

Recycling of Additively Printed Rare-Earth Bonded Magnets

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Abstract

In this work, we describe an efficient and environmentally benign method of recycling of additive printed Nd-Fe-B polymer bonded magnets. Rapid pulverization of bonded magnets into composite powder containing Nd-Fe-B particles and polymer binder was achieved by milling at cryogenic temperatures. The recycled bonded magnets fabricated by warm compaction of ground cryomilled coarse composite powders and nylon particles showed improved magnetic properties and density. Remanent magnetization and saturation magnetization increased by 4% and 6.5% respectively, due to enhanced density while coercivity and energy product were retained from the original additive printed bonded magnets. This study presents a facile method that enables the direct reuse of end-of-life bonded magnets for remaking new bonded magnets. In addition to magnetic properties, mechanical properties comparable to commercial products have been achieved. This research advances efforts to ensure sustainability in critical materials by forming close loop supply chain.

Keywords: Recycling; Bonded magnets; Cryomilling; Waste management; Additive manufacturing; Nd-Fe-B

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1. Introduction

Near-net shaped polymer bonded rare-earth transition-metal (RE-TM) gap magnets with intermediate performance between ferrites and sintered Nd-Fe-B magnets could have many important technological applications in sensors and electric drive technologies due to many unique advantages. (Bollero et al., 2017; Coey, 2002; Kurth and Drummer, 2014). Bonded magnets are comprised of two components; hard magnetic powder and non-magnetic polymer binder. Hard magnetic powders can be permanent magnet materials such as Nd-Fe-B, Sm-Co, Sm-Fe-N and Ferrite (ceramic magnets). Typical binders used for the fabrication of bonded magnets include nylon, polyphenylene sulfide (PPS), and thermoset epoxies (Wano-wari et al., 2015). Thermoplastic binders can be used to form complex shapes by injection molding or extrusion processes. In a typical compression molding process, isotropic Nd-Fe-B powders are coated with a thermoset epoxy binder and compacted into a simple shape using an uniaxial hot press (Ma et al., 2002).

An exciting processing technology that is revolutionizing the manufacturing sector, and is beginning to advance the production of bonded magnets is additive manufacturing (Gandha et al., 2018; Huber et al., 2016; Huber et al., 2017; Li et al., 2017). Additive manufacturing (AM) (commonly recognized as 3D printing) processes enable the joining of materials layer by layer to make objects using computer-aided design (CAD) models. It offers a cost-effective and time-efficient approach to manufacture products with complex geometries, advanced material properties and multifunctionality (Meng et al., 2018). According to Wohlers report 2018, 21% growth rate was reported for the AM industry with market share exceeding \$7.3 billion for the year 2017 (Ian Campbell, 2018). Compared with conventional manufacturing processes, AM technologies should potentially produce minimum or no wastes, and are more energy efficient. Nevertheless, some

amount of wastes are generated during purging, and in some cases the amount is inadvertently much higher than expected due to human, design and machine errors (Rejeski et al., 2018; Song and Telenko, 2017). For example, laser sintering (LS) – additive manufacturing generates up to 44% waste, and fused deposition modeling (FDM) printers generates nearly 34% of the plastic waste (Faludi et al., 2015; Song and Telenko, 2017). Considering the exponential growth and high demand for 3D printing technologies in both near and long terms, residues generated during AM process is bound to significantly increase, and therefore efficient recycling is inevitable from environmental and waste management standpoints. Most importantly, recycling will help to ensure additional economic benefits for the sustainability of AM processes without limiting other benefits.

Recently, recycling of magnets containing expensive rare earth elements (REE) have drawn considerable attention due to the concern over potential disruptions in the supply of REEs. To address this challenge, various recycling processes are developed to recover the REEs from pre-consumer (manufacturing) scrap and from complex post-consumer end-of-life Nd-Fe-B permanent magnet scrap (Gutfleisch et al., 2013; Kumari et al., 2018; Okabe et al., 2003; Sheridan et al., 2012; Walton et al., 2015; Yang et al., 2017; Yue et al., 2018; Zakotnik et al., 2008; Zakotnik and Tudor, 2015). Recycling processes of Nd-Fe-B sintered magnets into bonded magnets have also been reported (Gutfleisch et al., 2013; Itoh et al., 2004). However, research and development to recycle polymer bonded magnets have shown little progress because the recovery of rare earth magnetic powder from polymer composites is challenging. For instance, recovery of magnet powders from rare earth bonded magnets by a dissolution process at high temperature (230°C) have been reported in which chemicals for dissolution use at least one solvent from a group comprising tetralin, naphthalene, 1,4-hydroxynaphthalene, naphthol, biphenyl, etc. (Yamagata and Yamashita, 2003). Another recycling method in which magnet powders are separated by heating

