Composites Science and Technology 118 (2015) 154–162

Contents lists available at ScienceDirect

Composites Science and Technology

journal homepage: http://www.elsevier.com/locate/compscitech

Life cycle assessment of nanocellulose-reinforced advanced fibre composites

Martin Hervy [1], Sara Evangelisti, Paola Lettieri*, Koon-Yang Lee*, [1]

Department of Chemical Engineering, University College London, Torrington Place, WC1E 7JE, London, United Kingdom

A R T I C L E I N F O

Article history:
Received 27 May 2015
Received in revised form
14 August 2015
Accepted 30 August 2015
Available online 3 September 2015

Keywords:
Nanocellulose
Nanocomposites
Polymer-matrix composites (PMCs)
Mechanical properties
Modelling

A B S T R A C T

The research and development of nanocellulose-reinforced polymer composites have dramatically increased in recent years due to the possibility of exploiting the high tensile stiffness and strength of nanocellulose. In the work, the environmental impact of bacterial cellulose (BC)- and nanofibrillated cellulose (NFC)-reinforced epoxy composites were evaluated using life cycle assessment (LCA). Neat polylactide (PLA) and 30 wt.-% randomly oriented glass fibre-reinforced polypropylene (GF/PP) composites were used as benchmark materials for comparison. Our cradle-to-gate LCA showed that BC- and NFC-reinforced epoxy composites have higher global warming potential (GWP) and abiotic depletion potential of fossil fuels (ADf) compared to neat PLA and GF/PP even though the specific tensile moduli of the nanocellulose-reinforced epoxy composites were higher than neat PLA and GF/PP. However, when the use phase and the end-of-life of nanocellulose-reinforced epoxy composites were considered, the "green credentials" of nanocellulose-reinforced epoxy composites were comparable to that of neat PLA and GF/PP composites. Our life cycle scenario analysis further showed that the cradle-to-grave GWP and ADf of BC- and NFC-reinforced epoxy composites could be lower than neat PLA when the composites contains more than 60 vol.-% nanocellulose. This suggests that nanocellulose-reinforced epoxy composites with high nanocellulose loading is desirable to produce materials with "greener credentials" than the best performing commercially available bio-derived polymers.

1. Introduction

The growing awareness on the consequences of depletion of fossil resources and the increasing demand for more environmental friendlier products have led to the development of sustainable and/or renewable composites to replace petroleum-derived plastics [1], particularly in the automotive industry [2]. In this context, nanometre-scale cellulose fibres, or nanocellulose, are emerging green nano-reinforcements for polymers [3]. The major driver for utilising nanocellulose as reinforcement for polymer is the possibility of exploiting the high tensile stiffness and strength of the cellulose crystals [4]. Theoretical predictions [5–7] and numerical simulations [8–10] estimated the tensile moduli of single cellulose crystal to be between 58 and 180 GPa. X-ray diffraction [11–13] and Raman spectroscopy [14–16] further showed experimentally that a single cellulose nanofibre possess tensile moduli of between 100 and 160 GPa. The tensile strength of a single cellulose crystal has also been estimated theoretically to be between 0.3 and 22 GPa [17–20]. The high tensile modulus and strength of nanocellulose fibres suggest that they could potentially serve as replacement for glass fibres given their low toxicity and low density (1.5 g cm^{-3} for nanocellulose versus 2.5 g cm^{-3} for glass fibres). As a result, numerous research effort has been poured into the use of nanocellulose as reinforcement for polymers [21,22].

Nanocellulose can be produced via two approaches: top-down and bottom-up. In the top-down approach, (ligno)cellulosic biomass, such as wood fibres, is disintegrated into nanofibres. The earliest report on the preparation of wood-derived nanocellulose from (ligno)cellulosic biomass was presented by Wurhmann et al. [23]. The authors disintegrated ramie, hemp and cotton fibres using high intensity ultrasound into their respective elementary fibrils with diameters as small as 6 nm. Later, Herrick et al. [24] and Turbak et al. [25] used high pressure homogenisers to reduce the diameter of wood pulp to 10 nm. Wood-derived nanocellulose,

---

* Corresponding authors.
E-mail addresses: p.lettieri@ucl.ac.uk (P. Lettieri), koonyang.lee@imperial.ac.uk (K.-Y. Lee).

[1] Current address: *The Composites Centre*, Department of Aeronautics, Imperial College London, South Kensington Campus, SW7 2AZ, London, United Kingdom.

http://dx.doi.org/10.1016/j.compscitech.2015.08.024

herein termed nanofibrillated cellulose (NFC), can also be produced using stone grinders [26], whereby the high shearing forces generated by the static and rotating grinding stones fibrillates micrometre-sized pulp fibres into cellulose nanofibres. The bottom-up approach, on the other hand, utilises the fermentation of low molecular weight sugars using cellulose-producing bacteria, such as from the *Acetobacter* species, to synthesise nanocellulose [27]. Nanocellulose synthesised by bacteria, herein termed bacterial cellulose (BC), is pure cellulose without impurities that are typically present in wood-derived nanocellulose, such as hemicellulose, pectin and traces of lignin. In addition to this, both BC and NFC possess cellulose-I structure [28]. Even though BC possesses higher degree of crystallinity compared to NFC (72% for BC and 41% for NFC, respectively, based on area under the X-ray diffraction spectra), both BC and NFC were found to possess similar reinforcing ability for polymers [29].

