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Synergistic effects of guanidine-grafted CMC on enhancing antimicrobial activity and dry strength of paper

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ABSTRACT

In order to improve the strength property and antimicrobial activity of paper simultaneously, we prepared a novel multifunctional agent based on carboxymethyl cellulose (CMC) by a simple two-stage method. The first stage was the oxidation of CMC to obtain the dialdehyde CMC (DCMC), and the second stage was the graft of guanidine hydrochloride (GH) onto DCMC to obtain DCMC-GH polymer. The strength property and antimicrobial activity of DCMC-GH-coated copy paper have been studied by the tensile test and inhibition zone method, respectively. The results showed that the dry strength index could increase about 20% after the paper was coated with DCMC-GH. The coating of DCMC-GH on paper also resulted in excellent antimicrobial activities against *Escherichia coli* and *Staphylococcus aureus*, and the inhibition zone became larger as the GH content grafted on DCMC increased. The novel DCMC-GH polymer would be a multifunctional coating agent for food packaging paper.

1. Introduction

Cellulosic material obtained from forestry and agriculture has been considered to be renewable and inexhaustible resources for the production of paper and paperboard (Andresen, Johansson, Tanem, & Stenius, 2006; Nakagaito & Yano, 2008; Liu et al., 2013). Nowadays, paper and paperboard have taken up around 40% of the global packaging market, which are also the main material for food packaging (Ho, Zimmermann, Ohr, & Caseri, 2012). However, cellulose does not have any antimicrobial activity, and most molds and bacteria can multiply rapidly in cellulose paper under the proper conditions. Therefore, different kinds of natural and synthetic antibacterial agents have been studied and widely used in the food packaging paper process (Qian, Li, Sun, & Xiao, 2011; Sun et al., 2010; Qian, Xiao, Zhao, & He, 2011; Qian, Guan, He, & Xiao, 2008a). For example, Zhang applied the poly(hexamethylene guanidine hydrochloride) derivatized beeswax latex particles to copy paper and demonstrated that the coated paper has excellent antimicrobial activity against *Escherichia coli* even at a low dosage of the beeswax latex derivatives (Zhang & Xiao, 2013). Obviously, the guanidine used in the cellulosic materials has excellent antimicrobial activity against harmful bacteria.

In addition of antimicrobial activity, strength property is also an important attribute for food packaging paper. A number of strength agents have been used to improve the paper strength in recent years (Ghasemian, Ghaffari, & Ashori, 2012; Kumar & Singh, 2008; Kaushik, Singh, & Verma, 2010). Among these agents, carboxymethyl cellulose (CMC) with good improvement for paper strength property has attracted increasing attention due to its lower cost and easy availability from pulping industries. For example, Fatehi used the modified chitosan and carboxymethyl cellulose to improve paper strength and found that the significant improvement of paper strength was attributed to the extra development of fiber bonding (Fatehi, Qian, Kititerakun, Rirksomboon, & Xiao, 2009). However, coating agents that can improve strength property while maintaining antimicrobial activity for food packaging paper are very few.

In this work, we developed a simple approach of grafting guanidine hydrochloride (GH), an effective antimicrobial agent, onto the carboxymethyl cellulose. Janjic et al. reported a preparation method of a chitosan-coated lyocell fiber, in which the lyocell fibers were first oxidized by periodate to increase their aldehyde content and then chitosan was grafted onto the oxycellulose fibers by the Schiff base reaction (Janjic et al., 2009). According to the same mechanism, the corresponding Schiff base can be formed by the reaction of the aldehyde group of dialdehyde carboxymethyl cellulose and the free amino group of guanidine hydrochloride. The corresponding reaction scheme is illustrated in Fig. 1. To our knowledge, this is the first time that the carboxymethyl cellulose grafted

