# PREPARATION OF CATIONIC SOFTWOOD KRAFT PULP FIBRES AS RETENTION ADDITIVE TO PRODUCE RECONSTITUTED TOBACCO SHEET *VIA* PAPER-MAKING

# CHAOWEI WU,<sup>\*</sup> JIN LI,<sup>\*</sup> LITAO ZHANG,<sup>\*</sup> WENLING WANG,<sup>\*</sup> CHONG LUO,<sup>\*</sup> XIAOHUI TIAN,<sup>\*</sup> YANGYUAN TIAN,<sup>\*</sup> XIAOLONG ZHANG,<sup>\*</sup> CHEN WANG,<sup>\*</sup> RUNAN WANG,<sup>\*</sup> JIANKAI LI,<sup>\*</sup> XIAODONG YANG<sup>\*</sup> and YANGBING WEN<sup>\*\*</sup>

\*Henan Cigarette Industry Tobacco Sheet Ltd., Xuchang, Henan, 461100, China
\*\*Tianjin Key Laboratory of Pulp and Paper, Tianjin University of Science and Technology,
29, 13<sup>th</sup> Avenue, Tianjin Economic and Technological Development Area, Tianjin 300457, China
© Corresponding author: C. Luo, lc19871122@126.com,
Y. Wen, yangbingwen@tust.edu.cn

Received March 18, 2020

In this study, cationic modification of northern bleached softwood kraft (NBSK) pulp fibres was conducted through esterification with N-(2-3-epoxypropyl) trimethylammonium chloride (EPTMAC) for the production of cationic fibres that can be utilized as retention aid to decrease the content of harmful components in reconstituted tobacco sheets (RTS). For the modification process, EPTMAC loadings of 0.10 to 1.0 g/g fibre were investigated to produce pulp fibres with different contents of cationic groups. The obtained cationic NBSK pulp fibres were characterized by Fourier transform infrared spectroscopy (FT-IR), as well as in terms of their degree of polymerization (DP) and fibre diameter. The properties of the produced cationic NBSK pulp fibres were also investigated during the production of RTS. The results demonstrated that, compared to the control (containing original NBSK pulp fibres and commercial cationic guar gum retention aid), the addition of cationic NBSK pulp fibres substantially increased the retention of fillers and drainage property during the RTS production process. Moreover, the increase of cation group content increased the retention value of precipitated calcium carbonate (PCC) particles and tobacco fines. The application of cationic NBSK pulp fibres could also enhance the bulk of RTS, while maintaining the tensile strength.

Keywords: cationization, NBSK pulp fibres, reconstituted tobacco sheet, retention, tensile strength

### **INTRODUCTION**

Reconstituted tobacco sheet (RTS), produced from broken tobacco leaf, leaf stalk, fillers, and softwood or hardwood pulps, is widely recognized as an additive or tobacco alternative for the production of cigarettes, to reduce harmful components and process costs in cigarette manufacturing.<sup>1-5</sup> However, the high amount of fines/short fibres in the RTS pulp suspension normally results in a high level of dissolved polymers and colloidal substances (DCS) in the paper-making system and low strength of the sheet; thereby decreasing the efficiency.<sup>6-8</sup> Therefore, retention reagents that could improve

the retention of fillers and fines, while increasing the sheet strength are generally required during the production of RTS. The retention aids that are commonly utilized in producing RTS are cationically modified guar gum and chitosan, due to their characteristics of food safety and high efficiency.<sup>9-12</sup> The associated challenge of the two common retention aids used in the production of RTS is the reduction of the introduction of exogenous substances during cigarette combustion.<sup>13,14</sup> Thus, the elimination of polymer retention aids during the production of RTS is of great importance.

