



Potential of municipal solid waste paper as raw material for production of cellulose nanofibres



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ABSTRACT

When aiming for higher resource efficiency, greater utilization of waste streams is needed. In this work, waste paper separated from mixed municipal solid waste (MSW) was studied as a potential starting material for the production of cellulose nanofibres (CNFs). The waste paper was treated using three different techniques, namely pulping, flotation and washing, after which it was subjected to an ultrafine grinding process to produce CNFs. The energy consumption of the nanofibrillation and nanofibre morphology, as well as properties of the prepared nanofibres, were analysed. Despite the varying amounts of impurities in the waste fibres, all samples could be fibrillated into nanoscale fibres. The tensile strengths of the CNF networks ranged from 70 to 100 MPa, while the stiffness was ~ 7 GPa; thus, their mechanical strength can be adequate for applications in which high purity is not required. The contact angles of the CNF networks varied depending on the used treatment method: the flotation-treated networks were more hydrophilic (contact angle 52.5°) and the washed networks were more hydrophobic (contact angle 72.6°).

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1. Introduction

The amount of household waste generated by European households was over 200 million tonnes in 2014 (Eurostat, 2017). Despite effective collection and recycling systems for paper and board, part of this waste ends up as mixed household waste, i.e. as mixed municipal solid waste (MSW). The composition of mixed MSW varies by country and location, but it has been reported to contain from 10% to 39% paper and board; other typical components of MSW are food and gardening wastes, plastics, glass, metal, textiles, electronics and miscellaneous waste (Edjabou et al., 2015; Liikanen et al., 2016). Thus, a large amount of paper waste is generated, instead of being recycled into new products.

In order to transform the European Union (EU) into a more sustainable circular economy, the European Commission (EC) has set targets requiring that 55% of MSW should be recycled by 2025 and 65% by 2035, and that the amount of landfilled MSW should be reduced to 10% by 2035 of the total MSW generated (European Commission, 2018). Stricter rules are also set for recycling packaging waste: 75% of paper and cardboard packaging must be recycled by 2020 and 85% by 2030. Fulfilling these goals will be

a difficult task, which is why increasing the use of waste materials as raw materials is needed. Ultimately, the better use of secondary raw materials will result in higher resource efficiency avoiding the resource depletion and unnecessary disposal of wastes.

The most obvious use for the waste paper and board separated from MSW would be further use in the manufacture of new fibrous products, i.e. paper and board. On the other hand, not all fibres are suitable for papermaking. For example, repeated recycling shortens the fibres and results in strength reduction and inferior paper quality (Danial et al., 2015). The waste paper fibres separated from MSW are also subjected to microbes and contaminants, and previous study has shown that the papermaking potential of the fibres separated from mixed MSW is limited as their strength and optical properties are poor (Ojala, 2014). Therefore, other uses than papermaking could be more suitable for such waste paper fibres.

Cellulose nanofibres (CNFs) are long, flexible and entangled nanoscale fibres which can be separated from lignocellulose-containing plants. Due to their great strength, stiffness, low weight, biodegradability and renewability (Siró and Plackett, 2010), CNFs are studied for utilization in a variety of applications, such as electronics, packaging and nanocomposites (Ferrer et al., 2017; Jonooi et al., 2010; Rajala et al., 2016). CNFs are produced in commercial or pre-commercial scale in e.g. Finland, Norway, Japan and USA (An et al., 2017), and some commercial applications have already

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emerged the markets, such as milk cartons and hygiene products containing CNFs. Especially due to the needs of packaging, paper-board and paper industry, the global markets for nanocellulose are estimated to have a 33.9% CAGR from 2013 to 2024 (Global Market Insights, 2018; Transparency Market Research, 2015).

CNFs are currently produced from virgin cellulose pulp, but using waste paper and board fibre as raw material for CNF could be an innovative way to reuse these lower quality fibres, which are not of a high enough quality for paper products. Previous studies have shown that separately collected waste paper materials such as mixed office paper (Wang and Zhu, 2015) and recycled newspapers (Josset et al., 2014) can be utilized in the manufacture of CNF after disintegration into a slurry. Corrugated paper waste pulp has also been used as a starting material for CNFs by combination of chemical and mechanical treatments (Wang et al., 2013).

