β-Cyclodextrin Grafted Cellulose and Cationic Starch for Antibacterial Paper Products: A Comparative Study

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Two kinds of macromolecules applied in papermaking were modified with β-cyclodextrin (β-CD) and loaded with ciprofloxacin hydrochloride (CipHCl) in an attempt to compare their potential applications in antimicrobial paper. β-CD grafted cellulose (β-CD-Cel) and β-CD grafted cationic starch (β-CD-CS) were prepared by grafting β-CD onto cellulose fiber and cationic starch using citric acid (CA) and epichlorohydrin (ECH) as crosslinking agents, respectively. β-CD-Cel and β-CD-CS were both loaded with an antimicrobial agent (CipHCl) to form inclusion complexes, namely β-CD-Cel-CipHCl and β-CD-CS-CipHCl. Furthermore, the inclusion complexes were added to the pulp to prepare antibacterial paper. The antimicrobial activity and physical properties of the paper were investigated. The results showed that the paper with both inclusion complexes exhibited excellent antibacterial activity, and the antibacterial activity with β-CD-CS-CipHCl was higher than that with β-CD-Cel-CipHCl. Moreover, the addition of both β-CD-Cel-CipHCl and β-CD-CS-CipHCl affected the tensile and tear strengths of the paper. The paper with β-CD-CS-CipHCl had better physical properties than that with β-CD-Cel-CipHCl because the CS acts as a reinforcing agent in papermaking. These prepared antibacterial paper sheets may be useful for preventing wound and nosocomial infections in the medical and packaging fields.

Keywords: β-cyclodextrin; Cellulose fibers; Cationic starch; Grafting; Antibacterial property

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INTRODUCTION

Cyclodextrins (CDs) are a series of cyclic oligosaccharides composed of 6-8 D-glucose units and named α-, β-, and γ-CD, respectively. The D-glucose units are covalently bonded together *via* α-1,4-linkages to form torus-like structures with lipophilic inner cavities and hydrophilic outer surfaces. Every D-glucose unit has three free hydroxyl groups that differ both in their functions and reactivities. All of the secondary hydroxyl groups at the 2- and 3-positions of the D-glucose units are on one side of the torus, and all of the primary hydroxyl groups at the 6-positions of the D-glucose units are on the other side of the ring (Gonil *et al*. 2011). They are considered biocompatible and, in general, they do not elicit an immune response and have low toxicities. Furthermore, it is well-known that CDs possess a remarkable ability to form inclusion complexes with various small molecules *via* non-covalent interactions in their hydrophobic cavities. However, a limiting factor of CD is the absence of mucoadhesive properties (Sajomsang *et al*. 2013). To improve the mucoadhesive property of the CDs, the CDs can be introduced into a bioadhesive polymer. CD-based polymers with versatile functions are attracting more and more attention in various fields.

Here, some β-CD grafted polysaccharides were prepared and characterized in an attempt to apply them in the papermaking process. Polysaccharides such as cellulose and its derivatives and starch and its derivatives occupy a strategic position in papermaking and in other industries (Hebeish *et al*. 2010). Cellulose is the most abundant natural macromolecule on Earth. It is virtually an inexhaustible source of raw material that is environmentally friendly (non-toxic), biocompatible, renewable, and modifiable, which makes it one of the most promising feedstocks for future industrial use (Ioelovich *et al*. 2010; Medronho *et al*. 2013). Starch is composed of two polymers of α-glucose: linear amylose and a highly branched amylopectin. Amylose molecules consist of 200 to 20,000 glucose units that form a helix as a result of the bond angles between the them (Tang and Alavi 2011).

