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Microfibrillated cellulose from agricultural residues. Part I: Papermaking application

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ABSTRACT

Due to their abundance, nano-scalecellulose fiber materials such as microfibrillated cellulose, serve as promising candidates for nanocomposite production. Such new high-value materials are the subject of continuing research of new products from the pulp and paper industry sector. Microfibrillated cellulose (MFC) is generally considered to be composed of fibrils with diameter in the range of 10–100 nm liberated from larger plant based cellulose fibers and have a broad range of potential applications. In this work, we evaluated the preparation of MFC from biomass wastes using a new chemical pretreatment method before applying the traditional mechanical method. Rice straw, bagasse and cotton stalk biomass wastes used in this study were chemically treated with sodium hydroxide-sodium sulphite in order to remove lignin, hemicelluloses and to isolate cellulose, which was our starting material for the preparation of MFC. MFC was isolated from the bleached materials by mechanical treatment and the resulting materials were named MFCE30. The resulting MFCE30 were characterized by optical and scanning electron microscopies (SEM), Fourier Transform infrared spectroscopy (FTIR) and X-Ray diffraction. Composite paper sheet samples were prepared from bleached rice straw and bagasse pulps by adding different percentages of MFCE30 and both mechanical and optical properties of the resulting papers were studied. These properties were compared with those of paper sheets conventionally prepared in the pulp and paper Egyptian industry by adding 20% wood pulp as additive.

1. Introduction

Cellulose is the most important renewable natural resource on Earth. Previously, the image of cellulose was that of a hard crystalline material fibrous enough to be used for paper, textile, cloth, string, sanitary goods, etc. Cellulosic materials can be utilized with or without lignin and hemicelluloses, and are commonly used as lumber, textiles, and paper and packaging. Cellulosic fibers, as paper and paperboard, have traditionally been used in packaging for a wide range of food categories such as dry food products, frozen or liquid food and beverage and even fresh food (Lee et al., 2014; Andersson, 2008).

A new class of cellulosic material, microfibrillated cellulose, has emerged recently as a potential packaging material because it exhibits many of the barrier and mechanical properties required in packaging, in addition to the utilization of an abundant raw material, renewability and biodegradability (Restuccia et al., 2010). Cellulose is an important polysaccharide, and it is also a highly crystalline polymer which occurs with various crystalline structures. The degree of polymerization (DP) of natural cellulose may be as high as 15,000 for cotton, giving a molecular weight of 2.4 Mg mol^{-1} and a contour length of 7.7 μm. As cellulose elementary fibril is joined or coated with other wood or plant constituents, such as hemicelluloses, proteins, pectin or lignin, a microfibril is formed (Jacques et al., 2014). The preparation of microfibrillated cellulose (MFC) derived from wood was introduced by Turbak et al. (1983) more than three decades ago. Microfibrils are 10–100 nm thick and have a length of several μm, and can thus be regarded as nanofibers. Their strength, flexibility and aspect ratio suggest a host of tentative possibilities to utilize MFC in large-scale applications. For example, its exceptionally large specific surface area implies potential increased interactivity with secondary components/materials. This could create innovative breakthroughs in use of this material in

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Table 1
Chemical analysis for raw, pretreated and bleached materials: rice straw, bagasse and cotton stalk.

Agricultural waste	Holocellulose (wt%)			Lignin (wt%)			Ash (wt%)			Yield after pretreatment (%)	Yield after bleaching (%)
	Raw material	Pretreated	Bleached	Raw material	Pretreated	Bleached	Raw material	Pretreated	Bleached		
Rice straw	68.10	79.12	90.05	15.40	8.12	3.17	14.50	9.93	6.16	53.00	69.60
Bagasse	73.70	81.63	94.17	24.07	13.69	2.30	1.51	1.13	1.08	61.06	89.60
Cotton stalk	71.00	78.60	92.54	25.75	17.32	5.35	2.93	1.33	1.02	44.00	88.20

areas such as e.g. nanocomposite, packaging, coating and dispersion technology (Stenstad et al., 2008; Lin and Dufresne, 2014).

Herrick et al. (1983) and Turbak et al. (1983) first reported on a method for producing MFC. The MFC typically consists of disintegrated microfibril aggregates with a lateral dimension in the scale of tens of nanometers. The method was to pass a dilute cellulosic wood pulp fiber/water suspension through a mechanical homogenizer, where a large pressure drop facilitates microfibrillation. When a cellulosic pulp fiber suspension is homogenized, the procedure is often repeated several times in order to increase the degree of fibrillation (Spence et al., 2011). Also to facilitate disintegration, one may first reduce the fiber length by mechanical cutting or the fiber cell wall. If perfect cellulose fibrillation is achieved during the refining and homogenizing processes, the resulting cellulose fibrils can have diameters in the nanoscale (Nakagaito et al., 2005; Nakagaito and Yano, 2005; Iwamoto et al., 2005; Oksman et al., 2006; Yano and Nakahara, 2004). Depending on the source of cellulose, individual fibrils can be about 5–10 nm in diameter with lengths varying from 100 nm to several micrometers (Malainine et al., 2005). Within cellulose microfibrils, the cellulose molecules are organized in a crystalline order resulting from a regular network of inter-molecular hydrogen bonds (Goussé et al., 2005). MFC systems have already been intensively studied from a number of viewpoints and have revealed interesting and promising properties which may make this material suitable for use in a number of industrial fields and products (Dufresne, 2008; Hubbe et al., 2008; Nogi et al., 2009). These applications include absorbent sheets, filtering materials, rheology modifiers for food and cosmetics and additives to enhance the mechanical properties of paper and paperboard through the application of nano-coatings or nano-barriers (Minelli et al., 2010; Siro and Placket, 2010; Lavoine et al., 2012; Chang and Wang, 2013; Brodin et al., 2014).

Pulp and paper industry comprises manufacturing enterprises that convert cellulose fiber into a wide variety of pulps, papers and paperboards. Cellulosic fibers, as paper and paperboard, have traditionally been used in packaging. The most important raw material for the production of pulp is wood, softwoods and hardwoods being preferred. Only 7–8% of pulp production is based on annual plants, which are mainly employed when wood is not available. It is the case in Egypt where agricultural residues such as bagasse and rice straw are used. However, using agricultural residues, as a source for pulp and paper, results in low quality paper. Therefore, wood pulp is imported in Egypt, since there is no forest for wood source, and added at a level of 20% to both rice straw and bagasse pulps to improve the paper properties.

Thus, the aim of the present study was to replace imported wood pulp by MFC prepared from Egyptian agricultural by-products to enhance the mechanical properties of the produced paper. MFC is currently manufactured from a number of different cellulosic sources. Wood is obviously the most important industrial source of cellulosic fibers, and is thus the main raw material used to produce MFC (Lavoine et al., 2012). However, interest in other sources such as agricultural crops and their by-products, is increasing. They offer environmental benefits owing to their renewable nature and their low energy consumption in production (Alemdar and Sain, 2008a). Our research will show innovation in using MFC from the waste and will impact new activities to the paper companies, not only in Egypt, but also in other countries that are rare in wood pulp. In this study, MFC from rice straw, bagasse and cotton stalk fibers was prepared by mechanical disintegration of cellulosic materials after pretreatment with sodium hydroxide and sodium sulfite. The number of passes required for our study was easily tuned to 30. The resulting MFC was characterized with an optical microscope, scanning electron microscopy (SEM) and X-ray diffraction and was