# Enhancement of Delignification by Ionic Liquids Pretreatment and Modification of Hardwood Kraft Pulp in Preparation for Bleaching

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Ionic liquids (ILs) pretreatment is shown to improve the pulp bleachability and modify the physical strength of the hardwood pulp during the oxygen-chlorine dioxide-hydrogen peroxide (ODP) bleaching. The pretreatment enhanced lignin removal in ODP bleaching, resulting in decreased lignin content of the pretreated pulp. The brightness and strength properties were improved. In particular, the folding strength was increased by 153% relative to the control when the pulp was pretreated by [TEA][$HSO_4$]. The analysis of scanning electron microscopy, Fourier transform infrared spectroscopy, and X-ray diffraction confirmed the enhancement of delignification in ILs pretreatment by the removal of hemicellulose and lignin on pulp fibers' surface, and the well-preserved fiber framework of the hardwood pulp fibers during ODP-bleaching.

*Keywords: Bleachability; Hardwood pulp; Ionic liquids; Pretreatment; Properties*

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## INTRODUCTION

Elemental chlorine free (ECF) bleaching has been widely adopted because of its environmental, economic, and social benefits (Zhu *et al*. 2017). The ECF bleaching can reduce pollution by replacing chlorine and hypochlorite-based chemicals (Kaur *et al*. 2018). Various ECF oxidants, such as oxygen (O) (Li *et al*. 2016), chlorine dioxide (D) (Acharjee *et al*. 2017), and hydrogen peroxide (P) (Loureiro *et al*. 2011) have therefore been commonly used. They efficiently degrade lignin by attacking all the phenoxy units (Wu *et al*. 2017). Yet, an undesired reduction in physical strength has been widely reported due to carbohydrate degradation (Zhao *et al*. 2018). Other approaches, for example, laccase pretreatment, have been applied to enhance brightness, but a reduction in physical strength and viscosity occur (Ravalason *et al*. 2012). In general, a loss of physical strength is noticed while elevating the bleachability of pulp. Therefore, the development of a novel approach improving the bleachability while modifying the physical properties of hardwood pulp is desirable.

During ECF bleaching, chlorine dioxide can selectively cleave lignin fragments by imparting hydrophilic carboxylic groups that allow for dissolution of lignin (Sjostrom 1993; Karim *et al*. 2011). Hydrogen peroxide enhances brightness *via* lignin-chromophore oxidation reactions (Brogdon and Lucia 2005). However, lignin compounds in plant fibers are not only on the surface but also inside. Thereby, during the penetration and diffusion of the oxidizing agent, unnecessary carbohydrate degradation always takes place.

Therefore, it is important to develop an approach to loosen the lignocellulosic fiber structure, promote the accessibility of bleaching agent to lignin compounds, and ultimately reduce the influence on fiber strength. Various studies have been reported for this purpose such as using enzyme, ionic liquids, and ultrasonication (Yu *et al*. 2018).

Ionic liquids (ILs) are attractive "green solvents" on account of their good solubility, chemical stability, and recyclability (Anthony *et al*. 2002; Khan *et al*. 2017). Thus, they are good candidates to pretreat lignocellulose. Their strong coulombic and H-bonding ability disrupts the H-bond network within plant fibers, facilitating lignin removal (Da *et al*. 2013; Xu *et al*. 2017). In previous work of Peng *et al*. (2019), [$HSO_4^-$]-based IL pretreatment was applied to protect the fibers from depolymerization by improving pulp viscosity during the bleaching of eucalyptus kraft pulp. These ILs pretreatments notably reduced kappa number and improved the physical strength. In addition, they reduced fines content, decreased water retention value, and reduced the amorphous regions of the pulp fibers. [BMIM][$HSO_4$] has been reported for the processing of plant fibers, presenting mild modification over compositions (da Costa Lopes *et al.* 2013) and enhancing bleachabilities (Peng *et al*. 2019). [TEA][$HSO_4$] was recently reported to present similar lignin removal effect yet with higher tolerance to the water content. It presented similar effectiveness with up to 20% of water, which allows a significantly reduction in the cost of IL synthesis. A promising investigation by Brandt *et al.* (2017) reported that the cost of [TEA][$HSO_4$] reached the same level as to methanol ($1/kg), yet with high efficiency processing lignocellulosic biomass. Thereby in this work, enhancement of [BMIM][$HSO_4$] and [TEA][$HSO_4$] ILs pretreatment on the bleachability and properties of hardwood kraft pulp during ODP bleaching was studied through a comprehensive overview of the advantages of the ILs pretreatment in ODP bleaching. The objective was to use ILs pretreatment to swell the structure of lignocellulose and modify the bleachability and strength properties in ODP bleaching thereby improving the pulp quality.