Starch has attracted much attention and industrial use in recent years because of its relatively low price, renewability, and biodegradability. However, the utilization of native starch is limited due to its physicochemical properties. Therefore, starch is usually modified physically or chemically to improve its functionality for industrial applications and thus to extend its usefulness. Cationic starch, a chemically modified starch, is widely used as an additive, for example in papermaking to improve mechanical strength, retention of fines, dyes, and fillers, faster drainage, runability of the paper machine, and to decrease biological oxygen demand of paper mill effluent (Kuo and Lai 2007). To combine the advantages of both CDs and polysaccharides, the CD grafted polysaccharides, such as cellulose and cationic starch, have gained interest because they can be widely used in various fields (Buschmann *et al*. 1998; Gao and Zhao 2004; Martel *et al*. 2002; Ozmen and Yilmaz 2007; Xiao *et al*. 2007; Grigoriu *et al*. 2011; Cusola *et al*. 2013). Therefore, the introduction of CD moieties into the polysaccharide backbone may lead to a molecular carrier that possesses the cumulative effects of inclusion, size specificity, and transport properties of CDs as well as the biocompatible and renewable properties of the polymeric matrix.

In this work, β-CD was first grafted onto cellulose fiber and cationic starch, and then the cavities of β-CD were loaded with ciprofloxacin hydrochloride (CipHCl) to impart antimicrobial activity to the polysaccharides. Moreover, the obtained polysaccharide inclusion complexes were added to the pulp to prepare antibacterial paper. The antimicrobial activity and physical properties of the paper were also investigated.

EXPERIMENTAL

Materials

Cellulose fibers of softwood were provided by Liangmianzhen Co. Ltd., Guangxi, China. The fibers were treated with 1.5 M sodium hydroxide solutions for 2 h at 50 °C, and then the pretreated pulp fibers were refined in a PFI (model Mark V1; Pulp and Paper Research Institute of Norway) to a beating degree of 35 °SR. Cationic starch was supplied by Liangmianzhen Co. Ltd., Guangxi, China; its degree of substitution (DS) was 0.034. *E. coli* (ATCC 8739), *S. aureus* (ATCC 6538), LB agar, and LB broth were purchased from Huankai Microbial Sci. & Tech. Co. Ltd. (Guangdong, China). Citric acid (CA), epichlorohydrin (ECH), β-cyclodextrin (β-CD), sodium hypophosphite (SHP), anhydrous ethanol, sodium hydroxide, and ciprofloxacin hydrochloride (CipHCl) were of analytical grade and used without further purification.

Preparation of β-CD Grafted Cellulose (β-CD-Cel) and its Inclusion Complex

A mixture of 3.0 g of β-CD, 1.1 g of CA, 0.29 g of SHP, and 1.8 g of distilled water was reacted at 100 °C for 1.5 h. The sample was purified by washing with ethanol using a soxhlet for 6 h, followed by drying at 60 °C for 24 h to obtain the purified CA-β-CD. Then, 0.3 g of CA-β-CD and 0.1 g of SHP were dissolved in distilled water (20 mL), followed by the addition of pretreated cellulose fiber (1 g, oven dry) to obtain sufficient swelling. The reaction proceeded at 160 °C for 15 min. The obtained fibers (β-CD-Cel) were washed with distilled water and dried at 60 °C for 24 h. The grafting ratio of β-CD onto the cellulose fibers was calculated by the weight difference between the ungrafted and grafted cellulose fibers (Wang and Cai 2010), which was 9.7% in this study.

Grafted cellulose fibers (1 g) were immersed into 50 mL of CipHCl aqueous solution (100 mg/g oven dry fiber) at room temperature for 4 h under stirring. The obtained samples were washed with distilled water and dried at 60 °C for 24 h in a vacuum drying oven. The amount of loaded drug was calculated as the difference between the initial and the final amounts in the solution using a UV-visible spectrophotometer (DR 5000; HACH) at 275 nm (Peila *et al*. 2012). The loading amount of CipHCl was about 13.6 mg/g β-CD-Cel.