To achieve the goal of eliminating the utilization of retention aids, the cationic modification of wood fibres added in the RTS pulp suspension is considered to be a potential strategy.<sup>6</sup> The cationic charges on the surface of wood fibres could create an electric attraction force with ionic charged fillers and fines/short fibres of the tobacco waste, thus improving the filler retention value and the paper-making process.<sup>15,16</sup> Moreover, cationized cellulose fibres or cellulose derivatives have also been investigated to partially or completely replace the polyelectrolytes in the pulp and paper industry to reduce their environmental impact.<sup>17</sup> For example, Aguado et al. reported the cationic modification hardwood fibres of with (3-chloro-2-hydroxypropyl)trimethylammonium chloride to increase the retention of fillers and fines in the paper and board manufacturing industry.<sup>16</sup> In addition, Liu et al. investigated the addition of cationically modified cellulose nanofibres in the process of producing RTS and achieved a high filler (precipitated calcium carbonate) retention.<sup>18,19</sup> However, because of their high processing costs and gel-like characteristics, the large-scale application of cellulose nanofibres in the RTS production process is limited. Thus, to address these concerns, we hypothesize that cationically modified wood fibres could have a promising potential to serve as a retention aid, which can improve the paper-making process during the production of RTS.

In the work described in this paper, northern bleached softwood kraft (NBSK) pulp fibres were subjected to cationic modification with N-(2-3-epoxypropyl) trimethylammonium chloride (EPTMAC) to introduce a positive charge into the negatively charged fibres. Subsequently, the modified NBSK pulp was added into the tobacco pulp slurry for the preparation of RTS. The effect of cationic modification on the properties of the obtained NBSK pulp fibres was investigated in terms of degree polymerization, fiber morphology and cation group content. Moreover, the modified NBSK pulp fibres were also utilized in the preparation of RTS. The filler retention rate, pulp drainage property and physical properties of the produced RTS were also investigated. Our results have demonstrated that the cationically modified NBSK pulp fibres have potential to be used as an

alternative retention aid for the production of RTS through the paper-making process.

# EXPERIMENTAL

## Materials

NBSK pulp was obtained from a local paper mill (Shandong, China). The obtained pulp sheets were torn into pieces and soaked in deionized water for 72 h. Then, the soaked pulp was subjected to mechanical treatment in a PFI refiner, according to TAPPI standard T248 sp-00. A 24 g (equivalent to oven dried) pulp sample was refined at 10% (w/v) pulp consistency to a Canadian Standard freeness of 525 mL. The refined pulp was stored at 4 °C for further N-(2-3-epoxypropyl) experimentation. trimethylammonium chloride (EPTMAC), isopropanol and ethanol were purchased from Tianjin Chemical Reagent Co., Ltd., China. Cationic guar gum (CGG) and tobacco waste materials were provided by Henan Cigarette Industry Tobacco Sheet Ltd. (Xuchang, China).

## Cationic modification of cellulose fibres

The cationic modification of the refined pulp fibres with EPTMAC was performed in a 5000 mL beaker. A series of EPTMAC loadings of 0-1.0 g/g fibre was explored. For each modification experiment, 100 g (equivalent to oven dried) of the NBSK pulp fibres, 15 g NaOH, and 2 Lisopropanol (70% w/w) were mixed in the beaker and placed into the water bath (Ningbo Xinzhi Electrical Technology Co., Ltd, China) at 25 °C and 300 rpm for 2 h. Afterwards, the temperature of the reaction was increased to 60 °C. Then, EPTMAC was slowly added (10 mL/min) into the reaction system for the cationization. After mixing at 300 rpm for 4 h, the NBSK pulp fibres were thoroughly washed with deionized water to remove the unreacted chemicals and the cationically modified fibres were obtained and stored at 4 °C for further experimentation.

### Preparation of reconstituted tobacco handsheet

The preparation of the RTS was performed according to TAPPI standard T 205 sp-02. Briefly, 65 g of refined tobacco leaf and stem, 20 g of original or cationically modified NBSK pulp fibres, 15 g of precipitated calcium carbonate and 20 L of deionized water were mixed and disintegrated at a solid consistency of 0.5% (w/v) to prepare the pulp slurry. Then, the pulp slurry was applied to a rapid handsheet former (PTI, Vorchdorf, Austria) to prepare RTS with a basis weight of  $60 \pm 2$  g/m<sup>2</sup>. The wet sheet was air-dried in a constant temperature (23  $\pm$  1 °C) and humidity (50  $\pm$  2%) room for 24 h to get dry sheets for further experiments. For the RTS with the addition of original NBSK pulp, 0.15% cationic guar gum (based

on the total weight of pulp) was added into the suspension to improve the retention of fillers (precipitated calcium carbonate).