## EXPERIMENTAL

### Materials

Hardwood chips were obtained from a paper mill in China. 1-Butyl-3-methylimidazolium hydrogen sulfate ([BMIM][$HSO_4$], >95%) was obtained from Rhawn Chemical Co. Ltd. (Shanghai, China).

Triethylammonium hydrogen sulfate ([TEA][$HSO_4$], 83% in water solution) was synthesized following the method of Brandt-Talbot *et al.* 2017. The structure and purity of both ILs were confirmed with NMR. The $ClO_2$ (1 vol%) was obtained from Dahua Special Environmental Engineering Co. Ltd. (Shandong China), $H_2O_2$ (30%) and $H_2SO_4$ (98%) were purchased from Chemical Co. Ltd. (Laiyang China). Triethylamine (99%), anhydrous magnesium sulfate (99%), NaOH (>96%), and $Na_2S$ (>98%) were obtained from Fuyu Fine Chemical Co. Ltd. (Tianjin China). All chemicals used were analytically pure.

### Methods

*Pulp preparation*

The hardwood pulping conditions used were 21% active alkali, 25% sulfidity, 170 °C, and 90 min. The pulp chemical composition was: cellulose = 81.77 wt%, lignin = 15.30%, and hemicellulose = 3.52%. The properties of the pulp were kappa number of 20.04, brightness of 28.24 %ISO, and pulp viscosity of 1015 mL/g.

*Ionic liquid pretreatment*

Approximately 25 g of oven-dried pulp was mixed with 2.5 g of ILs in a sealed polyethylene bag, then water was added into the mixture to keep 10% of consistency. The pretreatment was conducted in a water bath at 60 °C for 60 min.

*ODP bleaching*

First, the pretreated pulp was adjusted to 10% of consistency in a polyethylene bag, and bleached in an ODP bleaching sequence. The conditions of ODP bleaching sequence were 0.5 MPa of oxygen pressure, 4% of $Na_2O$, 100 °C for 60 min in an oxygen stage (O); 9 kg/t pulp of $ClO_2$ dosage, pH value in the range 2 to 3, 70 °C for 30 min in chlorine dioxide stage (D), 12 kg/t pulp of $H_2O_2$ dosage, 5 wt.% of NaOH dosage for adjusting pH to the range 11 to 12, and 90 °C for 60 min in hydrogen peroxide stage (P).

*Ionic liquid recovery*

The effluent after oxygen delignification stage was collected and filtered to remove insoluble content. The filtrate was collected and dehydrated under rotary evaporation, followed by centrifugation and filtration. The filtrate was again collected and dehydrated under reduced pressure with Schlenk line at 70 °C for 24 h. The structure and purity of the recycled ILs was checked by NMR.

*Pulp analysis*

The content of cellulose, hemicellulose, and lignin were determined according to TAPPI T 201 wd-76 (2004), T 223 cm-01 (2004) and T 222 om-11 (2011), respectively.

Analysis of pulp viscosity was obtained using the method of ISO 10650 (1999), and the degree of polymerization (DP) of pulp was calculated based on the pulp viscosity ($\eta$). The DP was calculated according to the Eq. 1 (Mazumder *et al*. 2000):

$$DP^{0.905}=0.75[\eta] \quad (1)$$

Pulp yield was calculated as the mass ratio of pulp sample before and after bleaching.

Kappa number of pulp was determined according to the ISO 302 (2004) method.

