Carbohydrate Polymers 174 (2017) 330–336

Contents lists available at ScienceDirect

Carbohydrate Polymers

journal homepage: www.elsevier.com/locate/carbpol

Rapid fabrication of transparent film directly from wood fibers with microwave-assisted ionic liquids technology

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ARTICLE INFO

Article history:
Received 11 March 2017
Received in revised form 17 May 2017
Accepted 15 June 2017
Available online 16 June 2017

Keywords:
Ionic liquid
Microwave
Transparency
Mechanical properties

ABSTRACT

Presently flexibly transparent film or nanopaper from all cellulose was mostly fabricated by assembling cellulose nanofibers disintegrated from macroscopic wood fibers which mostly suffers from potential environmental toxicity or high cost. In this work, we firstly reported an all-cellulose transparent film fabricated by a novel microwave-assisted ionic liquids technology (MILT). The use of MILT for treating the original all-cellulose paper brings nearly 2.6 fold-increases in optical transmission, and 2.0 fold-increases in tensile property compared to those without microwave assistance. More importantly, by contrast with the partial dissolution of cellulose in typical DMAC/LiCl, ILs, NaOH/urea, the MILT is extremely time-saved with responding to the highest increase in mechanical property because the high efficient surface dissolution and welding bind individual sheets together under a micro environment.

1. Introduction

The highly transparent film or nanopaper has been the subject of intensive research during the last couple of decades due to its wide applications in flexible electronics and devices (Fang et al., 2014; Huang et al., 2016; Nakagaito, Nogi & Yano, 2010; Zhu, Xiao, Liu, Li, Weadock, & Fang, 2013). Compared to the transparent polymer film, the transparent all-cellulose film or nanopaper was widely considered to be an ideal material for the use as the flexible substrate because of its outstanding mechanical properties, optical transparency, economic production as well as environmental friendliness. In past decades, the transparent film has been successfully developed by disintegrating macroscopic wood fibers into cellulose nanofibers via enzyme (Pääkkö et al., 2007), acid hydrolysis (M. Akira, Tsuguyuki & Hayaka, 2011), or chemical pretreatments followed by aggressively mechanical processes (Zhang, Tsuzuki & Wang, 2015). But most of them suffer from potential environmental toxicity, the poor efficiency and high energy consumption. A high yield and sustainable process for directly processing macroscopic cellulose fiber into flexibly transparent film or nanopaper were becoming a hot topic (Sun, Wu, Ren & Lei, 2015).

Among known strategies for fabricating transparent film from all-cellulose matrix, the dissolution of cellulose in matrix by cellulose solvents is extremely attractive due to its high efficiency and low energy consumption. It can be achieved through mainly two different approaches: (1) the transparent cellulose films were prepared by firstly completed dissolution of cellulose in solvents followed by regenerating them in water or ethanol to form transparent cellulose films; (2) the other transparent cellulose films were prepared by surface partial dissolution of cellulose in solvents followed by the similar regeneration process (Nishino & Peijs, 2014). Compared with the first approach of completing dissolution of cellulose, the partly dissolution of cellulose has some advantages of simplifying the working procedure, time-saving, and also cost efficiency. For example, transparent cellulose films with good mechanical strength were prepared using the filter paper which was immersed in LiCl/DMAc for 12.0 h in order to partly dissolve cellulose and the undissolved fibers were bonded together to enhance the physical and structural strength of the films by Professor N. Takashi and the coworkers (Nishino & Arimoto, 2007a; Soykeabkaew, Arimoto, Nishino & Peijs, 2008; Soykeabkaew, Sian, Gea, Nishino & Peijs, 2009a; Soykeabkaew, Nishino & Peijs, 2009). The maximum stress of transparent cellulose films fabricated from hemp fibers reaches to 211 Mpa, however, the optical transparency is still very low and the fabricating process is highly time-consuming. As all we know, it is difficult to dissolve cellulose in organic and inorganic solvents due to its stiff fibril structure, strong intermolecular hydrogen bonding between cellulose molecules and also a high degree of crystallization. Research efforts over the past few decades have found several solvents of cellulose

