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Close-looped recycling of polylactic acid used in 3D printing: An experimental investigation and life cycle assessment

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ABSTRACT

The objective of this work is to investigate the potential of close-looped recycling of polylactic acid (PLA) that used in 3D printing, from the perspectives of material properties and environmental performance. A commercial grade of PLA was extruded into filament, then was subjected to fused deposition modelling (FDM) 3D printing process. The printed products were later shredded and re-extruded for repeating 3D printing cycle. Samples were taken from each repeating cycle, were characterised in terms of mechanical, rheological, molecular, thermal and morphological properties. In the experiment, the material can only be reprocessed for two 3D printing cycles, as the reprocessed material can no longer be further processing. Although little changes are observed in the mechanical properties, the viscosity measurements suggest that PLA deteriorates significantly due to the repeated FDM 3D printing cycles. The reduced viscosity values are compatible with the decrements in average molecular weights and thermal stability, the increments of carbonyl bonds, crystallinity and pinholes in the materials' surfaces. Virgin PLA was blended into the recycled material to improve the viscosities, which enables the blends 3D printable. The results of life cycle assessment approach show that the environmental burdens associated with close-looped recycling are lower than those of placing the 3D printed products in incineration or in landfill. Incineration might be another environmental alternative, due to the material embodies a relatively high calorific value; however, the material is not recuperated, thus incineration cannot satisfy the more stringent environmental legislations, in which recovery and reuse of materials are rigorously required.

1. Introduction

Three-dimensional (3D) printing is a typical additive manufacturing (AM) technique for fabricating products of various structures and geometries (Ngo et al., 2018). Owning to its capacity in highly customisable products, 3D printing has been widely applied in many industries, including construction (Wu et al., 2016), energy (Zhang et al., 2017), food (Godoi et al., 2016), medical (Jang et al., 2018), prototyping (Ngo et al., 2018) and manufacturing (Upadhyay et al., 2017). It is predicted that 50% of final products will be manufactured in 3D printing processes (Berman, 2012). Proliferation of 3D printing brings emerging environmental problems such as emissions and waste disposal. Compared to extensive efforts that paid on characterising emissions from 3D printing (see Stephens et al., 2013; Kim et al., 2015; Azimi et al., 2016; Steinle, 2016; Mendes et al., 2017; Stabile et al., 2017) and mitigating this problem (Kwon et al., 2017; Rao et al., 2017), little research concerns about the sustainability issue of 3D printing materials, that is, disposal of waste 3D printed products. Although 3D printing possesses higher material efficiency (Gebler et al., 2014), material sustainability is an issue that can no longer be ignored due to wide adoption of 3D printing. Close-looped recycling can be a promising measure in tackling this particular problem due to the following four reasons. First, despite intensive efforts have been paid on developing new materials for 3D printing,

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only a limited number of materials are commercially viable (Gebler et al., 2014; Ngo et al., 2018). Second, the global production of 3D printing materials like polylactic acid (PLA) is still limited, though PLA production has enjoyed a substantial growth during the past decade (Beltrán et al., 2018). Third, mechanical recycling of waste plastics is becoming a common and environmental practise in mitigating solid plastic waste problems (Andreoni et al., 2015; Gu et al., 2017), and recycled plastics have comparable performance to their virgin counterparts (Gu et al., 2016a, 2016b) and have even been applied on manufacturing high-value products such as automobiles (Gu et al., 2017). Fourth, high-quality recycling includes close-looped recycling is more sustainable than downcycling (Di Maria et al., 2018) and other alternatives like landfill (Bensadoun et al., 2016).

Yet, to the best of our knowledge, there is little literature that investigates this particular topic of close-looped recycling of 3D printing material, and the extant research concentrates on examining changes in mechanical properties that associated with repeated 3D printing cycles (Sanchez et al., 2017; Tian et al., 2017). Sanchez et al. (2017) studied the mechanical performance of PLA during repeated 3D printing processes and found that there were significant deteriorations in most mechanical properties after five recycling cycles. Tian et al. (2017) achieved material recovery rates of 100% for continuous carbon fibre and 73% for PLA matrix in recycling 3D printed continuous carbon fibre reinforced PLA composites. The recycling potential of 3D printing materials is only investigated from the perspective of mechanical performance. Although mechanical performance is crucial in the applications of 3D printing, there are also other properties that should be considered during recycling. For example, viscosity is a particularly important characteristic for fused deposition modelling (FDM), the most common and frequently studied 3D printing process. FDM 3D printers can only adopt materials which have viscosity in a limited range, as the viscosity should be high enough to provide structural support and low enough to enable extrusion (Wang et al., 2017). From material sustainability perspective, applying life cycle accounting techniques like life cycle assessment (LCA) to calculate the overall environmental impacts of the whole recycling process, would provide quantitative evidence that ensures the best route of disposing the plastic waste and demonstrates the gain of using the recycled polymers (Ragaert et al., 2017). However, the data on environmental impact of disposing 3D printed waste is in scarcity or even non-existed, as the studies on recycling 3D printing materials are also extremely limited.

