Deconstruction and Dissolving of a Fibrous Sodium Carboxymethyl Cellulose Network in a Water Solution

Hongjie Zhang,* Yufeng Hou, Pinyu Yu, Wenhui Zhang, and Qun Li

Sodium carboxymethyl cellulose (CMC) has received a tremendous amount of attention because of its outstanding solubility in water. In this study, the deconstruction behavior of a fibrous CMC network in water was expressed with kinetic models. The experimental results suggested that the water adsorption process was the key factor in the deconstruction of a fibrous CMC network, and it was controlled by the physical adsorption, which was confirmed by the high correlation coefficient ($R^2 > 0.95$) of the pseudo-first-order kinetic model. The dissolving behavior of a fibrous CMC network in water was monitored with the focused beam reflectance measurement technique, which showed that the whole process included the network deconstruction and dissolution of the fibrous CMC itself. The CMC network dissolving process consisted of three periods: a deconstruction period (t_0 to t_1), deconstruction and partial dissolution period (t_1 to t_2), and partial dissolution period (after t_2). The first deconstruction period (t_0 to t_1) was the key period that controlled the dissolving efficiency of the water-soluble handsheets. The deconstruction period was 17 s for the CMC with a degree of substitution (DS) of 0.80, while it was 34 s for the CMC with a DS of 0.50.

Keywords: Carboxymethyl cellulose; Deconstruction behavior; Adsorption kinetics; Dissolving process; Solubility

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INTRODUCTION

Sodium carboxymethyl cellulose (CMC), one of the most important functionalized cellulose derivatives, is prepared by its reaction with caustic alkali and chloroacetic acid and plays a significant role in many technical, industrial, and medical applications (Liu *et al.* 2014). Generally, CMC is a white or milky, non-toxic, fibrous powder or particles. Due to the superior performance as colloidal stabilizers, emulsifiers, and flow controllers, CMC has served well as part of regulation system for various industries, such as detergent, food, pharmacy, oil-drilling, and paper industries (Su *et al.* 2010). Moreover, fibrous CMC with special fibre-like morphology has attracted much attention in the field of lamellar soluble functional products, *e.g.*, “water-soluble paper”. Until now it has been possible to be used as electrical components, as well as the manufacture of degradable film in agriculture and packaging bags, by adjusting its solubility (Polcin and Eliasova 1975; Mizuai *et al*. 1989). Many products are soluble and, hence, the general water adsorption mechanism of fibrous CMC-based materials has to be addressed.

The quality of the soluble functional products is highly dependent upon the physical properties and solubility of the fibrous CMC network itself. The solubility of the fibrous CMC network depends on the structural and chemical properties, including the morphological characteristics (fiber length, width, crystallinity, *etc.*) and functional groups present on the CMC skeleton (Scallan 1983; Hubbe and Rojas 2008). As one of the most important functional groups on the CMC skeleton, the carboxymethyl group, which is characterized by a degree of the substitution (DS), plays an important role in many CMC properties (water-uptake behavior, solubility, *etc.*) (Derecskei and Derecskei-Kovacs 2006). The carboxymethyl groups can dissociate and contribute to the CMC solubility when CMC is suspended in water (Mondal *et al.* 2015). An effective process for increasing the dissociation activity of carboxymethyl groups in fibrous CMC networks was reported by Peng *et al.* (2012) and involved controlling the acid-base conversion efficiency of the fibrous CMC under certain conditions (75 °C, 4 to 6 s, *etc.*). In contrast, it has been shown that the dissociation activity of carboxymethyl groups is inhibited by aqueous solutions that contain high valence cationic electrolytes (Ca^{2+}, Fe^{3+}, *etc.*) (Zhang and Xu 2002), which limit the solubility of fibrous CMC networks by forming insoluble salts.