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http://dx.doi.org/10.1016/j.carbpol.2017.06.057
0144-8617/

to rapidly dissolve cellulose, such as N-methyl Morpholine, N-methylmorpholine-N-oxide(NMMO) or LiCl/DMAc or NaOH/urea aqueous solutions or PF/DMSO (Cui-Zheng, 2002; Cui-Ling & Xin-Ping, 2004; Hao, Jin, Jun Zhang & He, 2005; Yousefi, Nishino, Faezipour, Ebrahimi & Shakeri, 2011b; Zhu et al., 2006). Despite its excellent strengths, those solvents suffer from several major drawbacks: poor physical and optical properties with the materials, high toxic, uneconomic and unrecyclable (Kinstle & Irving, 1983). Compared to the above mentioned solvents, ionic liquids (ILs) are a novel type of green solvent since its excellent dissolving capacity, recyclable, high thermal stability and other advantages (Hameed, Xiong, Salim & Guo, 2013). Benoît J.C. Duchemin et al. (2009) prepared the cellulose films using ILs as the solvent and the filter paper as the original paper (Duchemin, Mathew & Oksman, 2009). However, they focused on the mechanical properties without more detailed studies about the optical performance of the treated paper. The nano-welding technology between cellulose was firstly put forward by Professor H. Yousefi et al. (2009) (Yousefi, Nishino, Faezipour, Ebrahimi & Shakeri, 2011a). It was proposed that the process of immersing rape straw in BMIMCl ionic liquids at 85 °C followed by soaking in methanol significantly improved the original paper, but results in dissatisfied optical properties. Heating chemical reactions by microwave irradiation has become advantageous technology since the first published reports by the groups of Gedye (Giguere, Bray, Duncan & Majetich, 1986; Gedye et al., 1986). More importantly, compared to the conventional heating methods, microwave heating has been shown to dramatically reduce reaction times, increase product yields, and enhance product purities by reducing unwanted side-reactions.

In this paper, we reported a highly transparent all-cellulose film by part dissolution of the cellulose fibers using ionic liquids (ILs) as the cellulose solvent and microwave assistance as the source of heating. Our all-cellulose film have the following advantages over previously reported films: (1) it has perfect interfacial compatibility because both fiber and matrix are composed of cellulose and the ILs has great efficient in dissolving cellulose; (2) the immersion time by MILT is only about 2 min, which is a great advantage compared to other similar treating technologies such as DMAC/LiCl, NaOH/urea; (3) tensile property and optical transmission have been greatly improved after the technology of MILT, the film has a 2.0 fold rise in tensile property and nearly 2.6 fold rise in optical performance by contrast with those without microwave assistance.

2. Materials and methods

2.1. Materials

Commercial eucalyptus dissolving pulp used as the raw cellulose source material was purchased from Guangzhou Chenhui Paper Co., Ltd. (China). Dichloromethane (99.8%), tetrahydrofuran (THF, GC – 99.5%), were provided by Shanghai Sigma-Aldrich Co., Ltd. (China); 1-ethyl-imidazole (analytical reagent grade, 98%), dimethyl phosphite (analytical reagent grade, 98%), supplied by Aladdin-Reagent Co., Ltd. (Shanghai China). Dichloromethane, tetrahydrofuran, 1-ethyl-imidazole, dimethyl phosphite, were used to synthesize ILs. All the reagents were used as received without further purification.

2.2. The synthesis of $EminMeOPO_2H$ ILs

The synthesis of 1-ethyl-2-methylimidazolium phosphorous methyl ester ($EminMeOPO_2H$) ILs was carried out by according previously reported literatures (Ruan, Chen, Liu, & Xia, 2014; Zhu et al., 2013). Dichloromethane, tetrahydrofuran, 1-ethyl-imidazole and dimethyl phosphite are raw material in the synthesis of ILs ($EminMeOPO_2H$). 80.0 ml tetrahydrofuran, 1-ethyl-imidazole and dimethyl phosphite were respectively added to a three-necked flask filled with argon, and the solvent was stirred by magnetic agitator (Shanghai Yingdi Automation Inc., China) at 85 °C for 48 h. In rotary evaporation (Shanghai Yingdi Automation Inc., China) a vacuum distillation apparatus was used to remove tetrahydrofuran from the reagent. The excess impurities were removed by extraction with ether as solvent. After firstly dissolving in dichloromethane and subsequently filtering through a neutral activated alumina, the reagent was treated with vacuum distillation to remove excess dichloromethane and ether. By doing this, the ionic liquid ($EminMeOPO_2H$) can be synthesized after continuous vacuum drying at 80 °C for 48 h.

