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The preparation and antibacterial activity of cellulose/ZnO composite: a review

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Abstract: The infectious diseases caused by various bacteria pose serious threat to human health. To solve this problem, antibacterial agents have been widely used in people's daily life to deactivate or kill these bacteria. Among the antibacterial agents, ZnO is one of the most promising metal oxide antibacterial agents due to its non-toxic nature and safe properties. To expand its application, many composites of ZnO have been widely studied. Cellulose, as one of the most abundant biopolymers, has many merits like biodegradability, biocompatibility and low cost. Thus, many studies focus on synthesized cellulose/ZnO. The synthetic strategy includes both chemical and physical methods. Many of them have been shown that cellulose/ZnO composites have excellent antibacterial activity and are environment-friendly and have many applications for example food packing, antibacterial fibers and so on. This review mainly discusses the preparation methods of cellulose/ZnO and their effect on the morphology and properties.

Keywords: ZnO, Cellulose, Antibacterial activity, Composite, Method

1 Introduction

The use of antibacterial agents is necessary to prevent the microorganism growth and reduce the harmful effects in our life at the same time [1,2]. Today, inorganic antimicrobial agents are promising such as metal salts, nano-sized metals and metal oxides [3,4]. In the metal oxide, CuO, TiO_2, ZnO, Al_2O_3, SiO_2, Fe_2O_3 and CeO_2 are frequently-used as antibacterial agents [5-18]. Among them, ZnO has excellent antibacterial activities and its application field is very broad. Thus, different methodology is applied to preparation of ZnO [19-21]. The gelatin/ZnO nanoparticle (NPs) composite films could be applied for food preservation due to their excellent antibacterial activity against both gram-positive and gram-negative foodborne pathogenic bacteria [22]; the ZnO/GO composites could be used in surface coatings on various substrates to effectively inhibit bacterial growth, propagation and survival in medical devices [23]; filters functionalized with ZnO nanorods (NRs) possess high air filtration efficiencies and high antibacterial activities and could be applied in industrial gas purification devices and indoor air cleaning systems [24].

In addition to its antibacterial agent, ZnO is proven to be a promising material due to its excellent mechanical properties, excellent chemical stability and heat resistance [25]. The application fields of ZnO nanoparticles are very broad such as electrodes, piezoelectric devices, optics, optoelectronics, photodiode devices, sensors, light-emitting diodes photocatalysts and so on [26-34]. The core–shell nylon 6,6-ZnO nanofiber mats could be quite applicable as a filtering/membrane material for treatment of organic pollutants for water purification due to their efficient photocatalytic properties, structural flexibility and stability [35], for example.

Cellulose, one of the most abundant biopolymers, has been widely used as a reinforcing material for fiber-thermoplastic composite materials [36] and is considered to be an almost inexhaustible source of raw material for the increasing demand for biodegradable and biocompatible products [37,38]. Cellulose derivatives also have many advantages, such as environmentally friendly, biocompatible, sustainable and cost-effective sources of carbon-based polymers and substrates for the development of sophisticated nanocomposite materials [39]. Cellulose acetate (CA) is one of the cellulose derivatives and has many exceptional properties, such as non-toxic, renewable, low

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cost and biodegradable [40]. CA is an important bio-based polymer that has been used in a broad field of applications such as plastics, lacquers, photographic films and textile [41-44]. Carboxymethyl cellulose (CMC) is derived from raw cellulose materials by alkalization and acidification. Different from natural cellulose, CMC is a kind of water-soluble cellulose ether [45,46] and is widely applied in many fields, such as chemical, geological, light industry, petroleum, drug, food and pharmaceutical [47-49].

However, cellulose does not have any antibacterial activity, which limited its application for food packaging. To endow cellulose with antibacterial properties, functional nanomaterials are widely used to compose with cellulose [50-53]. Several metal and metal oxide nanostructures, such as TiO_2, ZnO, Fe_2O_3, Ag and Cu etc., have been incorporated into paper products [54-56]. One of the effects of ZnO NPs on paper and applications have been investigated: coatings containing ZnO nanoparticles improved resistance to microbial attack [57] and ZnO is considered to be non-toxic [58] which can be used in food packaging [59].

