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OBTAINING OF ECOLOGICAL COMPOSITES FROM WOOD WASTE USING IONIC LIQUIDS

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Abstract: *The paper presents an eco-friendly method for obtaining wood composites, as an alternative to traditional fibreboard composite obtaining processes. Our proposed method comprises two steps: namely, dissolving wood in ionic liquids, followed by precipitation by water addition. The precipitated cellulose and lignin act as a natural binder for the wood particles thus avoiding the use of toxic organic resins as adhesives. The obtained composites present good water stability and compressive mechanical strength.*

Key words: *wood composite, 1-butyl-3-methylimidazolium chloride, compression strength, water adsorption, dimensional stability.*

1. Introduction

Wood is a fibrous material that represents both a transport medium for nutrients, as well as in conferring mechanical resistance and leaf support in some superior plant species. It is mainly comprised of cellulosic fibres that are embedded in a lignin matrix, which confers the wood water resistance and remarkable compression strength [13-15], [20].

Wood has been extensively used, either as fuel or as a construction material. The most commonly used source of wood consists in the stems (secondary xylem) of both coniferous and dicotyledonous trees [20].

However, excessive deforestation causes extinction of benign flora and fauna, changes to climatic conditions, and desertification Also, it is a major driver of global warming, responsible for up to 20% of global greenhouse gas emissions [7], [19].

During the manufacturing process of furniture, lumber and paper, the wood industry generates enormous quantities of wood waste. Nearly 50% of a tree is used to manufacture a product; the rest represents an unused resource [20], which could be recycled and embedded in new composite materials, such as *fibreboard*. Fibreboards, such as OSB (Oriented Strand Board) and MDF (Medium Density Fibreboard) are cheaper than pieces manufactured from massive wood, the manufacturing process is simpler and the end product has mechanical properties similar to lumber [3], [15], [20].

Albeit the process is very attractive economic-wise, the process uses non-ecologic and carcinogenic organic compounds as adhesives for the wood particles, among which formaldehyde-based resins present the biggest concern [16]. Also, the use of organic solvents for adhesives,

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such as toluene, acetone, or ethanol-acetone mixtures, is economically inefficient and posses an environmental risk, also by their capacity to transform their selves in the atmosphere, under the influence of UV radiation and the catalytic action of the nitrogen oxides, in more aggressive species with carcinogenic potential. As consequence, the replacement of the traditional volatile solvents from wood industry with less toxic and volatile solvents became an important aim for the scientists working both in material science and in environmental protection [8-12], [15], [18].

These issues can be overcome by using a new class of organic solvents for wood, such as ionic liquids.

Ionic liquids [IL] are a group of new organic salts that exist as liquids at a relatively low temperature (< 100 ^{0}C) [1], [2], [8], [11-18]. Ionic liquids have a series of proprieties like thermal stability, good electric conductivity, are fireproof, and have negligible vapour pressures. They can dissolve a great range of organic compounds [3-6] among which lignocellulosics have been of greatest interest [9], [10], [17], [18], [21].

The aim of this paper is to obtain and characterize wood composites, obtained by sawdust dissolution in ionic liquids at 100 ^{0}C, in an amount higher than that corresponding to its solubility limit, followed by the precipitation by addition of water. The precipitated cellulose and lignin act as a natural adhesive for the undissolved wood particles, thus eliminating the use of formaldehyde-based resins.

2. Materials and Methods

2.1. Materials

1-buthyl-3-methylimidazolium chloride ionic liquid (BMIMCl) (Figure 1), has been purchased from IoLiTec Ionic Liquids Technologies GmbH, Germany, and it is a white crystalline compound, with 99.5% purity.

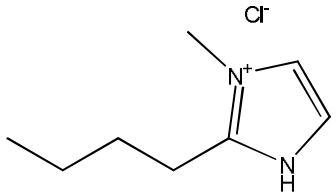

Fig.1. *Structure of the used ionic liquid*

As wood source, spruce (*Picea* sp.) flour (FM) (finely ground sawdust,) has been used.

