POTENTIAL APPLICATIONS OF IONIC LIQUIDS IN WOOD RELATED INDUSTRIES

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The use of ionic liquids (ILs) has provided a new platform for efficient utilization of wood. In this paper, applications of ILs in wood-related industries are reviewed. First, the dissolution of wood in ILs and its application are described. Then the ILs used for wood preservation and improvement of wood anti-electrostatic and fire-proof properties are illustrated. Finally, "green" wood processing with ILs is discussed. Although some basic studies of ILs, such as their economical syntheses and toxicology are eagerly needed and some engineering problems still exist, research for application of ILs in wood-related industries has made great progress in recent years.

Keywords: Application; Ionic liquid; Wood related industries

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INTRODUCTION

Wood is one of the world's most widely-used renewable biomaterials. Apart from its direct use, it can also be converted into numerous useful products, such as paper, cellulosic fibres, chemicals, fuels, *etc*. (Carter 2007). Wood and its derivative products have been closely involved with our lives, and their comprehensive utilization has drawn much attention from researchers and governments. To enhance the performance of wood, extensive research has been carried out on wood processing, treatment, preservation, and its conversion (Carter 2007; Li *et al.* 2004; Pernak *et al.* 2005; Zhu *et al.* 2006; Zhu 2008; Liebert and Heinze 2008). However, environmental problems sometimes can be caused due to the fact that hazardous chemicals are used or formed during its utilization. For example, solutions of sodium hydroxide and sodium sulfide are used, and large amounts of wastewater are formed during the widely used kraft pulping process. Therefore, many efforts are still needed for efficient and environmentally acceptable utilization of this renewable resource (Carter 2007; Zhu *et al.* 2006; Zhu 2008; Liebert and Heinze 2008).

Ionic liquids (ILs) are defined as organic salts that melt below about 100 ^{o}C. They are composed entirely of ions, typically large organic cations and small inorganic anions. Some of the most widely-used ILs are $[R_1R_2R_3IM]X$, $[R_1Py]X$, $[R_1R_2R_3R_4N]X$ and $[R_1R_2R_3R_4P]X$. Their structures are shown in Fig. **1**.

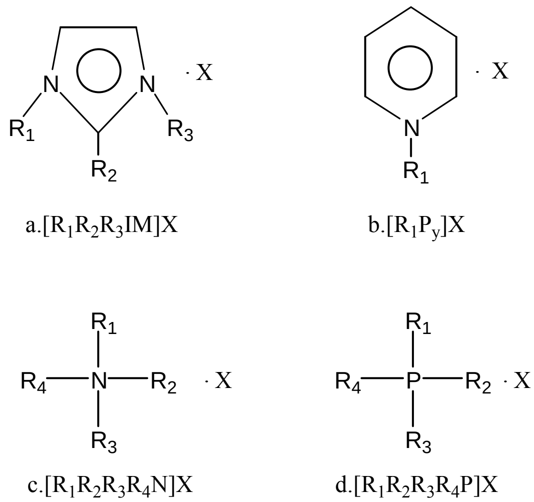

Fig. 1 Structure of some most widely-used ILs
R_1, R_2, R_3, R_4=H, alkyl, etc.
X=Cl^-, Br^-, BF_4^-, PF_4^-, $CF_3SO_3^-$, $CH_3OSO_3^-$, Tf_2N^-, etc.

Interest in ILs has grown in recent years because they have such excellent properties, such as chemical and thermal stability, inflammability, high conductivity, low melting point, and undetectable vapor pressure. Compared with conventional molecular solvents, they emit no volatile organic compounds to pollute the atmosphere, and their properties can be changed to match the end use application by varying their anions and cations (Rogers *et al.* 2003; Li 2005; Liebert and Heinze 2008). Although recent research on their toxicity and biodegradability indicate that some of them are not as eco-friendly as expected (Wells and Coombe 2006; Ke *et al.* 2007), the ILs are still considered as a potential alternative for green chemistry (Rogers *et al*. 2003; Ke *et al*. 2007). To date they have been widely used in chemistry and its related areas, and there are many books and articles which introduce ILs and their applications (Rogers *et al.* 2003; Li 2005; Parvulescu and Hardacre 2007; Martins *et al*. 2008). Relatively speaking, research on applications of ILs in wood-related industries is still scarce, and little published literature can be found. As a potential alternative to decrease environmental pollution in wood utilization by using ILs, more research efforts should be made in this area. In this paper, studies related to applications of ILs in wood-related industries have been reviewed. Also, the challenges and future prospects of applications of ILs in wood-related industries are discussed.