2.3. Preparation of the original all-cellulose paper

Raw cellulose fibers used for the preparation of original paper were pretreated by a beating process with a PFI mill and the beating degree of pulp is 27°SR (Hamjern Maskin 621, Dongguan International Material Tester) (China). The paper formation machine (MESSMER 255, Clausen Inc.) (American) was used for getting the original paper. After sandwiching prepared wet paper between two stacks of regular filter paper, we placed it under a pressure of 0.4 MPa and dried at 110 °C for 10 min. A piece of original all-cellulose paper was obtained.

2.4. Preparation of transparent film by MILT

The method of preparing transparent film is briefly described as below. The original paper with base density of 37 g/m^2 and an area of 28 cm^2 was first placed in a vacuum drying at 80 °C for 30 min to remove the residual water. The dried original paper was coated with 1.0 ml ILs at 72 °C within 5 s, and subsequently was sandwiched between flat plates of glass for starting a domestic microwave (P70F20CL-DG(B0) Guangdong Galanz Electric Manufacturing Co., Ltd.) (China) with effective 700.0 W for 2 min. The distilled water and anhydrous ethanol were respectively utilized to remove the residual ILs in the resulting paper. In order to further strengthen the pretreating paper, it was sandwiched between two stacks of PET (polyethylene terephthalate, ~ 60 μm) membrane and dried under 2.0 MPa and 80 °C for 10 min to obtain Film-2 sample. The Film-1 sample was obtained using the above mentioned method without microwave assistance.

2.5. Characterizations

The structure of the original paper, Film-1 samples and Film-2 samples were studied with a scanning electron microscope (SEM). JEM-100CXII at an accelerating voltage of 10 kV. Prior to the examination, the surface of the specimen was coated with a thin layer of gold, ~20 nm.

X-ray diffraction (XRD) patterns of the Film-1 and Film-2 samples were carried out on a X-ray diffractometer (D8 ADVANCE, Bruker Inc., Germany) with area detector operating at a voltage of 40 kV and a current of 40 mA using Cu Kα radiation (λ = 0.154 nm). The scanning scope of 2θ was from 4° to 50° at room temperature.

The mechanical strength of all the samples was measured using a universal tensile tester (Instron5565, Instron instruments Inc. USA). Samples were firstly cut to 15.0 mm × 50.0 mm and placed in constant temperature and humidity chamber at (50 ± 1) % relative humidity (RH) and (23 ± 1) °C for 24 h to ensure the stabilization of their water content before characterization and ten replicate specimens were tested from each film type.

UV–vis spectrometer with an integrating sphere (UV-9000 Shanghai Yuanyi Inc. China) with an integrating sphere was used to measure the total transmittance of the original paper, Film-1,

Film-2 samples and PET (polyethylene terephthalate, ~60 μm) in a wavelength range of 400 nm to 900 nm.

Atomic force microscopy (AFM) analysis (Bruker Instruments, Germany) was performed to study the surface topography of the transparent Film-2 samples. All of the samples were studied in contact mode using silicon cantilevers.