When the chain-structured CMC molecule dissolves into water, it separates into two parts, which are sodium cations and polymer anions. These ions in a solution can interact with each other by electrostatic forces. These interactions promote the solubility of CMC (Kumsah *et al.* 1976). Meanwhile, the functional groups (carboxymethyl and hydroxyl groups) also exhibit a polar attraction with water molecules, which contributes to the separation of CMC chains by creating considerable electrostatic interaction forces (weak hydrogen bonding) (Yang and Zhu 2007). However, the possible correlation with the dissolving process of fibrous CMC networks has been neglected by many researchers. This kind of polar attraction contributes in a major way to the swelling of fibrous CMC (Torgnysdotter and Wågberg 2003), which is responsible for the deformation of fibrous CMC (Yan and Li 2013). An increased deformation of fibrous CMC improves the dissolving efficiency of the fibrous CMC network (or paper sheets). Furthermore, the effect of the water sorption behavior needs to be thoroughly explored for a better understanding of the deconstruction and dissolving process of fibrous CMC networks in water.

In this study, the deconstruction mechanism of a fibrous CMC network (or handsheets) was examined through the sorption dynamic kinetics, including the pseudo-first-order and pseudo-second-order models. Also, the swelling ability and dynamic angle of the fibrous CMC network were measured. The main objectives of this study were to assess the action of water penetration and adsorption into the CMC network, better understand the deconstruction mechanism, and further optimize the properties of fibrous CMC-based products. Additionally, the dissolving process of the fibrous CMC network was full-scale recorded with the focused beam reflectance measurement (FBRM) process to evaluate the dissolving efficiency and obtain and analyze the key period that contributes to the dissolving process of a CMC network.

EXPERIMENTAL

Materials

Three fibrous CMC samples with different DS values were obtained from several cellulose factories in Shandong Province, China and were used without any further treatment. The characteristics of the samples are given in Table 1. Chemicals of analytical grade, including H_2SO_4, $NaHCO_3$, ethanol, and HCl, were obtained from local chemical companies in Tianjin, China.

Table 1. Characteristics of the CMC used in this Study

Number	DS	DP	Crystallinity (%)	Purification (%)
1	0.50	372 ± 1	25.6 ± 0.2	95
2	0.68	412 ± 3	24.1 ± 0.1	95
3	0.80	325 ± 1	23.5 ± 0.1	95

Note: DP – the Degree of Polymerization of CMC.

Swelling Ability Analysis

The swelling ratio was measured using the method described by Cuissinat and Navard (2006). Briefly, the CMC fibers were dispersed and adhered onto slides with glue. Then, an appropriate amount of deionized water was sucked from one end of the slide glass by a pipette and the water flowed to the other end under capillary force. The swelling and dissolution processes of the fibers were observed with a Nikon camera (DS-Fi2, Tokyo, Japan). The swelling ratio (γ) was calculated with Eq. 1,

$$\gamma = \frac{D_{swelling}}{D_{dried}} \tag{1}$$

where D_{dried} is the diameter of the dry fibers (cm) and D_{swelling} is the diameter of the fibers after swelling and moisturizing (cm).

Handsheet Preparation and Testing Methods

The handsheets were made without alkalization, in accordance with Zhang and Xu (2002). Figure 1 shows that acidification (25% H_2SO_4, 35 °C, and 2 h) was required to transition the solution to an insoluble fibrous HCMC state because of the solubility of the fibrous CMC. Then, the fibrous HCMC was used to prepare the handsheet samples, which were referred to as HCMC handsheets. In this study, the data for the deconstruction process was obtained at 25 °C using an Automatic COBB Tester (ACT 2500, FIBRO SYSTEM AB, Stockholm, Sweden), which is a real-time online monitoring program that documented the whole water adsorption process.