Recently the food packing industry has gained much more attention in many countries [60]. However, food bacterial contamination is a common problem in this easy-cooked food industry [61] which can lead to cross contamination, discoloration, stinky odor, and food borne illness [62,63]. Antimicrobial packaging is a type of active packaging which interacts with the product or the headspace inside to reduce, inhibit or retard the growth of microorganisms [64]. ZnO is currently listed as generally recognized as safe (GRAS) material by the Food and Drug Administration and is used as food additive [58]. ZnO in the nanoscale has shown antimicrobial properties and potential applications in food preservation [58]. Moreover, ZnO is a source of zinc and has essential micronutrient and serves important roles in growth, development and well-being in humans and animals [65].

2 The source of cellulose and the preparation of cellulose/ZnO

With the fast development of nanotechnology, high-quality nanomaterials have been fabricated successfully through different physical and chemical strategies during the past two decades [66]. Cellulose/ZnO composites can be prepared by the following methods: electrospinning [67,68], microwave [69,70], sol-gel [71], ultrasonic [72-74], hydrothermal [54,75], and precipitation [76,77] and so on. Compared with physical methods, chemical methods have shown some distinct advantages for the synthesis of ZnO nanoparticles, including easy scale-up, low reaction temperature and inexpensive equipment [50]. In this section, the main methods are chosen to discuss such as electrospun method, microwave method, sol-gel method, hydrothermal synthesis method, ultrasonic method and precipitation method. All synthesis methods of cellulose/ZnO composites mentioned in this work can also be conducted on a laboratory scale.

The source of cellulose is varied such as cotton fiber, cellulose acetate (CA), bacterial cellulose (BC), soft wood pulp, microcrystalline cellulose (MCC), carboxymethyl cellulose (CMC) as shown in Table 1. The source of the zinc ion is usually ZnO directly or synthesized indirectly by using $Zn(Ac)_2{\cdot}2H_2O$, $ZnCl_2$ and $Zn(NO_3)_2{\cdot}6H_2O$. Some groups prepared though simple, green and facile method [69,73] but some used organic agents [67,71].

Different sources of cellulose cause different morphology as shown in Figure 1. Figure 1a shows the smooth texture of the pristine cotton bandage [74], Figure 1b a plain cellulose acetate film exhibiting a smooth surface [75], Figure 1c the pristine BC sheet shows a nonwoven network structure of nanofibrous cellulose, with fiber diameter 55.00 ± 10.54 nm [72] and in Figure 1d, the morphology of the bleached softwood cellulose fibers is not smooth [54].

According to these studies, we conclude that different source of cellulose could be prepared as films (cotton fiber, cellulose acetate), paper (wood), hydrogels (CMC), foams (BC) and this would affect the morphology of the cellulose/ZnO. The morphology of the cellulose/ZnO included 1): ZnO nanoparticles grow on the cellulose fibers; 2): ZnO microparticles deposited on the surface of cellulose; 3): ZnO particles mixed with cellulose. The size of ZnO is affected by the preparation methods: the size of ZnO at 20~40 nm was usually prepared by precipitation, hydrothermal, ultrasonic, eletrospun, in situ formation and sonochemical method; while the size of ZnO more than 1 μm are usually prepared by microwave, hydrothermal method.

3 The properties of cellulose/ZnO

Phase and thermal stability have been widely used to analyze the cellulose/ZnO. XRD results show that both cellulose I and II types can be used to prepared the cellulose/ZnO. While different content of ZnO and the crystalline in the cellulose/ZnO would affect 2θ of the strongest diffraction peak, which means the major

Table 1: The source of cellulose, materials and method of synthesis.

