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Chemical Sciences Division
Advanced Manufacturing Office

Additive Manufacturing of Near-net Shaped Permanent Magnets

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1. Abstract

The technical objective of this technical collaboration phase I proposal is to fabricate near net-shaped permanent magnets using alloy powders utilizing direct metal deposition technologies at the ORNL MDF. Direct Manufacturing using the POM laser system was used to consolidate $Nd_2Fe_{14}B$ (NdFeB) magnet powders into near net-shape parts efficiently and with virtually no wasted material as part of the feasibility study. We fabricated builds based on spherical NdFeB magnet particles. The results show that despite the ability to fabricate highly reactive materials in the laser deposition process, the magnetic coercivity and remanence of the NdFeB hard magnets is significantly reduced. X-ray powder diffraction in conjunction with electron microscopy showed that the material experienced a primary $Nd_2Fe_{17}B_x$ solidification due to the undercooling effect (>60K). Consequently the presence of alpha iron phase resulted in deterioration of the build properties. Further optimization of the processing parameters is needed to maintain the $Nd_2Fe_{14}B$ phase during fabrication.

2. Statement of Objectives

This phase I technical collaboration project (MDF-TC-2016-65) was started on July 31, 2015 and was completed on July 31, 2016. The collaboration partner, Arnold Magnetic Technologies (Arnold) traces its roots back to the late 1800'**s.** Arnold is one of the only western companies that produces alnico, RECOMA® brand samarium cobalt, ferrite bonded magnets, and neodymium iron boron bonded magnets (both injection molded and compression bonded). In addition they provide assembly capabilities from prototype to production of assemblies from grams to hundreds of kilograms. Arnold is one of the most modern and is the largest U.S. based magnetics company. Today the three main business units are: Precision Magnets and Assemblies Group – magnets, assemblies and reprographics; Precision Thin Metals – ultra-thin metal foil and strip for both magnetic and non-magnetic uses; and Flexmag Industries – flexible bonded magnets (sheet and strip) for the advertising and OEM industries. The main objective of this project is to use additive manufacturing technologies to fabricate near-net shaped permanent magnets with fully dense sintered magnet properties. The magnetic and microstructural properties of the magnets have been characterized and fully evaluated.

3. Benefits to the Funding DOE Office's Mission

The use of permanent magnets in green energy technologies has increased dramatically. Rare-earth based permanent magnets are the key components in environmentally friendly technologies such as electric transportation (automotive, marine and aerospace) and direct or hybrid drive wind turbines. The high cost of rare earth raw materials and issues related to adequate and dependable supply of heavy rare earths, e.g. dysprosium, are holding back the adoption or expanded use of these technologies. Considering that a large portion of US rare-earth magnet manufacturing capability was transferred to China in the last decade [1], novel additive manufacturing techniques that avoid or minimize the use of rare-earth elements especially the heavy rare earth elements, need to be developed to reduce US vulnerability to uncertain rare earth material supply and high and volatile pricing [2-7]. Conventional manufacturing methods to make permanent magnets are energy inefficient partly because of the traditional sintering technology but also due to processing material that turns into waste product at or near the end of the manufacturing process. Recycling the waste material is not straight forward. The rare-earth elements tend to oxidize during the manufacturing process (grinding, diamond saw slicing, core drilling, etc.). Therefore, alternative ways need to be explored that not only require less energy during manufacturing but also generate less waste material, especially the fine partially oxidized waste material. One promising technique is a laser-based DMD (Direct Metal Deposition) processing system, where an optical heat energy source such as an industrial dye laser is used to directly fabricate metal parts [2]. The potential impacts of research include: developing near net-shape magnets without tooling; and the Critical Materials Institute (CMI) hub additive manufacturing project will benefit from this feasibility study. Overall, the advancement of the utilization of the additive techniques could lead to the adoption of additive manufacturing technology by magnet manufacturing companies and lead to job growth and higher US global manufacturing competitiveness.