Preparation of β-CD Grafted Cationic Starch (β-CD-CS) and its Inclusion Complex

Five grams of β-CD was dissolved in 20 mL of sodium hydroxide solution (300 g/L), followed by the addition of 2.47 mL ECH; the mixture was then reacted at 40 °C for 0.5 h. Then, 2.5 g of gelatinized cationic starch was added. After reacting at 40 °C for 2 h, absolute ethanol was added and a white flocculent precipitate was obtained. The precipitate was collected by centrifugation and then washed with an ethanol solution (50%, v/v). β-CD-CS was obtained by vacuum drying at 75 °C for 24 h. The DS and β-CD content of β-CD-CS was determined by GB 12091-89 and UV spectrophotometry, respectively. The DS of β-CD-CS was 0.027, and the grafting ratio of β-CD in β-CD-CS was 14.3%.

The complexes were prepared by a kneading method according to the procedures described by Arias *et al*. (1997) and Xin *et al*. (2010). β-CD-CS (1 g) and distilled water (2 mL) were mixed together in a mortar to obtain a homogeneous paste. CipHCl (0.2 g) was added slowly to the above mixture. The mixture was then ground uniformly for 1 h at room temperature. The obtained paste was dried in a vacuum oven at 50 °C for 24 h and then washed with cold ethanol. The complex was then dried again and pulverized into fine powder. The amount of loaded drug was determined by UV spectrophotometry. The loading amount of CipHCl was 25 mg/g β-CD-CS.

Characterization of β-CD-Cel and β-CD-CS

FTIR spectra of β-CD, β-CD-Cel, and β-CD-CS were obtained using a Fourier transform infrared spectrophotometer (Vector 33; Bruker) in a wavenumber range of 4000 to 500 cm^{-1} with the KBr disk method.

CP/MAS ^{13}C-NMR spectra of β-CD and β-CD-Cel were obtained using a Bruker II Advance-300 spectrometer (Karlsruhe, Germany) operating at resonance frequencies of 300.14 MHz for ^{1}H and 75.47 MHz for ^{13}C, using a spinning Bruker 4.0 mm MAS probe. Spinning speed of the samples was set to 5 kHz at room temperature. Each spectrum was obtained with an accumulation of 5000 scans. The delay time was 60 s, the proton 90° pulse width was 9 μm, and the contact time for cross polarization was 2.5 ms.

^{1}H NMR spectra of β-CD and β-CD-CS were recorded on a Bruker AV 300 MHz spectrometer operating at 500.13 MHz in D_2O. The chemical shift was normalized to an external sample of TMS (δ=0.00) in a capillary.

Preparation of Antibacterial Paper and its Physical Properties

Pulp with various ratios of β-CD-Cel-CipHCl to virgin fiber was diluted to 0.5% consistency. After agitating for 15 min, handsheets (60 g•cm^{-2}) were made in an Australia hand-sheet former according to the ISO 5269/2 standard.

Various dosages of β-CD-CS-CipHCl solutions based on oven-dry weight of the pulp were added to a 1% suspension of pulp. The mixture was agitated for 15 min at room temperature and then diluted with water to 0.5% consistency before forming the handsheets. After drying, all paper sheets were placed for conditioning at 52% relative humidity and a temperature ranging from 23 to 24 °C. The tensile index of the paper sheets was measured using an Alweron TH1 tensile tester (model CE064, AB Lorentzen & Wettre). The tear index was performed in accordance with ISO 1974 with a tearing tester (model 83-10, Lorentzen & Wettre).