The source of cellulose	Materials(mainly)	Method	Composite
Cotton fiber	Cotton linter pulp; ZnO; NaOH; Na_2SO_4	Microwave method and One-step coagulation method	RCZ [69]
	Plain-woven cotton fabric; $Zn(Ac)_2{\cdot}2H_2O$; NaOH; TEOS; HNO_3; NH_3(25%); Ethanol	Sol-gel method	ZnO/SiO_2 hybrid nanocomposite [71]
	Cotton fibers; $Zn(NO_3)_2$; SA; NH_4OH	Precipitation method	ZnO-SACNF [76]
	Cotton bandage; $Zn(Ac)_2{\cdot}H_2O$; $NH_3{\cdot}xH_2O$; Ethanol	Sonochemical method	ZnO-Fabric nanocomposite [74]
CA	CA; $Zn(Ac)_2{\cdot}2H_2O$; DMF; Acetone	Eletrospun method	ZnO embedded CA membrane [67]
	CA; $ZnCl_2$; NaOH; DMF	Hydrothermal method	ZOLCA [75]
BC	BC; $Zn(Ac)_2{\cdot}2H_2O$; NH_4OH; Ethanol	Ultrasonic-assisted synthesis method	ZnO particles-incorporated BC sheet [72]
	Paper; ZnO; NH_4OH	Ultrasonic method	ZnO nanoparticles coated paper [73]
Wood	Bleached softwood cellulose fibers; $ZnCl_2$; Na_2CO_3; Ethanol	Hydrothermal method	Z-P [54]
	Filter paper; $Zn(Ac)_2{\cdot}2H_2O$; NaOH; Ethanol; Sulfuric acid; $AgNO_3$	Precipitation method	ZnO-Ag/CNCs Nanoparticles [77]
	Softwood; ZnO; PDDA; PSS	Eletrospun method	NFC/ZnO [78]
P-cellulose, α-Cellulose	$Zn(Ac)_2{\cdot}2H_2O$; $AgNO_3$; HMT; NH_3aq(25-29 wt%); P-cellulose; α-Cellulose; Polyvinyl chloride; PVC RB3 resin	Microwave assisted synthesis method	Ag/ZnO modified cellulose fillers [70]
MCC	MCC; HCl; Citric acid; $NH_3{\cdot}H_2O$; $Zn(NO_3)_2{\cdot}6H_2O$; NaOH	Simple precipitation through electrostatic interaction	CNC/ZnO nanohybrids [79]
	MCC; PHBV; HCl; Citric acid; Chloroform; Ethanol; $Zn(NO_3)_2{\cdot}6H_2O$; NaOH	Eletrospun method	CNC-ZnO [68]
CMC	CMC; ECH; $Zn(NO_3)_2{\cdot}6H_2O$; NaOH; HCl	In situ formation	CMC/ZnO nanocomposite hydrogels [80]
	CMC; Chitosan; Glycerol; Oleic acid; Tween 80; ZnO NPs	Casting method	CMC-CH-OL-ZnO [81]

TEOS: Tetraethylorthosilicate; SA: Sodium alginate; DMF: Dimethyl formamide; HMT: Hexamethylenetetramine; MCC: Microcrystalline cellulose; PHBV: poly(3-hydroxybutyrate-co-3-hydroxy-valerate); ECH: Epichlorohydrin; PDDA: poly(diallyldimethylammonium chloride); PSS: poly(sodium 4-styrenesulfonate); RCZ: cellulose based ZnO nanocomposite; ZnO-SACNF: nano zinc oxide-sodium alginate cellulose fibres; Z-P: ZnO nanowire-immobilized paper matrices; NFC: nanofibrillated cellulose; CNC: cellulose nanocrystal; CMC-CH-OL-ZnO: carboxymethyl cellulose-chitosan-oleic acid incroporated with zinc oxide nano particles; ZOLCA: ZnO nanoparticles loaded cellulose acetate;

phase in the cellulose/ZnO would be influenced by the ZnO content, as shown in Figure.2. In Figure. 2a, the two broad peaks and one intense peak appear in the cellulose/ZnO near to the 2θ angles of 14.8°, 16.8°, 22.2° and 34.1°, which are characteristic of the (101), ($10\bar{1}$), (002) and (040) reflections of cellulose I, respectively [54,71,82,83]. In Figure 2b, the diffraction peaks at 2θ = 12°, 20°, and 22° are ascribed to the ($1\bar{1}0$), (110), and (020) planes of the cellulose II crystalline type, respectively [69,84]. In comparison to the strong intensity diffraction peak of ZnO the diffraction peak of cellulose in RCZ are very weak [69] which could be attributed to ZnO growing on the surface of cellulose in the cellulose/ZnO which makes it harder to collect the diffraction data of cellulose during the XRD test. In additional, the types and ratio of precursors, the pH of solution (basicity) and the preparation method will affect the major phase of cellulose/ZnO (analyzed by XRD) and the related data is shown in Table 2. The intensity of ZnO diffraction peak in composites will increase when the increasement of the ZnO content in composites. Moreover, the good crystalline of cellulose will also obtain high diffraction intensity. Due to the crystallinity character of ZnO and cellulose, the content of ZnO in cellulose/ZnO is the primary factor which determine major phase in composites.