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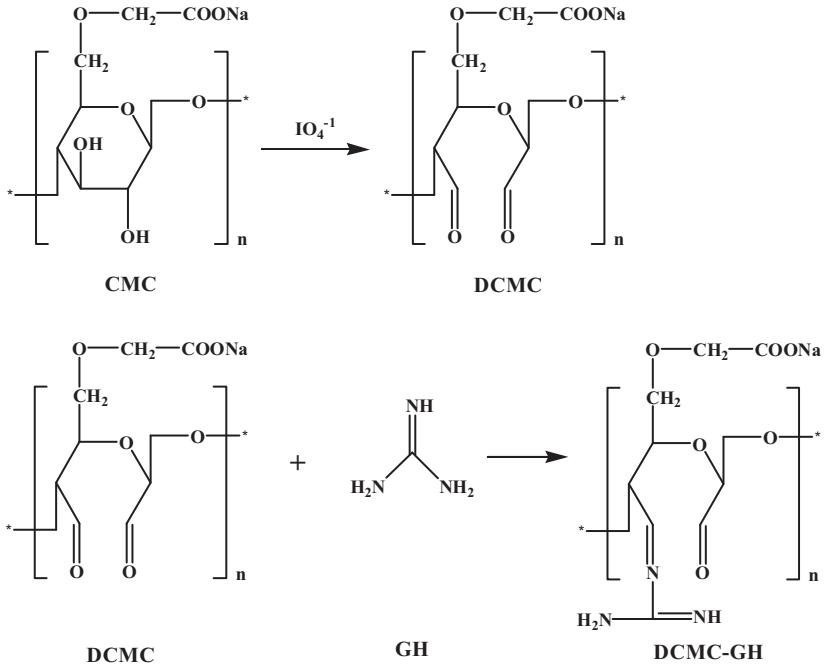

Fig. 1. Schiff base reaction of DCMC and guanidine hydrochloride.

with GH is prepared and used for the improvement of the strength property and antimicrobial activity of food packaging paper.

2. Materials and methods

2.1. Materials

Carboxymethyl cellulose sodium (CMC), guanidine hydrochloride (GH), and sodium periodate were purchased from Aladdin Reagent Co., Ltd (China). The viscosity of the 2% (w/v) CMC solution at 25 °C was 300–800 mPa s. All other chemicals were of analytical grade and used without further purification.

2.2. Oxidation of CMC with sodium periodate

The DCMC was prepared by oxidation of CMC according to the reference (Mu, Guo, Li, Lin, & Li, 2012): about 5 g CMC powder was dissolved in 100 mL distilled water at room temperature, 50 mL of 0.11 g/mL sodium periodate solution was then added into the above CMC solution under stirring. The pH of the mixture was adjusted to about 3.0 with 1 M sulfuric acid solution. After reaction for 4 h at 35 °C in the dark, the reaction was stopped by pouring the solution into a large amount of ethanol. The precipitate was centrifuged and washed with ethanol, followed by deionized water for three times to remove all the iodic compounds. The DCMC was obtained after the precipitate was freezing dried for 12 h.

2.3. Preparation of guanidine hydrochloride-grafted dialdehyde carboxymethyl cellulose (DCMC-GH)

About 1 g DCMC was dissolved in 20 mL distilled water under mechanical stirring at 50 °C. Guanidine hydrochloride was then added into the above solution and adjusted to about pH 5.0 with 1 mol/L NaOH solution. The molar ratios of guanidine hydrochloride and DCMC were 1:1, 1:4, 1:8, 1:12, and 1:16, respectively. After reaction at 60 °C for 2 h, the reaction was stopped by pouring the solution into a large amount of ethanol. The precipitate was centrifuged at 4,000 g for 15 min. The DCMC grafted with different GH contents was obtained after the precipitate was freezing dried for 24 h.

2.4. Characterization of DCMC-GH

The FTIR spectra of carboxymethyl cellulose, guanidine hydrochloride, and DCMC-GH were recorded using a Fourier transform infrared spectroscopy (Thermo Nicolet 360) at a resolution of 4 cm^{-1} in the spectral region of 500–4000 cm^{-1}, by making a KBr pellet with sample. Thermogravimetric analysis (TGA) of carboxymethyl cellulose, guanidine hydrochloride, and DCMC-GH were performed on a TG-DTA instrument (Netzsch STA 449F3). Approximately 5 mg of sample was weighed and heated from room temperature to 700 °C at a heating rate of 10 °C/min under nitrogen flow rate of 20 mL/min.

