of this method is that it reduces the consumption of chemicals. This approach is illustrated in Fig. 3.

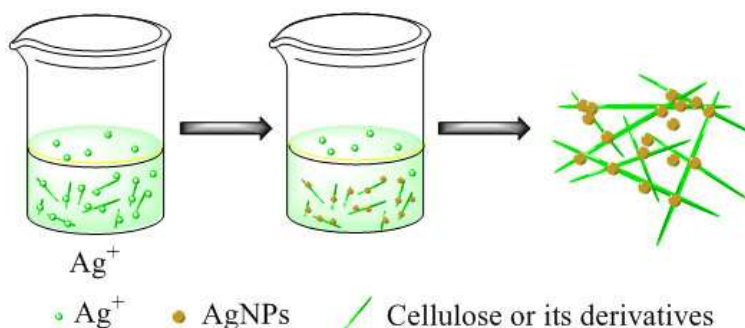


Fig. 3. *In situ* chemical reduction method, in which cellulosic materials serve as both a matrix and a reducing agent

Due to the intrinsic reducibility of cellulose, either natural cellulose, RC, or some of cellulose derivatives can be employed as a reducing agent without any processing. For example, the AgNPs embedded cellulose films or microspheres were fabricated by immersing the materials in an aqueous AgNO_3 solution at 80 °C for 24 h (Wu *et al.* 2012). Wu *et al.* (2014a) developed a similar method to synthesize and impregnate AgNPs onto

bacterial cellulose (BC) nanofibers *via* immersing the BC nanofibers into a silver ammonia solution at 80 °C for 10 min. Kolarova *et al.* (2017) prepared AgNPs-cellulose composite film by reduction of silver nitrate by methyl cellulose. Elayaraja *et al.* (2017) generated AgNPs by immersing TEMPO (2,2,6,6-tetramethylpiperidine-1-oxyl radical)-oxidized BC in AgNO₃ solution and keeping it the dark over night at 40 °C. The prepared AgNP deposited BC was a promising alternative to control the shrimp pathogen. Because the reducibility of natural cellulose is usually very weak, additional energy, *e.g.*, microwave heating (Oluwafemi *et al.* 2016; Xu *et al.* 2016) or water bath heating (Yang *et al.* 2012a; Emam *et al.* 2014; Wu *et al.* 2014b), may be necessary during the redox reaction between cellulose and silver ions.

Natural cellulose can be introduced in specific reductive groups for the *in situ* reduction of silver ions. Wu *et al.* (2008) fabricated dialdehyde cellulose *via* a periodate oxidation reaction, and reductive aldehyde groups were successfully introduced. Cheng *et al.* (2013) prepared AgNPs/cellulose compounds using aminocellulose as a combined reducing and capping reagent.

Summary

In physical methods, nano-silver colloid has to be prepared in advance. However, there is no need for such preparation before the *in situ* chemical reduction method. Compared with physical adsorption, *in situ* chemical reduction is simpler, and the silver loading ratio is improved to a certain extent. Furthermore, the antibacterial endurance of AgNPs-containing cellulosic materials is effectively enhanced due to the internal loading of AgNPs within cellulosic materials. Especially, when cellulose itself plays a role as reducing agent, the process is conducted without using ordinary reductants, which often are hazardous.

COVALENT BONDING METHOD FOR THE PREPARATION OF AgNPs-CELLULOSE COMPOSITES

Because the fixation ratio of AgNPs and the antibacterial endurance of AgNPs-containing cellulose fibers are influenced by the interaction between fibers and particles, a strong and effective bridge should be established to enhance the combination between cellulosic materials and AgNPs. Thus, the covalent bonding method has been examined in a preliminary study. The procedures of the covalent bonding method are as follows: i) AgNPs are immobilized by a suitable dendrimer; and ii) the covalent bonds between polymer-parceled AgNPs and modified cellulose are formed during the crosslinking reaction process.

Zhang *et al.* (2013) firstly prepared amino functional AgNPs by a one-step reaction between silver nitrate and amino-terminated hyperbranched polymer (HBP-NH₂). And then the amino functional AgNPs were grafted on the NaIO₄ oxidized cotton fabric. The preparation process is shown in Fig. 4. The AgNPs grafted oxidized cotton fabric showed excellent antibacterial property and laundering durability. After exposing to 50 consecutive home washing conditions, the Ag content of AgNPs grafted oxidized cotton fabric only decreased from 149.88 to 116.08 mg/kg, and the bacterial reduction was maintained over 96% against both *S. aureus* and *E. coli*.