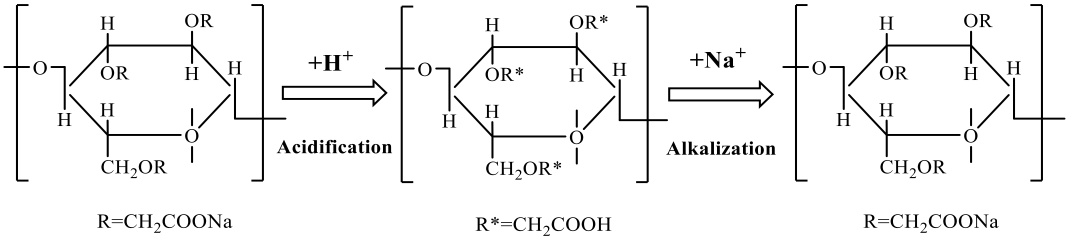

Fig. 1. Chemical structure transformation of the HCMC and Na-CMC

After the HCMC handsheets were formed, they were alkalized (10% $NaHCO_3$) by a spraying method to obtain the resulting samples (water-soluble handsheets). Then, the dynamic angle of the resulting handsheets was determined with a dynamic contact angle tester (PGX, FIBRO SYSTEM AB, Stockholm, Sweden) at 25 °C.

Dissolving Process Analysis

The handsheets (network of fibrous CMC with a DS of 0.80) were cut into square chips (30 mm × 30 mm). After being wetted in water for 2 s, the square chips were re-dried with a Freezing Dryer (FD-A-50, Boyikang, Beijing, China) at -50 °C under vacuum conditions for 4 h. The functional groups were characterized (dried and re-dried fibrous CMC networks) with attenuated total reflection Fourier transform infrared (ATR-FTIR) spectroscopy at 25 °C and a 65% relative humidity. The measurement was performed with a ceramic infrared light source at a scanning speed of 2 mm/s. For each sample, the range was 4000 cm^{-1} to 3000 cm^{-1}, with a resolution of 1 cm^{-1}. Then, each of the square pieces were placed in a beaker filled with 100 mL of deionized water under certain conditions (25 °C and 400 rpm), and the dissolving process was monitored with FBRM (PI-9.5-14/206 Probe, Mettler Toledo, Zurich, Sweden).

RESULTS AND DISCUSSION

Deconstruction Mechanism of the Fibrous CMC Network

Kinetic analysis

The fibrous CMC network (or handsheet), when used as a fiber matrix, is formed by hydrogen bonds and the combination of fibrous CMC windings, which contain a large number of polar groups (carboxyl group and hydroxyl group). Thus, there is a great potential for water penetration and adsorption into fibrous CMC networks.

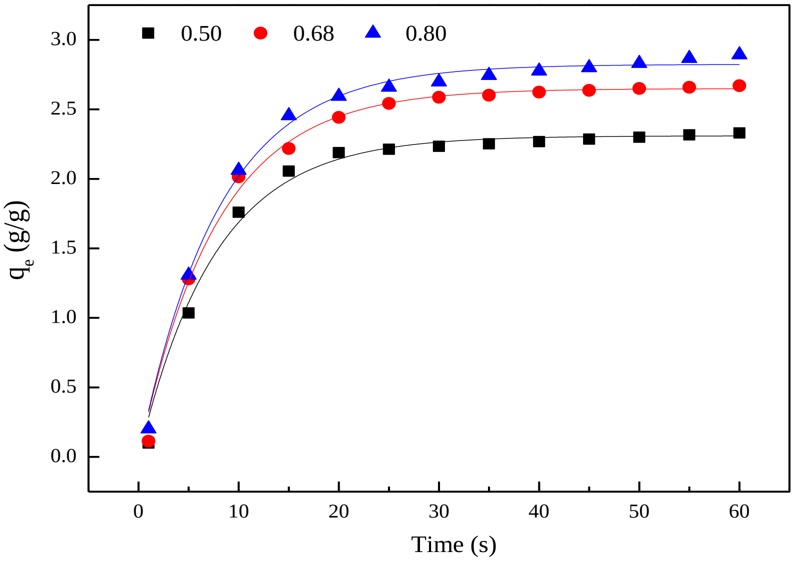

Fig. 2. Water adsorption into the CMC handsheet as a function of time

To investigate the deconstruction process of the fibrous CMC network, popular kinetic models, including pseudo-first-order and pseudo-second-order models, were utilized (Azizian 2004). Figure 2 shows the adsorption of water into the fibrous CMC network as a function of time. The pseudo-first-order kinetic model was used to model the sorption process and is shown below as Eq. 2,

$$\log(q_e - q_t) = \log(q_e) - \frac{k_1}{2.303}t \tag{2}$$

where q_e (mg/g) and q_t (mg/g) are the amounts of water absorbed into the fibrous CMC network at equilibrium and reaction time t (s), respectively, and k_1 (g/g·s) is the sorption rate constant of the pseudo-first-order kinetic model.