APPLICATIONS

Dissolution of Wood in ILs and Its Application

The main chemical components in wood are cellulose, hemicellulose, and lignin. Cellulose is a homo-polymer of β-1,4 –glucose units with a very highly regular H-bonded network between its layers, especially in the case of crystalline cellulose. Hemicellulose is a carbohydrate hetero-polymer with different monomers. Lignin is an irregular, cross-linked polymer network, which is composed of randomly cross-linked phenylpropanoid units. The three-dimensional lignin network that binds the wood components together makes it practically impossible to dissolve wood in its native form in conventional solvents. The insolubility of wood in common solvents has severely hampered the development of efficient methods for its utilization and analysis (Kilpelainen *et al.* 2007; Zhu 2008). Dissolution of wood in ILs have the potential to provide a new platform to address such challenges (Kilpelainen *et al.* 2007; Argyropoulos 2008a; Zhu 2008).

In recent years there have been lots of reports on dissolution of cellulose in ILs and its applications (Zhu *et al.* 2006; Liebert and Heinze 2008), but dissolution of wood in ILs is far more complicated due to wood's complex structure from the three-dimensional lignin network (Kilpelainen *et al.* 2007). To our knowledge, the ILs used to dissolve wood are limited in the imidazole-based ones, that is $[R_1R_2R_3IM]X$, whose structure has been shown in Fig. **1**. Table **1** summarizes the ILs used for wood dissolution according to various published sources.

Table 1 The ILs Used for Wood Dissolution According to Published Acrticles

Reference cited	ILs used for wood dissolution
Myllymäki and Akasela (2005)	1-butyl-3-methylimidazolium chloride
Xie and Shi (2006)	3,3 prime -ethane-1,2-diylbis(1-methyl- 1H-imidazol-3-ium) dichloride and 3,3 prime -ethane-1,2-diylbis(1-methyl- 1H-imidazol-3-ium) dichloroaluminate
Fort *et al.* (2007)	1-butyl-3-methylimidazolium chloride
Kilpelainen *et al.* (2007)	1-butyl-3-methylimidazolium chloride, 1-allyl-3-methylimidazolium chloride, 1-benzyl-3-methylimidazolium chloride, etc

Kilpelainen *et al.* (2007) have given a detailed discussion on the mechanism and affecting factors for dissolution of wood in ILs based on the Abraham solvation equation, which had been successfully used by Anderson *et al* (2002) to characterize the polymer solvation capacity of ILs. They pointed out that dissolution of wood in ILs is not only needed to disrupt the H-binding interaction present crystalline cellulose in wood but also to interact and solvate the aromatic character of lignin. The presence of water and its relative amount in wood will also have a great effect on its dissolution. Their experiments have confirmed this mechanism of wood dissolution in ILs and proven that wood can be readily dissolved in ILs. Dissolution of wood in ILs has opened a new avenue in wood research and its efficient utilization in the following areas:

1) Extraction of cellulose from wood wastes

The kraft pulping process is by far the most widely used method to extract cellulose from wood wastes. The kraft process involves the semi-chemical degradation of the lignin/hemicellulose matrix in wood by treatment with solutions of sodium hydroxide and sodium sulfide at high temperatures and pressures. In principle, the use of these toxic and chemicals and intensive processing conditions poses hazards in terms of air and water pollution. Research has indicated that the use of ILs for pulping can avoid the use of toxic and hazardous chemicals, and the process can be carried out under mild conditions (Myllymäki and Akasela 2005; Upfal *et al.* 2005; Fort *et al.* 2007; D'andola *et al.* 2008; Edye and Doherty 2008). Myllymäki and Akasela (2005) first claimed in their patent that they successfully delignified wood wastes for a pulping process by dissolution of wood in 1-butyl-3-methylimidazolium chloride (BMIMCl) and recovered the cellulosic fiber from the BMIMCl solution, but they didn't describe how the cellulosic fiber was recovered in detail. Fort *et al.* (2007) made further study on dissolution of wood in ILs and found that wood chips could be partially dissolved in BMIMCl. They successfully recovered cellulosic fiber from the BMIMCl solution in fair yields by the addition of a variety of precipitating solvents and found that the recovered cellulosic fiber was free of lignin and hemicellulose. Unfortunately, how to efficiently use the remaining hemicellulose and lignin in the BMIMCl solution and recover the BMIMCl is still an aggravating problem for its industrial use.