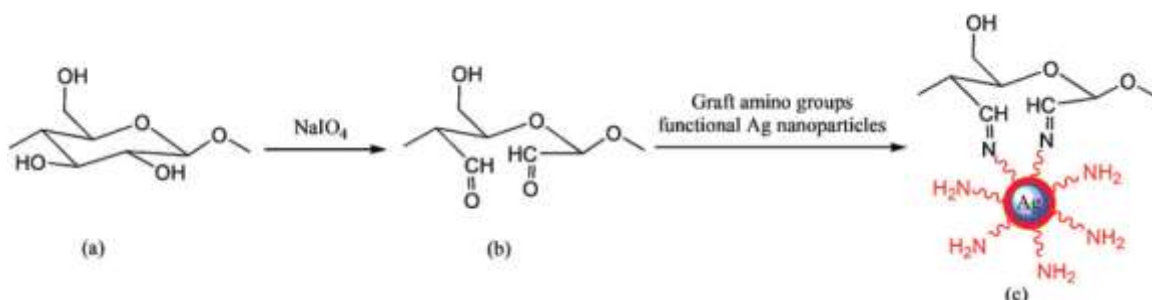


Fig. 4. The preparation of AgNPs grafted oxidized cotton fabric (reprinted with permission from Zhang *et al.* 2013)

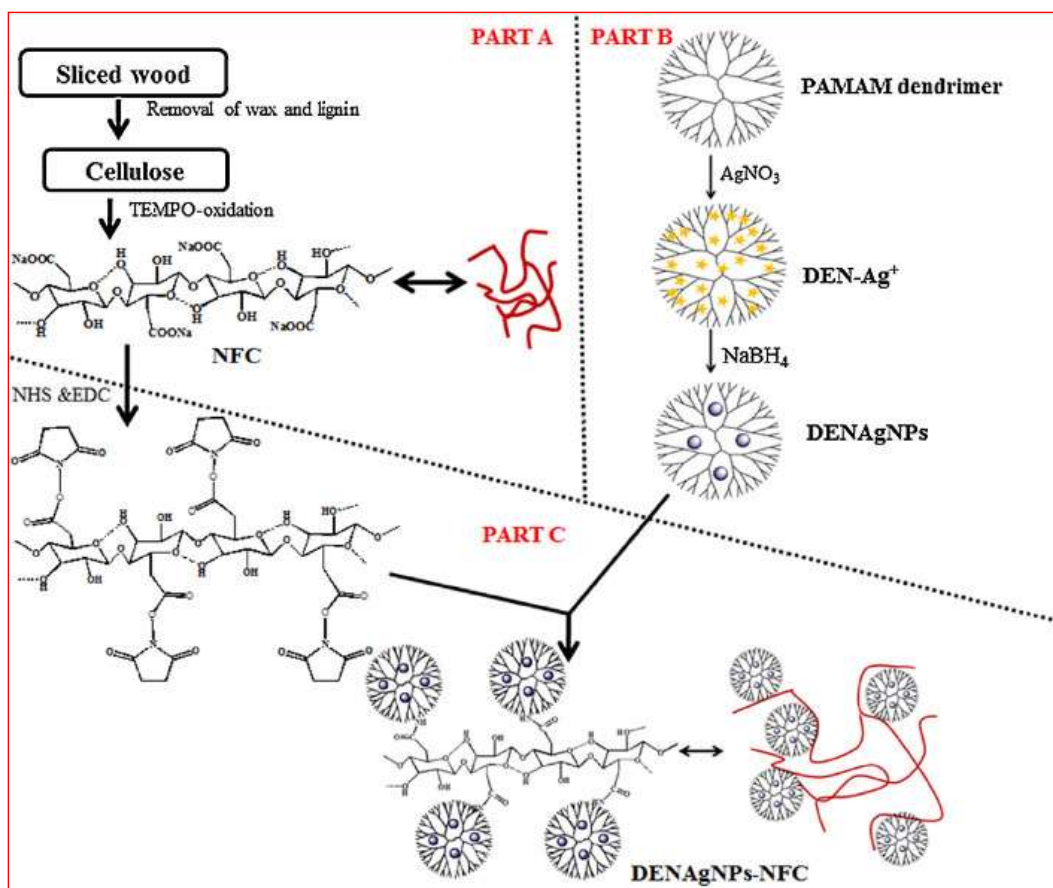


Fig. 5. The amidation reaction between NFC and DENAgNPs (reprinted with permission from Ramaraju *et al.* 2015)