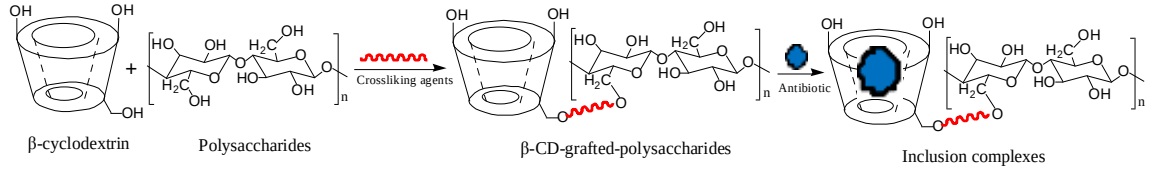
Antibacterial Activity of the Paper Sheets

The antibacterial activities of the prepared paper sheets against *E. coli* and *S. aureus* were assessed using the disc diffusion method (Scott and Higham 2003). First, 200 μL of *E. coli* (2.5×10^7 CFU mL^{-1}) or *S. aureus* (1.5×10^7 CFU mL^{-1}) suspensions were uniformly spread on agar discs and circles of paper with diameters of 6 mm were put on them. After the discs were incubated overnight at 37 °C, the bactericidal activity was observed by visual inspection of the clearance of the bacterial lawn as well as the size of the clearance as measured by calipers across an average diameter. Experiments were done in triplicate.

RESULTS AND DISCUSSION

Characterization of β-CD-Cel and β-CD-CS

The process of preparing β-CD grafted polysaccharides and the inclusion with antibiotics are shown in Scheme 1. Both β-CD and polysaccharides contain several reactive hydroxyl groups that can be linked by crosslinking agents. CA is a polycarboxylic acid with three carboxylic groups, which are capable of forming ester linkages with hydroxyl groups of both cellulose fibers and β-CD, thus binding the β-CD to cellulose fibers through the formed ester linkage with CA (Gawish *et al*. 2009; Dong *et al*. 2014). ECH has been used as a crosslinking agent for β-CD that can react with hydroxyl groups of β-CD. Additionally, CS can react with ECH because it also contains several hydroxyl groups in its structural unit (Liu *et al*. 2007).



Scheme 1. Schematic representation of β-CD grafted onto polysaccharides and its inclusion with antibiotics

The FTIR spectra of β-CD, β-CD-Cel, and β-CD-CS are shown in Fig. 1. In the FTIR spectrum of β-CD (Fig. 1a), the extremely broad band at 3387 cm^{-1} and the band at 2930 cm^{-1} can be attributed to O-H stretching and C-H stretching vibrations, respectively. Meanwhile, the bands at 1159, 1084, and 993 cm^{-1} are characteristic of C-O stretching vibrations. In the case of β-CD-Cel, a new band at 1738 cm^{-1} can be associated with the C=O stretching vibration for an ester bond in the reaction between carboxylic acid from CA-β-CD and hydroxyl groups from cellulose fibers. Moreover, the FTIR profile of β-CD-CS was similar to that of β-CD because β-CDs are formed from enzymatic degradation of starch by bacteria. Besides the characteristic peaks of the starch backbone, an additional band at 1460 cm^{-1} is assigned to the C-N stretching vibration of the substituted cationic group from the cationic starch. This band is not present in β-CD and thus is evidence of the grafting of β-CD onto the cationic starch.

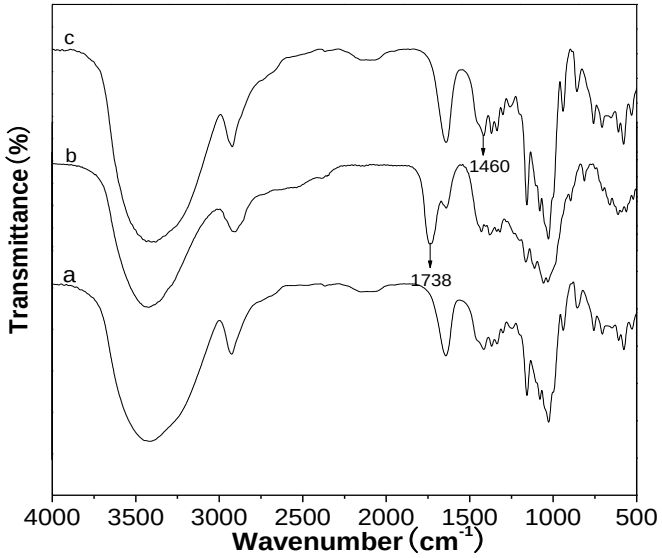


Fig. 1. FTIR spectra of β-CD (a), β-CD-Cel (b), and β-CD-CS (c)