Thermal degradation of cellulose involved depolymerization, dehydration, and decomposition of glycosyl units followed by formation of a charred residue [77]. Compared with the CNCs, the thermal degradation curves with a single degradation peak shifted to higher

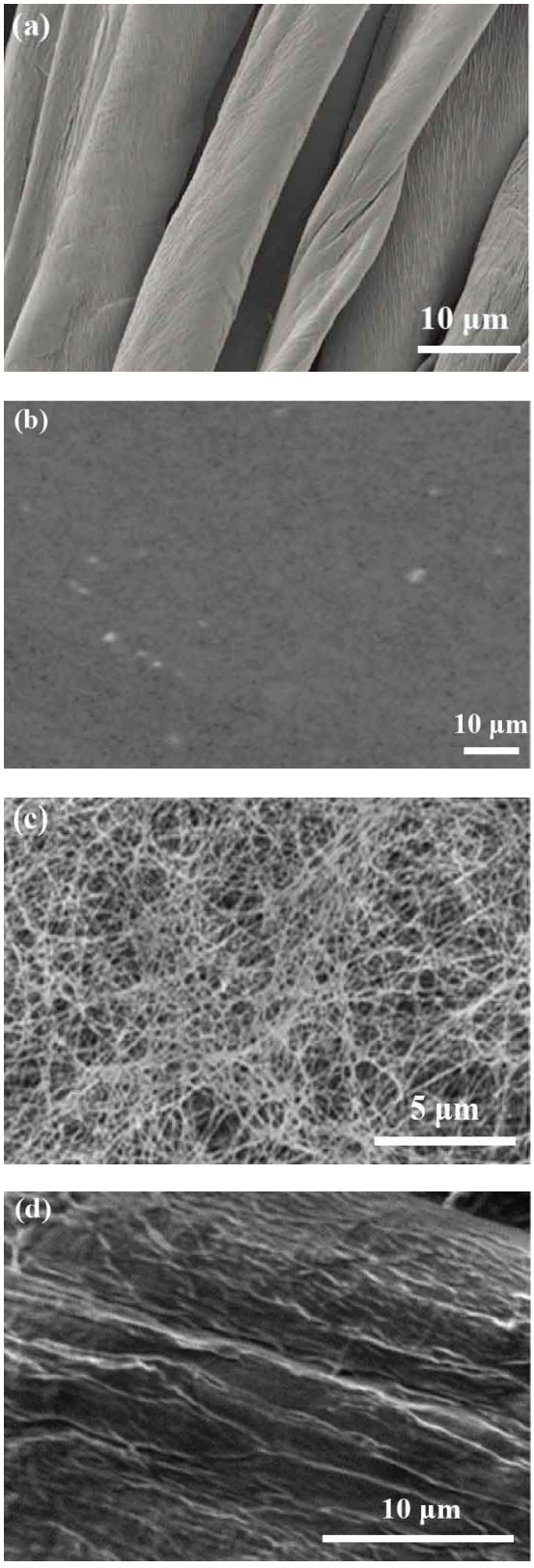

Figure 1: The SEM of cellulose a: pristine bandage fabric [74]; b: plain cellulose acetate [75]; c: pristine bacterial cellulose [72]; d: bleached softwood cellulose fibers [54].

temperature for the CNC/ZnO nanohybrids in Figure 3a. These results indicated that the thermal stability of the nanohybrids was better than that of the CNCs. This was ascribed to the stronger interactions between oxygen atoms of the CNCs and ZnO nanoparticles, thus providing a thermal barrier for the cellulose skeleton by absorbing the heat [77,79]. However, ZnO also reduced thermal stability of the cellulose in some composites as shown in Figure 3b [71]. ZnO reduced thermal stability is likely because ZnO has high thermal conductivity and cellulose a very low conductivity but direct interaction of ZnO nanoparticles and cellulose make heat transportation much easier which resulted in the lower thermal decomposition temperature [85].

In conclusion, different prepared method will also affect the thermal stability of the cellulose/ZnO. ZnO prepared by precipitation method will increase thermal stability in the cellulose/ZnO, but ultrasonic method and sol-gel method will reduce the thermal stability in the cellulose/ZnO.