Ramaraju *et al.* (2015) prepared AgNPs-containing cellulosic antibacterial material using this method (Manna *et al.* 2001; Bendi and Imae 2013). The preparation process included three procedures: i) a silver nitrate solution and NH_2 -terminated fourth generation poly (amido amine) dendrimer (PAMAM) solution were mixed together, and sodium borohydride solution was slowly added to form a PAMAM containing AgNPs (DENAgNPs); ii) NFC, suspended in water, was reacted with equimolar coupling reagents, 1-ethyl-3-(3-dimethylaminopropyl)-carbodiimide (EDC) and N-hydroxy-succinimide (NHS) to form a stable NHS-carboxylated NFC; and iii) an aqueous DENAgNPs solution was added into the NHS-carboxylated NFC suspension to form DENAgNPs-NFC *via* an amidation reaction. The amidation reaction is shown in Fig. 5.

Summary

There are some disadvantages with the covalent bonding method, such as the larger chemical consumption and the prolonged preparation period. Nevertheless, the greatest benefit of this method is the relative strength and permanence of the covalent bond between the cellulosic materials and the AgNPs-wrapped polymers. The AgNPs are tightly immobilized on the cellulosic materials, endowing permanent antibacterial activity. Although this preparation method is in the early stages, it may be the focus of future research.

CYTOTOXICITY ANALYSIS OF AgNPs

Since AgNPs are widely used as an antibacterial agent, which can kill microorganisms cells, their biological activities on human cells are worth considering. Some research has been focused on this subject and has been reported.

Travan *et al.* (2009) indicated that AgNPs did not show any cytotoxic effect toward three different eukaryotic cell lines, *i.e.*, mouse fibroblast-like (NIH-3T3), human hepatocarcinoma (HepG2), and human osteosarcoma (MG63) cell lines. They thought that this was due to the fact that AgNPs could exert their antimicrobial activity by simple contact with the bacterial membrane, while they could not be taken up and internalized by eukaryotic cells. Panáček *et al.* (2009) found that AgNPs exhibited no cytotoxic effects on human fibroblasts at silver concentrations of 0.05 mg/L to 54 mg/L. However, the research of Greulich *et al.* (2009) showed that AgNPs exhibited cytotoxic effects on human mesenchymal stem cells at high concentrations but also induced cell activation at high but nontoxic concentrations of AgNPs. Recently Shaheen and Fouda (2017) investigated the cytotoxicity of AgNPs, and their research demonstrated that AgNPs were safe for eukaryotic human cell represented by HepG2, Mcf7 and BHK cellline.

Through the research we can know that AgNPs exhibit distinguishing toxicity to different human cells. Nevertheless we can conclude that AgNPs are relatively nontoxic to human cells. Consequently, AgNPs can be regarded as sufficiently safe to merit further investigations in the applications of medicine, wound dressing, food packaging, and some other fields.

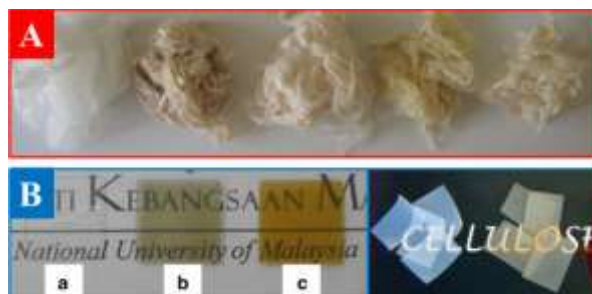


Fig. 6. The photographs of AgNPs-containing (A) modal fibers (reprinted from Pivec *et al.* 2017) and (B) RC membranes (reprinted with permission from Cai *et al.* 2009. "Nanoporous cellulose as metal nanoparticles support," *Biomacromolecules* 10(1), 87-94; Copyright (2018) American Chemical Society and with permission from Chook *et al.* 2014).