The CP/MAS ^{13}C NMR spectra of native cellulose and β-CD-Cel are shown in Fig. 2A. For cellulose, the signals between 55 and 110 ppm are attributed to various carbon atoms in the glucose; there are six peaks corresponding to ^{13}C chemical shifts of cellulose carbons, C1 (105.8 ppm), C4 (78 ppm), C2/C3/C5 (70-80 ppm), and C6 (66.2 ppm) of anhydroglucose units of cellulose (Sathitsuksanoh *et al*. 2011). Compared to the spectrum of cellulose fibers, a new signal at 173.8 ppm is assigned to the carbonyl group of modified cellulose fibers for an ester bond, which confirmed the reaction between carboxylic acid from CA-β-CD and hydroxyl groups from cellulose fibers. In addition, an obvious signal at 45.1 ppm appeared due to $-CH_2-$ group of CA. This also confirmed the reaction between CA-β-CD and cellulose; therefore β-CD was grafted onto cellulose fibers with citric acid as a bridge.

The ^{1}H NMR spectra of β-CD and β-CD-CS are shown in Fig. 2B. In the ^{1}H NMR spectrum of β-CD, the peaks at 4.83, 3.39, 3.82, 3.30, 3.49, and 3.65 ppm are attributed to H-1, H-2, H-3, H-4, H-5, and H-6 of D-pyran glucose units; the peaks recorded between 5.0 and 5.5 ppm are ascribed to the hydrogen atoms of hydroxyl groups, and the peak at 3.6 ppm is attributed to the O-H group (Guan *et al*. 2014). Compared to

β-CD, the β-CD-CS exhibited an additional peak at nearby 3.19 ppm, which can be ascribed to the CH_3-N^+ group substituted cationic group from cationic starch (Wang and Xie 2010). The existence of this signal resulting from the hydrogen atom of the substituent indicated that CS was linked with β-CD and the etherification reaction occurred.

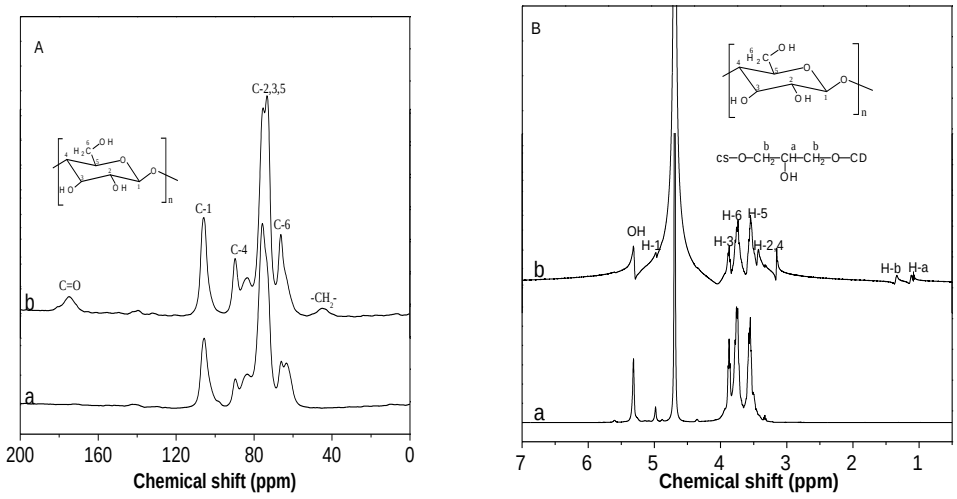


Fig. 2. CP/MAS ^{13}C NMR spectra (A) of native cellulose (a) and β-CD-Cel (b); ^{1}H NMR spectra (B) of β-CD (a) and β-CD-CS (b)