Table 2 shows that the coefficient of determination (R^2) for the pseudo-first-order kinetic model was high (almost 1), which indicated that the water sorption process onto the fibrous CMC network (or deconstruction of the fibrous CMC network) was well fitted by the pseudo-first-order kinetic model.

Table 2. Parameters of the Pseudo-first-order and Pseudo-second-order Models for Water Sorption onto the Fibrous CMC Network

Parameter	Pseudo-first-order Kinetics			Pseudo-second-order Kinetics		
DS	q_e (g/g)	k_1 (g/s)	R^2	q_e (g/g)	k_2 (g/s)	R^2
0.50	2.31	0.131	0.988	3.44	0.137	0.853
0.68	2.65	0.129	0.991	3.76	0.0147	0.849
0.80	2.82	0.126	0.994	4.04	0.0138	0.842

A pseudo-second-order kinetic model was also used to express the water sorption process (or deconstruction of the fibrous CMC network). The pseudo-second-order water sorption kinetic model is shown below as Eq. 3,

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e}t \tag{3}$$

where k_2 (g/s) is the rate constant of the pseudo-second-order kinetic model.

The R^2 for the pseudo-second-order kinetic model was relatively low compared with that of the pseudo-first-order model, which suggested that the pseudo-second-order kinetic model does not fit the process of water sorption into the fibrous CMC network as well. The pseudo-first-order model was able to explain the water sorption mechanism of the fibrous CMC network better, which indicated that the deconstruction of the fibrous CMC network was controlled only by physical adsorption (Fatehi *et al.* 2013a,b).

Effect of different DS values on the contact angle of the CMC-based handsheets

The contact angle was measured to assess the surface wettability of the fibrous CMC network (Fig. 3). The main factors affecting the contact angle were the structure and chemical composition or functional groups of the network (Šikalo *et al.* 2005). Under identical experimental conditions, the CMCs with DS values of 0.80, 0.68, and 0.50 had similar initial contact angles, which were 65.4°, 65.2°, and 65.0°, respectively. This meant that the structure of the network may have had a small impact on the contact angle. Thus, the main factor affecting the wettability or dynamic contact angle was the chemical composition or functional groups.

In this study, a droplet could penetrate the inner space of the fibrous CMC network completely within 2 s because of the strong mutual polar attraction between the water molecules and polar groups (-OH and -COOH) on the CMC molecular skeleton. Herein, the changing rate of the dynamic contact angle was linearly fitted within 0.6 s, which showed that the changing rate (|*K*|) of the dynamic contact angle increased from 2.86° to 4.66° when the DS of the fibrous CMCs increased from 0.50 to 0.80, which is shown in Fig. 3. These results revealed that the electrostatic capacity was strengthened when more carboxyl groups were substituted on the fibrous CMC molecular chains.