The water retention value (WRV) of pulp was determined according to the ISO 23714 (2013) method. The weight mean length, weight mean width, and fines content of pulp fibers were measured using a fiber quality analyzer (FQA-LDA02, OpTest Equipment Inc., Ontario, Canada).

*Handsheet preparation*

Pulp handsheets were prepared according to the literature (Chen *et al*. 2017). The grammage of the prepared pulp handsheet was 80 g/m$^2$, and they were conditioned at 23 °C and 50% humidity for 24 h prior to analysis. The properties include of tensile index, tear index, folding endurance, and brightness of the pulp handsheets were measured according to the TAPPI standard methods TAPPI T494 om-01 (2004), TAPPI T414 om-04 (2004), TAPPI T511 om-02 (2004) and TAPPI T452 om-02 (2004), respectively.

*Scanning electron microscopy (SEM)*

The surface observations of pulp fibers were done using a scanning electron microscope (Regulus 8220, Hitachi, Tokyo, Japan). The oven dried samples were thoroughly scanned at 5 kV accelerating voltage and 2000 times magnification.

*Fourier transform infrared spectroscopy (FTIR)*

The FT-IR analysis of the pulp samples were conducted using a Bruker FT-IR (Vertex70, Bruker, Karlsruhe, Germany) and the scanning range was 4000 to 500 $cm^{-1}$, with a resolution ratio of 4 $cm^{-1}$ and a scanning speed of 32 $s^{-1}$.

*X-ray diffraction (XRD) analysis*

The degree of crystallinity of the pulp was evaluated using a X-ray diffraction (D8-Advance, Bruker, Karlsruhe, Germany). The scan mode used was $2\theta$ at 35 mA. Angles ranged from 5° to 60°. The crystallinity index of various samples was determined according to (Segal *et al*. 1959) Eq. 2:

$$CrI\ (\%) = 100 \times (I_{002} - I_{am})/I_{am} \quad (2)$$

where $CrI$ was the crystallinity of cellulose, $I_{002}$ was the scattering intensity of the diffraction of 002 plane, $I_{am}$ was the diffraction intensity at $2\theta$ =15.6°.

## RESULTS AND DISCUSSION

### Effect of ILs Pretreatment on the Chemical Compositions and the Bleachability of the Hardwood Pulp

Effects of ILs pretreatment on the composition of bleached pulp during ODP bleaching are shown in Table 1, while the effect of ILs pretreatment on the bleachability of the kraft pulp during ODP bleaching process is listed in Table 2. Table 1 shows that ILs pretreatment protected the pulp fibers by increasing the cellulose content and viscosity, and enhanced lignin removal during ODP bleaching. The lignin content and kappa number of the pulp pretreated by [TEA][$HSO_4$] were decreased by 18% and 69%, while the brightness increased by 1.33 %ISO compared to the control. Thereby ILs pretreatment can enhance and modify bleachability. As shown in Table 2, the ILs pretreatment also increased pulp yield, demonstrating the protection effect of ILs pretreatment for pulp fibers, which did not influence the drainability as shown in a stable water retention value (WRV).

**Table 1.** Compositions of the Pretreated and Unpretreated Pulp during ODP Bleaching Process

| ILs Pretreatment | Cellulose (%) | Hemicellulose (%) | Lignin (%) |
|---|---|---|---|
| -- | 95.27±0.21 | 2.44±0.08 | 1.97±0.07 |
| [BMIM][$HSO_4$] | 95.45±0.13 | 1.87±0.13 | 1.83±0.06 |
| [TEA][$HSO_4$] | 96.23±0.12 | 2.25±0.12 | 1.62±0.03 |

**Table 2.** Effect of ILs Pretreatment on the Bleachability of Hardwood Pulp during ODP Bleaching Process

| ILs Pretreatment | Brightness (%ISO) | Kappa number | Yield (%) | WRV (g/g) |
|---|---|---|---|---|
| -- | 79.17±0.05 | 1.54±0.08 | 96.76 | 1.50±0.04 |
| [BMIM][$HSO_4$] | 79.80±0.03 | 0.67±0.08 | 97.09 | 1.41±0.05 |
| [TEA][$HSO_4$] | 80.50±0.02 | 0.48±0.08 | 97.23 | 1.49±0.02 |