Antibacterial Activity of the Prepared Paper Sheets Loaded with CipHCl

The antibacterial activity was determined based on the diameter of the growth inhibition zone. Photographs of antibacterial paper and the inhibition zone values are shown in Fig. 3 and Table 1, respectively. To confirm the antimicrobial activity of β-CD-Cel and β-CD-CS, two control samples containing 10% β-CD-Cel and 2.5% β-CD-CS unloaded with CipHCl were prepared as well. No inhibition zone was found around the control samples (Figs. 3a, 3b, 3e, and 3f), whereas a distinct inhibition zone can be observed around the paper sheets containing CipHCl (Figs. 3c, 3d, 3g, and 3h) against *E. coli* and *S. aureus*. This indicates that neither β-CD-Cel nor β-CD-CS showed any self-antimicrobial activity, and paper sheets containing CipHCl exhibited a remarkable antibacterial activity against both gram-negative and gram-positive organisms. Moreover, the diameter of the inhibition zone increased with a higher content of β-CD-Cel-CipHCl and β-CD-CS-CipHCl due to an increase in the amounts of CipHCl incorporated into the paper sheets. In addition, the inhibition zones of the paper sheets prepared with β-CD-Cel-CipHCl against *E. coli* and *S. aureus* were 17.1 and 15.3 mm, respectively, when the content of β-CD-Cel-CipHCl was 10%; that is, the adding amount of CipHCl was 2.56 mg/per paper, whereas that of the paper sheets prepared with β-CD-CS-CipHCl against *E. coli* and *S. aureus* were 16.9 and 15.8 mmm, respectively, when the content of β-CD-CS-CipHCl was 2.5%, that is, the adding amount of CipHCl was 1.18 mg/per paper. For two inclusion complexes of β-CD grafted polysaccharides with CipHCl, paper sheets with β-CD-CS-CipHCl exhibited similar antimicrobial activity to that with β-CD-Cel-CipHCl at lower adding amounts of CipHCl. Therefore, a conclusion can be drawn that the antibacterial activity of the paper with β-CD-CS-CipHCl is higher than that of the paper with β-CD-Cel-CipHCl at the same adding amounts of CipHCl during the papermaking

process. This is mainly because a large proportion of CipHCl molecules in β-CD-Cel-CipHCl are physically adsorbed onto the surfaces of the fibers or entrapped in the interstices of the fiber wall, most of which can be desorbed and flowed away with white water during the papermaking process.