Many studies focus on the preparation of hydrophobic and super hydrophobic surface by appropriate surface modification of the sample. Contact angle measurements are carried out to evaluate the wetting properties of the cellulose/ZnO and the results are summarized in Table 3. The water contact angle of RCZ films varies from 87.5° to 102.0° as the ZnO content increase, which is significantly larger than that of the regenerated cellulose (RC) film (79.7°) [69]. In the case of pure CA fibrous membrane, the measured contact angle is found to be 47° initially and the ZnO embedded CA is about 124°, which is much higher than that of the pure CA fibers so that the wetting property of the CA has changed from hydrophilic to hydrophobic when ZnO is impregnated into it [67]. The hydrophobic properties of the cellulose/ZnO is much better than cellulose which may due to the interspaces among the ZnO particles trapping air whose water contact angle is considered as 180°. Therefore, the trapped air could be served as part of the surface, resulting in a solid/air composite surface to increase the hydrophobicity of composite [67,69].

RC film has good tensile strength (σb) and Young's modulus (E), with values of 40.6 MPa and 2.5 GPa, respectively. The ZnO content from 2.7 wt % (RCZ4) to 7.4 wt % (RCZ8) led to a slight decrease in the elongation at break (ε_b) compared with the RC film. However, the E and σb values slightly increase and reach 3.1 GPa and 57.1 MPa with 7.4 wt % ZnO loaded which are comparable with the cellulose–carbon nanotube film. The σb values of the films decrease with increased ZnO content over 7.4 wt %. This can

Table 2: The influence factors on major phase and thermal stability of cellulose/ZnO.

	The major phase of cellulose/ZnO		The thermal stability	
	Cellulose	ZnO	increase	reduce
Reference	[54, 71, 73, 79, 86-88]	[69, 84][85]	[77, 79, 84]	[71, 73]
Affecting factor	Source of cellulose, basicity in reaction system; the ratio of precursors		Preparation methods	

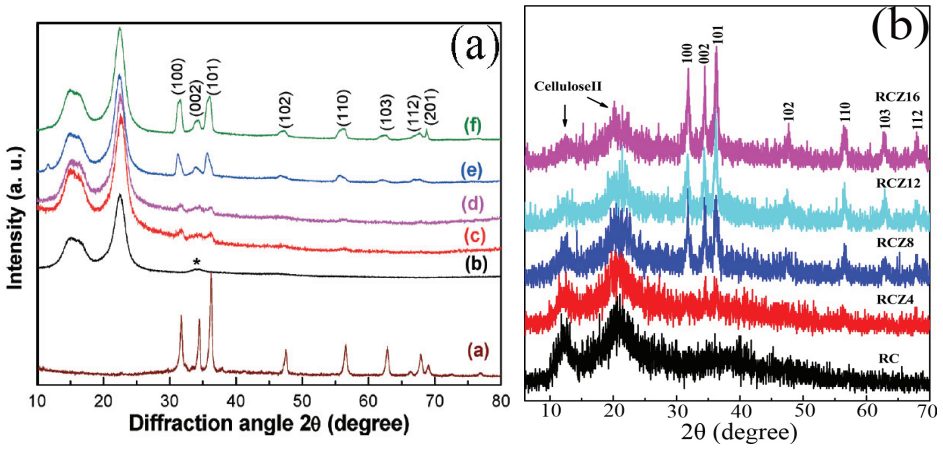

Figure 2: The XRD plots of cellulose/ZnO (a): Cellulose as major phase in composites [54]; (b): ZnO as major phase in composites [69].