### Effect of ILs Pretreatment on Fiber Qualities and Strength Properties of the Pulp

Effect of ILs pretreatment on fiber qualities and strength properties of the pulp during ODP bleaching are shown in Tables 3 and 4. Table 3 showed an increase of length, width, viscosity, and DP of the pretreated pulp fibers. As can be seen, the viscosity and DP increased by 17.8% and 19.8% , respectively, while fines content decreased compared to the control. Table 4 showed that the tensile index, tear index, and folding endurance of the pretreated pulp during ODP bleaching were all improve. In particular, the folding endurance of the pulp pretreated by [BMIM][$HSO_4$] and [TEA][$HSO_4$] increased by 100% and 153%, respectively, compared to the control pulp. These results are consistent with increase of length, viscosity, and DP of the pretreated pulp fibers showing an enhancement of ILs pretreatment on the bleachability and strength properties of hardwood pulp.

Both ILs pretreatment in this work facilitated the dissolution of lignin during the bleaching process, which contributed to the swelling of the pulp fibers, and possibly led to the higher fiber width within pretreated samples. It should be admitted that only minor difference in lignin content was found in the final ODP bleached samples, which could be attributed to the mild pretreatment conditions. ILs dispersed into the vast aqueous phase allowing a desirable mild modification over the pulp fibers, by the meantime avoided ILs attaching on the surface of pulp fibers. Therefore the recycling of the ILs was easily achievable.

On the other hand, the IL pretreatment dissolved hemicellulose component and modified the micro-fiber compositions. The mild process conditions allowed the modification of micro-fiber to take place while keeping the cellulose framework untouched. Hence, the fiber length of pulp increased, resulting in an increase of viscosity and DP of the pulp fibers. All these effects mentioned above contributed to the enhancement of the physical strength in the handsheets.

In addition, the ionic liquids used in the pretreatment process could be recycled by collecting the effluent after oxygen delignification stage, where up to 98% of ILs could be recovered. These ILs could be reused after purification to process handsheet with similar physical strength upto 3 cycles.

**Table 3.** Effect of IL Pretreatment on Pulp Fibers Qualities during the ODP Bleaching Process

| ILs Pretreatment | Fiber length (mm) | | Fiber width (μm) | Fines (%) | Viscosity (mL/g) | DP |
|---|---|---|---|---|---|---|
| | $L_c$ (n) | $L_c$ (w) | | | | |
| -- | 0.631 | 0.793 | 13.50 | 4.19 | 684±3 | 988 |
| [BMIM][$HSO_4$] | 0.632 | 0.796 | 13.61 | 4.02 | 721±4 | 1047 |
| [TEA][$HSO_4$] | 0.639 | 0.795 | 13.60 | 3.24 | 806±4 | 1184 |

Definitions: $L_c$ (n) = number mean length; $L_c$ (w) = weight mean length

**Table 4.** Effect of IL Pretreatment on the Physical Strength of Pulp Handsheet

| ILs Pretreatment | Tensile index ($N·m·g^{-1}$) | Tear index ($mN·m^2·g^{-1}$) | Folding endurance (times) |
|---|---|---|---|
| -- | 52.75±0.25 | 6.93±0.24 | 126±12 |
| [BMIM][$HSO_4$] | 57.13±0.50 | 7.60±0.06 | 252±6 |
| [TEA][$HSO_4$] | 60.13±0.13 | 7.87±0.13 | 319±10 |

## SEM Analysis

The fibers morphology of the ODP bleached pulp were accessed from SEM analysis, as shown in Fig. 1. According to Fig. 1(a), the control pulp fibers showed a smooth surface during bleaching illustrating the removal of the wax and encrusting substances on the fiber surface (Mohtar *et al*. 2017). Figures 1(b) and 1(c) displayed cracks and holes on fiber surface including small fragments were observed after ILs pretreatment, which validated successful removal of hemicellulose and lignin on pulp fibers' surface.