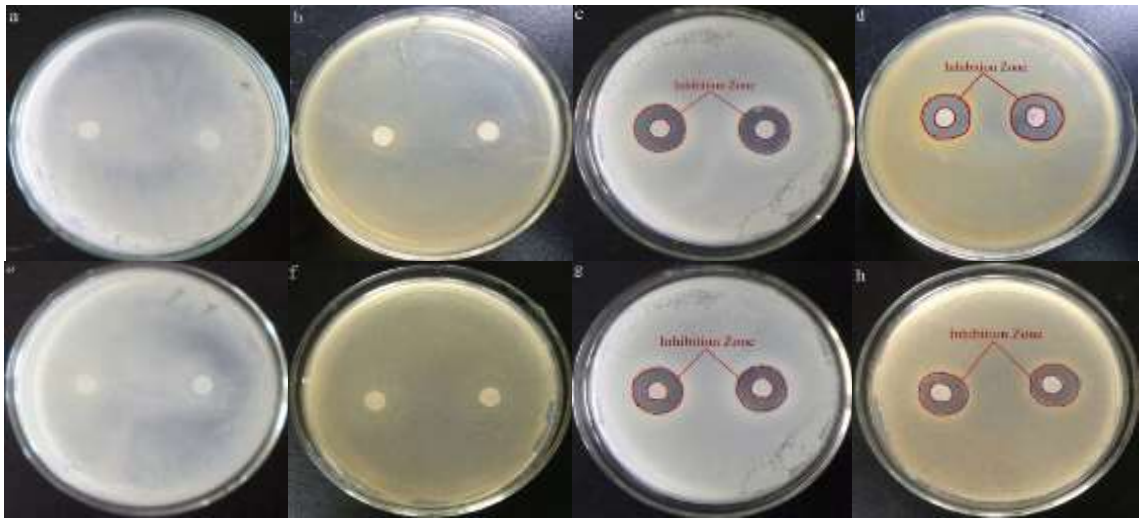


Fig. 3. The antimicrobial activities of paper sheets containing 10% β-CD-Cel (unloaded with CipHCl) against *E. coli* (a) and *S. aureus* (b); paper sheets containing 10% β-CD-Cel-CipHCl against *E. coli* (c) and *S. aureus* (d); paper sheets containing 2.5% β-CD-CS (unloaded with CipHCl) against *E.coli* (e) and *S.aureus* (f); paper sheets containing 2.5% β-CD-CS loaded with CipHCl against *E. coli* (g) and *S. aureus* (h)

Table 1. Antimicrobial Activity of Paper Sheets Made from Inclusion Complex of β-CD-Cel/β-CD-CS and CipHCl against *E. coli* and *S. aureus* by Diffusion Method

Samples	Content of β-CD-Cel-CipHCl/β-CD-CS- CipHCl (wt% to oven dry pulp)	Loading amount of CipHCl (mg/ per paper)	Diameter of inhibition zone against *E.coli* (mm)	Diameter of inhibition zone against *S.aureus* (mm)
β-CD-Cel-CipHCl	Control sample-1	0	-	-
	10	2.56	17.1	15.3
	25	6.40	21.2	18.6
β-CD-CS-CipHCl	Control sample-2	0	-	-
	2.5	1.18	16.9	15.8
	5.0	2.36	20.9	18.7

Physical Properties of the Prepared Paper Sheets Loaded with CipHCl

The physical properties of paper sheets with various contents of β-CD-Cel-CipHCl are shown in Fig. 4A. The tensile and tear strengths of the control paper were 43.7 N•m/g and 15.4 mN•m^2/g, respectively. With increasing content of β-CD-Cel-CipHCl, the tensile and tear strengths of the paper sheets dropped dramatically. This is

the combined result of two opposing actions. On the one hand, tensile and tear strength of cellulose fibers decreased due to the cellulosic degradation when they were cured at a high temperature in the acidic medium; on the other hand, the strength was improved due to the enhancement in the crosslinking reaction of cellulose through esterification with CA-β-CD. It is clear from the results that the loss in tensile and tear strengths due to cellulosic fiber degradation overcomes the improvement due to the crosslinking reaction; the net result is that the tensile and tear strengths decreased with increasing content of β-CD-Cel-CipHCl. Strengthening agents, such as cationic starch and cationic polyacrylamide, could be added to overcome the decrease of paper strength resulting from the use of β-CD-Cel-CipHCl in an actual papermaking process. Furthermore, the brightness of the control paper was 82.5%, while that of paper made from 100% β-CD grafted cellulose fibers was only 74.2%. The lower brightness of grafted fibers compared to virgin fibers is due to the fact that cellulose fibers become yellow under the high temperature in the grafting reaction.