3. Results and discussion

3.1. Reaction principle analysis

Cellulose ($C_6H_{10}O_5$) n is a β-1, 4 linked linear polymer of glucose units with good chemical and biodegrading properties. The multi-step and prolonged processes are needed to dissolve cellulose in traditional organic solvents due to its stiff structure. Compared to other cellulose solvents, ILs (EminMeOPO$_2$H) are recently attracting great attentions due to its high-efficient, unique properties and special characters. Microwave heating which is a new type of heating method was utilized to improve the efficiency of cellulose dissolution (Dr, 2004). The dried original paper with base density of 37 g/m^2 and an area of 28 cm^2 was firstly coated by the 1.0 ml EminMeOPO$_2$H ILs in an over-dried microwave oven chamber at 72 °C within 5 s (Fig. 1a). The ILs rapidly penetrated into cell wall of cellulose and broke the original hydrogen bonds between or/and inner the cellulose chains due to the attack of cation and anion from the ILs. When starting the microwave oven, the strong polar cation and anion from ILs were respectively vibrated with ultra-top speed in electromagnetic field (Fig. 1b). Meanwhile, the vibrating process promoted its temperature which rapidly accelerated the permeating process of ILs into the cell wall of cellulose, and greatly promoted the efficiency of breaking hydrogen bonds. Due to the insufficient addition of ILs, the dissolution of cellulose was mostly occurred on the surface of cellulose fibers. In this condition, the main skeleton of cellulose fiber still remained in situ to be connected together with the partly dissolved cellulose. After complete treating in microwave oven, the resulting paper was peeled from the glass plate for washing in distilled water and anhydrous ethanol. The partly dissolved cellulose on the surface of cellulose was instantaneously regenerated due to the reconstruction of hydrogen bond with the removal of ILs. Regeneration of the dissolved cellulose on the surface of cellulose fiber was considered as the adhesive to bond the neighboring cellulose fibers. The schematic diagram of treating the paper from experiments in which the system was kept dry and an experiment with microwave assistance present for the sake of comparison (Fig. 1c). Once the washing process had completed, hot-pressing was applied and maintained for 10 min under 2.0 MPa and 80 °C after sandwiched between two stacks of PET membrane. By applying hot pressing the partly dissolving cellulose liquid became plasticization and easier fluidization which further fills in the pores between or/and inner the cellulose fibers (Fig. 1c). The fill of partly dissolved cellulose liquid replaced the air in the pores between or/and inner the cellulose fibers and the regenerated cellulose further flows and levels the coarse pores between or/and inner the resulting cellulose fibers under high temperature and pressure. The replacement of cellulose for air in pores yielded the films with proportionable refractive index along the whole cellulose matrix, which provided the optical transparency of the treating paper. Rich regeneration of the dissolved cellulose on the surface of cellulose fiber enabled facile formation of effective bonding, both intrachain, interchain and also the neighboring cellulose fibers.

3.2. Properties of transparent films

In order to investigate the properties of transparent film, the structure of the paper samples was examined (Fig. 2). The differences from the ideal structure of original paper to the treated paper samples (Original paper, Film-1 sample and Film-2 sample) were distinctly observed from the SEM images by scanning cross-sections and fracture surface. The original paper was loosely composed of large pores and cellulose fibers. The distinct integration between fibers is not observed after assembling them only by hydrogen bond. The difference in refractive index between air in pores and the basic cellulose results in the opaque of the original paper (Soykeabkaew, Sian, Gea, Nishino & Peijs, 2009b). Compared to the porous and loose structures of the original paper, the Film-1 sample treated only by EminMeOPO$_2$H ILs is denser while maintains fibrous profile because the partly dissolved cellulose binds individual fibers together (Fig. 2c,d). In particular, the replacement of air in pores by cellulose is highly desirable for achieving good optical and mechanical properties of the resulting paper. Among known strategies for heating assistance, the electromagnetic field generated by microwave, is arguably the most versatile and easily scalable method. Yet, much as in structure of the original paper samples, such coarse pores and fibrous structure was completely removed by developing appropriate microwave process that translates electromagnetic field into the part dissolution of cellulose by ILs. For this reason, the work presented herein is devoted to overcoming the challenges presents in part dissolution of cellulose. At the same conditions, applying by MILT yielded the films without any pores or fibrous profile as can be seen (Fig. 2e,f). An interesting result in this regard was that the fracture surface of Film-2 sample resulted from MILT presented no layer structure compared to the Film-1 sample resulted from the hot-pressing. The results verified that how to combine ILs and microwave field to make uniform films with much higher smoothness, optimal optical and mechanical properties. More importantly, compared to the pure hot-pressing process, the results indicated the much higher dissolving efficiency of cellulose by MILT because of the potentials effects of electromagnetic field on the process of part dissolution on the surface of cellulose fiber matrix. This result was also demonstrated by the previously reported literatures (Fang & Fang, 2011; Li, 2007; Liu, Wang, Cheng, Shui & Zeng, 2005).