2.5. Determination of GH content grafted on carboxymethyl cellulose

The content of nitrogen in DCMC-GH was determined by the Kjeldahl method. The content of nitrogen was calculated by the formula (Hou, Liu, Liu, Duan, & Bai, 2008):

$$N\% = \frac{V \times C \times 14 \times 10^{-3} \times 100}{W}$$

where V is the volume of hydrochloric acid solution (HCl) used in titration for DCMC-GH (mL), C is the concentration of standard HCl solution (mol/L), W is the weight of testing sample (g).

The GH content grafted on carboxymethyl cellulose was calculated as follows:

$$GH\% = \frac{N\%}{14} \times 59$$

Table 1 lists the GH contents of DCMC-GH, which are prepared at the conditions of different molar ratios of GH/DCMC.

Table 1
The GH contents of DCMC-GH prepared at the different molar ratios of GH/DCMC.

	DCMC-0	DCMC-2	DCMC-4	DCMC-7	DCMC-8	DCMC-12
GH/DCMC (molar ratio)	–	1:16	1:12	1:8	1:4	1:1
GH content (%)	0	2.04	4.01	6.95	8.39	12.66

2.6. Coating of DCMC-GH onto paper

About 2 g DCMC-GH was added into 40 mL distilled water, and heated at 40 °C with mechanical stirring until the DCMC-GH was dissolved completely. After the DCMC-GH solution was cooled to room temperature, the samples of the copy paper (70 g/m^2) with width of 150 mm and length of 150 mm were immersed into the above solution for 10 and 30 min, respectively, and then dried at 70 °C for 20 min in a plate dryer. The amount of DCMC-GH coated on paper was about 30 mg/g paper.

2.7. Scanning electron microscope (SEM)

The morphology of the cross-section of the DCMC-GH-coated copy paper and blank copy paper was investigated using a scanning electron micrograph (Nova Nano SEM 230, FEI). The paper was cryofractured in liquid nitrogen. The fractured surface was then coated with gold on an ion sputter coater, and operating voltage of SEM was 5 kV.

2.8. Tensile strength measurement

The dry tensile property of the DCMC-GH-coated copy paper and blank copy paper were measured using a strength testing machine (DCP-KZ 300, Sichuan Changjiang Paper Instrument Co., Ltd., China). Dry strength was measured using TAPPI Test Method T494. For each sample, at least ten replicates were tested and the results were presented as the average of the tested samples.

2.9. Characterization of antimicrobial activity

To examine the antimicrobial effect of DCMC-GH-coated copy paper, *Escherichia coli* (*E. coli*) ATCC 8739 and *Staphylococcus aureus* (*S. aureus*) ATCC 6538, a Gram-negative and Gram-positive organism, respectively, were used as the test organisms. The bacteria were grown in LB liquid medium (10 g L^{-1} peptone, 5 g L^{-1} yeast extract, 10 g L^{-1} NaCl, and pH 7.0) for 12 h at 37 °C and further diluted to get concentration of about 10^5 CFU/mL with NaCl solution (0.85%, w/v). Then diluted suspensions (0.1 mL) of *E. coli* or *S. aureus* were distributed into the LB agar medium (10 g L^{-1} peptone, 5 g L^{-1} yeast extract, 10 g L^{-1} NaCl, and 15 g L^{-1} agar). A disk (8.5 mm diameter) was punched out of DCMC-GH-coated copy paper or blank copy paper, and was centrally placed on the solid agar plates, previously seeded with individual test bacteria. After incubation at 37 °C for 24 h, the zones of inhibition were observed.