The physical properties of paper sheets with various contents of β-CD-CS-CipHCl are shown in Fig. 4B. With the increase of β-CD-CS-CipHCl, the tensile strength of the paper firstly increased and then dropped. When the adding amount of β-CD-CS-CipHCl was 2.5%, the tensile and tear index were increased 36.8% and 9.0%, respectively. It is believed that β-CD-CS-CipHCl adsorbs to cellulosic fibers during wet end processing, thereby enhancing bonding strength between the fibers during the process of sheet forming (Eriksson *et al*. 2005; Hubbe 2007). It is well known that the adsorption of CS on fibers will reach a saturation condition and any further increase of the adsorption amount beyond the saturation point is impossible. Therefore, the amount of β-CD-CS-CipHCl on cellulose fibers is limited by its maximum adsorption amount. When the adding amount was more than 5.0%, the positively charged starch may also cause fiber flocculate and poor paper formation uniformity, resulting in the decreased tensile strength (Shen *et al*. 2009; Yoon and Deng 2006). The tear strength of paper sheet behaved in a manner similar to that of the tensile strength with respect to β-CD-CS-CipHCl addition.

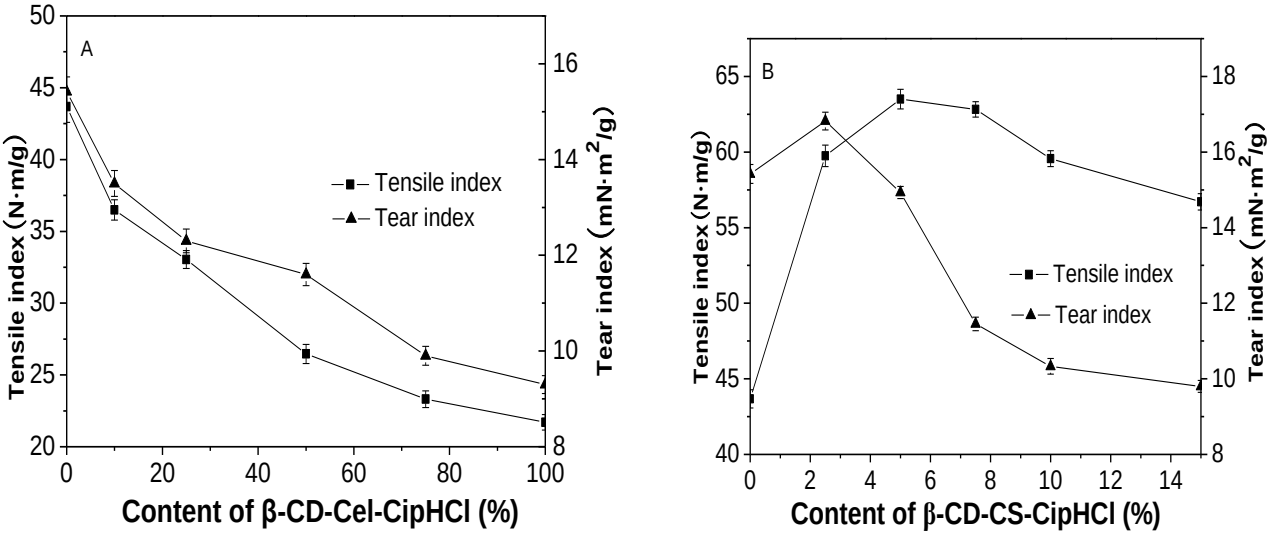


Fig. 4. Physical properties of paper sheets with various contents of β-CD-Cel-CipHCl (A) and β-CD-CS-CipHCl (B)

CONCLUSIONS

1. β-CD grafted polysaccharides were obtained by grafting β-CD onto cellulose fiber and cationic starch with crosslinking agents of citric acid (CA) and epichlorohydrin (ECH), respectively.
2. The paper sheets with added β-CD-Cel-CipHCl and β-CD-CS-CipHCl showed excellent antibacterial activity, and the antibacterial activity of paper with β-CD-CS-CipHCl was higher than that with β-CD-Cel-CipHCl.
3. With increased content of β-CD-Cel-CipHCl, the tensile and tear strengths dropped dramatically, whereas the tensile and tear strengths first increased and then decreased for β-CD-CS-CipHCl.

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