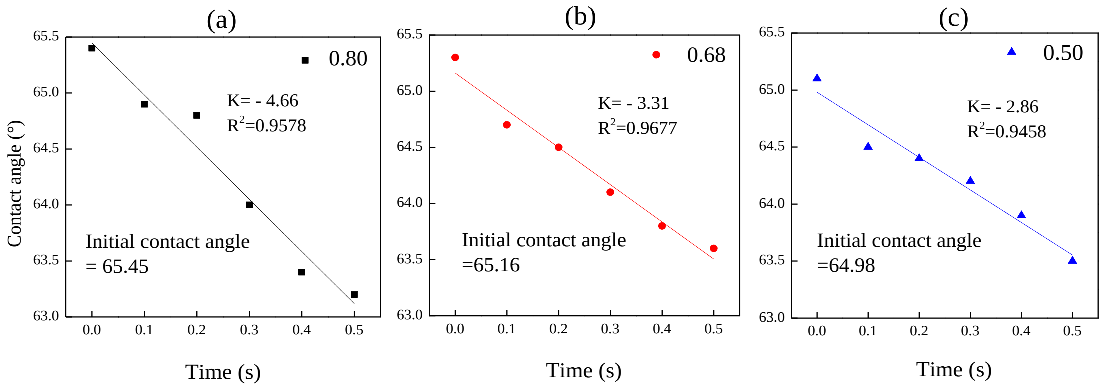

Fig. 3. Changing rate of the dynamic contact angle for DS values of: (a) 0.80, (b) 0.68, and (c) 0.50

Dissolving Process of the Fibrous CMC Network

The dissolving process of the fibrous CMC network determines the properties of the final products, which is strongly related to the deconstruction mechanism or water adsorption mechanism into the network. Thus, the swelling ability of the fibrous CMC was evaluated by the swelling ratio. Also, the dissolving process of the fibrous CMC network was investigated based on the physical adsorption mechanism discussed above.

Fibrous CMC swelling ability

The swelling ratio ($D_{\text{swelling}}/D_{\text{dried}}$), which is an estimation of water molecules penetrating the cellulose amorphous region, was used to characterize the swelling ability of the fibrous CMC (Cuissinat and Navard 2006). The $D_{\text{swelling}}/D_{\text{dried}}$ values are given in Table 3. The fiber swelling performance was affected by the bulk dissociated groups in the fibers (Zhao *et al.* 2016). Because of the polar attraction of water molecules and functional groups contained on the CMC molecular skeleton, the water molecules entered the amorphous region of the fibrous CMC chains, which resulted in fibrous CMC deformation and an increase in the distance between the fibrous CMC chains.

Table 3. Swelling Ratios of the CMC Fibers

Number	DS	$D_{\text{swelling}}/D_{\text{dried}}$
1	0.50	2.07 ± 0.02
2	0.68	2.43 ± 0.03
3	0.80	2.87± 0.05

Furthermore, the carboxyl groups present on the CMC skeleton promoted the swelling ability because they have a higher polarity than hydroxyl groups. This is because of the osmotic pressure caused by the counterions in water (Ueno *et al.* 2007). The CMC swelling ratio increased from 2.07 to 2.87 with an increase in the DS from 0.50 to 0.80, which suggested that the swelling ability of the CMC was enhanced with an increase in the DS. Considering the promoting relationship between the swelling capacity and solubility of the CMC, an increase in the DS had a positive effect on the CMC solubility.

Dissolving process of the fibrous CMC network

The swelling of the fibrous CMC led to the final deconstruction and dissolving of the fibrous CMC network. The FBRM process, which is an efficient tool mainly related to the characterization and monitoring of crystals, oil-water emulsions, and cell and bacterial systems, was used to monitor the dissolving process of the alkalized fibrous CMC network under certain conditions (25 °C and 400 rpm) (Heath *et al.* 2002).

Figure 4 shows the variation in the fines (< 10 µm) that existed in the solution as a function of time. This reflected the dissolving process of the CMC network, which involved a deconstruction period (t_0 to t_1), deconstruction and partial dissolution period (t_1 to t_2), and partial dissolution period (after t_2).