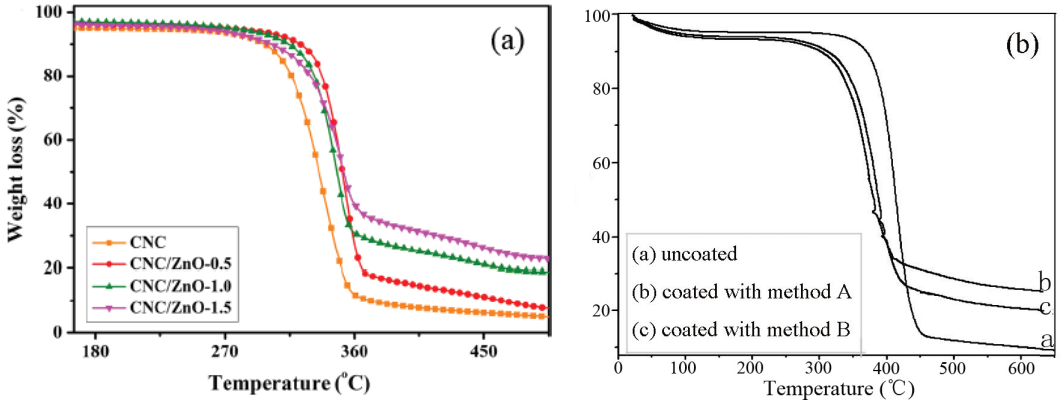

Figure 3: TG curves of cellulose/ZnO (a): the thermal stability increased compared to cellulose [79]; (b): the thermal stability reduced compared to cellulose [71].

be attributed to the tendency of ZnO nanoparticles to form larger agglomerates at higher content, leading to relatively poor dispersion in the cellulose matrix. However, all of the RCZ films display higher tensile strength than RC films, which further confirm the strong interactions between ZnO nanoparticles and the cellulose matrix [69]. The ε_b, σb and E of ZnO–SACNF fibers are depicted in Table 3. The results indicate that the ZnO–SACNF could be utilized for the longer duration of use without any significant damage or breakage [76].

The hydrophobic and super hydrophobic surface was usually used MCC, CA and cotton fiber and prepared by electrospinning, microwave and the solvent evaporation method and the contact angle over 100°. Along an increase in the ZnO content in the cellulose/ZnO, the contact angle and the tensile strength were also increased.

Many analytical methods have been adopted to evaluate the antibacterial activity of ZnO nanoparticles. The common methods were colony forming count method, disc diffusion method and so on. In order to quantitatively determine the antibacterial activity, the colony forming count method was applied. Gram-negative Escherichia coli (E. coli) and Gram-positive Staphylococcus aureus (S. aureus) are mainly chosen as model bacteria to evaluate

the antibacterial activity of ZnO nanoparticles. The obtained bacterial suspension of S. aureus and E. coli had cell concentrations around 10^8 CFU·mL^{-1}. The bacterial suspension accompanying with the sample then incubated at 37°C. After incubation, taking a certain amount of the cell suspension diluted with 0.85-0.9% saline solution and then spread on sterilized petri-plates and incubated at 37°C. The number of the bacterial colonies referred to the number of the bacterial cells which survived. The reduction in viable bacterial cells (R %) was calculated by the following equation: R % = (Viable cells at 0 h − Viable cells after test) / (Viable cells at 0 h) × 100% [54, 56, 72, 86, 87]. The results were shown in Table 4.

Table 3: Comparison of the A_w, ε_b, E, σ_b properties of the cellulose/ZnO.

Sample	A_w (deg)	ε_b (%)	E (GPa)	σ_b (MPa)
RCZ4 [69]	87.5	10.1	3.0	55.0
RCZ8 [69]	96.2	10.1	3.1	57.1
RCZ12 [69]	103.4	10.6	2.7	49.0
RCZ16 [69]	102.0	8.6	2.6	43.3
ZnO-SACNF (0.5% SA) [76]		22.1	0.275	29.5
ZnO-SACNF (1.5% SA) [76]		24.9	0.379	46.5
ZnO embedded CA membrane [67]	122.4-124.2			

40 wt% Zn: zinc oxide bionanocomposite foam by a bacterial cellulose mediated with a ZnO loading of 40 wt%; 70 wt% Zn: zinc oxide bionanocomposite foam by a bacterial cellulose mediated with a ZnO loading of 70 wt%; ZnO-BC: nanocrystalline ZnO particles into bacterial cellulose pellicle; G-ZnO: glucose-ZnO; S-ZnO: sucrose-ZnO; St-ZnO: starch-ZnO; AA-ZnO: alginic acid; 0.75% ZnO: bandage coated with 0.75% ZnO nanoparticles; ZCB: ZnO-coated cotton bandage.

For the agar diffusion method, the samples were cut into disc shapes with a certain diameter. The fresh strain was diluted, then obtained a certain concentration

Table 4: Viable cell reduction activity of samples on E. coli and S. aureus.

