

Alignment of Magnetic Particles in Anisotropic Nd-Fe-B Bonded Magnets

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Abstract

High performance anisotropic bonded magnets need high volumetric filling fraction of magnetic powder in a polymer binder and their good alignment while possessing good enough mechanical properties. The correlation between the degree of alignment (DOA) and the volumetric filling fraction of magnetic powder, the type of binder and the processing parameters have been studied using Nd-Fe-B anisotropic bonded magnets. Thermomagnetic analysis and differential scanning calorimetry (DSC) results show the magnetic powder can be well aligned in a bonded magnet under an external magnetic field at a temperature higher than the binders' melting point by 20-60 K, which depends on the type of magnetic powder and kind of binder. The DOA results mainly from the interplay between the Zeeman energy ($E_{app.}$) and the inter-particle static energy ($E_{stat.}$) in bonded magnets at the chosen alignment temperature and the magnetic fields. A good alignment can be achieved by an alignment magnetic field which is about twice as its coercivity in an anisotropic Nd-Fe-B bonded magnet, which is confirmed by our experimental and modeling results.

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1 Introduction

Nd-Fe-B bonded magnets have been widely applied in data storage and consumer electronic devices.[1–4] To produce bonded magnets, powders are mixed with thermoset or thermoplastic-polymers for compression or injection molding methods, respectively. With appropriate processing design, both compression and injection molding can produce magnets to a near-net shape. Recently, Additive Manufacturing (AM) is becoming a promising technique for bonded permanent magnet (PM) production.[5–12] As opposed to conventional subtractive methods of manufacturing, AM has the advantage of components being made near the net shape without any required tooling and limits wasting of raw materials.

Like full density sintered magnets, there are two types of bonded magnets, i.e. isotropic and anisotropic magnets. The magnetic easy-axis orients randomly and along a preferential direction in isotropic- and anisotropic magnets, respectively. Theoretically, the maximum energy product of anisotropic magnets is four times as that of isotropic magnets.[13] There are two main kinds of anisotropic Nd-Fe-B magnetic powders commercially available for producing bonded magnets. One is MQA (Magnequench Anisotropic) Nd-Fe-B anisotropic magnetic powders, produced by hot-deformation (or die-upsetting) of powders derived from melt-spun Nd-Fe-B ribbons. [14,15] The other one is Magfine Nd-Fe-B anisotropic magnetic powder prepared using a Hydrogenation-Disproportionation-Desorption-Recombination (HDDR) route.[16,17] Each particle of the anisotropic magnetic powder of MQA- or Magfine- Nd-Fe-B contains several millions of $\text{Nd}_2\text{Fe}_{14}\text{B}$ grains. However, the distribution of magnetic easy axes of $\text{Nd}_2\text{Fe}_{14}\text{B}$ grains (or [001] crystallographic orientation) is not completely aligned along a single direction in each particle[15,18]. The degree of alignment (DOA) is generally evaluated by remanence ratio (M_r/M_s).

The DOA of magnetic powders determine the maximum possible DOA value and maximum energy product of a final bonded magnets. To distinguish between the DOA of Nd-Fe-B particles and that of final bonded magnets, we name the DOA of magnetic powder as intrinsic DOA (Φ_i) and that of final bonded magnet as total DOA (Φ_t) in this paper.

In addition to the performance of magnetic powder itself, a higher remanent magnetization and energy product can be achieved with a higher loading proportion of magnetic powder and good magnetic alignment in polymer matrix for anisotropic bonded magnets. The magnetic remanence is proportional to the volume fraction of magnetic powder and total DOA of bonded magnets. The maximum energy product is proportional to the square of magnetic remanence or square of the volume fraction of the magnetic powder. However, the maximum loading fraction of magnetic powder is limited by the requirement for sufficient mechanical properties of bonded magnets. Typically, in injection molding methods, up to 65 volume % of magnet fractions are filled in a polymer; however, in compression molding methods, up to 80 volume % of magnet fraction are filled [19,20]. The mechanical properties depend on the type and content of binder and its distribution in bonded magnets. DOA is intimately related to the type of magnetic powder, binder, temperature and applied magnetic field for alignment.[21,22] To achieve good magnetic and mechanical properties in bonded magnets, establishing optimized processing conditions is very important.