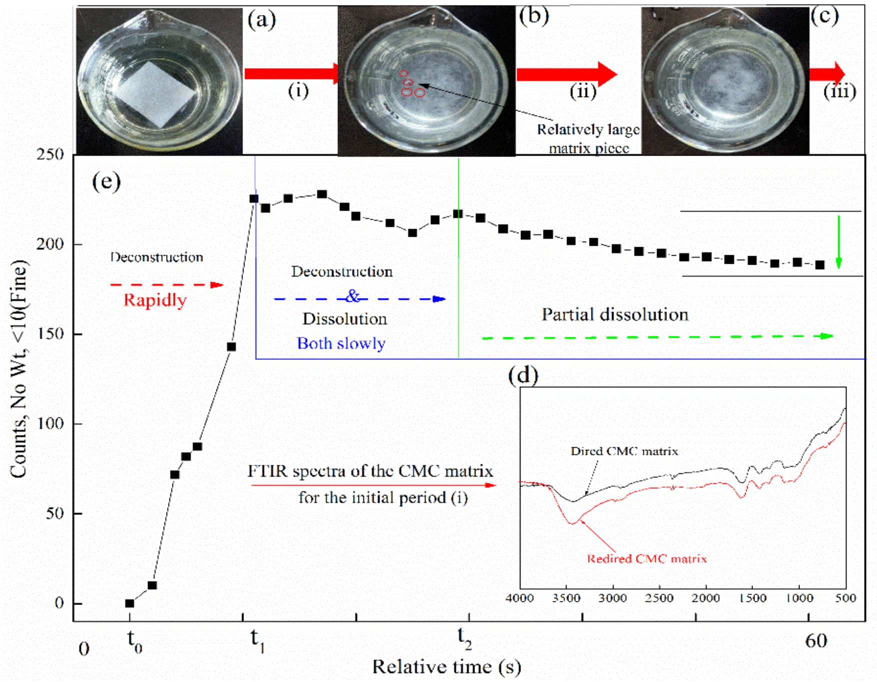

Fig. 4. Model of the fibrous CMC network dissolving process according to the FBRM process (25 °C and 400 rpm) (a) the state of the fibrous CMC network at the beginning of deconstruction period; (b) the state of the fibrous CMC network after the deconstruction period; (c) the state of the fibrous CMC at the end of the dissolving process; (d) the FTIR analysis for the dried and redried CMC matrix; (e) the whole dissolving process monitored by FBRM

In the deconstruction period, because of the physical interaction between the water molecules and functional groups (hydroxyl and carboxyl groups) in the fibrous CMC network, the fibrous CMC absorbed water rapidly to form a "water-bridge" state between the fibrous CMCs. This meant that contact points between the fibers were diminished. The fibrous CMC network was deconstructed because of a weakening and reduction of the hydrogen bonds. According to the FTIR spectra of the fibrous CMC network (Fig. 4d), the vibration peak (3420 cm^{-1}) of the hydroxyl group was enhanced after wetting, which indicated that hydrogen bonds were substantially eliminated by water molecules. Then, the main structure of the CMC network (Fig. 4a) was deconstructed from the outside in.

The second period incorporated the phenomena of deconstruction and partial dissolution. With the further deconstruction of the main structure, many CMC network pieces with a comparatively large size appeared quickly and the amount of fibrous CMC increased, which is shown in Fig. 4b. The CMC network pieces were not broken down completely. At the same time, more fibrous CMC and fines were dispersed in the water. Further, some fibrous CMC and fines were swollen enough to dissolve into the water. Therefore, both phenomena (deconstruction and dissolution) occurred in this period (t_1 to t_2), which caused a fluctuation in the total measured fines content.

The third period consisted only of partial dissolution. The large-sized network pieces were deconstructed completely and suspended as single CMC fibers in the water. Simultaneously, the total amount of fines was reduced, which caused continuous swelling and partial dissolution of the fibrous CMC chains in the water, which is shown in Fig. 4c.

For the whole dissolving process of the fibrous CMC network, the deconstruction period was the key period that controlled the dissolving efficiency. This mode was applied to the three samples, which is shown in Fig. 5, and the curves fit well to this dissolving mode.