The first use of nanocellulose as reinforcement in polymers, namely polypropylene, polystyrene and polyethylene, was reported by Boldizar et al. [30]. The authors found that the tensile modulus increased from 2.4 GPa for neat polystyrene to 5.2 GPa for polystyrene reinforced with 40 wt.-% NFC. Yano et al. [31] reported tensile modulus and strength of up to 20 GPa and 325 MPa, respectively, for BC-reinforced epoxy composites containing 65 wt.-% BC loading. Tensile modulus and strength of up to 8 GPa and 202 MPa, respectively, have also been obtained for NFC-reinforced hydroxyethyl cellulose composites containing 68 wt.-% NFC loading [32]. It is therefore evident that high performance BC- and NFC-reinforced polymer composites can be produced. However, one major question still remains: “*Are nanocellulose-reinforced polymer composites truly environmentally friendly compared to commercially available renewable polymers or engineering materials?*” Li et al. [33] recently used life cycle assessment (LCA) to analyse the environmental impact associated with the production of NFC using high pressure homogenisation and high intensity sonication. TEMPO-oxidised and chloroacetic acid-etherified kraft pulp were chosen as the starting materials in their LCA model. The authors found that NFC produced from high-pressure homogenisation of TEMPO-oxidised kraft pulp has the lowest environmental impact among all the NFC production routes studied. In a recent LCA study conducted by Hohenthal et al. [34], the non-renewable energy consumption associated with the production of NFC nanopaper was estimated to be 107.5 MJ kg^{-1}. This is significantly higher than the energy consumed for the production of polylactide (PLA), which is estimated to consume only 42 MJ kg^{-1} [35] of energy.

With increasing demand for environmental friendlier materials, it is timely to investigate the environmental impact associated with the manufacturing of nanocellulose-reinforced polymer composites. Therefore in this work, we quantify the environmental impact associated with the manufacturing of BC- and NFC-reinforced polymer composites through a life cycle assessment approach, starting from the production of nanocellulose (i.e. the cradle) to the end-of-life (i.e. the grave) of the nanocellulose-reinforced polymer composites. Joshi et al. [36] have suggested that natural fibre composites could potentially compete environmentally with glass fibre-reinforced polymer composites in most applications. In this work, a comparison of environmental impact between BC-/NFC-reinforced polymer composites and glass fibre-reinforced polymer composites is also reported.

2. Methodology

LCA is the internationally recognised method used to assess of the environmental performance of a product. ISO 14040:2006 (E) states that LCA should “*consider the entire life cycle of a product from raw material extraction and acquisition, through energy and material production and manufacturing, to use and end-of-life treatment and final disposal*”. Any LCA study should start with a carefully defined scope of the study, functional unit and system boundaries. These elements are the guideline that will help reaching a conclusion that answers the initial objective of the LCA; whether it is the comparison of different products or the identification of a “hotspot”. Following this step, a life cycle inventory (LCI) consisting of the materials and energy used in every step of the life cycle is compiled. Using this LCI, life cycle impact assessment (LCIA) can then be conducted. All the inputs and outputs of the system are allocated to the different environmental impact categories. Finally, the outcome of the LCA can be given in the form of interpretations and recommendations to lower the environmental burden of the system [37].

2.1. Goal and scope definitions

The aim of this study is to evaluate the environmental impact of high performance BC- and NFC-reinforced epoxy composites through a cradle-to-grave LCA including their manufacturing, use phase and end-of-life. Two commercially available benchmark materials were chosen for comparison: (i) 30 wt.-% randomly oriented glass fibre-reinforced polypropylene (GF/PP) composites and (ii) polylactide, which is considered the best performing bio-derived polymer [3]. The system boundary for our nanocellulose-reinforced epoxy composites and our benchmark materials is shown schematically in Fig. 1. A distinction is made between the foreground system, which is defined as the processes of main importance in regards to the study (direct measurements can often be taken), and the background system, which is defined as the processes used to support the foreground system (supply of energy and materials) [38].

2.2. Description of the polymer and composite manufacturing processes

The manufacturing of BC- and NFC-reinforced epoxy composites modelled in our system is based on our previous work [29]. This study was chosen as high loading of nanocellulose, a pre-requisite to producing high performance nanocellulose-reinforced polymer composites [3], was achieved using conventional vacuum assisted resin infusion (VARI), a widely used composite manufacturing technique. Briefly, never-dried bleached birch kraft pulp (*Betula pendula*) was passed through a Supermass Colloider (Masuko Sangyo Co., Kawaguchi, Japan) seven times and the final obtained consistency of NFC in water was approximately 2 wt.-%. The NFC suspension was diluted to 0.4 wt.-% prior to nanopaper manufacturing. Similarly, BC pellicles were first cut into small pieces and blended (Breville BL18 glass jug blender, Pulse Home Products Ltd., Oldham, UK) for 2 min at a consistency of 0.1 wt.-%. The manufacturing of both BC and NFC nanopaper closely resembles that of conventional paper manufacturing process, whereby the BC and NFC suspension were vacuum filtered, wet pressing under a 10 kg weight, followed by drying at 55 °C to obtain BC and NFC nanopapers. To manufacture BC- and NFC-reinforced epoxy composites, 11 nanopapers were sandwiched between two PTFE coated glass fabrics (FF03PM, Aerovac, West Yorkshire, UK) and placed on top of a polyester porous flow medium (15087B, Newbury Engineer Textile, Berkshire, UK). Another polyester porous flow medium was placed on top of the PTFE coated glass fabric. A heat stabilised Nylon 6 vacuum bag (Capran 519, Aerovac, West Yorkshire, UK) was then used to cover the whole set up and vacuum sealant tape (SM5127, Aerovac, West Yorkshire, UK) was used to seal the set up. A vacuum was then applied (~15 mmHg) and the liquid epoxy resin (PRIME 20ULV, Gurit Ltd, Isle of Wight and Hamble, UK) was fed at room temperature from the bottom of the