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Closed-loop recycling of polyamide12 powder from selective laser sintering into sustainable composites

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ABSTRACT

A considerable amount of powder residue is generated during the selective laser sintering (SLS) process. Processing the powder residue into filaments for extrusion-based additive manufacturing (EAM) provides an opportunity to recycle SLS residue without significantly reducing its value. In this study, the feasibility of using a milled carbon fiber (mCF)/recycled polyamide12 (rPA12) composite filament for EAM was demonstrated. A microscopic study on the morphology of mCF showed that mCF is a short and smooth fiber. The mCF did not significantly change the melting temperature, crystallization temperature or crystallinity of rPA12 as measured by differential scanning calorimetry (DSC). A morphological study on the fracture-surface of the composites revealed that there was moderate interfacial interaction between the mCF and PA12. Tensile strength, tensile modulus, flexural strength, flexural modulus and impact strength of rPA12 were improved by 35%, 163%, 61%, 138% and 23%, respectively with the addition of 30 wt.% mCF.

1. Introduction

During the selective laser sintering (SLS) process, a layer of powder is applied on the printing bed before the laser melts and fuses the powders into a complete layer (Wendel et al., 2008). There is considerable residue generated in the SLS manufacturing process. The unsintered powder (80–90%) is a potential residue stream for other applications (Dotchev and Yusoff, 2009). Using the power residue without adding new powder in subsequent sintering steps results in parts with a rough surface texture. This is because the molecular weight of the residue powder is increased and the morphological properties for SLS are negatively impacted (Dotchev and Yusoff, 2009). The addition of new powder often replaces about 50% of the residue in SLS processing. The increase in molecular weight of residue powder is not an issue if it is reused for other plastic processing techniques like injection and compression molding. Because powders used in SLS have to meet specific requirements, polyamide12 (PA12) powders designed for SLS cost around $150/kg, while the cost of PA12 pellets for conventional plastics processing is below $3/kg. Therefore, using PA12 powder residue from SLS directly for injection molding is not justified in terms of its value. To use this expensive residue, a relatively high-value product should be targeted. The cost of PA12 filament for extrusion-based additive manufacturing (EAM) is approximately $100/kg where fiber addition can further increase its value. Hence, turning SLS powder residue into polymer filaments for EAM can be one of the solutions to its successful reuse (Mägi et al., 2015; Kumar and Czekanski, 2017).

One shortcoming of EAM is it lowers mechanical properties of 3D-printed parts compared to their injection molded counterparts (Wang and Gardner, 2017). Many efforts have been implemented to reduce the gap of mechanical properties between 3D-printed and injection molded parts through process control and fiber reinforcement. We previously studied the effect of printing parameters on the impact strength of polypropylene and polylactic acid (Wang and Gardner, 2017; Wang et al., 2017a). The impact strengths of

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these two polymers can be improved by using proper printing parameters. Carbon fibers are frequently employed to strengthen the mechanical properties of parts from the EAM process. Commercial EAM printers have a typical nozzle diameter of 0.4–0.5 mm. To prevent the fibers from clogging the nozzle, short fibers are often favored by researchers. Short carbon fibers (SCF) were compounded with acrylonitrile-butadiene-styrene (ABS) to make filaments for EAM (Tekinalp et al., 2014). The 10 wt.% SCF-filled-ABS composites displayed similar tensile properties to a compression-molded equivalent. Using an innovatively modified EAM printer, continuous CF was simultaneously printed with ABS filament (Yang et al., 2017). The flexural and tensile strengths of 3D-printed CF/ABS composites with 10 wt.% fiber-loading level were much higher than 3D-printed ABS and approached the strength values of injection-molded CF/ABS composites. There are only a few studies reported using recycled PA12 (rPA12) powder for EAM (Mägi et al., 2015; Kumar and Czekanski, 2017). Virgin and rPA12 powders from the SLS process were compounded with short aramid fibers or thermoplastic polyurethane (TPU) to make filaments for EAM (Mägi et al., 2015). TPU was reported to improve the tensile properties of rPA12. Tungsten carbide was blended with rPA12 from SLS to form filaments (Kumar and Czekanski, 2017). The addition of filler increased rPA12's glass transition temperature, melt flow index and mechanical properties. Both research papers reported on small objects that were built using rPA12 composite filaments.