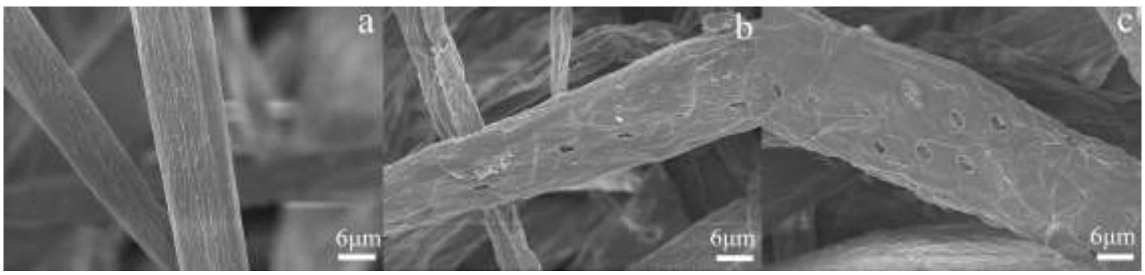


**Fig. 1.** SEM images of the ODP bleached pulp: (a) Control pulp; (b) [BMIM][$HSO_4$] pretreated pulp; and (c) [TEA][$HSO_4$] pretreated pulp.

## FTIR Analysis

The FTIR spectra of the pulp samples are shown in Fig. 2, where the peak at 3400 $cm^{-1}$ corresponded to stretching vibration of O-H groups, the absorbance at 2940 $cm^{-1}$ to 2850 $cm^{-1}$ for C-H stretching vibration (Mohtar *et al*. 2017), the peaks at 1437 $cm^{-1}$ and 1335 $cm^{-1}$ were characteristic absorption peaks of lignin (Bouchard and Douek 1993), the absorbances at 1160 $cm^{-1}$ and 1060 $cm^{-1}$ were associated with C-O-C tensile vibrations and C-O-C pyranose ring skeletal vibration, and the peak at 898 $cm^{-1}$ was a β-type glycosidic bond vibration, indicating that the xylan units were linked by a β-type glycosidic bond (Sun *et al*. 1996). As shown in Fig. 2, there was no new characteristic peak appearing for the pretreated pulp fibers, which confirmed that ILs pretreatment can only change the physical structure of pulp fibers, but did not alter the chemical structure such as functional groups.

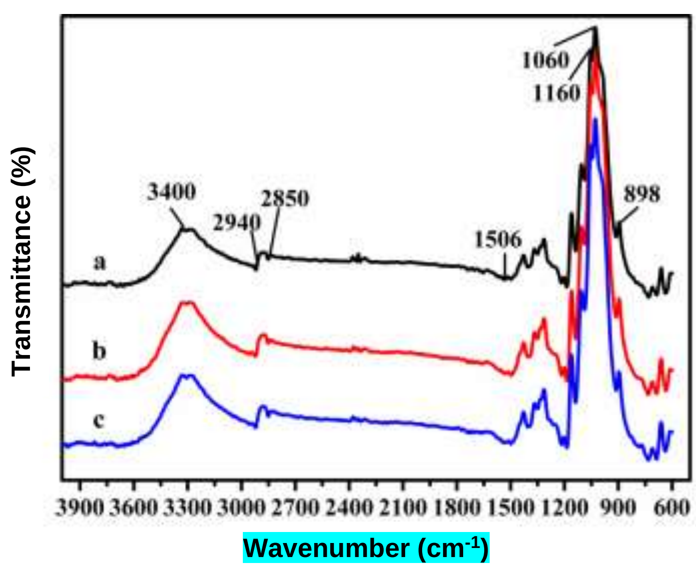


**Fig. 2.** FTIR analysis of the ODP bleached pulp: (a) Control pulp; (b) [BMIM][$HSO_4$] pretreated pulp; and (c) [TEA][$HSO_4$] pretreated pulp