#### Fourier transform infrared (FT-IR) spectroscopy

FT-IR spectra of NBSK pulp fibres were collected using a PerkinElmer FT-IR spectrophotometer (Spotlight 400 FT-IR Imaging System, PerkinElmer Co., USA), in the frequency range of 4000-400 cm<sup>-1</sup>. Approximately 10% potassium bromide (based on the dry weight of fibres) was used to prepare a transparent pellet for FT-IR analysis.

#### Cation group content measurement

In the cationic modification, the amount of EPTMAC that reacted with the fibers was equal to the nitrogen content of cationized softwood kraft pulp fibres. The cation group content of the cationically modified cellulose fibres was determined using elemental analysis (EA, Vario EL cube, USA) to determine the N content in the cationically modified fibres. The content of cation groups was calculated following Equation 1:<sup>20</sup>

$$DS = \frac{N_{\%}}{14 - 151.6 \times N_{\%}^{9}} \times 162$$
 (1)

where N% denotes the percentage of dry elemental nitrogen, 14 is the atomic weight of nitrogen, and 151.6 is the molecular weight of the EPTMAC.

#### **Analytical methods**

The intrinsic viscosity ([ $\eta$ ]) of the original and oxidized NBSK fibres was determined by dissolution in cupriethylenediamine (CEM) at 25 °C, using an Ubbelohde viscometer, following TAPPI standard method T 230 om-08. The DP of cellulose was



#### Figure 1: Cationic modification of NBSK fibres with EPTMAC and adsorbed PCC particles

calculated based on the intrinsic viscosity value by Equation  $2^{:21}$ 

$$DP^{0.905} = 0.75[\eta] \tag{2}$$

Surface morphology of RTS was examined with scanning electron microscopy (SEM) (SU9000, Hitachi, Japan). Prior to subjecting the samples to SEM analysis, they were mounted on aluminum stubs and coated with a thin layer of gold. An accelerating voltage of 15 kV was used to scan the samples.

The modified fibres were soaked in deionized water for 24 h, and the fibre concentration was adjusted to 0.1% (w/v). The mean fibre length was measured by an L&W Fibre Tester (FT, code 912, Lorentzen & Wettre Co.).

The handsheets were tested for tensile strength and bulk, using TAPPI T 220 sp-01 and TAPPI T411 om-15, respectively.

## **RESULTS AND DISCUSSION** Cationic modification of NBSK pulp fibres

Figure 1 shows the reaction mechanism for the cationization of cellulose with EPTMAC. During the cationization process, the etherification reaction between the hydroxyl groups of cellulose and the epoxy groups of EPTMAC occurred, thereby introducing 0.0664 to 0.432 (DS) cation group into the cellulose structure. Moreover, alkali is generally required for the modification process, as it could not only activate the hydroxyl groups of cellulose, but also swell the cellulosic fibres for improving the penetration of EPTMAC into the fibre.<sup>15,16,22</sup>



Figure 2: Effect of EPTAC loading on the cationic group content of NBSK pulp fibres



Figure 3: FT-IR spectra of NBSK fibres in wavenumber ranges of (a) 4000-400 cm<sup>-1</sup>, (b) 2000-1000 cm<sup>-1</sup>

Following cationic modification, the presence of cation groups on the surface of cellulose is expected to improve the retention of the negatively charged fillers (PCC) *via* charge For the application of cationically modified

NBSK pulp fibres as a retention aid, the content of cation groups is considered to be an important parameter. Figure 2 shows the effect of EPTMAC loading on the cation group content of the NBSK pulp fibres.

As shown in Figure 2, with the increase of EPTMAC loading, the content of cation groups increased. For example, at an EPTMAC loading of 0.20 g/g fibre, the DS of cation group on the NBSK pulp fibres was 0.137, while increasing the EPTMAC loading to 1.0 g/g, the DS of cation group content of the pulp fibres rose to 0.432. These results indicate that the cationic modification process occurred efficiently. Figure 2 also shows the Zeta potential of the modified NBSK pulp fibres. following cationic modification with different EPTMAC loadings. Similar to the trend of the cation group content, the Zeta potential of the fibres also increased with increasing the EPTMAC loading, indicating the successful grafting of quaternary ammonium salt groups of EPTMAC into the cellulose structure. Due to the presence of positive charge, the modified pulp fibres could be utilized to improve the retention value of PCC during the production of RTS.