bonded magnets at temperatures as high as 1200°C to decompose resins, have also been reported (Ofuku, 1998). These methods for magnet powder recovery are typically complex and often involve the use of harsh chemicals and/or high temperatures. The complexity of these processes suggests that they may be cost-prohibitive to implement, especially when REEs prices are low. As a result, they cannot be deployed as routine recycling processes. Thus there is a need for developing cost-effective and process-efficient method for reusing and recycling REEs.

Developing deployable magnet recycling technology requires processes that are energy efficient, potentially profitable when deployed commercially and have less detrimental impact on the environment (Nlebedim and King, 2018). For bonded magnets, these conditions are easier to meet if minimal reprocessing is applied to enable reuse, rather than elemental recovery of the REE contents; which requires additional investments in energy and other resources. In fact, elemental recovery is unlikely to result in products that will be used in applications other than permanent magnets because REEs are often recovered as mixed oxides of constituent elements. In general, there needs to be a decision to reuse without much processing or elemental recovery of the REEs with minimal reprocessing. In our recent study, Sm-Co powder collected from industrial swarfs (wastes generated from post-manufacturing processing of magnets such as grinding, polishing, etc.) was studied for use in magnetic filaments for 3D printing of bonded magnets (Khazdozian et al., 2018).

In this work, additively printed Nd-Fe-B bonded magnets were used to develop a recycling process for bonded magnets, enabled by cryogenic pulverization (cryomilling) and subsequent remanufacturing of the bonded magnets. We are unaware of any previous study in which cryomilling enabled the recycling of bonded magnets, particularly those derived from additive manufacturing. Thermal, magnetic and mechanical properties have been studied to evaluate the

performance of the recycled magnets. The newly developed method is exemplified here by recycling of nylon bonded Nd–Fe–B magnets.

2. Experimental procedure

The experimental schematic of the recycling process is shown in Scheme 1. In this work, we utilized bonded magnets generated by a Big Area Additive Manufacturing (BAAM) process. Although the BAAM system is known to produce minimal or no waste (Li et al., 2016), the process reported in this work can easily be transferred to recycling end-of-life bonded magnets or wastes generated via any AM bonded magnet systems. The material used (Fig. 1a) was a product of purging the BAAM system before transitioning to a different material composition. The AM system deposits high-performance engineered thermoplastics and customized thermoplastic composites via melt extrusion processing, which enables rapid prototyping of parts completely unbounded in size and shape with minimal waste (Li et al., 2016).

The starting AM bonded magnets used contained 65 vol.% isotropic Nd-Fe-B powder (MQP-B+ -10118-070) and 35 vol.% Nylon-12. To achieve nearly fully dense magnet by compaction, it is critical to start with a composite powder that contain small and narrowly distributed particle sizes. Considering that 35 vol.% of the materials is composed of highly plastic nylon, it is extremely difficult to achieve pulverization by conventional grinding at room-temperature. Hence, to pulverize the AM bonded magnet effectively and to overcome plastic deformation, it was subjected to milling at liquid nitrogen temperatures (-196°C) for 30 min, using a 6870D Freezer/Mill-SPEX Sample Prep. Fig. 1(b) shows bonded magnet powder after cryomilling. For cryomilling, about 20 g of AM bonded magnets (plus bonded magnet filaments) were cut into small pieces (2-5mm is size) and placed in a polycarbonate jar with stainless steel cap along with

a stainless steel impactor. The jar was dipped in liquid nitrogen and allowed to cool down for 20 min before milling was commenced. Milling was performed for 30 min at the impact rate of 10 cps. In order to make recycled bonded Nd-Fe-B magnets, the cryomilled powders were compression molded in a cylindrical die using a uniaxial pressure of 10 MPa and at three different temperatures: 200 °C for specimen #1, 185 °C for specimen #2 and 175 °C for specimen #3. Fig. 1c shows the images of 9.6×19.25 mm (*l*×*d*) cylindrical size magnets produced. This experiment was not performed in an inert atmosphere since the nylon-12 binder helped limit oxidation, which simplifies the recycling process.