## XRD Analysis

Figure 3 shows the XRD patterns of the ODP bleached pulp. In Fig. 3, the cellulose fraction indicated two peaks corresponding to ($I_{am}$), ($I_{002}$), which appeared at 15.6° and 22°, respectively. As shown in Fig. 3, the crystallinity of the control pulp was 36.2%, while that of the pulp pretreated by [BMIM][$HSO_4$] and [TEA][$HSO_4$] were 48.8% and 50.3%, respectively. In other words, an increase of crystallinity by 35% and 39%, compared to the control pulp was observed, which further indicated the enhancement of ILs pretreatment on the bleachability and properties of hardwood kraft pulp due to removal of hemicellulose and lignin during ODP bleaching. Meanwhile, the protection of ILs on the pulp fibers resulted in retaining cellulose with a higher degree of crystallinity (Rosli *et al*. 2013).

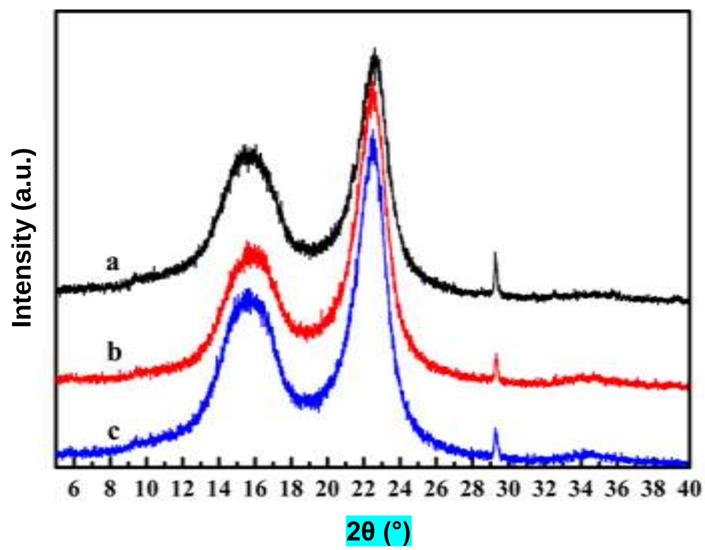


**Fig. 3.** XRD analysis of the ODP bleached pulp: (a) Control pulp; (b) [BMIM][$HSO_4$] pretreated pulp; and (c) [TEA][$HSO_4$] pretreated pulp

## CONCLUSIONS

1. Ionic liquids pretreatment enhanced the bleachability and properties of hardwood kraft pulp during ODP bleaching.
2. A decrease of lignin content and increase of brightness and strength properties of the pretreated pulp were obtained compared to the control; especially the folding strength of the pulp pretreated by [TEA][$HSO_4$] considerably increased by 153%.
3. The crystallinity of the pulp pretreated by [BMIM][$HSO_4$] and [TEA][$HSO_4$] increased by 35% and 39%, respectively.
4. The analysis of SEM and FTIR confirmed the enhancement of delignification in ILs pretreatment by the removal of hemicellulose and lignin on pulp fibers surface, and the well-preserved fiber framework of the hardwood pulp fiber during ODP-bleaching.

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## REFERENCES CITED

Acharjee, T. C., Jiang, Z., Haynes, R. D., and Lee, Y. Y. (2017). "Evaluation of chlorine dioxide as a supplementary pretreatment reagent for lignocellulosic biomass," *Bioresource Technol.* 244, 1049-1054. DOI: 10.1016/j.biortech.2017.08.038

Anthony, J. L., Brennecke, J. F., Holbrey, J. D., Maginn, E. J., Mantz, R. A., Rogers, R. D., Trulove, P. C., Visser, A. E., and Welton, T. (2002). *Physicochemical Properties of Ionic Liquids. Ionic Liquids Synth*, Wiley‐VCH Verlag GmbH Co. KGaA, DOI:10.1002/3527600701.ch3

Bouchard, J., and Douek, M. (1993). "Structural and concentration effects on the diffuse reflectance FTIR spectra of cellulose, lignin and pulp," *J. Wood Chem. Technol.* 13(4), 481-499. DOI: 10.1080/02773819308020530