2.2. Obtaining of the composites

The wood flour was mixed with ionic liquids in an appropriate amount, corresponding to 40% wt, reported to the ionic liquid at 100 ^{0}C, under continuous mechanical stirring.

The obtained raw material has been allowed to cool at room temperature, after which determined amounts have been added in a circular mould (10 mm in diameter) and pressed with a force of 50 atmospheres for 5 minutes, by the means of a hydraulic press.

The compressed circular-shaped wood-ionic liquid mixture was immersed in distilled water at room temperature (25 ^{o}C), in order to eliminate the IL from the pellet.

The distilled water has been replaced periodically, in order to allow a constant concentration gradient and to eliminate the IL in a more efficient manner. The complete removal of the ionic liquid has been monitored by electrical conductivity measurements of the washing water. Usually, this process takes between 72 and 100 hours.

After the complete removal of the ionic liquid, and the precipitation of wood components, the final step has been the drying of the pellets at 105 ^{o}C for five hours, until the materials reached constant weight. For further tests they have been reconditioned for a week at 55% relative humidity and 22 ^{0}C, until they reached

constant weight. For comparison, neat wood flour (without ionic liquid) has been pressed and conditioned in the same manner as the composite.

The photographical image of the reference and of the obtained composite has been illustrated in Figure 2.

Fig. 2. *Reference pressed wood flour (left) and obtained wood composite (right)*

2.3. Moisture absorption experiments and dimensional change monitoring

The conditioned wood samples were immersed in closed glass recipients with a relative humidity of 86%, ensured by a supersaturated K_2SO_4 solution, and their mass and dimensions (diameter and thickness) were determined at precise time intervals until a constant value has been reached (10 days).

Also, the same worksteps have been performed by immersing the composite and the reference in 50 mL of distilled water. The equilibrium moisture content of the samples at time t has been calculated by using Eq. (1) [19]:

$$EMC = \frac{m_t}{m_{d\,ry}} \cdot 100, \qquad (1)$$

where: m_t - the mass of the pellet at time t; m_{dry} - initial mass of the conditioned pellet.

2.4. Mechanical compression strength determination

Compression tests of the conditioned wood composite and blank has been performed on a Zwick Z020 universal testing device (Zwick Roell GmbH), by compressing the pellet to 80% of its original thickness. The stress-compressive strain curves were obtained and processed with the instrument's software.

All presented values are the mean value of 3 parallel measurements.

3. Results and Discussion

The results regarding the water sorption kinetic of the obtained wood composite and reference, in contact with the 86% relative humidity atmosphere is presented in Figure 3:

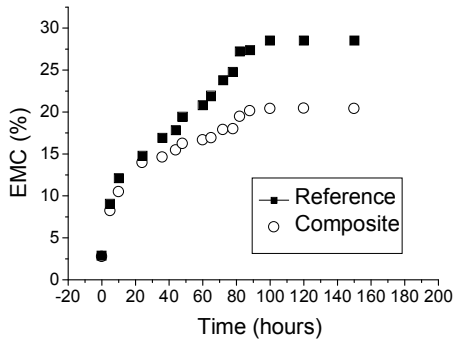

Fig. 3. *Water sorption kinetic of the obtained composite and reference in the atmosphere with 86% RH*

The results regarding the water sorption kinetic of the obtained wood composite is presented in Figure 4.

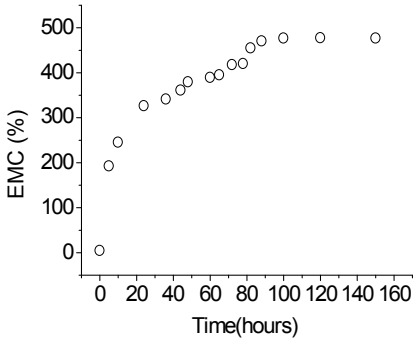

Fig. 4. *Water sorption kinetic of the obtained composite*

As it can be seen from Figure 3, the equilibrium moisture content in the case of the compressed wood flour reference is 30% bigger than in the case of the composite. This fact may be due to the precipitated lignocellulose which acts as a binding agent for the sawdust particles, also reducing the water absorption compared to the pressed sawdust without the IL.