To further confirm the successful modification of cellulose, the FT-IR spectra of the NBSK pulp fibres were recorded (Fig. 3). As shown in Figure adsorption during the production of RTS, thus reducing or even eliminating the requirement of cationic polymer additives.

3, the absorption peaks at approximately 3400 cm<sup>-1</sup> and 2882 cm<sup>-1</sup> were assigned to the O-H stretching and symmetrical stretching of C-H in the glucose ring, respectively; these two absorption peaks were observed in all the fibre samples, indicating the cationic modification process maintained the basic structure of cellulose. Compared to the spectrum of the original NBSK pulp fibres, the cationically modified samples had two new peaks at 1428 and 1371 cm<sup>-1</sup>, which were attributed to the stretching of methyl groups in ammonium and the C-N stretch in EPTMAC structure, respectively.23,32 The results of FT-IR analysis further suggest that the EPTMAC was successfully grafted onto the cellulose structure.

# **Characterization of cationic NBSK fibres**

Up to this stage, the results indicated the successful modification of NBSK pulp fibres with EPTMAC. To evaluate the effect of cationization on the properties of cellulosic fibres, the DP and fibre diameter were determined (Fig. 4). As shown in Figure 4 (a), the increase in the cation group content of cellulose slightly decreased its DP following cationic modification. For example, when the DS of cation group was 0.432, the DP of cellulose was about 1017, which was slightly lower than that of the original pulp cellulose ( $\Box$ 1237). As the hydrolysis of cellulose

during cationic modification could be neglected, the reduction in the cellulose DP following modification might be attributed to the grafting of cation groups onto the cellulose structure, which slightly reduced the viscosity of the cellulose solution after dissolving in CED.<sup>15,16,23-25</sup>



Figure 4: Properties of cationically modified NBSK; (a) Degree of polymerization, (b) fiber mean length, and (c) fiber mean width

Figure 4 (b and c) shows the length and width of fibres following cationic modification. As shown in Figure 4 (b), the fibre length (2.04 mm) was essentially constant throughout the cationic process – this modification means the modification mainly occurred on the surface of fibre or microfiber. The preservation of fibre length is favorable for the production of RTS by increasing the physical strength of the paper. Different from fibre length, the fibre width increased from 27.5 µm to 40.1 µm by increasing the DS of cation group to 0.432 (Fig. 4 (c)). One likely explanation might be that the grafting of EPTMAC onto the cellulose structure increased the hydrophilicity of NBSK fibres, which facilitated the penetration of water into the fibre, thereby resulting in the swelling of the fibre cell wall and increasing of the fibre width.<sup>24,26</sup>

# Application of cationic NBSK pulp in RTS production

During the production of RTS *via* the paper-making process, retention additives and wood pulp fibres that could improve the retention of fillers and increase the physical strength of the RTS are normally required. In this study, instead of using conventional wood pulp fibres and CGG (a commercial polymer additive), cationically modified NBSK pulp fibres were utilized for the production of RTS *via* the paper-making process. We hypothesized that the cation groups in the cellulose structure can not only adsorb the PCC

particles (a filler used in RTS production), but also improve the strength properties of the produced RTS. Figure 5 shows the effect of the addition of cationically modified NBSK pulp fibres on the retention value (PCC retention value and single-pass retention value) and filtration volume of the pulp slurry for the production of RTS.