In this work, waste paper separated from mixed MSW was studied as a raw material for the production of CNF. To the best of our knowledge, this is the first time mixed waste which is not separately collected has been used in the manufacture of CNF. The challenges in using this kind of material as CNF raw material arise from its varying composition which is ranging from printed newspapers to plastic film-coated liquid packaging as well as from the contaminants originating from their use or from the waste recycling system (food residues, plastics, sand etc.). Thus, in order to purify the material, the waste paper was pre-treated using pulping, flotation or washing, after which the waste fibres were fibrillated using ultrafine grinding, while monitoring the fibrillation process and its energy consumption. The mechanical and physical properties of prepared waste nanofibres were studied and compared to those prepared from virgin pulp in order to assess their potential in CNF applications.

2. Materials and methods

2.1. Materials

The waste paper separated from mixed MSW was obtained from Fortum Recycling and Waste Solutions, Riihimäki, Finland. The waste material (Fig. 1) had gone through mechanical pre-sorting to remove most of the non-paper materials containing different paper and board grades, such as newspaper, tissue paper, milk cartons and cardboard. Some remaining pieces of metal, foil, plastic and textiles were manually removed from the waste before further treatment. Stora Enso Mill in Oulu, Finland kindly supplied the bleached softwood sulphate pulp used as a reference material (REF). All the chemicals used in the study, namely oleic acid, calcium chloride, sodium silicate, hydrogen peroxide, acetone,



Fig. 1. The waste paper separated from mixed MSW.

sodium hydroxide and sulphuric acid, were of reagent grade and purchased from VWR International Oy (Helsinki, Finland).

2.2. Waste paper treatment

The waste paper was treated using three different techniques: (1) pulping (pulped waste fibres [PWF]), (2) pulping and flotation (flotated waste fibres [FWF]) and (3) pulping and washing washed waste fibres [WWF]). First, the waste paper was defibrated into fibres following the pulping procedure of the INGEDE 11 method (INGEDE Method 11, 2012) for printed paper products. The amount of chemicals used in pulping are presented in Table 1. The pulping chemicals, which included sodium hydroxide, oleic acid, calcium chloride and sodium silicate, were dissolved in hot water (~ 90 °C) before mixing with 2.5 kg of paper waste (dry content). The pulping was done with a Hobart H600 mixer (Hobart GmbH, Germany), using 15 wt% dry content at 45 °C with a pulping time of 60 min. Hydrogen peroxide was added to the mixer after 10 min of pulping in the mixer.

After pulping, one-third of the pulped waste paper was further treated using flotation according to the INGEDE 11 method. Flotation is typically used in paper recycling to remove hydrophobic particles, such as ink and dirt specks, as well as fillers from recycled pulp (Schabel and Holik, 2010). The flotation was done using a Delta 25 laboratory flotation cell (VOITH, Germany) and waste paper pulp with 0.8% dry content in 45 °C for 12 min using an air flow rate of 10 L/min and a pressure of 3 bar. After flotation, the flotation accept was collected, and its water content was reduced by filtration for further treatment. One-third of the pulped waste paper was washed with tap water using a Somerville type screen fractionator (Lorentzen & Wettre, Sweden) equipped with a 50-mesh size screen. The screen was used to remove larger contaminants from the waste pulp as the washed fibres passed through the screen and were collected into an 80 µm pore size filter bag.

2.3. Ultrafine grinding

Ultrafine grinding was used to prepare CNFs from the different fibre fractions. First, pulp suspensions with a 2 wt% dry content were prepared and mixed using an overhead stirrer for 30 min at 500 rpm, while the pH of the suspensions was adjusted to 7.0. After mixing, the suspensions were ground using an MKCA6-2J grinder (Masuko Sangyo Co., Ltd., Japan) at a speed of 1500 rpm. The grinding gap was reduced gradually from 0 gap (contact mode) to -90 µm while the pulp was passing through the grinding stones. The viscosity of the pulp suspension was monitored continuously during the grinding, and after reaching the -90 µm gap, the grinding was continued until the viscosity of the suspension reached its peak value and no longer increased.

The energy consumption of the ultrafine grinding process was measured with an iEM3250 Schneider-Electric energy meter. Energy consumption (E) was calculated as kWh/kg based on the dry weight of the fibres:

$$\text{Energy consumption (kWh/kg)} = \text{power(kW)} \times \text{time(h)}/\text{mass(kg)}. \quad (1)$$

Table 1
The chemicals used in pulping of paper waste.

Chemical	Amount based on dry paper (%)
Sodium hydroxide, NaOH	1.2
Sodium silicate, Na ₂ SiO ₃	1.8
Oleic acid, C ₁₈ H ₃₄ O ₂	0.9
Calcium chloride, CaCl ₂	0.2
Hydrogen peroxide, H ₂ O ₂	0.8

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