2) Pretreatment of wood wastes for the production of chemical feed-stocks and biofuels

Pretreatment is a key procedure in the production of chemical feed-stocks and biofuels from wood wastes through fermentation. Because the micro-structure of cellulose, especially its crystalline degree can be manipulated during its regeneration from its ILs solution, pretreatment of wood wastes using ILs can greatly decrease the crystalline degree of the restructured cellulose, which will benefit the subsequent enzymatic hydrolysis process for the production of chemical feed-stocks and fuels through fermentation. Some studies (Liu and Chen 2006; Zhu *et al.* 2006; Dadi *et al.* 2006; Stegmann and Maase 2007; Argyropoulos 2008b; Balensiefer 2008; Balensiefer *et al.* 2008) have indicated that pretreatment of wood wastes using ILs can greatly increase the enzymatic hydrolysis rate and yield. Although use of ILs is a potential alternative pretreatment approach in production of chemical feed-stocks and biofuels, there is still lack of concrete evidence that this process is superior to the traditional pretreatment methods, for example, the steam explosion pretreatment.

3) Preparation of wood derivatives and composites

Preparation of wood derivatives and composites can greatly extend the scope of wood applications. Dissolution of wood in ILs makes it possible to prepare wood derivatives under homogeneous and mild conditions (Xie *et al.* 2007, 2008; Argyropoulos and Xie 2008). Xie *et al.* (2007, 2008) have done extensive research in this area. They successfully prepared the highly substituted wood derivatives with high yield under mild conditions (2 h, 70 °C) by reacting wood dissolved in ionic liquids with acetyl chloride, benzoyl chloride, and acetic anhydride in the presence of pyridine. Their further studies indicated that these highly substituted wood derivatives can be used as fillers for thermoplastics and have good interfacial miscibility with synthetic polymers, such as poly(styrene) and poly(propylene). Apart from preparation of wood thermoplastic composites, the homogeneous derivation of wood in ILs solution also makes analysis of wood components and its structure much convenient, as we will discuss later.

4) Analysis of wood components and its structure

Analysis of wood components and its structure is an indispensable part in wood chemistry research. However, the traditional analytical procedures are often technical and laborious because wood can't dissolve in conventional solvents. Moreover, its native structure is always been destroyed for fractionating its various components in the course of analysis. Dissolution of wood in ILs has offered a variety of new possibilities for convenient analysis of wood components and its native structure. Fort *et al.* (2007) directly analyzed the cellulosic material and lignin content in the wood by means of conventional ^{13}C NMR techniques via dissolution of wood in the BMIMCl. Kilpelainen *et al.* (2007) put forward a convenient approach for its native structural and macromolecular characterization in the wood by its direct derivation in ILs solution instead of the prior isolation of its individual components.

Wood Preservatives

Wood is a widely-used, economical, and renewable building resource. However, untreated wood is subject to attack by insects and micro-organisms, for example, decay, stain, mould fungi, and bacteria. To ensure long-term structural performance, wood must be protected from biotic wood degradation factors. Preserved wood can be defined as lumber or plywood that has been treated with wood preservatives to protect it from termites and fungal decay. Many preservatives have been used in wood industry, and still many efforts have been made to develop new preservatives. The reason is that most of the wood preservatives in current use are chemicals that might be harmful to human health or are not eco-friendly. ILs have shown great potential in becoming the eco-friendly, as well as free of both arsenic and pentacholorophenol, and yet they also show effectiveness as wood preservatives. Pernak *et al.* (2004, 2005, 2006) have done extensive research on this subject. They found some ILs with a nitrate counterion, such as 3-alkoxymethyl-1-methyl-imidazolium tetrafluoroborates and hexafluoro-phosphates, didecylmethyl-ammonium and benzalkonium nitrates, exhibited good fungicidal activity and could be potential candidates of wood preservatives. Stasiewics *et al.* (2008) also found such pyridinium-based ILs as 1-alkoxymethyl-3- dimethylaminopyridinium chlorides, 1-alkoxymethyl-4-dimethyl-aminopyridinium chlorides, 1-alkoxymethyl-3-dimethylamino-pyridinium acesulfamates, and 1-alkoxymethyl-4-dimethylaminopyridinium acesulfamates could be effective components of potential wood preservatives.

Wood Electrostatic Control and Fire Retardant Treatment

Wood products having anti-electrostatic or fire retardant properties have wide applications in many fields (Carter 2007). However, wood itself is flammable and it does not have anti-electrostatic ability. As we know, ILs are inflammable, and some of them are excellent conductors for electric current. These properties provide the possibilities for use of ILs in wood electrostatic control and fire retardant treatment. Li *et al.* (2004) reported that some ILs such as 1-butyl-3-methylimidazolium tetrafluoroborate, 1-butyl-3-methylimidazolium hexafluorophosphate, 1-butyl-3-methylimidazolium chloride, 1-ethyl-3-methylimidazolium tetrafluoroborate, and 1-ethyl-3-methylimidazolium hexafluorophosphate were effective anti-electrostatic agents for pine and maple. The University of Alabama carried out studies on use of ILs as wood fire retardants and found some ILs can be used to take the place of the currently widely-used phosphorus fire retardant compounds (Rogers *et al*. 2003). Similar research also has been conducted in our laboratory, and the preliminary results are encouraging (Zhu *et al*. 2007).