3. Results and discussion

3.1. FTIR analysis

According to the reaction scheme shown in Fig. 1, the novel multifunctional coating agent based on carboxymethyl cellulose was prepared by two reaction steps. The first step was the oxidation of the carboxymethyl cellulose to obtain the dialdehyde carboxymethyl cellulose (DCMC), and the second step was the graft of guanidine hydrochloride (GH) onto the DCMC to obtain the DCMC-GH. The corresponding reaction process can be demonstrated by the FTIR spectra shown in Fig. 2. The FTIR spectrum of CMC showed the characteristic peaks at 1624 and 1418 cm^{-1} due to the symmetric and asymmetric modes of stretching vibration of carboxylic groups (COO$^-$) (Yadav, Rhee, Jung, & Park, 2013; Luna-Martinez et al., 2011; Su, Huang, Yuan, Wang, & Li, 2010). When the CMC was oxidized by periodate, the C2—C3 bond of glucose residues was cleaved. This specific cleavage led to the formation of two aldehyde groups per glucose unit, thus a characteristic peak was appeared at 1741 cm^{-1}(C=O stretch) in the spectrum of DCMC. Another characteristic peak at 897 cm^{-1} was attributed to the formation of hemiacetal bonds between the aldehyde groups and neighbor hydroxyl groups (Li, Wu, Mu, & Lin, 2011; Mu et al., 2012).

After the guanidine hydrochloride was grafted on DCMC, the major change in the spectrum for DCMC-GH was that an intensive characteristic peak at 1598 cm^{-1}(N–H bending vibration) and 3392 cm^{-1} (N–H stretching vibration) were found. The results indicated that guanidine hydrochloride was successfully grafted onto DCMC via the Schiff base reaction.

3.2. Thermogravimetric analysis (TGA)

The thermogravimetric analysis of CMC, GH, and DCMC-GH has been performed, and the results were shown in Fig. 3. It can be seen from Fig. 3a that 8.4, 36.9, and 14.5 wt% degradation of CMC during the first, second, and third step, respectively were found. The weight loss of CMC before 200 °C during the first step was mainly due to water evaporation, which may be from the free water of CMC, and the second mass loss of about 36.9% above 200 °C was ascribed to the thermal decomposition of CMC (Liu et al., 2011; Li, Sun, & Wu, 2009). After the GH was grafted onto DCMC, the DCMC-GH (DCMC-12, 7, and 2) also showed three main stages of mass loss. The first mass loss was found from 22 to 176 °C, possibly due to the loss of free water in CMC. The second mass loss was observed at temperature 176–308 °C, mainly attributed to the decomposition of CMC. For the third mass loss appeared at 308–700 °C was mainly due to the decomposition of both CMC and GH (Ma, Chang, & Yu, 2008; Mu et al., 2012).

In addition, Fig. 3b showed the derivative thermogravimetric (DTG) curves of CMC, GH, and DCMC-GH. It was found that all the DCMC-GH had lower peak degradation temperature than did the CMC and GH. It was very likely that CMC had a lower peak

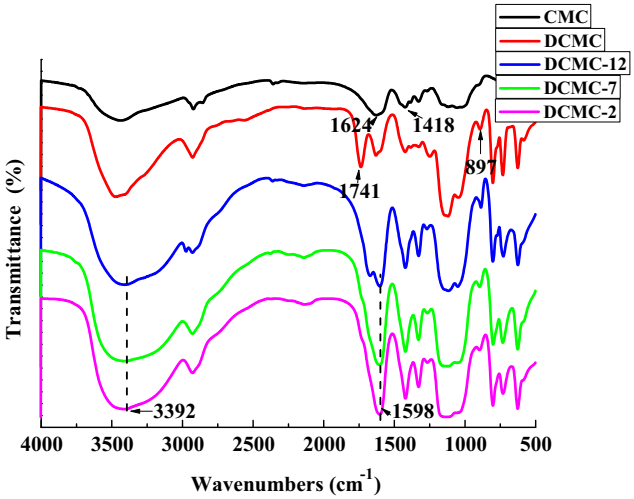

Fig. 2. FTIR spectra of CMC, DCMC, DCMC-12, DCMC-7, and DCMC-2.