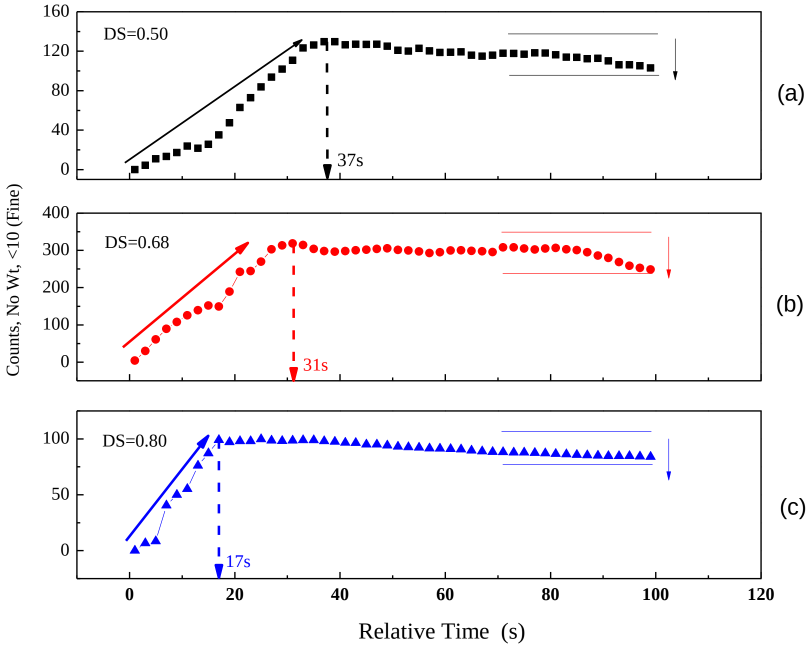

Fig. 5. Dissolving process of the different CMC materials (25 °C and 400 rpm) with DS values of: (a) 0.50, (b) 0.68, and (c) 0.80

For the network formed by the fibrous CMC with a DS of 0.80, the time required for the fines to reach a maximum value in the solution was 17 s in the deconstruction period (t_0 to t_1). The fibrous CMC with a higher DS showed much stronger swelling and wetting abilities, as was discussed previously. The mutual attraction between the water molecules and functional groups was weakened because of the decrease in the amount of carboxyl groups on the fibrous CMC skeleton. Therefore, the speed of water molecules penetrating the fibrous CMC network slowed, which had a negative effect on the deconstruction and solubility of the CMC network. The deconstruction time (time for the fines in the solution to reach a maximum value) was 31 s for the CMC with a DS of 0.68 and 37 s for the CMC with a DS of 0.50. Obviously, when the DS value increased from 0.50 to 0.68, the dissolving efficiency increased by 16.2%; moreover, the dissolving efficiency increased 45.2% as the DS increased from 0.68 to 0.80, which supports the findings just discussed.

CONCLUSIONS

1. The deconstruction mechanism (or water adsorption mechanism) of the fibrous CMC network was fitted well by the pseudo-first-order kinetic model ($R^2 > 0.95$), indicating that the overall rate was mainly controlled by physical adsorption. The $D_{\text{swelling}}/D_{\text{dried}}$ and dynamic contact angle spreading rate confirmed that the water sorption and swelling ability were enhanced when more carboxymethyl groups were substituted on the CMC molecular chain.
2. The dissolving behavior of the fibrous CMC network, which was monitored by FBRM, consisted of three periods: a deconstruction period (t_0 to t_1), deconstruction and partial dissolution period (t_1 - t_2), and partial dissolution period (after t_2). The deconstruction period was the key period that controlled the dissolving efficiency of the fibrous CMC network. The potential application of this mechanism assisted by FBRM is to accurately judge the dissolving capability of fibrous CMC-based products, as well as to optimize the performance of fibrous CMC-based materials.

ACKNOWLEDGMENTS

The authors would like to acknowledge the financial support from the Key Projects of Natural Science Foundation of Tianjin (Grant No. 16JCZDJC37700).

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Article submitted: March 30, 2018; Peer review completed: July 7, 2018; Revised version received and accepted: September 3, 2018; Published: September 10, 2018.
DOI: 10.15376/biores.13.4.8132-8142