Green Wood Processing

During wood processing, such as production of wood composites, lumber, and paper, some unwanted by-products, for example, volatile organic compounds and hazardous pollutants, are formed. It is very difficult to deal with these by-products economically using current pollution treatment technologies because of their low concentrations. Milota and Li (2004) have put forward a new method to remove these by-products from wood processing exhaust gases by absorbing them into ILs (as shown in Fig. 2).

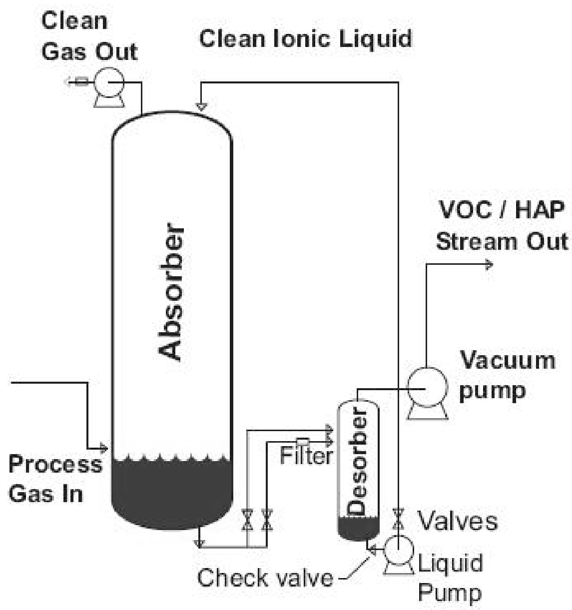

Fig. 2. Schematic process for application of ILs in treatment of wood processing exhaust gases

Milota and his group carried out extensive research on this process (Wang *et al.* 2007; Milota *et al.* 2007, 2008). In order to select an appropriate IL as the absorbent, they measured the Henry's Law constants for the main components of wood processing exhaust gases in four imidazolium-based and eight phosphonium-based ILs. The stability of the imidazolium-based ILs proved problematic. The constants for the phosphonium-based ILs were low, indicating that the ILs tested might be very good absorbents. They also conducted lab and pilot scale tests for this process using IL tetradecy (trihexyl) phosphonium dicyanamide as an absorbent. It is reported that this approach could significantly decrease the energy consumption and concentrate the by-products. The concentrated by-products could be burned for fuel or distilled to useful products, and the IL used to absorb the by-products could be easily recovered and reused. As a newsworthy event from the Oregon State University of April 2006, Milota *et al.* carried out the first mill trial of an IL absorption system designed to remove pollutants from wood dryer and press exhaust. During the three-day trial conducted on a Weyerhaeuser veneer dryer in Eugene, the system removed methanol and formaldehyde from dryer exhaust well enough to meet the Maximum Achievable Control Technology (MACT) standards that will be imposed in 2008.

FUTURE PROSPECTS

Use of ILs has provided a new platform for comprehensive utilization of wood, although many problems need to be further addressed. First, dissolution of wood in ILs has opened a new avenue for wood chemistry research and to improve the efficient utilization of wood. Second, the ILs can be used as wood preservatives or to improve wood's anti-electrostatic and fire-proof properties. This will extend the range of wood applications and reduce the use of some dangerous chemicals. Finally, use of ILs is also a competitive alternative exhaust treatment technology in the wood processing, which can reduce the energy consumption and waste of resources.

However, application of ILs in wood- related industry in industrial scale at present still faces some challenges. The main problem is the cost of ILs, which has hampered use of ILs at a commercial scale. Therefore, economical synthesis of ILs and their highly efficient recovery technologies will play vital roles in their industrial applications. Another problem is that basic studies, dealing with such as the biodegradability, toxicology, and thermodynamic data of ILs are still scarce.

There are also lots of engineering problems that need to be solved. The most challenging engineering problem is the high viscosity of ILs, which is expected to cause severe mass and heat transfer problems during their industrial use. Some measures have been taken to reduce their viscosity, such as development of low viscous ILs (Fukaya *et al.* 2006) and use of mixtures of ILs (Kilpelainen *et al.* 2007), but these measures can only partially resolve this problem. Although these challenges still exist, great progress has been made in research for application of ILs in wood-related industries in recent years. As mentioned earlier, some applications have been investigated at a pilot scale. Although many problems associated with application of ILs in wood-related industries need to be solved, it is quite clear that in the near future these applications will be implemented at a full industrial scale and society can be expected to benefit greatly from these applications.

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