To address this knowledge gap, the main objective of this work is to investigate the potential of close-looped recycling PLA that used in 3D printing, in terms of changes in properties and related environmental impacts. PLA is studied due to its limited global production (around 0.21 Mt in 2016) (Beltrán et al., 2018) and extensive application in 3D printing (Rao et al., 2017). As a type of biopolymers, PLA is made from crops and increasing its production could jeopardise the food security of poorer countries (Mülhaupt, 2013). Moreover, PLA is also used in other applications like packages (Beltrán et al., 2018). These characteristics justify the choice of material, as prolonging the life of PLA used in 3D printing is essential in reducing material consumption and waste generation. In this study, a commercial grade of PLA was extruded into filament and fed into an FDM 3D printer, and the printed products were shredded, extruded and then subjected to the 3D printing process again. The cycle was repeated till the material cannot be subjected to further 3D printing. During each cycle, a certain amount of sample was extracted for detailed characterisation with the purpose of identifying possible properties changes. The properties tested include: (1) mechanical performance in terms of tensile and flexural properties; (2) rheological properties which refer to the viscosities of PLA in different cycles; (3) molecular structures, including number average molecular weights (M_n) and weight average molecular weight (M_w) that measured by gel permeation chromatography (GPC), and functional groups that detected by Fourier transform infrared spectroscopy (FTIR); (4) thermal properties that tested via differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA); (5) morphological features that investigated via scanning electron microscopy (SEM). The selection of properties was determined in accordance with the conceptual framework for evaluating the quality of recycled plastics proposed by Vilaplana and Karlsson (2008). Virgin PLA was then blended with recycled PLA in different proportions, since compounding recycled plastics with virgin counterparts is a simple and effective route for improving desirable properties (Gu et al., 2014; Hopmann et al., 2015). Apart from this, blending recycled PLA with virgin materials for upgrading the performance has been pointed out as one of the future research directions in mechanical recycling of PLA (Badia and Ribes-Greus, 2016). The environmental performance was assessed using an LCA approach for comparing the close-looped recycling with other end-of-life alternatives such as landfill and incineration, since these three routes are most common in disposing plastic waste and are frequently compared in the previous LCA studies (Bensadoun et al., 2016; Gu et al., 2017, 2018).

The contribution of this study is threefold. First, the materials recycled from FDM 3D printed products of different cycles have been characterised in a comprehensive manner, while the extant literature tends to concentrate solely on mechanical properties (Sanchez et al., 2017; Tian et al., 2017). Second, the key performance issue, i.e., viscosity, in achieving close-looped recycling of 3D printing material was identified, and an attempt has been carried out to mitigate the problem via blending with virgin material. Third, environmental impacts of close-looped recycling of 3D printing material were first modelled and compared to other disposal routes, with the purpose to identify the most sustainable disposal route for 3D printed products.

The rest of this paper is organised as follows. The experimental sector in this study are illustrated in the next section. The experimental results are presented and discussed in Section 3, and Section 4 concludes this study.

2. Materials and methods

2.1. Materials

A commercial grade of PLA with a trademark of Ingeo™ 4032D, produced by *NatureWorks Co., Ltd.*, Guangdong, China was used in this work. The specific density of this PLA material is 1.24 g cm^{-3}. This type of material is frequently applied as raw material of 3D printing filament, see online 3D printing sites such as Stro3D (2018) and 3DprintWorks (2018).

2.2. Processing

Based on the selection of properties and simulated PLA recycling processes in the previous literature (Badia and Ribes-Greus, 2016; Sanchez et al., 2017), a scheme for close-looped recycling of PLA in 3D printing was adapted as illustrated in Fig. 1. In this experimental work, virgin PLA pellets were extruded into filament and then subjected to FDM 3D printing, and 3D printed products were shredded and re-extruded into filament; the properties of the materials were characterised during each cycle, as the samples were taken from the filaments, while only mechanical tests use 3D printed specimens. The experimental scheme enables the comparison of materials of different cycles and proportions, as specified