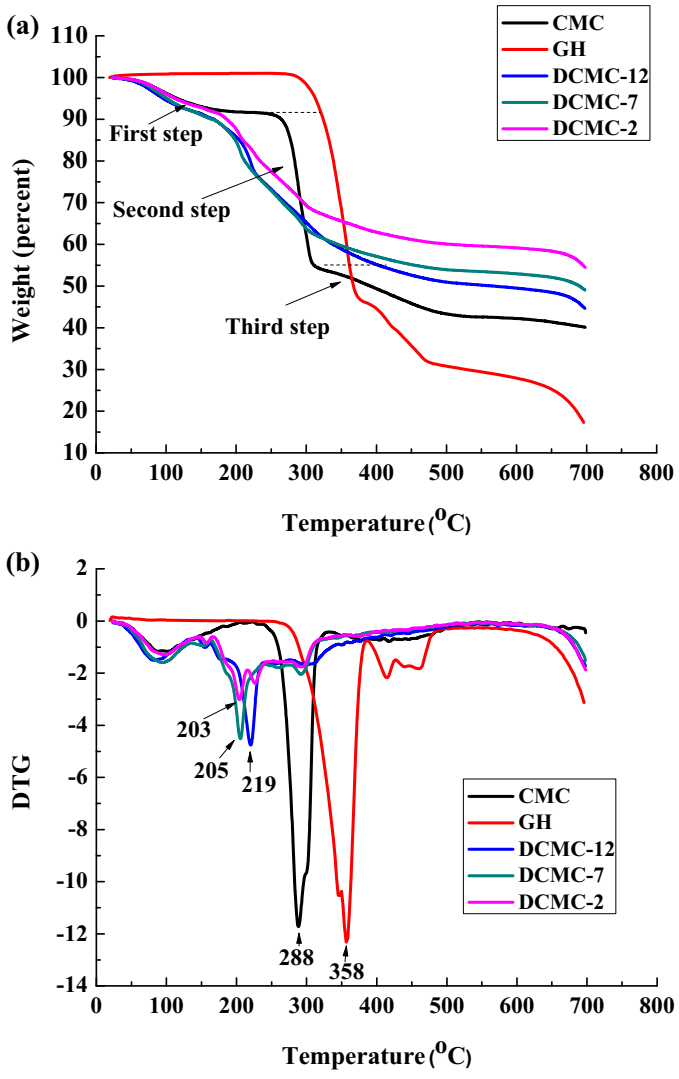

Fig. 3. The thermogravimetric (TG) (a) and derivative thermogravimetric (DTG) (b) curves of CMC, GH, DCMC-12, DCMC-7, and DCMC-2.

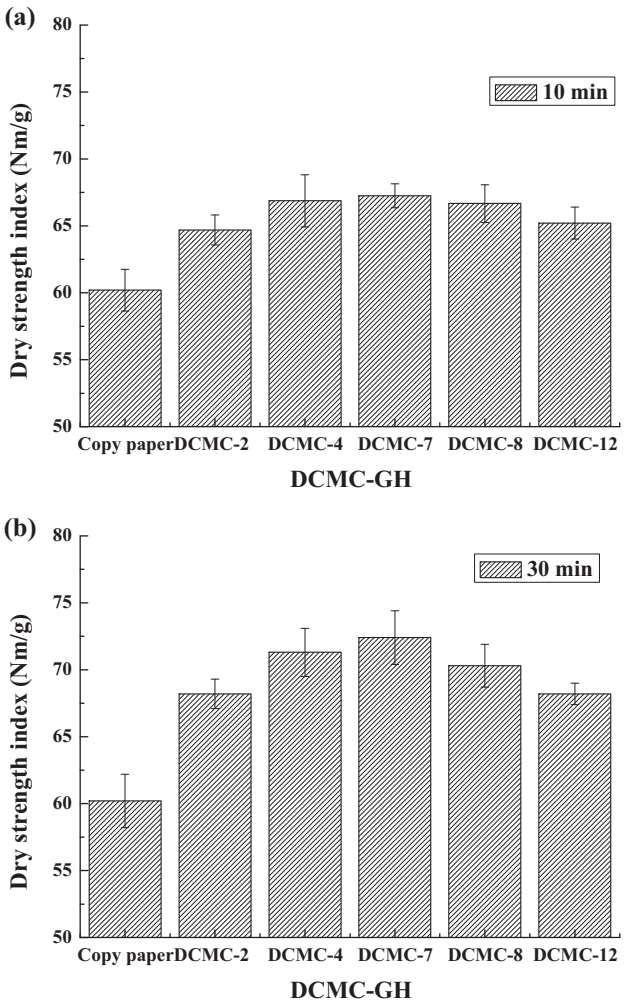

Fig. 5. Dry strength index of blank copy paper and DCMC-GH-coated copy paper: (a) immersed for 10 min, (b) immersed for 30 min.

degradation temperature than did the GH, thus exhibited the poor thermal stability. The decomposition of CMC component in DCMC-GH could accelerate the decomposition of the DCMC-GH, thus leading to the DCMC-GH had a lower peak decomposition temperature. Liu prepared the Zr-CMC/GPS composites and found the similar results (Liu et al., 2011). It should be noted that the DCMC-GH with the degradation temperature above 200 °C could satisfy the need of food packaging.