In our past work, we successfully prepared high performance isotropic and anisotropic Nd-Fe-B bonded magnet using additive manufacturing technology and reported the magnetic and mechanical properties.[7,23–25] As part of our endeavor to advance additive manufacturing of

high-performance Nd-Fe-B bonded magnets, we study different processing parameters that contribute to the resultant magnetic alignment and hence magnetic properties of anisotropic Nd-Fe-B bonded magnets. The processing parameters of interest include powder-loading fraction, rheological states of polymer binders, magnetic interaction between the particles and applied magnetic field during the alignment process. Controlling the resultant hard magnetic properties in anisotropic bonded magnets requires understanding the various physics interacting to produce such properties. The goal of this work is to elucidate the physics that drive orientation of magnetic easy axes to produce high energy density Nd-Fe-B bonded magnets. Ultimately, such understanding can be adopted in various approaches for producing bonded magnets, including injection and compression molding and additive manufacturing techniques.

2 Experimental methods

We selected two kinds of anisotropic Nd-Fe-B powders (MQA-38-15 and Magfine-15) to prepare bonded magnets. These are two key types of commercially available anisotropic Nd-Fe-B powders for bonded magnets manufacturing and will help to ensure that the findings of the present work can have a broader impact. To determine the intrinsic DOA (Φ_i) of magnetic powder, we measured the magnetic hysteresis loops. Slightly milled MQA and Magfine Nd-Fe-B powder (less than 0.1 mm) were mixed with paraffin wax, aligned under magnetic field of 30 kOe at 323 K (the melting point of paraffin wax is 312 K), then cooled to room temperature under field for the magnetic measurements. To achieve high magnetic alignment of powder, the volume of magnet powder was about 30% in liquid wax. The observed *DOA* reflects the degree of *c*-axis crystal texture of 2:14:1 phase in Nd-Fe-B powder, i.e. the intrinsic DOA (Φ_i). Figure 1 shows the normalized magnetic hysteresis loop. The remanence ratio or intrinsic DOA (Φ_i) for MQA and Magfine powder are 0.91

and 0.92, respectively. The bonded magnets made with MQA and Magfine are expected to have a total DOA (Φ_t) less than the corresponding DOA (Φ_i) values of powder.

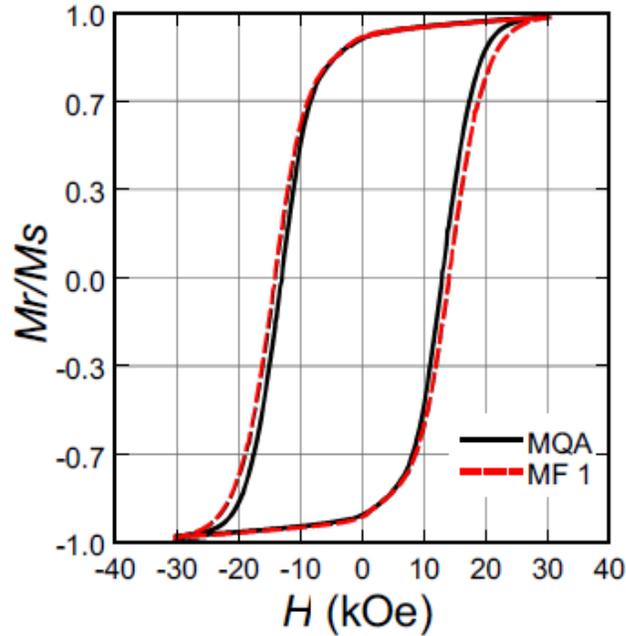


Figure 1. Magnetic hysteresis loop of MQA (black solid line, MQA) and Magfine (red dotted line, MF 1) anisotropic Nd-Fe-B magnetic powders.