FORMS AND APPLICATIONS OF AgNPs-CELLULOSE COMPOSITES

For various applications, AgNPs-containing cellulosic antibacterial materials should take different forms, as shown in Figs. 6 through 8. The general map of applications of AgNPs-cellulose composites is shown in Fig. 9. Natural cellulose fiber materials loaded with AgNPs are usually made into antibacterial paper products (Xu *et al.* 2015) or mixed with fluff pulp to manufacture antibacterial disposable hygiene products. AgNPs-containing NFC materials, which are normally fabricated into membrane, aerogel, or hydrogel products, are widely used in food packaging, chemical catalysis, environmental protection, and other fields (Dong *et al.* 2013; Das *et al.* 2015). AgNPs-containing MCC maintains the original powder morphology of MCC, and it is utilized as functional filler (Silva and Unali 2011). AgNPs-containing RC can be spun into silks for textile materials (Pivec *et al.* 2017) or cast into membranes for effluent treatment (Weis *et al.* 2005; Kallioinen *et al.* 2010; Puro *et al.* 2010). Singla *et al.* (2017a) prepared AgNPs and cellulose nanocrystals nanobiocomposites (NCs) in film and ointment forms. NCs were found to significantly enhance *in vivo* skin tissue repair by decreasing production of inflammatory cytokines and increasing fibroblast proliferation, angiogenesis, and finally tissue neo-epithelization and regeneration in less than 14 days by favoring collagen deposition. NCs may serve as potential candidates as antibacterial wound dressings for accelerating tissue repair and regeneration, such as serving for diabetic patients (Singla *et al.* 2017b).



Fig. 7. AgNPs contained in various products: (A) paper (reprinted with permission from Xu *et al.* 2015), (B) NFC membranes (reprinted with permissions from Dong *et al.* 2013 and Ramaraju *et al.* 2015), and (C) NFC gels (reprinted with permissions from Dong *et al.* 2013 and Das *et al.* 2015)

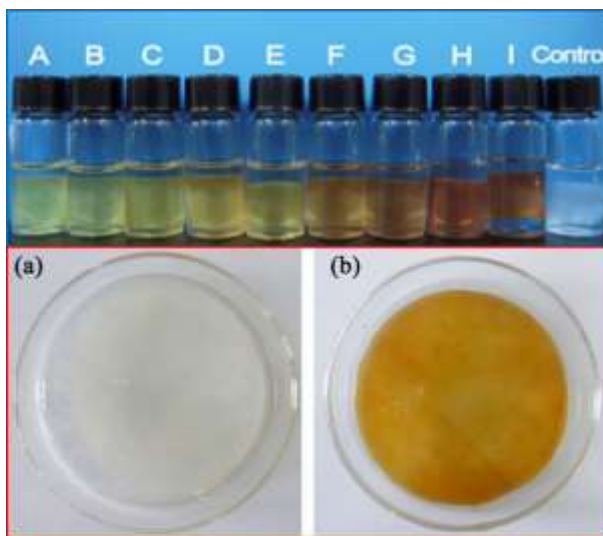


Fig. 8. AgNPs-containing BC membranes (reprinted with permissions from Yang *et al.* 2012b and Li *et al.* 2015)