To provide a database for future comparisons, the mechanical properties of injection-molded rPA12 parts using made the powder residues from SLS process is worth exploring. There are several studies that examined the properties of SCF-reinforced PA composites from injection molding (Molnár et al., 1999, Hassan et al., 2003; Karsli and Aytac, 2013; Feng et al., 2013; Liang et al., 2014; Sang et al., 2016). Through extrusion and injection molding, SCF/PA6 composites were formed to reveal the effect of SCF on the impact strength of PA6 (Molnár et al., 1999). The impact strength improved with a fiber content ranging from 5 to 16 vol.%. Two methods were applied to compound CF and PA6: 1) extrusion to yield short CF/PA6 feedstock and 2) pultrusion to yield long CF/PA6 feedstock for injection molding (Hassan et al., 2003). CF/PA6 composites from the pultrusion process displayed superior mechanical properties. In another study, SCF was compounded with PA6 and injection molded into test coupons (Feng et al., 2013; Liang et al., 2014). SCF increased the tensile strength and elastic modulus while the elongation at break of PA6 decreased. SCF functioned as a nucleating agent that facilitated heterogeneous nucleation, trans-crystallization and α-form crystal formation. Silane-surface treatment was applied to SCF to improve the interfacial compatibility between SCF and PA6 (Sang et al., 2016). After silane treatment, the tensile, impact and flexural strength of SCF/PA6 composites were significantly improved. One mechanical property of PA6 that was consistently reported to be impaired by CF was the elongation at break (Karsli and Aytac, 2013). The elongation at break of PA6 was lowered to 20% from 320% by the addition of 20 wt.% SCF.

Table 1
Compositions of mCF/rPA12 composites.

Designation	mCF (wt.%)	rPA12 (wt.%)
rPA12	0	100
mCF10rPA12	10	90
mCF15rPA12	15	85
mCF20rPA12	20	80
mCF30rPA12	30	70

Based on the above literature, to the best of our knowledge, there are few studies which address the mechanical properties of carbon fiber (CF)/recycled PA12 (rPA12) composites obtained by the injection molding process. In this study, mCF was added into the rPA12 to impart the composites with enhanced mechanical properties. The purpose of using mCF (a short CF) was in an effort to prevent nozzle clogging during the 3D printing of those composite filaments. Composites were made through the injection molding process and contained four different mCF contents: 10 wt.%, 15 wt.%, 20 wt.% and 30 wt.%. The feasibility of using the mCF/rPA12 composites for EAM was demonstrated by printing a fuel-line quick connector using the 15 wt.% mCF/rPA12 composite filament.

2. Materials and methods

2.1. Materials

The rPA12 was sourced from residue generated by the SLS additive manufacturing process, as a matrix polymer, used in sustainable automotive composites. Milled carbon fibers were kindly supplied by Toho Tenax America Inc. (Rockwood, TN, USA), having a density of 1.8 g/cm^3.

2.2. Material compounding and injection molding

The mCF and rPA12 powders were dried overnight (20 h) in an oven at 60 °C to a moisture content less than 1% using a conventional air oven. Drying temperature and duration were

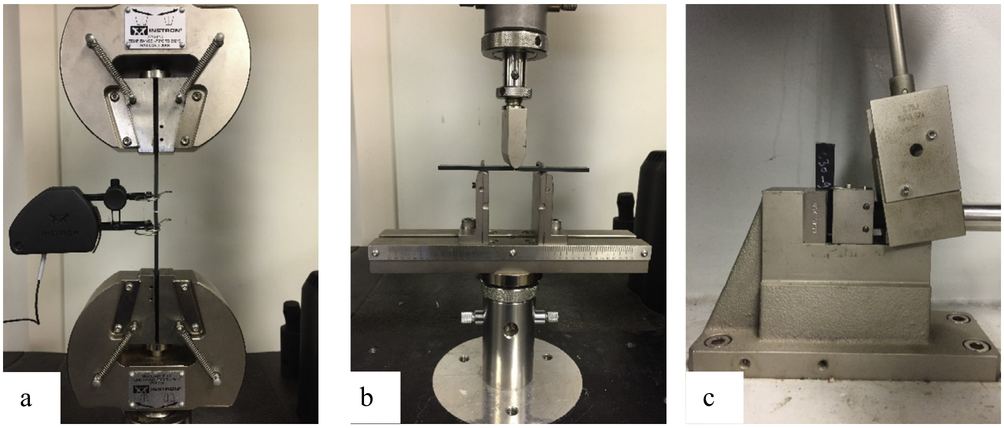

Fig. 1. Experimental setups for tensile (a), flexural (b) and Izod impact tests (c).