4. Technical Discussion of Work Performed by All Parties

Additive manufacturing (AM) is a near-net shape manufacturing technique, which can produce final parts with minimal material waste [8]. Several AM techniques are available and are broadly classified based on the nature of the process as *powder bed* and *powder blown* [9]. In this work a powder blown directed energy deposition technique, has been used. This process is based on the coaxial laser cladding process where the part is built in layers. The primary process parameters used to control the deposit geometry and part density are the laser power, travel speed and powder feed rate. So far only limited feasibility studies have been attempted to fabricate magnets using additive manufacturing. Since the additive manufacturing process experiences naturally rapid cooling rates, the interface velocities and the solidification structure can be significantly different from those of the conventional manufacturing process. In addition it has been shown that by increasing the solid-liquid interface velocities it is possible to obtain a $Nd_2Fe_{14}B$ microstructure. Due to the rapid interface velocities in the laser DMD processing it is hypothesized to shift the solidification mode from non-magnetic or soft phase such as primary γ iron to primary $Nd_2Fe_{14}B$ thereby maximizing the magnetic properties. Hence the aim of this work was to study the feasibility of fabricating NdFeB magnets using the laser powder-blown directed-energy deposition.

Commercial Magnequench anisotropic MQA-38-14 (designated as MQA) and isotropic spherical MQP-S-11-9 (designated as MQP) NdFeB powders were used for this study. Fabrication was performed using the DM103D laser directed-energy system with a 1kW high power diode laser operating at a wavelength of 910 nm. The depositions were performed under an argon gas blanket, where the oxygen content was <5 ppm, to avoid oxidation of the rare earth metals. In addition to the cover blanket a stream of argon was also delivered coaxially along with the powder to shield the melt pool from oxidation and contamination. The builds were fabricated using a laser power of 500W a powder feed rate of 5.3 g/min and a travel speed of 700 mm/min. A total of 25 layers were built and the approximate build height was about 12.5 mm with a height of 0.5mm/layer. Following fabrication of the builds, magnetization properties and phase identification of the builds were carried out [10].

As shown in Fig. 1, NdFeB builds were printed. Phase composition, microstructure, and magnetic flux were measured before and after printing to determine the effect of laser deposition on these properties. As shown Fig. 1 (c) and (d), the microstructure changed significantly after laser deposition. Fig. 2 shows the hysteresis loops for both as-received MQP powders and build after laser deposition. The magnetization for the build at 35 kOe reached about 127.2 emu/g whereas the starting isotropic MQP powder with a theoretical density of 7.43 g/cm^3 reached only 113.0 emu/g. There is a small normal hysteresis (H_{cB})of 500 Oe retained in this material, but the vast majority of the coercivity was lost after laser processing implying that hard magnetic phase did not survive. Normally the formation of magnetic properties is associated with the formation of the tetragonal $Nd_2Fe_{14}B$ phase. The poor magnetic properties could be attributed to the absence of $Nd_2Fe_{14}B$ or the creation of alternate, possibly soft phases. The poor magnetic properties could be attributed to the absence of $Nd_2Fe_{14}B$. XRD patterns obtained from the as-received MQA powder and the as-built sample are shown in Fig. 3. Fig. 3 shows that the as-received powder contained almost 100% $Nd_2Fe_{14}B$, while the as-built sample showed the presence of additional phases. Peaks belonging to $Nd_2Fe_{17}B_x$ and α-Fe phases were identified in addition to the $Nd_2Fe_{14}B$ phase. The lattice parameters of the constituent phases were obtained from the Rietveld refinements. Accordingly, the lattice parameter of alpha-Fe was found to be a=2.8701 Å, which is close to the literature value [11-