2.1 Materials Characterizations

Morphologies of the starting AM bonded magnets (Fig. 1a), cryomilled powder (Fig. 1b) and recycled magnet (Fig. 1c) were examined by scanning electron microscopy (FEI Teneo LoVac). The magnetic properties were obtained at 300K with a vibrating sample magnetometer up to a maximum applied magnetic field of 30 kOe. Thermal characteristics of the composite pellets were determined with a differential scanning calorimeter (DSC). The mechanical properties of the sample were evaluated by compression method (according to ASTM D695-15) using a cylindrical sample of 12.7 mm diameter and 25.4 mm height. The test was carried out at ambient conditions using Instron 5569 testing machine at a constant crosshead displacement rate of 1 mm/min. The crosshead separation was used to evaluate the sample displacement. The density of the fabricated magnets were measured by water displacement method.

3. Results and Discussion

Surface morphology of the starting AM bonded magnets are presented in SEM micrographs in Fig. 2a. The plate-shaped bright magnetic particle sizes in the range of 20–200 μm, separated by

dark nylon polymer binder matrix were observed. The particle size of magnetic powder plays an essential role in determining powder to nylon ratio, magnetic and mechanical properties (Grujić et al., 2010; Grujic et al., 2010). The morphology of the cryomilled powders is shown in Fig. 2b. It can be seen that after cryomilling for 30 min, the size of plate-shaped Nd-Fe-B is reduced to smaller dimensions, [$<100 \mu\text{m}$], compared to the starting AM magnets. Fig. 2c shows the surface morphology of recycled bonded magnets. Compared with the starting AM magnet material's morphology, more densely packed particles are observed in recycled bonded magnets. These small sized plate-like particles would result in higher packing density under the optimal compression conditions (Li et al., 2006; Zhang et al., 2009).

The thermal properties of the materials studied by DSC are shown in Fig. 3. $T_{m,onset} = 195^\circ\text{C}$ is the onset of the melting temperature (T_m) and provides guidance for the warm compaction process because knowledge of the melting characteristics is necessary for the successful consolidation of the powder-binder mixtures. If the melting requirements are not satisfied then the binder may not melt or over-melt; either of which will result in non-optimal compaction and possible deterioration of magnetic and mechanical properties. It can be seen that the $T_{m,onset}$ are similar for the starting AM bonded magnets and cryomilled powders (195°C) but slightly higher ($\sim 198^\circ\text{C}$) for the recycled bonded magnets. This may be related to higher packing fraction of the powder in the remade bonded magnets. Compression molding is known to result in higher packing fractions compared to injection molding which is more comparable to 3D printing. The peaks of the melting are similar for the starting AM and the recycled bonded magnets, compared to the cryomilled powder. Also, the DSC plot for the cryomilled powder did not show the shoulder immediately after $T_{m,onset}$ as observed for the other two samples. This is likely due to more uniform melting in

the less dense cryomilled powder, compared to the other two (Phang et al., 2004). It is likely that some of the melting characteristics are related to the effect of thermal cycling on the nylon binder.

The room temperature magnetic hysteresis plots of the starting and recycled magnets are shown in Fig. 4a and the corresponding magnetic properties shown in Table 1. The approach for obtaining the properties from the hysteresis plots and their significances have been explained (Ener et al., 2018; Gandha et al., 2014; Goll and Kronmüller, 2000; M. P. Paranthaman, 2016). For all the recycled bonded magnets, remanence (B_r or M_r), magnetization at maximum applied field (M_s), coercivity (H_c) and maximum energy product ($(BH)_{max}$) are within 96 – 104%, 100 – 106.5%, 90 – 96% and 86 – 100% respectively, compared to the starting AM bonded magnet. The energy stored in a permanent magnet can be obtained from the product of magnetic induction (B) and magnetic field (H), where $B = H + 4\pi M$ and M is the magnetization (Fig. 4a). $(BH)_{max}$ is the maximum energy that can be stored. These results indicate that the recycled bonded magnets retained the hard magnetic behavior of the original AM bonded magnets.