The XRD patterns from experiments for the sake of comparison between the original paper, Film-1 and Film-2 samples (Fig. 3). The XRD patterns of the original paper exhibits obvious diffraction peaks at 14.86° and 22.75°, which correspond to the crystalline form of cellulose-I. The Film-2 sample shows the diffraction peaks of cellulose II as indicated by the appearance of a board peak at 2θ = 21.2°, 20.2°. The results indicated that the transformation from cellulose I to cellulose II occurred after the process of MILT. The diffraction spectra were taken using the Segal method (Inoue, Yano, Endo, Sakaki & Sawayama, 2008). The definition of the CrI is

$$C_{rI}(\%) = [(I_{002} - I_{am})/I_{002}] \times 100 \quad (1)$$

in which I_{002} is the intensity of the crystalline peak at about 2θ = 22.5° and I_{am} is the intensity at 2θ = 18.0°. This method is usually applied to cellulose I but not for cellulose II. Here, we used the areas of the crystalline and amorphous regions in the XRD spectra. The Scherrer's equation was used for estimating the crystallite size:

$$D = K\lambda/\beta \cos\theta \quad (2)$$

Where λ is the wavelength of the incident X-ray (0.154056 nm), K is the constant of 0.89, θ is the diffraction angle corresponding to the planes, βis the X-ray crystallite size and D is the full-width at half maximum (FWHM) of the X-ray peak corresponding to the plane (Burton, Ong, Rea & Chan, 2009).The crystallinity index (CrI) of original paper, the Film-1 sample and the Film-2 sample are 73%, 44%, and 26%, respectively. The index of original paper is much higher than that of Film-2 sample, which indicated that MILT process had