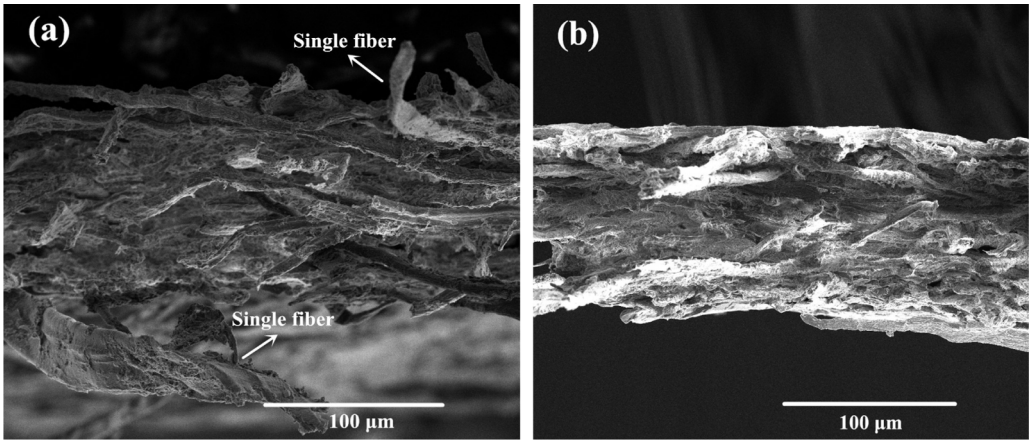

Fig. 4. SEM images of the fractured cross-sections of (a) blank copy paper and (b) DCMC-12-coated copy paper.

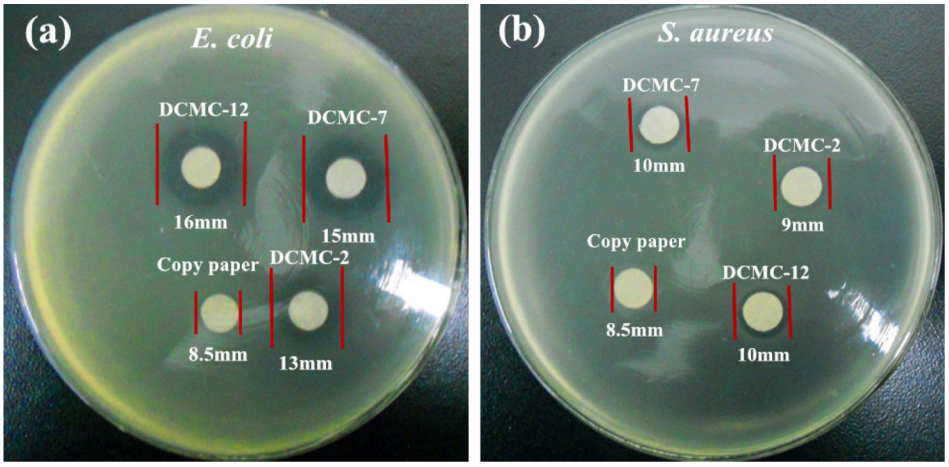

Fig. 6. The antimicrobial activities of blank copy paper and DCMC-GH-coated copy paper against *E. coli* (a) and *S. aureus* (b).

3.3. SEM analysis of DCMC-GH-coated copy paper

SEM images of the cross-section of the DCMC-GH-coated copy paper and blank copy paper are shown in Fig. 4. It can be seen from Fig. 4a that the fibers in blank copy paper were dispersed very loosely, the surface looked very rough, and some fibers were broken away from the surface. This would have an adverse impact on the paper strength. Whereas, the cross-sectional images of the DCMC-GH-coated copy paper in Fig. 4d showed a dense structure and a smooth surface, thus improving the bondability between fibers. This would be very important to the improvement of paper strength.