Fig. 4(b)-(d) are plots of the H_c , B_r and $(BH)_{max}$ of starting and recycled bonded magnets, comparing the performance of the magnets in the temperature range of 250 – 400 K. The H_c for all specimens are comparable to that of the starting AM bonded magnet, especially in the range of 300 – 400 K. 200 °C is the optimum processing temperature for the recycling process because at all measurement temperatures, specimen #1 processed at 200 °C has comparable $(BH)_{max}$ and better B_r than the starting material. This is likely because at 200 °C (compared to processing at 185 °C for specimens #2 and 175 °C for specimen #3), the nylon binder is most softened to promote better compaction. This is also reflected in the result that specimen #1 has the highest density. At optimum temperature, compaction molding should result in a better packing fraction than 3D printing, which compares more to extrusion molding.

Recycled bonded magnet can be used for applications only if they satisfy requirements of thermal, magnetic and mechanical properties. Having shown the thermal and magnetic properties, mechanical properties of the bonded magnets are presented next. Uniform particle distribution and good adhesion between polymer matrix and magnetic filler particles are crucial for superior mechanical properties. The compression stress-strain curves for three recycled bonded magnets (specimen #1, specimen #2, and specimen #3) are shown in Fig. 5. The inset shows picture of mechanical property testing set-up (left) with recycled bonded magnet and the image of the mechanical failure of bonded magnet after compression test (right). Compression test results indicate that there is a variation in the absolute strength and ultimate strain among the specimens, depending on the processing temperatures. The compression molding of specimen #1 at 200°C results in better molding due to decrease in viscosity which provide uniform cross-linking of polymer and less porosity compared to samples prepared at 185 °C specimen #2 and 175 °C specimen #3 (Sato et al., 1985). The mechanical behavior of all samples are typical of that of semi-crystalline thermoplastic polymer, where plastic deformation occurs before the failure of polymer. Highest compressive strength achieved was 85.4 MPa for specimen #1, which is comparable to current commercially available compression bonded Nd-Fe-B magnet (~80 MPa to 120 MPa) (Alliance, 2018). These results agree with previous explanation that specimen #1, processed at 200 °C, is better compacted compared to specimen #2 and specimen #3. Since the recycled magnets comprise ~65 vol% of magnet powder, the compressive strength can be further increased by using materials with higher magnet powder loading due to filler reinforcement effect. The Young's modulus is as high as 4.48 GPa and the ultimate strain ranges from 4-7%; similar to the literature values (Li et al., 2016).

4. Conclusions

A method for recycling additive manufactured bonded magnets to remake bonded magnets has been developed. In this process, no harsh chemicals were used and the potential for advancing manufacturing of recycled products has been demonstrated. A recycled magnet showed improved density and remanence, compared to the starting bonded magnets. The improved magnetic properties have been shown to be a result of optimized cryomilling of the powder, which did not result in any observable degradation of the magnet material. Comparability of the magnetic, thermal and mechanical properties of the recycling process at 200 °C with that of the original AM magnet indicates that this process is suitable for remaking bonded magnets for reuse in applications. In application, this process would also minimize processing complexities as it was performed in air, i.e. without the need for inert atmosphere. However, for other binders such as PPS, inert atmosphere may be necessary. The use of simple physical cryomilling, elimination of harsh chemicals and retaining of the original magnetic properties, all indicate that the process used is eco-friendly and can help to ensure sustainability in materials for permanent magnet production.

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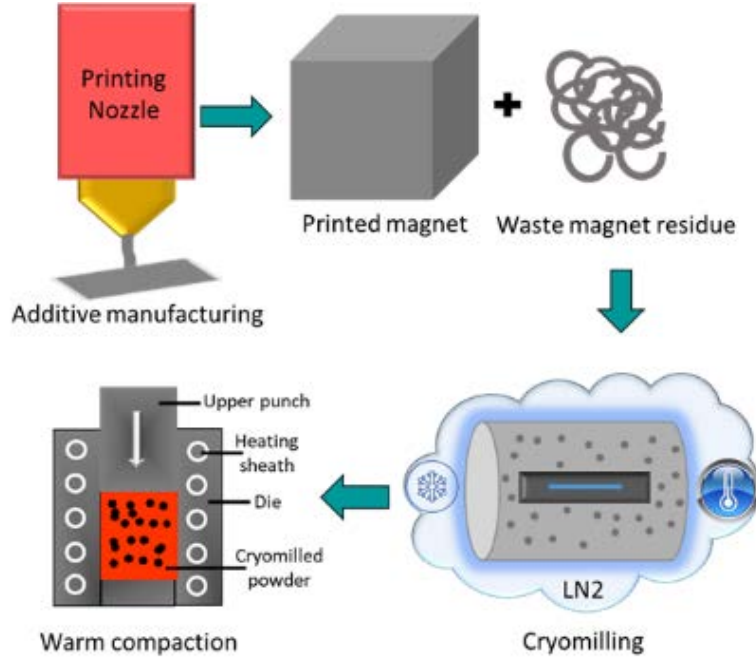
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Table 1: Summarized magnetic properties of the AM bonded magnet, cryomilled powder and recycled bonded magnet at 300 K.