As shown in Figure 5 (a), with the increase in the cation group content, the retention value increased. For example, when increasing the DS from 0.137 to 0.432, the PCC retention value increased from 42.2% to 58.5%, and single-pass retention value increased from 74.0% to 88.3%. Moreover, compared to the commercial retention aid of CGG, the cationically modified NBSK pulp fibres achieved a much higher PCC retention value when the DS of cation group was higher than 0.25 (Fig. 5 (a)). For example, when adding 0.15% CGG and 25% original NBSK pulp fibres for the RTS production, the PCC retention value was 42.2% and single-pass retention value was 74.0%. In contrast, the addition of 25% cationically modified NBSK pulp fibres with a DS of cation group of 0.252, the PCC retention value was 45.8% and single-pass retention value was 78.6%. These results are in accordance with previous findings on the utilization of cationic cellulose derivatives to improve the retention value of fillers.<sup>16,17</sup> The increase of the retention of PCC could be due to the positive surface charge of modified NBSK pulp fibres, which promotes the adsorption of anionic tobacco fibres, fines and PCC particles through electrostatic interaction.<sup>28,29</sup> Figure 5 (b) shows that the drainage property of the RTS pulp slurry was similar to the trend of the PCC retention value (Fig. 5 (a)), following the addition of cationic NBSK pulp fibres or commercial retention additive. This is due to the fact that the positively charged NBSK pulp fibres improve the adsorption of PCC particles and tobacco waste fines onto the fibre surface, thereby resulting in PCC flocculation and an improvement in the drainage property of the RTS pulp slurry during the paper-making process. To provide further insights into the flocculation of the PPC, the fiber surface of the samples was investigated using SEM imaging (Fig. 6).

As shown in Figure 6, significant differences were observed on the fiber surface between the original and cationically modified NBSK pulp fibres. Compared to the original NBSK pulp fibres, with the addition of 0.15% CGG (Fig. 6 (a)), the surface of the cationic NBSK pulp fibres adsorbed more PCC particles (Fig. 6 (b-f)). Moreover, with increasing the cation group content, more PCC particles were absorbed onto the fibre surface. For example, when the DS of cation group was 0.432, the fibre surface was almost covered by PCC particles and fines in the RTS pulp slurry.



Figure 5: Effect of cationic NBSK pulp fibres on (a) PCC retention value and (b) drainage property of RTS pulp slurry



Figure 6: SEM images of NBSK pulp fibres after addition into RTS pulp slurry; (a) Original pulp fibre with addition of 0.15% CGG, (b) DS = 0.0664, (c) DS = 0.137, (d) DS = 0.252, (e) DS = 0.335, (f) DS = 0.432 (magnification  $\times 1500$ )



Figure 7: Effect of cationic group on tensile strength (a) and bulk (b) of RTS prepared by the paper-making process

This further confirmed the statement that cation groups on the cellulose structure could absorb the anionic PCC particles *via* electrostatic interaction.

For the production of RTS through the paper-making process, the strength of the RTS is also a critical parameter. Figure 7 shows the effect of the addition of cationic NBSK pulp fibres on the tensile strength and bulk of the obtained RTS. As shown in Figure 7 (a), when increasing the DS of cation group from 0.0664 to 0.432, the tensile index of the RTS decreased from 13.4 to 10.5 N·m/g. This might be explained by the fact that the adsorption of a high amount of PCC particles and fines blocks or weakens the hydrogen bonding between tobacco fibres and wood fibres,

resulting in the reduction of RTS strength.<sup>28-30</sup> Moreover, when the DS of the cationic NBSK fibre was 0.432, the tensile index of the RTS was 10.5 N·m/g, it was only about 6% lower than that of the control RTS with the addition of original NBSK pulp fibres and commercial CGG retention additive (11.3  $N \cdot m/g$ ). This indicates that cationically modified NBSK pulp fibres could be a promising alternative to improve filler and fine retention, while maintaining the tensile strength of the RTS. Figure 7 (b) shows that the addition of cationic NBSK pulp fibres, instead of original NBSK pulp fibres and CGG, could improve the bulk of the RTS. As shown, the bulk of RTS increased from 2.08 cm<sup>3</sup>/g (with original NBSK pulp fibres and 0.15% CGG) to 2.34 cm<sup>3</sup>/g (the DS of cation group on the cationic NBSK fibre was 0.432); this is due to the retention of a higher amount of PCC particles when using cationic NBSK pulp fibres. The increase of bulk is favorable for the production of RTS because it could promote the cigarette burning rate and decrease CO release when smoking.<sup>31</sup> Taken together, the results of this study indicate that the introduction of cation groups on the NBSK pulp fibres could improve PCC particles retention and eliminate the utilization of CGG polymer retention additive during the production of RTS. Moreover, the addition of cationic NBSK pulp fibres can also maintain the tensile strength, while increasing the bulk of the produced RTS.