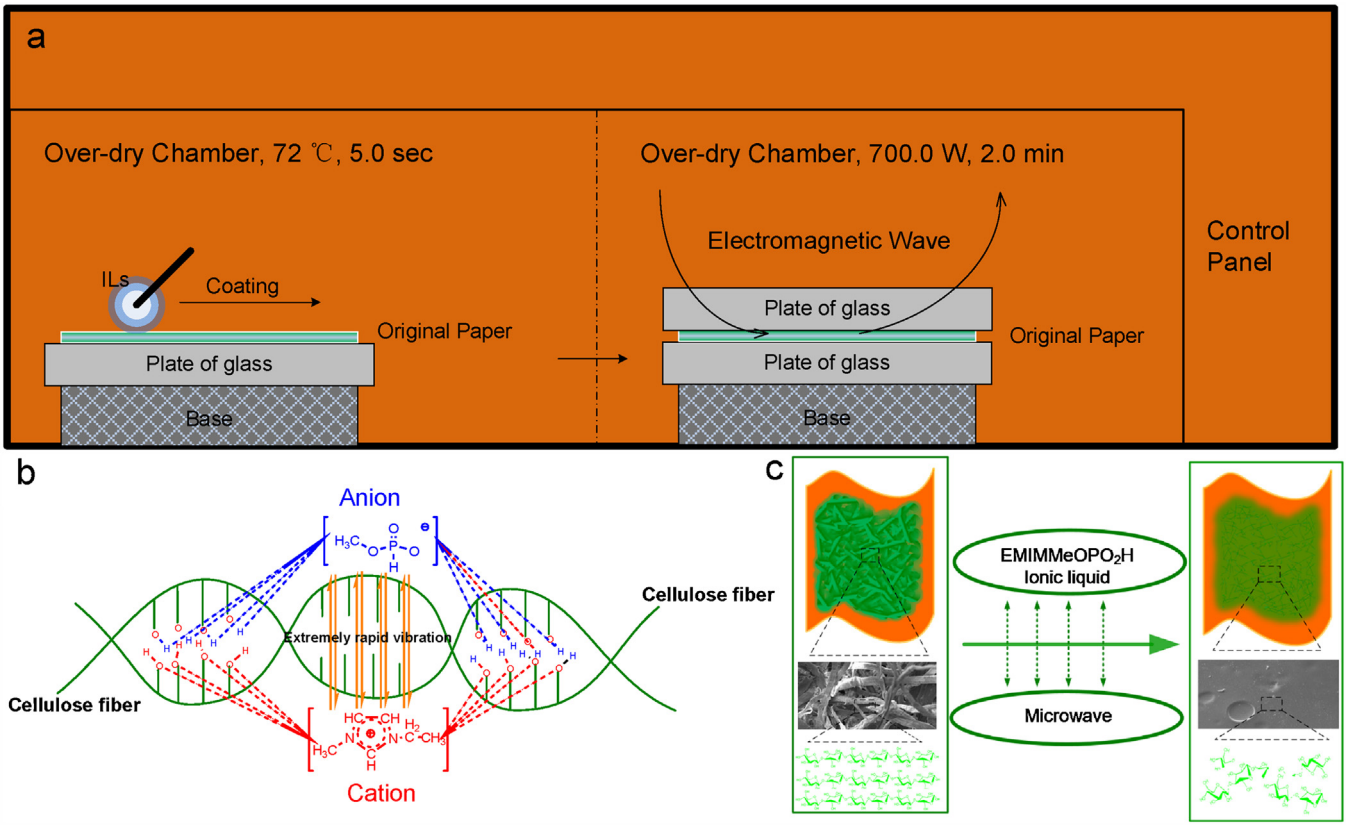

Fig. 1. (a) Scheme of the original paper during the treating process with ILs and microwave assistance; (b)Mechanism of extremely rapid vibration of strong polar EminMeOPO$_2$H ILs during the part dissolution of cellulose; (c)The schematic diagram of treating the original paper with coating ILs and subsequent microwave radiation in domestic microwave oven.

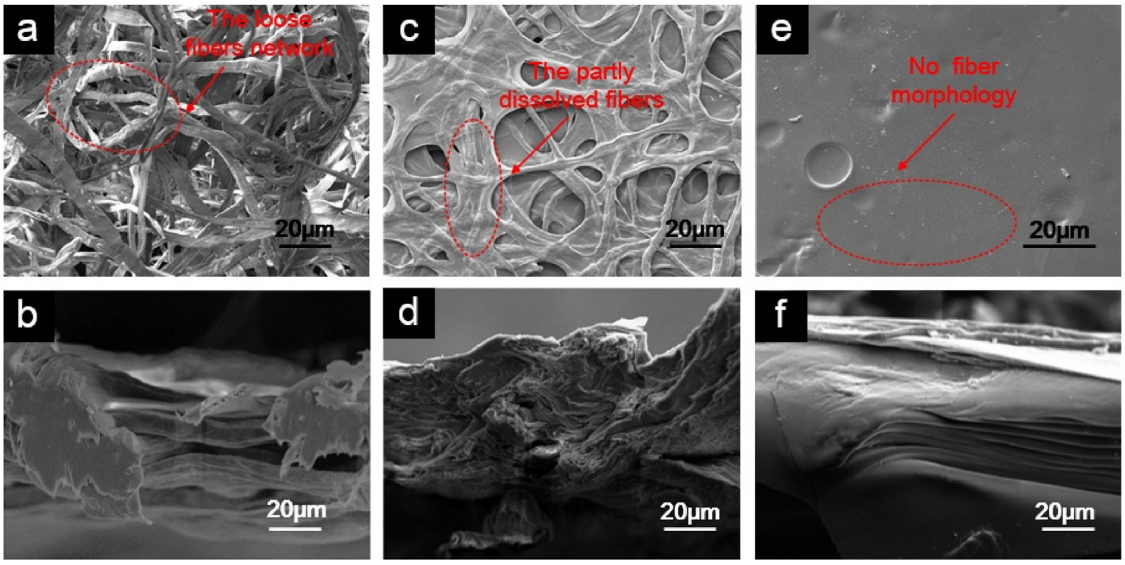

Fig. 2. SEM micrographs of cross-sections: (a) Original paper, (c) Film-1sample and (e) Film-2 sample; SEM micrographs of the fracture surface: (b) Original paper, (d) Film-1 sample and (f) Film-2 sample.

damaged the crystalline structures of cellulose fibers. This result is in agreement with the conclusion of the Sun (However, 2014).