Figure captions



Scheme 1. Experimental procedure for recycling additively printed bonded magnets.



Fig. 1. (a) Starting AM bonded magnets (65 vol.% Nd-Fe-B in nylon), (b) cryomilled powder, and (c) recycled bonded magnets.

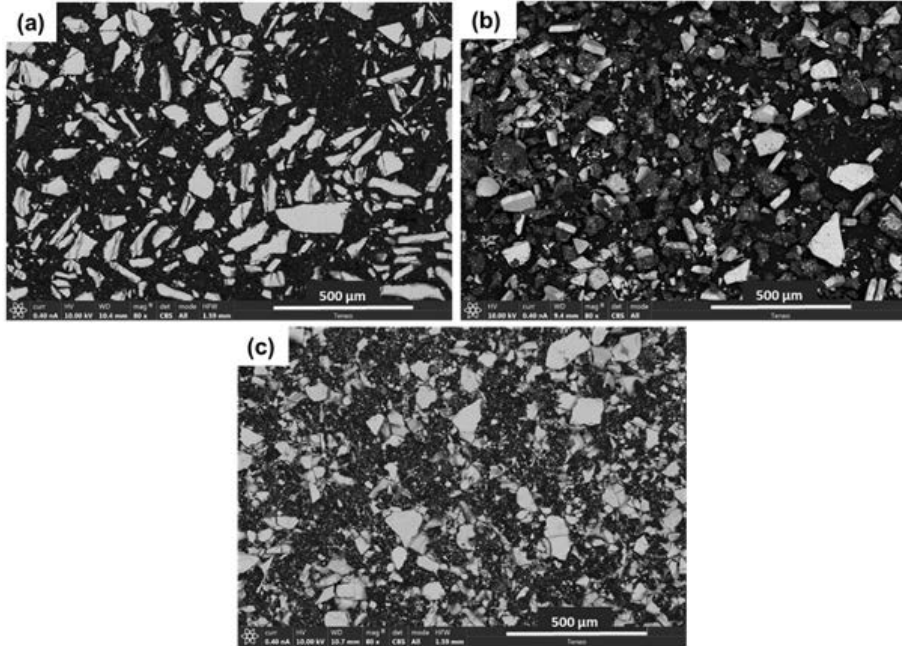


Fig. 2. SEM micrograph of (a) the starting AM bonded magnet, (b) cryomilled powder, and (c) recycled bonded magnet (scale bar 500 μm).