17]. On the other hand, the lattice parameters of the $Nd_2Fe_{14}B$ phase were calculated to be a=8.7963 Å, c=12.1923 Å and a=8.7974 Å, c=12.1509 Å for the as-received powder and the as-built sample, respectively. While the a-lattice parameter did not change much, the shift in the c-lattice parameter suggests that there may have been elemental composition change leading to the formation of a magnetically soft phase. To confirm if the shift in the lattice parameter is driven by compositional change atom probe tomography needs to be performed. The observed lattice parameters from the as-built sample agree with those reported for $Nd_2Fe_{17}B_x$[16]. The formation of secondary phases also leads to the diminishing of the desirable hard magnetic phase. The presence of alpha iron will shunt the field internally reducing H_{cJ}. Hence to rationalize the drop in properties the solidification sequence in the material needs to be understood during non-equilibrium conditions. Typically $Nd_2Fe_{14}B$ is expected to solidify by a primary γ iron formation and then subsequently a peritectic reaction occurs where the L+ γ iron transforms to the $Nd_2Fe_{14}B$ phase [18]. However due to the rapid cooling rates encountered during the laser deposition process, the peritectic reaction can be suppressed which results in the absence of the $Nd_2Fe_{14}B$ in the build [11, 13, 16].

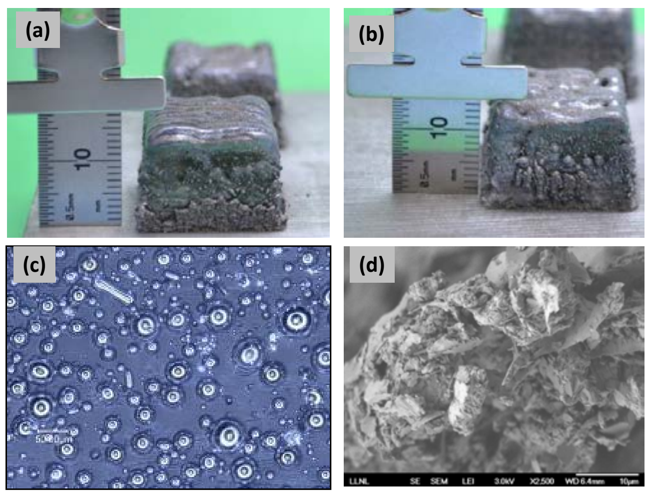

Fig. 1. Cross-sectional optical images of the laser deposited builds ((a) and (b)). SEM images of (c) as received commercial MQP-S-11-9 powders, and (d) Build after laser deposition.

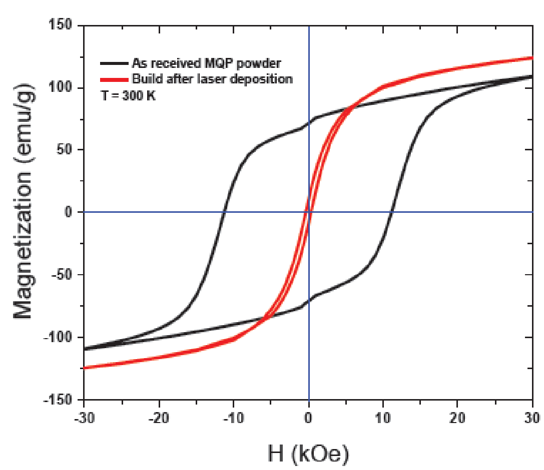

Fig. 2. Magnetization curve of the starting MQP powder and the build. Note the sharp drop in coercivity and remanence of the magnets after processing [10].