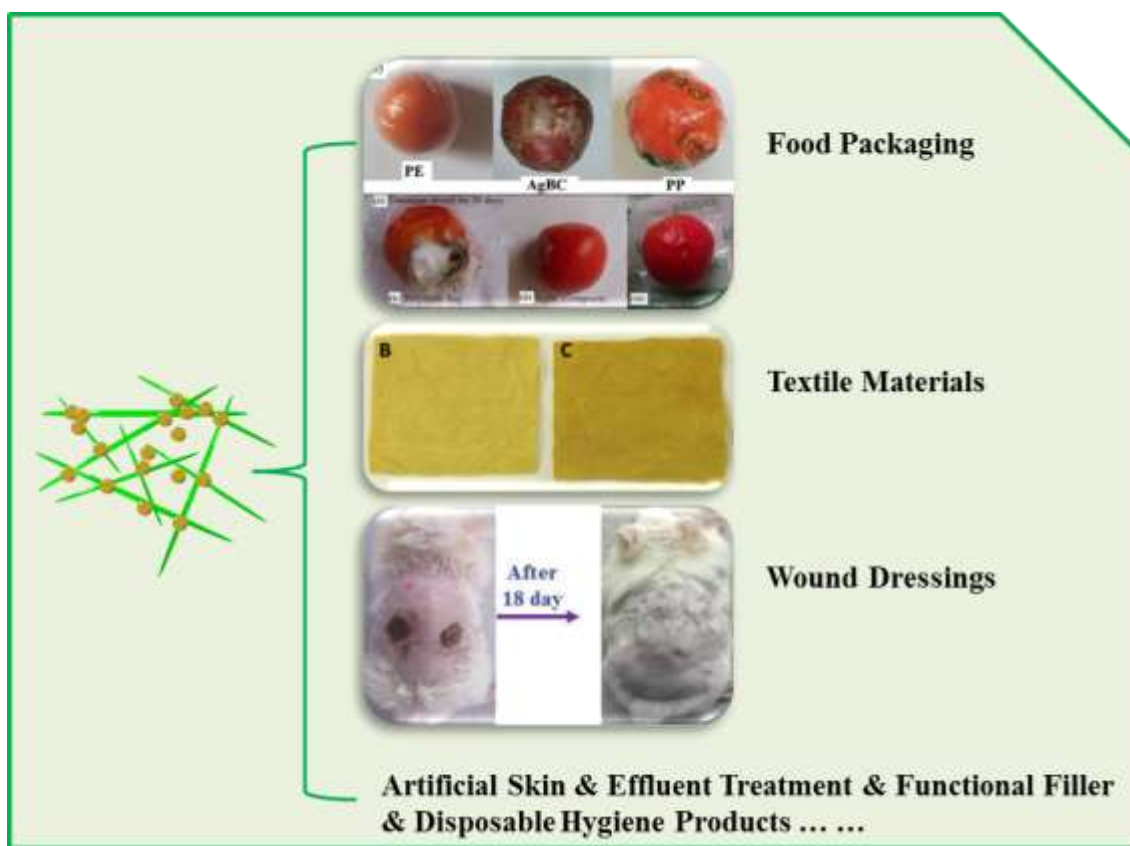


Fig. 9. The applications of AgNPs-cellulose composites (reprinted with permissions from Adepu and Khandelwal 2017; Tang *et al.* 2015; and from Singla *et al.* 2017b)

Because it is easy to shape, BC mostly has been formed into membranes. The AgNPs-containing BC membrane can be utilized as an antibacterial artificial skin to replace the defective skin, or as a medical antibacterial dressing to replace the traditional cotton wool, gauze, and bandages used to treat burns or chronic ulcerative disease (Czaja

et al. 2006, 2007; Fu *et al.* 2013). Adepur and Khandelwal (2017) fabricated BC-AgNPs antifouling materials. They found that food stuff was protected from microbial spoilage for 30 days when stored in BC-AgNPs nanocomposites having < 2% silver, whereas spoilage was noticed within 15 days for food stuff stored in regular polythene bag. Therefore, BC impregnated with AgNPs serves to be highly useful material for food packaging as well as healthcare systems. With its superior mechanical properties and excellent biocompatibility, the AgNPs-containing BC membrane has great potential in medicine and health care (Yang *et al.* 2012b; Li *et al.* 2015).

CONCLUSIONS

1. This paper reviewed three common techniques used to prepare AgNPs-containing cellulosic antibacterial materials, *i.e.*, the physical method, *in-situ* chemical reduction method, and covalent bonding method. The specific forms and application status of AgNPs-containing cellulosic materials prepared by the three methods are introduced.
2. Commonly, the physical method is convenient to operate, and the loading ratio of AgNPs to cellulosic material is relatively high. However, the physical adsorption force between the AgNPs and the cellulosic material is correspondingly weak, and this is a drawback of this method. Compared with the physical method, the operation process of the *in-situ* chemical reduction method is obviously simplified, and the antibacterial endurance of AgNPs-containing cellulosic materials is effectively enhanced due to the internal loading of AgNPs in cellulosic materials. Considering the final product performances, the covalent bonding method is superior to the above two methods due to the covalent bond combination between cellulosic materials and AgNPs-wrapped polymers. However, some disadvantages now exist, such as the larger chemical consumption and the prolonged preparing period.
3. The AgNPs-containing cellulosic antibacterial materials can be widely used in food packaging, chemical catalysis, environmental protection, functional materials, textiles, skin beauty products, medical items, health care, and other fields. To date, the exploration of the application of AgNPs-containing cellulose materials is still in progress. In general, the research on BC membranes and CA fibers is mature, and there have been commercial products in the market. Hence, the AgNPs-containing BC membranes and CA fibers are expected to be the earliest to achieve industrial production.

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