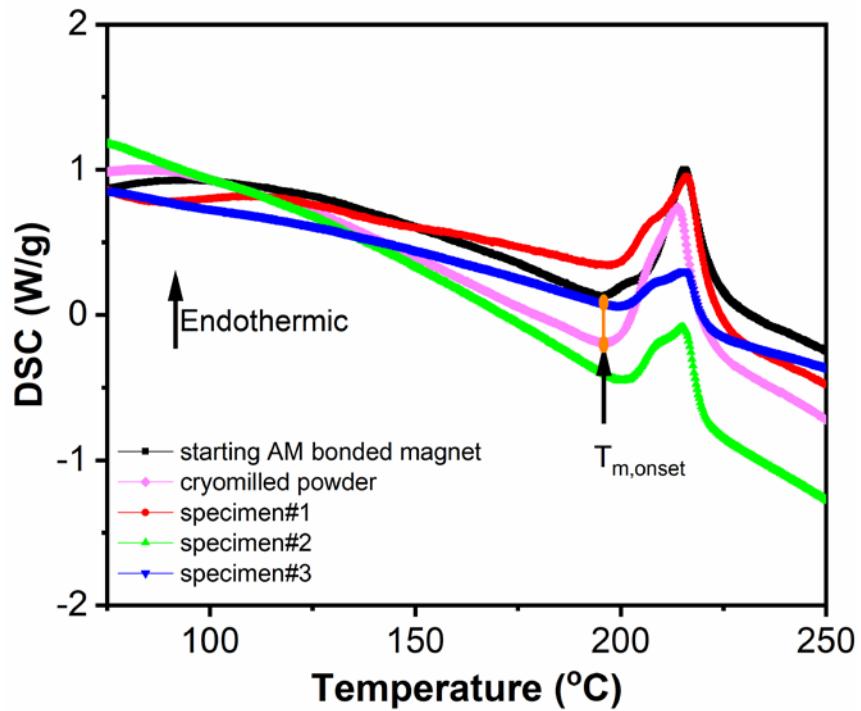


Fig. 3. DSC plot of starting AM bonded magnet, cryomilled powder and recycled bonded magnet.

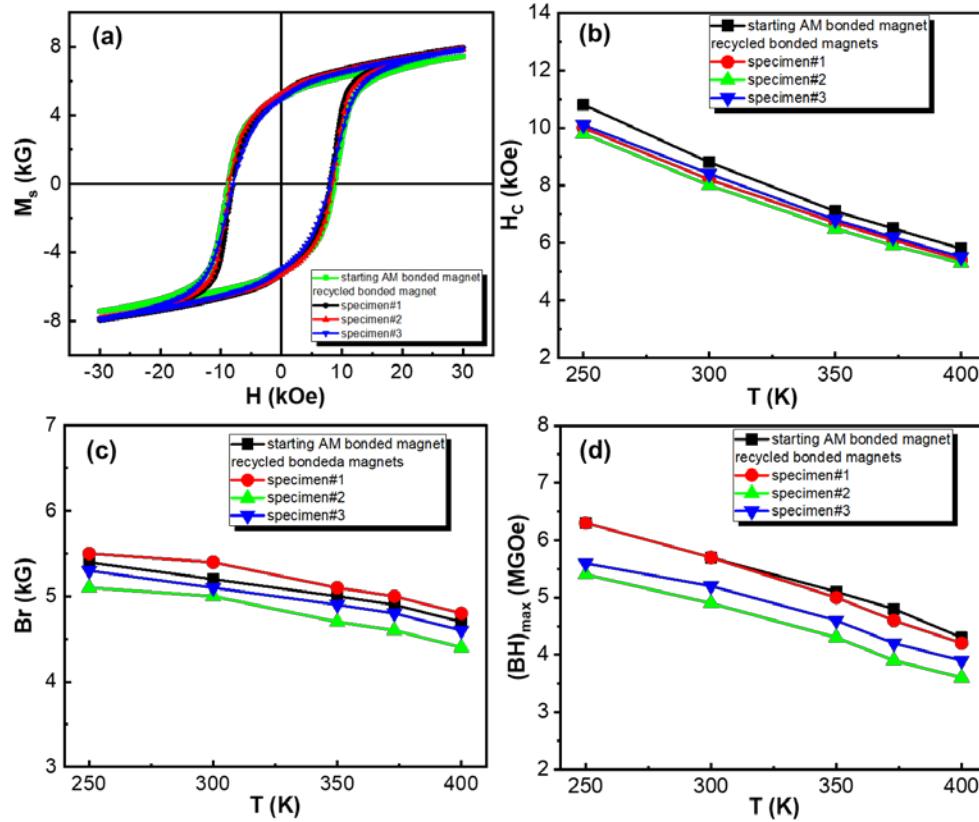


Fig. 4. (a) Room temperature hysteresis plot of starting AM bonded magnet and recycled bonded magnet, (b-d) magnetic properties of AM bonded magnet and recycled bonded magnet as a function of temperature.