3.4. Strength property of paper

The CMC has been used to improve the paper strength because it has similar structure to cellulosic fiber and can form a number of hydrogen bonds with fibers. In this work, the DCMC-GH was used to coat on paper, and the effects of DCMC grafted with different contents of GH on the paper strength have been studied, the results were shown in Fig. 5. It can be seen from Fig. 5a that the dry strength index of paper increased from 60.2 to 67.3 N m/g when the paper was immersed in DCMC-7 solution for 10 min. The short retention time may induce part of the inner fibers of copy paper not to form bonds with DCMC-GH, thus resulting in the small extent of improvement of paper strength. However, when the retention time increased to 30 min, the dry strength index increased from 60.2 to 72.4 N m/g as the DCMC-7 was used to coat, showing that DCMC-GH was an effective agent on the improvement of dry strength property. This should be the reason that a lot of bonds can form between the $-COO^-$ and $-NH_2$ groups of the DCMC-GH and the —OH group of the paper fibers (Salam, Lucia, & Jameel, 2013). In addition, the increased tightness of the DCMC-GH-coated copy paper compared to the blank copy paper also contributed to the improvement of paper strength. It should be noted that the dry strength index begins to decrease slowly when the DCMC grafted with high content of GH (DCMC-8 and 12) was used to coat on paper, this may be the reason that the excessive $-NH_2$ brought from DCMC-8 or 12 would prevent the hydrogen bonds between cellulose fibers during the drying process of paper, thus decreasing the paper strength (Zhong, Peng, Yang, Cao, & Sun, 2013).

3.5. Assay of antimicrobial activity

In addition of strength property, antimicrobial activity is also a very important character for food packaging paper. Antimicrobial agent was often used in paper products to prevent people from being attached by harmful bacteria in people's daily life. Therefore, the antimicrobial activities of the DCMC-GH-coated copy paper against *E. coli* and *S. aureus* have been tested by the inhibition zone method (Das, Ara, Dutta, & Mukherjee, 2011). Fig. 6 showed the test results of the antimicrobial activity. It can be seen from Fig. 6a that the diameter of inhibition zone for the DCMC-2, 7, 12-coated copy paper against *E. coli* was 13, 15, 16 mm, respectively, suggesting that the DCMC-GH-coated copy paper have excellent antimicrobial activity against *E. coli*, and the antimicrobial activity was improved as the increasing of the GH content grafted on the DCMC. This was the fact that guanidine can destruct the cell membrane of bacteria and cause the leakage of intracellular components from bacterial cells, thus has strong antimicrobial activity against *E. coli* (Qian, Guan, He, & Xiao, 2008b).

It was found from Fig. 6b that the DCMC-GH-coated copy paper also had a certain antimicrobial activity against *S. aureus*, but the diameter of inhibition zone against *S. aureus* was smaller than that of inhibition zone against *E. coli* at the same GH content grafted on DCMC. This can be explained by the reason that *E. coli* is a Gram-negative organism and can be adsorbed onto the positively charged surface of DCMC-GH containing a number of $-NH_3^+$ group through electrostatic interaction, which resulted in the larger inhibition zone (Zhou et al., 2009).

4. Conclusions

In summary, a novel multifunctional coating agent based on CMC was prepared for the first time. Herein, the CMC was first oxidized by periodate to form aldehyde groups and then guanidine hydrochloride was grafted onto the DCMC according to the Schiff base reaction. The coating of DCMC-GH onto copy paper resulted in the improvement of paper strength property because of the hydrogen bonds between CMC and paper fibers. Meanwhile, the DCMC-GH-coated copy paper exhibited excellent antimicrobial activities against *E. coli.* and *S. aureus* due to the antimicrobial activity of guanidine in DCMC-GH. According to the same mechanism, the guanidine can also be grafted onto other nature biopolymers with aldehyde groups, and resulting in high antimicrobial activity.

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