This fact can be also seen in the case of water sorption experiments, where the compressed wood flour completely disintegrates after the first 30 seconds of immersion in water, while the obtained composite preserves its dimensional stability better.

Figure 5 depicts the dimensional change of the composite and reference, at equilibrium water absorption, when in contact with the 86% relative humidity atmosphere.

As it can be remarked, by absorbing water from the atmosphere, swelling of the composite and reference occurred, which also owes for dimensional changes, the more water absorbed at equilibrium, the more pronounced of diameter and thickness variation.

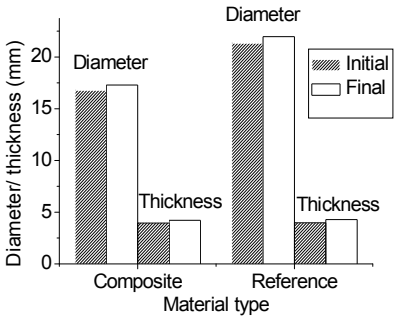

Fig. 5. *Dimensional changes of composite and reference in 86% RH atmosphere*

The surface of the pellet as well as the water in which the composites have been immersed during the water absorption test, were periodically checked for fungi. No occurrence of fungi has been observed by means of visual analysis of the sample and storing water.

Regarding the mechanical performance of the composite in initial (dry) state, it can be noticed from Figure 6 that no significant difference in compression strength at 80% deformation occurs.

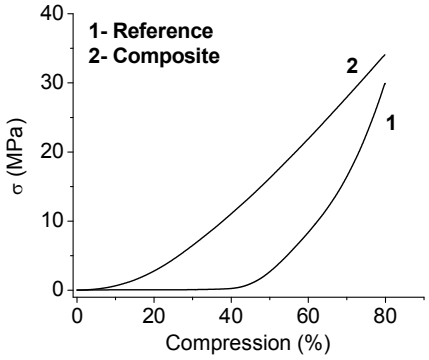

Fig. 6. *Stress - compression strain of wood composite and reference in dry state*

However, the reference probe (pressed wood flour) is less resistant to compression. Being compressed to nearly 60% of their original thickness these pellets counter the force with minimum resistance, fact that can be seen in the negligible variation of the compression stress.

Regarding the pellets obtained with the BMIMCl ionic liquid, one can see that they are more resistant to compression in the first stage of compression (up until 60% of their initial thickness), probably due to the fact that they are more compact, as a result of the precipitated lignocellulose that binds the wood particles together, thus the composite being able to oppose more resistance to compression.

Regarding the mechanical properties of the composite maintained in an 86% relative humidity (Figure 7), it can be observed that the compression strength decreases with only 15%, in comparison to the reference, which compression strength at 80% deformation decreases with more than 50% percent. This fact is most likely caused by a plastifying effect of water on the cellulose matrix.

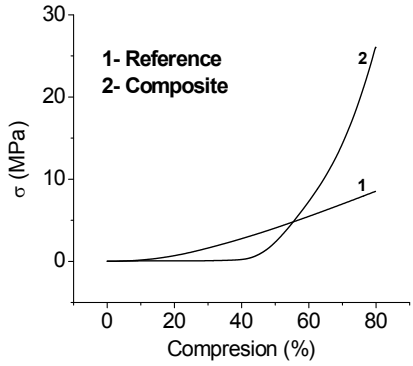

Fig. 7. *Stress- compression strain of wood composite and reference in contact with 86% RH atmosphere*

Also, water may contribute to the relative displacement of the wood particles in the reference, where no interparticle binding occurs.

4. Conclusions

A new composite material has been obtained by using a waste resulted from wood industry, namely spruce wood flour and an ecologic green solvent for lignocellulose, namely 1-butyl-3-methylimidazolium chloride.

The obtained composite maintains greater compression strength and dimensional stability when in contact with media of different humidity, than the reference.

Taking into account that the studied ionic liquid is non-volatile, our proposed method is more ecological than other traditional fiberboard obtaining processes, which are energetically inefficient and contribute to the pollution of the atmosphere.

Acknowledgements

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