The height AFM image of Film-2 sample where a closed fibrous structure is clearly observed (Fig. 4a).The image indicated that the Film-2 sample displayed smooth, highly uniform and dense surface. The surface RMS (Rq) roughness of the Film-2 sample was around 4.73 nm, which is lesser than the nanopaper (7.7 nm), regenerative cellulose (6.8 nm) and PET (7.0 nm) (Chen et al., 2015; Chinga-Carrasco et al., 2014; Wang et al., 2013). The surface roughness of the Film-2 sample are promising for future flexible transparent

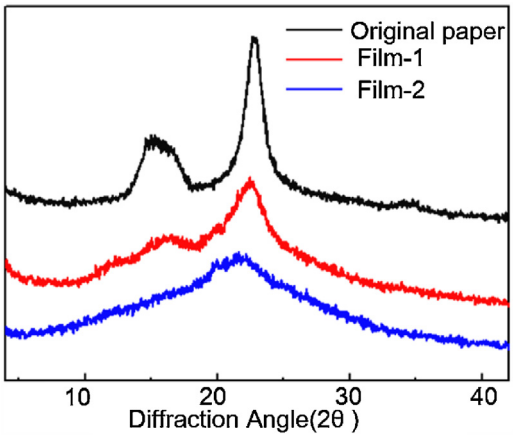

Fig. 3. XRD patterns of Original paper, Film-1 sample and Film-2 sample.

electronic devices. One of the intriguing properties of transparent paper substrates is their high optical transmittance. The optical properties of original paper, Film-1 sample and Film-2 sample were compared with a PET (polyethylene terephthalate) plastic substrate and nanopaper (Fig. 4c, d). The original paper made from long fibers is opaque, but when the air between or/and inner the cellulose fibers was replaced by the regenerated cellulose, the paper becomes transparent. Since the regenerated cellulose has a similar light refraction index ~1.5 with the regular fibers as opposed to air, the paper maybe tuned from opaque to transparent depending on the degree of pores filled with the regenerated cellulose. The transmittance of the Film-2 sample resulted from MILT at 550 nm reaches up to 82%, which is higher than the Film-1 sample resulted from the hot-pressing because more regenerated cellulose fills the pores to decrease the lighting scattering. The presence of high transmittance will be desirable for optoelectronic devices such as thin film solar cells. The results described above also proves the method of MILT is a useful method for fabricating the transparent film.

The mechanical properties of transparent film play an important role for the comprehensive device. To examine the suitability of the cellulose films for industry applications, the mechanical properties of the Original paper, Film-1 sample, and Film-2 sample were determined (Fig. 5a). It can be seen that the Film-2 sample treated with MILT caused the most excellent tensile strength, which was suggested by the highest tensile stress (46.0 MPa). In comparison, although the tensile strength value for the Film-1 sample is lower than the Film-2 sample, it was still higher than the original paper. Therefore, these results also indicated that the green $EminMeOPO_2H$ ILs had the great potential to be applied in the fabricating process of transparent film. And the high mechanical strength also gives Film-2 sample huge potential in roll-to-roll printed electronics. DMAC/LiCl, PEG/NaOH, NaOH/urea are three common solvent for cellulose fiber, and ILs is an emerging solvent. The ratio of maximum stress between the some paper treated with different solvent and original paper to reaction time curves were shown (Fig. 5b). By contrast with the typically similar DMAC/LiCl, PEG/NaOH, NaOH/urea treating technologies, MILT shows its great advantages and potential in fabricating the transparent films. The ratio of maximum stress between the treated paper and original paper of MILT is over tenfold higher than the other treating technologies. More importantly, the reaction time of the MILT is only around 2 min, which is significantly less than other methods. The treating technologies of MILT meets the requirements for transparent film fabrication due to higher efficiency and shorter reaction time.

4. Conclusions

In summary, the transparent films with optimal transmittance, excellent tensile strength and surface smoothness were successfully prepared using the treating technologies of MILT. The film shows much lower surface roughness, much higher transparency and tensile strength than original paper. We have compared the performance of several treating technologies, it was proved that