Two types of binders (Nylon12 and PPS) were selected to fabricate bonded magnets. Here, different loading fractions of Nd-Fe-B powder were mixed with binders and warm-compacted into bonded magnets with a diameter of 8mm and a length of 5mm. For the Nylon-12 and PPS binders, the temperatures of warm compaction were 450 and 550 K, respectively. The relative mass content of magnetic powder and binder were used to control the volume fraction of magnetic powders in the bonded magnets. Here, it is assumed that the binders and magnetic powders have their theoretical density and there is no porosity in bonded magnets. The correlation between the density of bonded magnets and the loading fraction of magnetic powders should be linear. As shown in

Figure 2, the linear relationship between density and loading fraction was confirmed for all the bonded magnets with the two kinds of magnetic powders and the two types of binders.

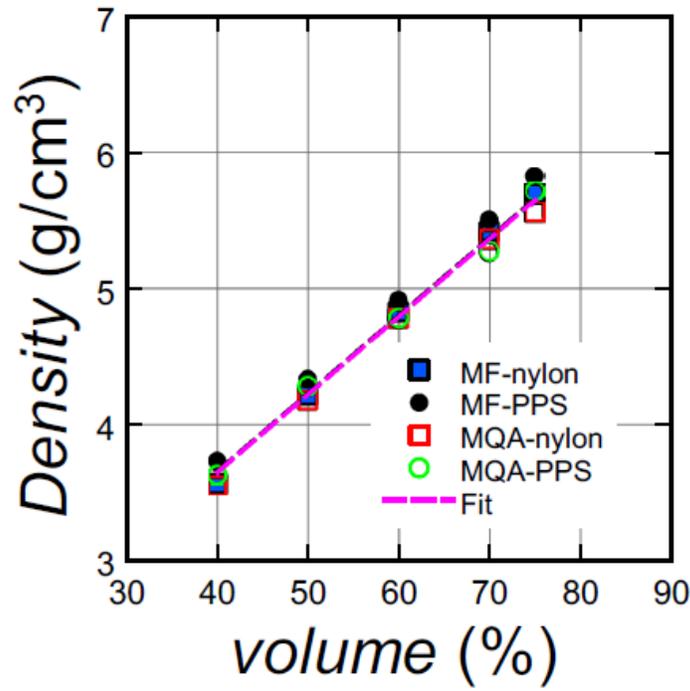


Figure 2. Correlation between density and loading fraction of magnetic powder in Nylon-(circle) and PPS- (square) Nd-Fe-B bonded magnets. Open and solid symbols are for MQA and Magfine powder, respectively.

Prior to magnetic alignment, magnetic hysteresis loops of the as-prepared magnet with randomly oriented particles were measured at 300 K. To magnetically align Nd-Fe-B powder in the as-prepared bonded magnets, a post-compaction magnetic field alignment was performed with an applied external magnetic field of up to 30 kOe. Samples were heated in magnetic fields from 300 to 530 K (Nylon-12) or 630 K (PPS) with a dwell time of 15 min at 530 K or 630 K, and then cooled down to 300 K. More details have been reported before. [24] After each alignment process, the magnetic hysteresis loops were measured again at 300 K. All the magnetic hysteresis loops were measured using a Quantum Design Versalab VSM magnetometer. The total DOAs (Φ_t) were determined using the remanent magnetization ratio of bonded magnets after magnetic alignment.

To determine the melting point of binder in bonded magnets, DSC measurements have been performed using a NETZSCH STA449F3 Jupiter thermal analyzer.