Samples	Crystalline size (nm)	ZnO contents (wt %)	Reduction in cell viability (%) or Antibacterial ratio (%)										
			E. coli						S. aureus				
			1h	2h	3h	5h	6h	9-24h	1h	2h	3h	5h	15-24h
2.0 Z-P [54]		2.0			62.58		95.30	97.48					
7.0 Z-P [54]	10-20	7.0			66.34		98.72	99.81					
10.0Z-P [54]		10.0			83.11		99.74	99.98					
18.0 Z-P [54]		18.0			95.88		99.97	99.99					
40 wt% Zn [87]	10-20	40	39.9		82.9	96.7							
70 wt% Zn [87]		70	42.8		90.0	98.7							
ZnO-BC-1 [72]	55	37.3±0.6						99.80±0.02					99.80±0.03
ZnO-BC-2 [72]	56	45.5±0.9						99.79±0.02					99.79±0.02
ZnO-BC-3 [72]	63	46.7±0.8						99.79±0.03					99.80±0.03
G-ZnO [56]	30.9			80.49						57.78			
S-ZnO [56]	28.3			85.37						66.68			
St-ZnO [56]	23.6	0.1		83.59						52.90			
AA-ZnO [56]	19.0			86.06						81.71			
0.75%ZnO [74]	30	0.75	99.84	100					66.40		99.93		
ZCB [86]	15	3.11				63						56	

Antibacterial activity of ZnO nanoparticles. The obtained bacterial suspension of S. aureus and E. coli had cell concentrations around 10^8 CFU·mL^{-1}. The bacterial suspension accompanying with the sample then incubated at 37°C. After incubation, taking a certain amount of the cell suspension diluted with 0.85-0.9% saline solution

Table 5: The mean value of the inhibition zone width of composites.

Samples	Crystalline size (nm)	ZnO contents (wt %)	W_{inh} (mm) E. coli	S. aureus
RCZ4 [69]	15.3	2.7	5.8	8.7
RCZ8 [69]	16.4	7.4	7.2	9.7
RCZ12 [69]	18.2	11.7	8.8	12.3
RCZ16 [69]	19.2	15.1	10.2	18.8
ZnO-BC-1 [72]	55	37.3±0.6	5.7±0.29	2.9±0.75
Z1 [80]	10-20		3.5±1	5±1.5
Z2 [80]			4.5±1	6.5±1
Z3 [80]			6±0.5	8.5±0.5
Z4 [80]			7.5±1	11.5±1
RBC-ZnO1 [84]	77	4.8	9.5	
RBC-ZnO2 [84]		8	13	
ZnO/SiO_2 hybrid nanocomposite (A) [71]		11.207	2.25	5.75
ZnO/SiO_2 hybrid nanocomposite (B) [71]		3.489	3.25	8.25
ZnC [89]	50	15	2.5	4.25

of bacteria suspension was around 10^8 CFU·mL^{-1}. After that, the bacteria suspension was spread on the plates uniformly and the samples were placed on the center of the plate and then incubated for 18-24 hours at 37°C. After incubation, a bacterial inhibition zone was formed around the samples and the width of the bacterial inhibition zone was measured and recorded as the antibacterial effect of composites. The width of the inhibition zone (W_{inh}) was calculated using the following equation: $W_{inh} = (d_1-d_2) / 2$ [69,74]. The results were shown in Table 5.

The agar diffusion and the colony forming count method could qualitative and quantitative analyse the antibacterial activity of cellulose/ZnO. Cellulose/ZnO show super antibacterial activity compared with both ZnO and cellulose. The higher percentage of ZnO incorporate, the better antibacterial activity of composites could be obtained, despite the source or the contents of cellulose in the cellulose/ZnO.

Cellulose/ZnO have advantages compared to ZnO with the antibacterial activity of the composite better than ZnO. According to Guo and her group's study [85], the cellulose/ZnO shows excellent antibacterial activity compared to ZnO. The width of the inhibition zone of cellulose/ZnO is 11.9 mm (against S. aureus) and 10.2 mm (against E. coli); while the width of inhibition zone of ZnO is 2.48 mm (against S. aureus) and 1.26 mm (against E. coli). Xu and his group got the same result from ZnO/BC by using BC (bacterial cellulose) as cellulose source. They found the ZnO/BC foam could make viability reduce to 96.7~98.7% when against E. coli, while ZnO powder is 93.6% [87].