Brandt-Talbot, A., Gschwend, F. J. V., Fennell, P. S., Lammens, T. M., Tan, B., Weale, J., and Hallett, J. P. (2017). "An economically viable ionic liquid for the fractionation of lignocellulosic biomass," *Green Chem.* 19(13), 3078-3102. DOI: 10.1039/c7gc00705a

Brogdon, B. N., and Lucia, L. A. (2005). "New insights into lignin modification during chlorine dioxide bleaching sequences (Ⅲ): The impact of modifications in the (EO) *versus* e stage on the D1 stage," *J. Wood Chem. Technol.* 24(3), 201-219. DOI: 10.1081/WCT-200038082

Chen, J., Jiang, Q., Yang, G., Wang, Q., and Fatehi, P. (2017). "Ultrasonic-assisted ionic liquid treatment of chemithermomechanical pulp fibers," *Cellulose* 24(3), 1483-1491. DOI: 10.1007/s10570-016-1180-y

Da, Costa Lopes, A. M., Joao, K. G., Morais, A. P. C., Lukasik, E. B., and Lukasik, R. B. (2013). "Ionic liquids as a tool for lignocellulosic biomass fractionation," *Sustain. Chem. Processes* 1, 1-3. DOI: 10.1186/2043-7129-1-3

ISO 302 (2004). "Pulps - Determination of Kappa number," International Organization for Standardization, Geneva, Switzerland.

ISO 5351(2009). "Pulps - Determination of limiting viscosity number in cupriethyleneiamine (CED) solution," International Organization for Standardization, Geneva, Switzerland.

ISO 23714 (2013). "Pulps - Determination of water retention value (WRV)," International Organization for Standardization, Geneva, Switzerland method.

Karim, M. R., Islam, M. N., and Malinen, R. O. (2011). "Response of eucalyptus camaldulensis and *Acacia mangium* kraft pulp in different ECF bleaching options," *Wood Sci. Technol.* 45(3), 473-485. DOI:10.1007/s00226-010-0338-2.

Kaur, D., Bhardwaj, N. K., and Lohchab, R. K. (2018). "Environmental aspect of using chlorine dioxide to improve effluent and pulp quality during wheat straw bleaching," *Waste. Biomass. Valor.* (1), 1-9. DOI: 10.1007/s12649-017-0193-6

Khan, A. S., Man, Z., Bustam, M. A., Nasrullah, A., and Muhammad, N. (2017). "Efficient conversion of lignocellulosic biomass to levulinic acid using acidic ionic liquids," *Carbohydr. Polym* 181, 208-214. DOI: 10.1016/j.carbpol.2017.10.064

Li, J., Zhang, C., Hu, H., and Chai, X. S. (2016). "Effect of shortening kraft pulping integrated with extended oxygen delignification on biorefinery process performance of eucalyptus," *Bioresource Technol.* 202, 119-124. DOI: 10.1016/j.biortech.2015.11.074

Loureiro, P. E. G., Evtuguin, D. V., and Carvalho, M. G. V. S. (2011). "The final bleaching of eucalypt kraft pulps with hydrogen peroxide: Relationship with industrial ECF bleaching history and cellulose degradation," *J. Chem. Technol. Biotechnol.* 86(3), 381-390. DOI: 10.1002/jctb.2527

Mazumder, B. B., Ohtani, Y., Zhou, C., and Sameshima, K. (2000). "Combination treatment of kenaf bast fiber for high viscosity pulp," *J. Wood Sci.* 46(5), 364-370. DOI: 10.1007/BF00776397

Mohtar, S. S., Zulaikha, T. M. B. T. N., Md Noor, A. M., Shaari, N., and Mat, H. (2017). "An ionic liquid treatment and fractionation of cellulose, hemicellulose and lignin from oil palm empty fruit bunch," *Carbohydr. Polym.* 166, 291-299. DOI: 10.1016/j.carbpol.2017.02.102

Peng, J., Qi, L., Yang, G., He, M., Xue, Y., and Chen, J. (2019). "Effect of ultrasonic-assisted ionic liquid pretreatment on the bleachability and properties of eucalyptus kraft pulp," *J. K. TAPPI* 51, 18-27. DOI: 10.7584/JKTAPPI.2019.04.51.2.16