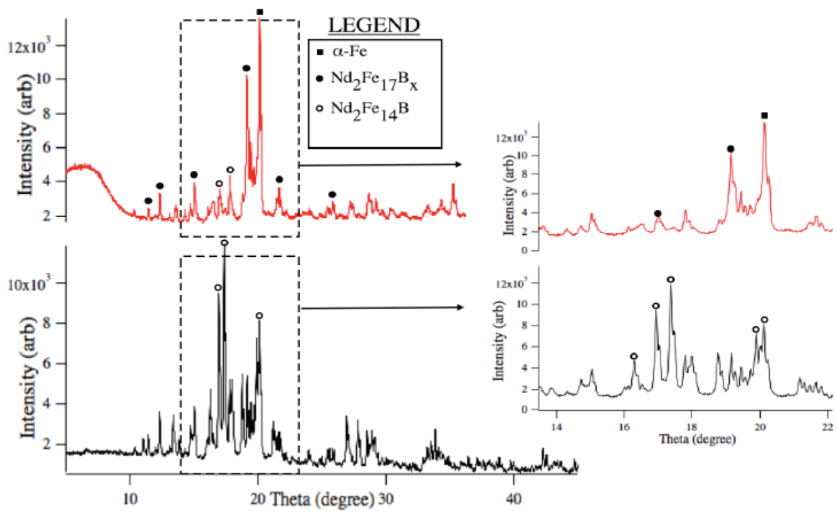

Fig. 3. XRD patterns obtained from the powder (shown below in black) and the build (shown above in red). Note the sharp decrease in the intensity of the peaks corresponding to the $Nd_2Fe_{14}B$ hard magnetic phase in the additively manufactured samples. Also note the corresponding increase in the intensity in the alpha iron peak after laser processing [10].

It is clear that to prevent the loss in the coercivity and remanence in the build it is necessary to retain the $Nd_2Fe_{14}B$ phase. Hence we need to engineer the solidification sequence so that the build solidifies as a primary $Nd_2Fe_{14}B$. This can be promoted by maintaining an undercooling of <60 K to facilitate nucleation. The undercooling can be controlled by careful selection of process parameters and by controlling the interface growth velocities. In addition post process thermal solutionizing may also help in decomposing the non-equilibrium $Nd_2Fe_{17}B_x$ and the formation of $Nd_2Fe_{14}B$ phase in the build.

5. Subject Inventions (As defined in the CRADA)

Further investigation of direct metal deposition is needed to explore the possibility of new invention. The following manuscript will be published based on some of the results from this research:

N. Sridharan, E. Cakmak, Fred A. List, Huseyin Ucar, Steve Constantinides, S.S. Babu, Scott K. McCall, and M. Parans Paranthaman, "Rationalization of solidification mechanism of neodymium-iron-boron magnet obtained after laser directed-energy deposition process," (2016) (submitted).

6. Commercialization Possibilities

Additively printed sintered magnets offer a better option for making low cost intricate shapes from magnet powder feed. Here we evaluated a novel alternate approach – direct metal deposition (DMD) - to fabricate near-net-shape isotropic NdFeB magnets. Magnetic and microstructural characterizations indicate that while magnetization ($4\pi M_{sat}$) was improved, the permanent magnetic properties degraded as a result of laser deposition. Further process improvements are needed in order to improve coercivity (H_{cJ}) to compete with sintered magnets.

7. Plans for Future Collaboration

Additive manufacturing offers significant advantages such as cost effectiveness (no tooling required), fast speed (simple procedure), and capability of producing parts unlimited in size and shape. Therefore, DMD provides an effective method in realizing arbitrary shape with minimum cost and waste, and has the potential to revolutionize large-scale industry production of sintered magnets. In the future work, the effect of laser powder and post annealing will be investigated. Following this work, ORNL plans to continue working with Arnold to develop an additive process towards the goal of achieving printed magnets with much improved magnet properties that are suitable for many industrial magnet applications.

8. Conclusions

In this work, we report the feasibility of using the directed energy additive manufacturing technique to handle reactive rare earth elements without any significant loss of alloying elements. Though builds were successfully fabricated the magnetic properties were significantly reduced from that of the starting alloy. X-Ray diffraction showed a large fraction of $Nd_2Fe_{17}B_x$ and some $Nd_2Fe_{14}B$ along with a significant fraction of α-Fe. Further process optimization is necessary to improve the magnetic properties.

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