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Palm rachis microfibrillated cellulose and oxidized-microfibrillated cellulose for improving paper sheets properties of unbeaten softwood and bagasse pulps

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ABSTRACT

Bleached palm rachis pulp, pretreated with xylanase enzymes, was used for isolation of microfibrillated cellulose (MFC) and TEMPO-oxidized MFC (TMFC) by ultrafine grinding. The isolated MFC and TMFC were used at ratios from 2.5% to 20% for improving strength properties of paper sheets prepared from unbeaten softwood and bagasse pulps. The retention of microfibrillated cellulose in paper sheets was also estimated. The results showed that MFC or TMFC brought about an increase in density, wet and dry tensile strength, tear resistance, and a decrease in air permeability of paper sheets prepared from unbeaten softwood or bagasse. However, usual beating of softwood fibers was much more effective in improving strength properties of softwood paper sheets than addition of microfibrillated cellulose. On the other hand, the improvement in strength properties of bagasse paper sheets as a result of MFC or TMFC addition was generally higher than that resulted from beating of bagasse pulp. Use of TMFC with unbeaten softwood or bagasse fibers resulted generally in better improvement in tensile strength (wet and dry) than in case of using MFC.

1. Introduction

Microfibrillated cellulose (MFC) isolated from lignocellulosic materials is a promising natural nano-structured polymer to be used for different industrial and biomedical applications (Siro and Planket, 2010). Nanopaper sheets prepared from MFC are characterized by high wet and dry tensile strength, transparency, flexibility, oxygen barrier properties, and low water absorption (Abdul Khalil et al., 2014, Aulin et al., 2010).

Different additives are used in paper making to improve the properties of produced paper sheets according to the end use. The most commonly used additives are strength promoters to improve strength properties of paper sheets in wet and dry conditions. The strength properties of paper are controlled by the fiber strength, the bonding degree of the fiber network and the strength of the bonds. Without bonding, no fiber network is formed.

In addition to the use of additives, an important process in paper making to improve the strength properties of produced paper sheets is what is called refining or beating of fibers. Beating is known to improve the flexibility and the bonding ability of the fibers due to fibrillation of the surface of the fibers. Beating also modifies other properties, such as sheet formation, absorbency, porosity, or optical properties (Page, 1989; Eklund and Lindsröm, 1991; Molin and Daniel, 2004).

There is a recent interest in using cellulosic nanomaterials, namely microfibrillated cellulose, in paper making. The physical properties of MFC differ from those of a traditional cellulosic pulp consisting of macroscopic fibers. In this context, microfibrillated cellulose was investigated to improve the strength of paper sheets made from different kinds of long fibers, i.e., softwood (Ahola et al., 2008; Eriksen et al., 2008; Taipale et al., 2010; Joseleau et al., 2012; Su et al., 2014), short fibers, i.e., hardwood (González et al., 2014, 2013, 2012; Madani et al., 2011), and agricultural residues (Hassan et al., 2011; Petroudy et al., 2014). However, for the best of our knowledge, little studies have been conducted so far to compare the improvement in properties of paper sheets as a result of beating the fibers before paper sheets making with the improvement

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resulted from adding microfibrillated cellulose to unbeaten fibers. For example, Sehaqui et al. (2013) studied mixing 10% microfibrillated cellulose with bleached softwood pulp fibers (90%) subjected to different number of beating revolutions. Effects from xyloglucan hemicellulose addition were also studied. Microfibrillated cellulose significantly enhanced strength of the paper hand sheets in the dry and wet states. Addition of 10% microfibrillated cellulose to wood fibers beaten to 1000 revolutions resulted in paper sheets with tensile strength comparable to that obtained after beating the softwood fibers to 4000 beating revolutions. However, fully beaten softwood pulp at 4000 beating revolution showed no improvement in tensile strength in case of adding 10% of microfibrillated cellulose.

González et al. (2012) used TEMPO-oxidized nanofibrillated cellulose for improving properties of eucalyptus (hardwood) paper sheets made from unbeaten or slightly beaten fibers, and compared the improvement occurred to that resulted from beating of the fibers. Up to 9% (based on weight of paper sheets) of the nanofibers were used. The results showed improvement in strength properties of paper sheets as a result of addition the nanofibers to pulp fibers but generally, strength properties of paper sheets were higher in case of the fully beaten pulp than those to which the nanofibers were added.

Su et al. (2013) studied the use of microfibrillated cellulose with eucalyptus hardwood fibers to produce high strength and low gas permeability cellulosic nanocomposites; up to 75 wt.% of microfibrillated cellulose was used in the nanocomposites. The effect of blending microfibrillated cellulose with hardwood fibers was compared to the direct refining of the fibers. The addition of microfibrillated cellulose resulted in better improvement in wet tensile strength and lower air permeability of paper sheets than in case of using refined fibers. However, use of refined fibers was more beneficial than adding microfibrillated cellulose, especially at high ratios of microfibrillated cellulose (≥25 wt.%), regarding drainage and sheet formation uniformity.