## CONCLUSION

In this study, cationic NBSK pulp fibres were successfully produced by cationization with EPTMAC. During the cationization process, increasing the EPTMAC loading increased the content of cation group of cellulose, while only slightly decreased the DP of cellulose. Moreover, the cationic modification process had little impact on the fibre diameter. When using the cationic NBSK pulp fibres in the production of RTS, the retention value of PCC particles and tobacco fines was significantly increased. Moreover, when comparing with the RTS prepared by the conventional process (adding original NBSK pulp fibres and commercial CGG retention aid), the addition of cationic NBSK pulp fibres also maintained the tensile index and improved the bulk of the RTS. The results demonstrated that the cationic fibres could be an alternative as a retention aid for the production of RTS through the paper-making process.

**ACKNOWLEDGMENTS**: The authors wish to acknowledge the financial support from the Natural Science Foundation of Tianjin, China (Grant No. 18JCYBJC86500). The valuable comments made by the anonymous reviewers are also sincerely appreciated.

## REFERENCES

<sup>1</sup> C. Wu, C. Luo, X. Fan, X. Wang and Y. Wen, *Cellulose Chem. Technol.*, **53**, 1009 (2019), https://doi.org/10.35812/CelluloseChemTechnol.2019. 53.99

<sup>2</sup> W. Gao, K. Chen, R. Yang, J. Li and F. Yang, *Cellulose Chem. Technol.*, **46**, 277 (2012), http://www.cellulosechemtechnol.ro/pdf/CCT3-4(2012) )/p.277-282.pdf

<sup>3</sup> L. Wang, Y. Wen, D. Sun, Y. Mao and Y. Yao, *Adv. Mater. Res.*, **314**, 2338 (2011), https://www.scientific.net/amr.314-316.2338

<sup>4</sup> A. Kumar and J. Gomes, U.S. Patent 6216706 (2001)

<sup>5</sup> M. Chen, Z. Xu, G. Chen, H. Wang, C. Yin *et al.*, *J. Anal. Appl. Pyrol.*, **105**, 227 (2014), https://doi.org/10.1016/j.jaap.2013.11.008

<sup>6</sup> W. Gao and K. Chen, *Cellulose*, **24**, 2581 (2017), https://doi.org/10.1007/s10570-017-1270-5

<sup>7</sup> H. Liu, H. He, C. Cheng, J. Liu, M. Shu *et al.*, *Appl. Microbiol. Biotechnol.*, **99**, 469 (2015), https://doi.org/10.1007/s00253-014-5960-8

<sup>8</sup> J. Li, J. Hu, S. Li, J. Li and J. Liu, *Carbohyd. Polym.*, **196**, 102 (2018), https://doi.org/10.1016/j.carbpol.2018.04.125

<sup>9</sup> M. Abdallah, *Portugaliae Electrochim. Acta*, 22, 161 (2004),

http://www.scielo.mec.pt/pdf/pea/v22n2/22n2a07.pdf

<sup>10</sup> N. Thombare, U. Jha, S. Mishra and M. Z. Siddiqu,
 *Int. Biol. Macromol.*, **88**, 361 (2016),
 https://doi.org/10.1016/j.ijbiomac.2016.04.001

<sup>11</sup> H. Chi, H. Li, W. Liu and H. Zhan, *Colloid. Surface A: Physicochem. Eng.*, **297**, 147 (2007), https://doi.org/10.1016/j.colsurfa.2006.10.039

 <sup>12</sup> F. Renault, B. Sancey, P. M. Badot and G. Crini, *Eur. Polym. J.*, **45**, 1337 (2009), https://doi.org/10.1016/j.eurpolymj.2008.12.027

<sup>13</sup> E. F. Litzinger, B. B. Chakraborty and W. R. Conway, U.S. Patent 5377698 (1995)

<sup>14</sup> E. F. Litzinger, B. B. Chakraborty and W. R. Conway, U.S. Patent 5765570 (1998)