3 Results and discussion

3.1 Effect of loading fraction on magnetic alignment and hard magnetic properties

Figure 3(a) displays a typical magnetic hysteresis loop of MQA Nd-Fe-B Nylon-bonded magnet with a magnetic powder loading fraction of 75 vol%. After magnetic alignment (at 30 kOe), the magnetic remanence increases from 4.0 to 8.2 kGs, while the intrinsic coercivity H_{ci} changes from 9.8 to 10.8 kOe. The enhancement in remanence is due to the magnetic alignment of the easy axis of the magnetic powder in bonded magnets. The alignment induced enhancement of coercivity is ascribed to the coercivity being mainly controlled by the nucleation of reversal magnetic domains in MQA- Nd-Fe-B powders [26]. Further, the inductive coercivity increases from 2.9 to 5.9 kOe (Fig. 3b), resulting from the increase in remanence B_r . The maximum energy product increases from 2.9 to 13 MGOe. As shown in Fig. 3c, the magnetization has a jump at a temperature of 490-500 K during annealing under external applied fields. This temperature is slightly higher than the melting point of Nylon-12 (450 K), which means the binder (Nylon-12) is melted and at a low viscosity state. The low viscosity of the binder reduces friction between binder and particle and facilitates the magnetic powder alignment under magnetic field. This is a necessary condition to achieve alignment of magnetic particles in Nd-Fe-B bonded magnets during magnetic alignment process (see details on sample and measurement in the section “Experimental method”). Similar situations are observed with Nd-Fe-B bonded magnets with PPS binder (See Fig. S1 in the supplemental materials).