In a previous work, microbrillated cellulose was isolated from date palm fruit stalks using enzyme-assisted ultrafine grinding (Hassan et al., 2014). Date palm (*Phoenix dactylifera*) is abundant in several areas of the world with about 105 million date palms are currently being grown (Agoudjil et al., 2011). Due to their high content of cellulose and fiber dimensions that are similar to hardwood fibers, the residues left from date palm cultivation and fruit harvesting such as rachis, leaves, and fruit stalks have been studied for paper making (El-Morsy et al., 1981; Khristova et al., 2005; Khiari et al., 2011; Ghosh and Nag, 2010, 2009). Recently, date palm rachis has found interest for preparation of cellulosic nanomaterials, namely cellulose nanofibers and cellulose nanocrystals (Boufi et al., 2014; Bendahou et al., 2010).

In the current work, the improvement in paper sheets properties as a result of adding different ratios of microfibrillated cellulose isolated from date palm fruit stalks using enzyme-assisted ultrafine grinding was compared to the effect of usually-practiced beating of long fibers (softwood fibers) and short fibers (bagasse fibers). Two types of microfibrillated cellulose were used: oxidized microfibrillated cellulose and non-oxidized microfibrillated cellulose.

2. Experimental

2.1. Materials

Bleached kraft bagasse pulp was kindly supplied by Qena Company for Pulp and Paper, Qena, Egypt. Average fiber length and width (as determined by *MorFi* analyzer, TECHPAP LB 01 Morfi equipment) of bagasse fibers were about 1.05 mm and 29.6 μm, respectively. Degree of freeness (°SR) of bagasse pulp was 23. The wood pulp was kindly delivered by Domsjö (Sweden) and corresponded to a mix between Spruce and Pinus (60% and 40%, respectively). Average fiber length and width of wood fibers were about 2.09 mm and 31.46 μm, respectively. °SR of wood pulp was 18. Both of bagasse and wood pulps were received as dried sheets.

Palm rachis was obtained from local field in Giza, Egypt, washed to remove dirt, and chopped in 2–3 cm-long pieces in a mill. Bleached palm rachis pulp was prepared as described before (Hassan et al., 2014). Briefly, palm rachis was treated using 15% NaOH at 150 °C for 3 h. The produced pulp was bleached using sodium chlorite/acetic acid mixture at 80 °C for 1 h (Wise et al., 1946). The chemical composition of bleached pulp was: α-cellulose 71.5%, pentosans 18.4%, DP 1264, and ash 0.64% as determined using standard methods (Browning, 1967).

Analytical grade sodium citrate and citric acid were used for preparation of citrate buffer (pH 5.3). 2,2,6,6-tetramethyl-1-piperidine oxoammonium salt (TEMPO), sodium hypochlorite and sodium bromide were reagent grade chemicals and used as received.

2.2. Enzymatic pretreatment of bleached palm rachis pulp

Enzymatic pretreatment of palm rachis pulp with xylanase enzymes was carried out to facilitate isolation of microfibrillated cellulose from the fibers as follows (Hassan et al., 2014): twenty grams of bleached bagasse pulp were treated with xylanase enzymes in citrate buffer (PH 5.3) in 500 ml conical flask at 10% consistency. The concentration of xylanase enzymes was 60 IU g^{-1}. The reaction mixture was kept under shaking condition (200 rpm) at 50 °C for 4 h. At the end of reaction the pulp was filtered and washed thoroughly with distilled water.

2.3. TEMPO oxidation of bleached palm rachis

The method used was based on that of Saito et al. (2007). Palm rachis pulp (3 g) was dispersed in distilled water (400 ml) with TEMPO (0.048 g, 0.3 mmol) and sodium bromide (0.48 g, 4.8 mmol). Then 30 ml of sodium hypochlorite solution was then added with stirring and the pH was adjusted to 10. At the end of reaction the pH is adjusted to 7 and the product was centrifuged at 5000 rpm. The product was further purified by repeated adding water, dispersion, and centrifugation. Finally the product was purified by dialysis for 1 week against de-onized water with 3500 MWCO Spectra/Por dialysis tubing.

2.4. Preparation of microfibrillated cellulose

The pulp was first disintegrated by a IKA high-shear mixer (T-T18 ULTRA TURRAX Basic) at low speed using pulp suspensions of 2% consistency. The fibers were then refined using ultrafine friction grinder or a so-called supermasscolloider (MKCA6-2, Masuko Sanguo, Japan) and were passed through the device up to 60 times in case of enzyme-treated fibers and for 20 times for TEMPO-oxidized pulp. The gap between the disks was adjusted to 9 μm. MFC was centrifuged at 10,000 rpm to reduce its water content and kept wet in the fridge.

2.5. Characterization of microfibrillated cellulose

Atomic force microscopy (AFM) was carried out using a Multimodal AFM (DI, Veeco, Instrumentation Group) with both tapping and conductive mode (C-AFM). The tips were Multi130 for tapping and MESP for C-AFM. A drop of microfibrillated cellulose suspension was deposited on a fresh mica substrate and left to air dry.

For surface charge determination, particle charge detector PCD 02 (Mütek, Germany) was used. Microfibrillated cellulose