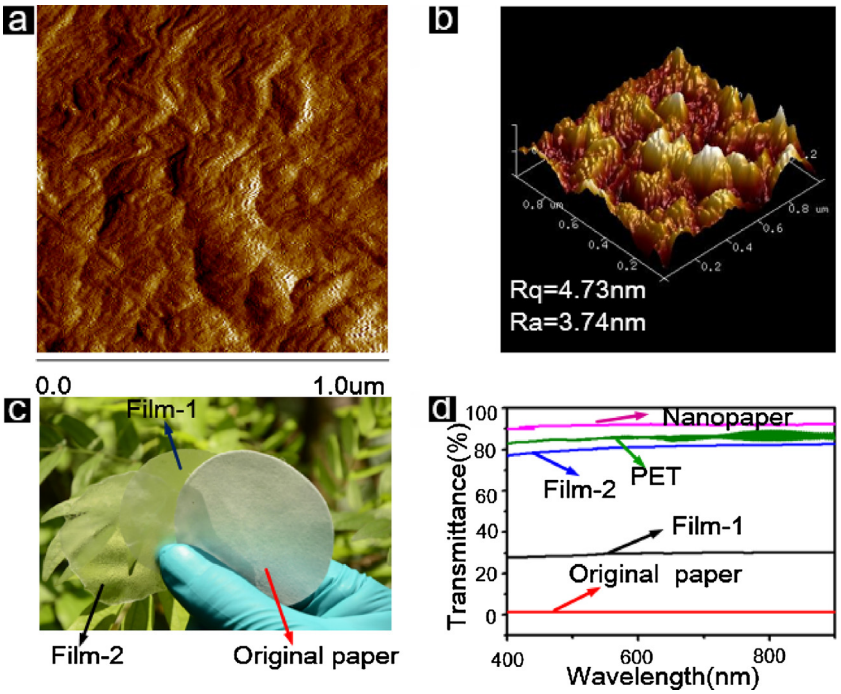

Fig. 4. Surface topography of Film-2 sample was observed by AFM. (a) Two-dimensional scanned amplitude and (b) 3D scanned height image of the transparent paper; (c) Optical transmittance plot for PET, nanopaper, original paper, Film-1 sample and Film-2 sample in the range from 400 to 900 nm; (d)The digital images of original paper, Film-1 sample and Film-2 sample in the left of the plot.

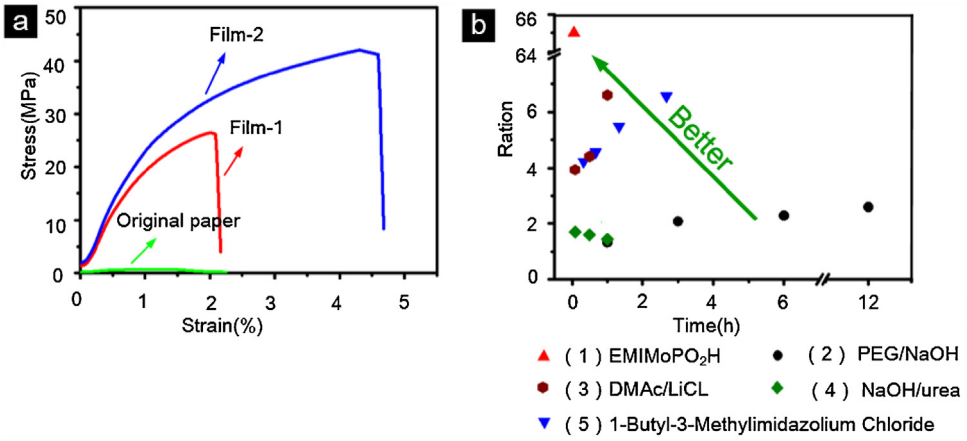

Fig. 5. Stress-strain curves of the Original paper, Film-1 sample, and Film-2 sample, together with the ratio of maximum stress between the paper treated with different solvent and original paper to reaction time curves of some reported paper. PEG/NaOH (Han & Yan, 2010), DMAC/LiCl (Nishino & Arimoto, 2007b), NaOH/urea (Dormanns, Schuermann, Müssig, Duchemin & Staiger, 2015), 1-Butyl-3-Methylimidazolium Chloride (Duchemin, Mathew & Oksman, 2009b).

the treating technologies of MILT has huge advantage in fabricating the transparent films. In particular, the transparent film is totally made by cellulosic resources, which indicated that this film is easily degrades and environmentally friendly. For device applications, these excellent optical, mechanical, and lower surface roughness suggest the great potential of transparent film in next-generation of flexible and transparent electronics and in a broad range of other cost-efficient and practical applications.

Acknowledgments

This work was supported by funding received from Guangzhou science and technology plan project (Grant No. 201704030066), Guangdong Province Youth Science and Technology Innovation Talents (Grant No. 2014TQ01C781),State Key Laboratory of Pulp and Paper Engineering (Grant No. 2016C08) and the Fundamental Research Funds for the Central Universities (Grant No. 2017ZD087) South China University of Technology.

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