<sup>15</sup> M. S. Atiq, A. Rehman, K. Iqbal, F. Safdar, A. Basit *et al.*, *Cellulose Chem. Technol.*, **53**, 155 (2019), http://doi.org/10.35812/cellulosechemtechnol.2019.53. 17

 <sup>16</sup> R. Aguado, A. Moral and A. Tijero, *J. Clean. Prod.*, 174, 1503 (2018),

https://doi.org/10.1016/j.jclepro.2017.11.053

<sup>17</sup> Z. Lu, X. An, H. Zhang, L. Liu, H. Dai *et al.*,
 *Carbohyd. Polym.*, **229**, 115434 (2020),
 https://doi.org/10.1016/j.carbpol.2019.115434

<sup>18</sup> H. Liu, Z. Liu, H. Liu, L. Hui, F. Zhang *et al.*, *Ind. Crop. Prod.*, **130**, 592 (2019),
 https://doi.org/10.1016/j.indcrop.2019.01.021

<sup>19</sup> H. Liu, Z. Liu, H. Liu, L. Hui, F. Zhang *et al.*, *Carbohyd. Polym.*, **210**, 372 (2019), https://doi.org/10.1016/j.carbpol.2019.01.065

<sup>20</sup> I. R. Jouybari, M. Yoosefi and M. Azadfallah, *BioResources*, **12**, 3890 (2017), https://ojs.cnr.ncsu.edu/index.php/BioRes/article/view/ BioRes\_12\_2\_3890\_Rashidi\_Jouybari\_Preparation\_C ationic\_CMP\_Softwood

<sup>21</sup> C. Wang, Z. Yuan, A. Wang, J. Qu, Z. Fang *et al.*, *Cellulose*, **27**, 2041 (2020),

https://doi.org/10.1007/s10570-019-02941-9

<sup>22</sup> M. Hasani, E. D. Cranston, G. Westman and D. G. Gray, *Soft Mat.*, 4, 2238 (2008), https://doi.org/10.1039/B806789A

<sup>23</sup> Y. Song, Y. Sun, X. Zhang, J. Zhou and L. Zhang, *Biomacromolecules*, 9, 2259 (2008), https://doi.org/10.1021/bm800429a

<sup>24</sup> T. Ho, T. Zimmermann, R. Hauert and W. Caseri, *Cellulose*, 18, 1391 (2011), https://doi.org/10.1007/s10570-011-9591-2

<sup>25</sup> F. Zhang, Z. Pang, C. Dong and Z. Liu, *Carbohyd. Polym.*, **132**, 214 (2015), https://doi.org/10.1016/j.carbpol.2015.06.055

<sup>26</sup> G. Sim, M. N. Ala and L. Godbout, *Cellulose*, **21**, 4595 (2014),

https://doi.org/10.1007/s10570-014-0425-x

<sup>27</sup> Y. Sang, M. McQuaid and P. Englezos, *BioResources*, **7**, 0354 (2012), <u>https://ojs.cnr.ncsu.edu/index.php/BioRes/article/view/</u> <u>BioRes 07 1 0354 Sang ME Prefloc PCC High F</u> iller /1311 <sup>28</sup> P. R. Charani and H. M. Mohammad, *Cellulose Chem. Technol.*, **53**, 767 (2019), https://doi.org/10.35812/cellulosechemtechnol.2019.5 3.75

3.75 <sup>29</sup> W. Xie, Z. Song, Z. Liu and X. Qian, *Carbohyd. Polym.*, **150**, 114 (2016), https://doi.org/10.1016/j.carbpol.2016.05.010

<sup>30</sup> W. K Mosse, D. V Boger, G. P Simon and G. Garnier, *Langmuir*, 28, 3641 (2012), https://doi.org/10.1021/la2049579

<sup>31</sup> S. Zhou, M. Ning, Y. Xu, Y. Hu, J. Shu *et al.*, *J. Anal. Appl. Pyrol.*, **100**, 223 (2013), https://doi.org/10.1016/j.jaap.2012.12.027

<sup>32</sup> M. Constantin, S. Bucatariu, L. Ursu, M. Butnaru, O. M. Daraba *et al.*, *Cellulose Chem. Technol.*, **53**, 695(2019),

https://doi.org/10.35812/cellulosechemtechnol.2019.5 3.68