Ravalason, H., Bertaud, F., HerpoeL-Gimbert, I., Meyer, V., Ruel, K., Joseleau, J. P., Grisel, S., Olive, C., Sigoillot, J. C., and Petit-Coni, M. (2012). "Laccase/HBT and laccase-cbm/HBT treatment of softwood kraft pulp: impact on pulp bleachability and physical properties," *Bioresource Technol.* 121, 68-75. DOI: 10.1016/j.biortech.2012.06.077

Rosli, N. A., Ahmad, I., and Abdullah, I. (2013). "Isolation and characterization of cellulose nanocrystals from *Agave angustifolia* fibre," *BioResources* 8(2), 1893-1908. DOI: 10.15376/biores.8.2.1893-1908

Segal, L., Creely, J. J., Martin, A. E., and Conrad, C. M. (1959). "An empirical method for estimating the degree of crystallinity of native cellulose using the x-ray diffractometer," *Text Res. J.* 29(10), 786-794. DOI: 10.1177/004051755902901003

Sjostrom, E. (1993). *Wood Chemistry: Fundamentals and Applications* (2nd ed.), Academic Press, San Diego.

Sun, R., Lawther, J. M., and Banks, W. B. (1996). "Fractional and structural characterization of wheat straw hemicelluloses," *Carbohydr. Polym.* 29(4), 325-331. DOI: 10.1016/s0144-8617(96)00018-5

TAPPI T201 wd-76 (2004). "Cellulose in pulp (Cross and Bevan method)," TAPPI Press, Atlanta, GA.

TAPPI T222 om-11 (2011). "Acid-insoluble lignin in wood and pulp," TAPPI Press, Atlanta, GA.

TAPPI T223 cm-01 (2004). "Pentosans in wood and pulp," TAPPI Press, Atlanta, GA.

TAPPI T414 om-04 (2004) "Internal tearing resistance of paper (Elmendorf-Type Method)," TAPPI Press, Atlanta, GA.

TAPPI T452 om-02 "Brightness of pulp, paper and paperboard (Directional reflectance at 457 nm)," TAPPI Press, Atlanta, GA.

TAPPI T494 om-01 (2004) "Tensile properties of paper and paperboard (using constant rate of elongation apparatus)," TAPPI Press, Atlanta, GA.

TAPPI T511 om-02 "Folding endurance of paper (MIT Tester)," TAPPI Press, Atlanta, GA.

Wu, W. J., Zhang, Y., Jiang, B., and Jin, Y. C. (2017). "Sulfite-formaldehyde pulping and oxygen delignification of wheat straw," *China. Bio. Mater. Pap.* 2(3), 12-17. DOI: 10.12103/j.issn.2096-2355.2017.03.002

Xu, J., Liu, B., Hou, H., and Hu, J. (2017). "Pretreatment of eucalyptus with recycled ionic liquids for low-cost biorefinery," *Bioresource Technol.* 234, 406-414. DOI: 10.1016/j.biortech.2017.03.081

Yu, X., Bao, X., Zhou, C., Zhang, L., Aea, Y., and Yang, H. (2018). "Ultrasound-ionic liquid enhanced enzymatic and acid hydrolysis of biomass cellulose," *Ultrason. Sonochem.* 41, 410-418. DOI: 10.1016/j.ultsonch.2017.09.003

Zhao, H., Li, J., and Zhang, X. (2018). "Fundamental understanding of distracted oxygen delignification efficiency by dissolved lignin during biorefinery process of eucalyptus," *Bioresource Technol.* 258, 1-4. DOI: 10.1016/j.biortech.2018.02.122

Zhu, H. X., Yao, S. Q., Chen, Y. M., Xin-Yu, L. I., Nie, S. X., and Qin, C. R. (2017). "Research progress of AOX reduction technology for ECF bleaching," *China Pulp Pap.* 32(2), 52-57. DOI: 1000-6842 (2017) 02-0052-0

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