4 Self-assembly and antibacterial mechanism

The self-assembly mechanisms of the cellulose/ZnO should be clarified. Hydrogen bonding was used to expound the self-assembly mechanisms of cellulose/ZnO in the previous reports[90]. Different from above, there were two driving forces in fabricating cellulose/ZnO by experiment and theory[85] - electrostatic attraction and hydrogen bonding. In the cellulose/ZnO, the Zn atoms of ZnO on its (001) plane would self-assemble to the Os atom of cellulose to stabilize system, which was driven by the electrostatic attraction and formed the morphology which was cellulose as matrix and ZnO crystal nucleus grow on it. The second driving force was the intermolecular hydrogen bonding, which has been well known in cellulose-involving materials.

The antibacterial mechanism of cellulose/ZnO was due to the highly reactive species such as superoxide, hydrogen peroxide and hydroxyl (O^{2-}, H_2O_2 and OH^-) which were formed on the surface of ZnO activated by both UV and visible light. Many researchers have proposed the generation of H_2O_2 on the surface of ZnO as the main

effect in the inhibition of bacteria growth [11,91]. The establishments of H_2O_2 as follow:

$ZnO + h\nu\ (\lambda < 388\ nm) \rightarrow e^- + h^+$

$H_2O + h^+ \rightarrow H^+ + HO\bullet$

$O_2 + e^- \rightarrow O_2^{\bullet-}$

$O_2^{\bullet-} + H^+ \rightarrow HO_2\bullet$

$HO_2\bullet + e^- \rightarrow HO_2^-$

$HO_2^- + H^+ \rightarrow H_2O_2$

5 Application and outlook

The cellulose/ZnO could be used in many fields such as food packaging, photoluminescent papers, antimicrobial and bioactive paper and functional paper which ZnO particles could not be used in these fields [51,54,56,92]. The composite could also be used in biomedical and healthcare, catalysis, and electronic fields just like ZnO particles [69,86]. The composite of cellulose/ZnO also has another advantage compared to ZnO. As powder ZnO is easy to mix with other materials but it is hard to made into a device or moulding by itself. While cellulose has the advantage to make into the shape of device because of its suppleness. The combining of ZnO and cellulose would enable the composite of suppleness, excellent antibacterial and fluorescence properties. Thus it will expand the application of both cellulose and ZnO.

The cellulose/ZnO will be used widely in the future due to its advantages like low cost, simple, environmental friendly and functional. Since cellulose/ZnO is mixed the merits of cellulose and ZnO, it can be used in many fields. After coating ZnO powders, the cellulose composite can possess super antibacterial activity and good fluorescence properties which could be used to prepare antimicrobial, bioactive paper, photoluminescent papers and functional paper. Due to its excellent properties and simple preparation method, cellulose/ZnO paper could be widely used as food packaging, biomedical application and healthcare.

6 Conclusions

This review article mainly discussed the preparation methods of cellulose/ZnO and its properties, especially the antibacterial activities investigated in recent years. Different sources of cellulose could be prepared as films (cotton fiber, cellulose acetate), paper (wood), hydrogels (CMC), foams (BC) and would affect the morphology of the cellulose/ZnO. The morphology of the cellulose/ZnO included 1): ZnO nanoparticles grown on the cellulose fibers, 2): ZnO microparticles deposited on the surface of cellulose, 3) ZnO particles mixed with cellulose. And the hydrogen bonding is used to expounded composite mechanisms of cellulose/ZnO.

The major phase in the cellulose/ZnO would be affected by factors of preparation method, pH of reaction system and the source of cellulose as well as the ratio of precursors. However, the contents of ZnO in the composites is key factor to dominate the major phase because of its high crystalline.

Both the thermal stability and antibacterial activity of cellulose/ZnO vary according to different preparation methods. High thermal stability of the cellulose/ZnO will be achieved if prepared with precipitation method. However, when the sol-gel method and ultrasonic method have been applied, composites with low thermal stability will be obtained. To antibacterial property of cellulose/ZnO composites, it only controlled by the contents of ZnO since cellulose itself don't have any antibacterial activity. The higher percent incorporation of ZnO in composites, the better antibacterial activity it could be obtained, despite the source or the contents of cellulose in the cellulose/ZnO.

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