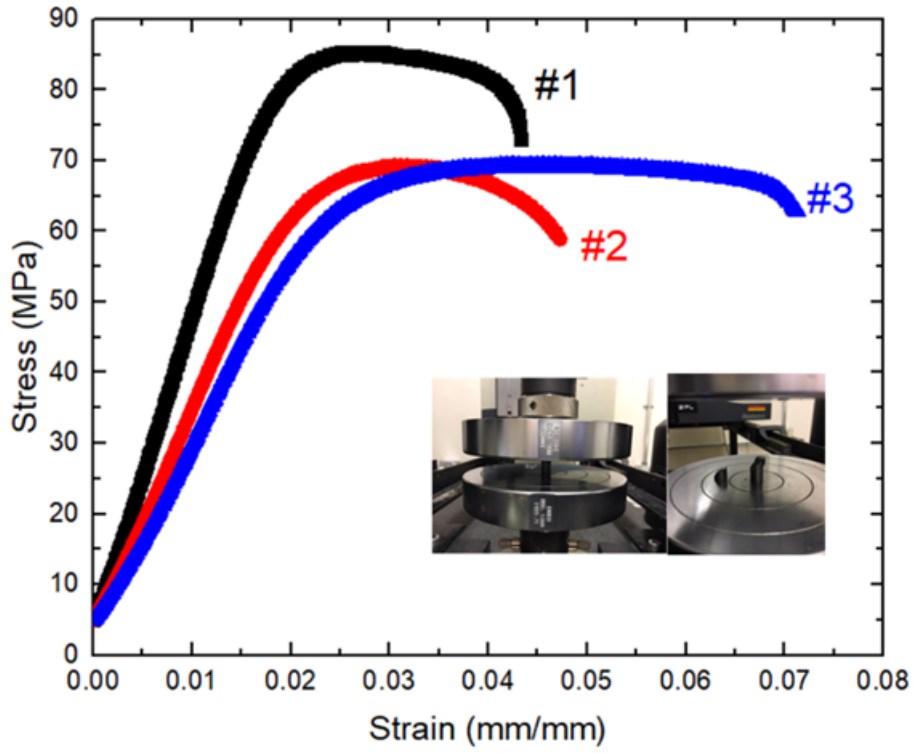
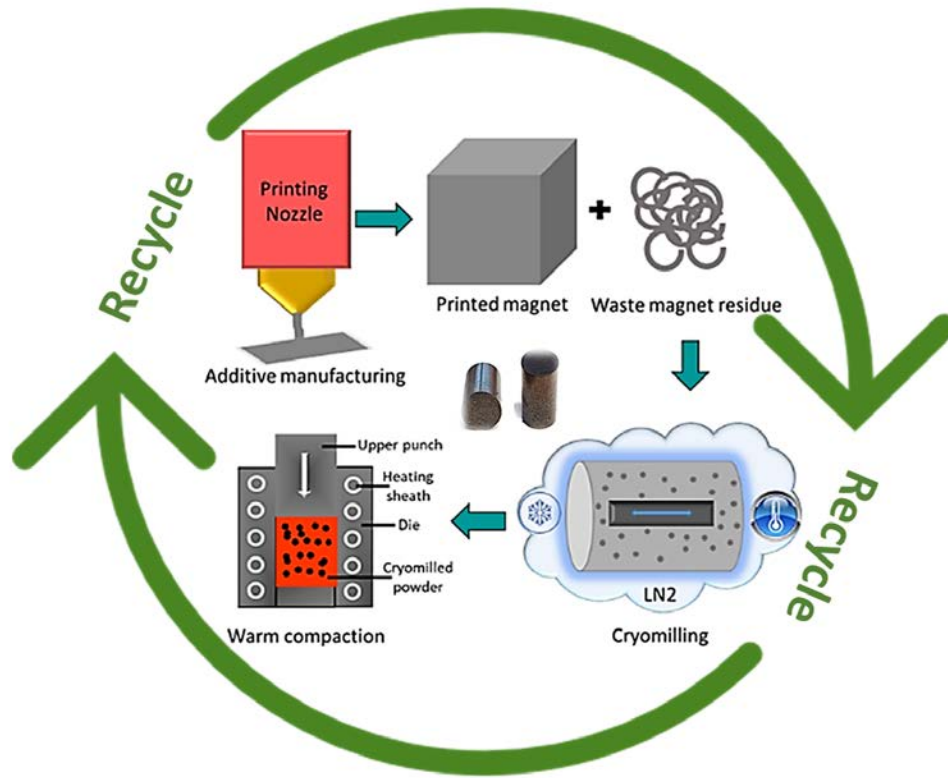


Fig. 5. Compression stress-strain curves of recycled bonded magnet warm compacted at 200 °C specimen #1, 185 °C Specimen #2 and 175 °C specimen #3.



Graphical Abstract