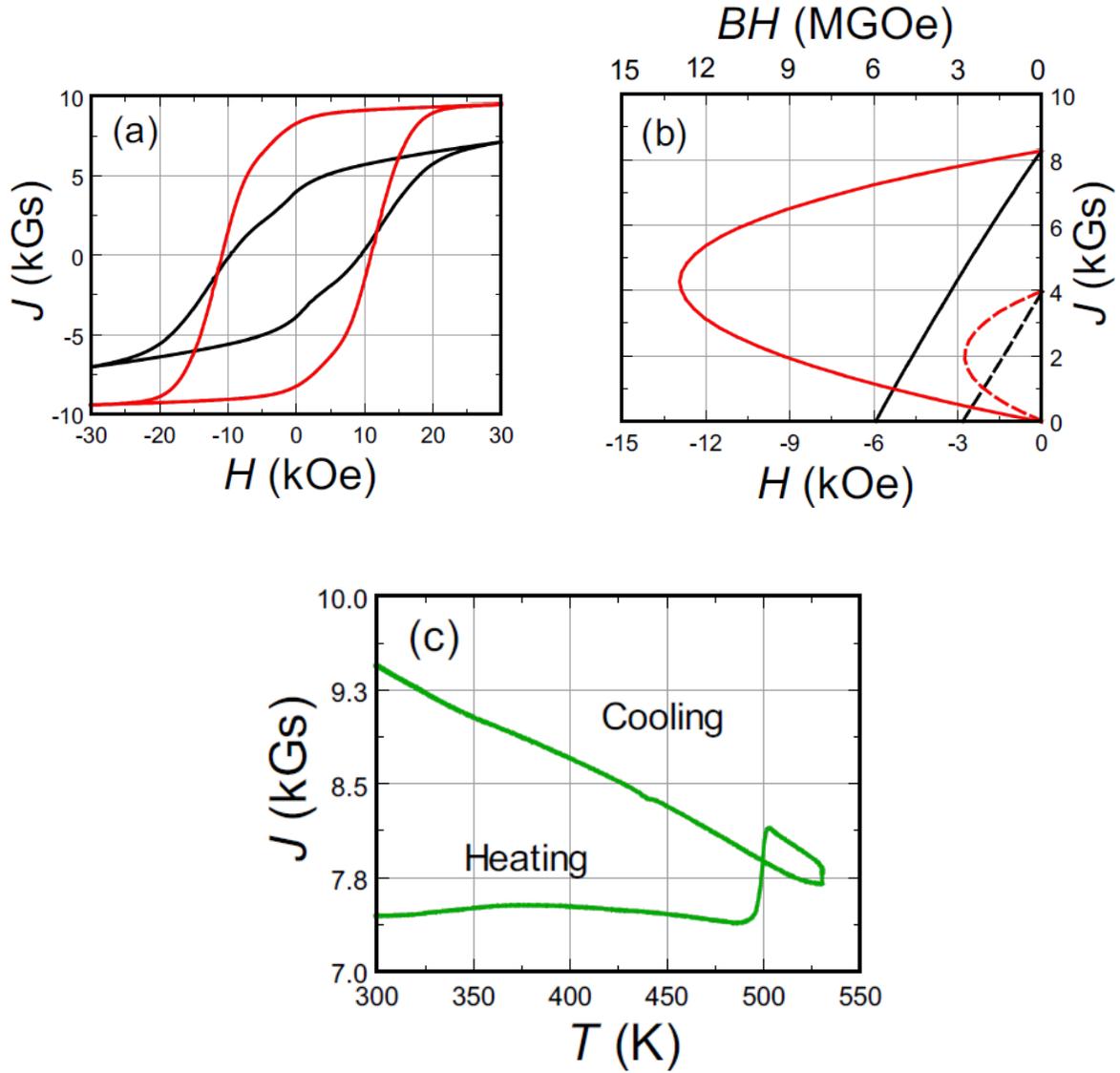


Figure 3. (a) Magnetic hysteresis plots, J - H (black and red lines are for non-aligned and aligned samples, respectively); (b) B - H curves (black) and energy product (red); solid and dash lines are for non-aligned and aligned samples, respectively; (c) Magnetization vs. temperature plot, J - T for the heating and cooling profiles. All data are for 75 vol% MQA Nd-Fe-B Nylon bonded magnets.

The magnetic remanence depends on the filling fraction of magnetic powder and their alignment in bonded magnets. The relationship can be expressed as,

$$B_r = f^* \Phi_p^* M_{r0} \quad (1)$$

Where B_r , f , and Φ_p are remanence of bonded magnets, volume fraction of magnetic powder, and degree of alignment (DOA) of magnetic particle in bonded magnet, respectively. M_{r0} is remanence of the magnetic powder, which is determined by its intrinsic DOA (Φ_i) and saturation magnetization (M_{s0}), i.e. $M_{r0}=\Phi_i*M_{s0}$. The observed total DOA (Φ_t) of the bonded magnet is contributed by the intrinsic DOA, Φ_i , (i.e. the degree of c -axis crystal texture of 2:14:1 phase of Nd-Fe-B) and the extrinsic DOA, Φ_p , (i.e. degree of particle alignment induced by an external magnetic field). Eq. (1) can be rewritten as

$$B_r=f*\Phi_p*\Phi_i*M_{s0} \quad \text{or} \quad B_r=f*\Phi_t*M_{s0} \quad (2)$$

We have investigated the loading fraction of Nd-Fe-B on the alignment and magnetic properties of bonded magnets. As shown in Fig. 4, the magnetic remanence B_r increases almost linearly while the remanence ratio M_r/M_s remains almost unchanged with increasing loading volume fractions from 40% to 75% in the aligned Nd-Fe-B bonded magnets. The ratio of M_r/M_s or Φ_t is about 0.88 in both Nylon- and PPS-bonded magnets, and the derived Φ_p is 0.97 based on the formula (2). In other words, the magnetic powders have been aligned by 97% under an external applied magnetic field of 30 kOe ($\Phi_p=0.97$). As seen from Eq. (2), the remanence B_r is directly proportional to the filling fraction of magnetic powder having fixed value of Φ_t (~ 0.88) in the bonded magnets prepared here. The intrinsic coercivity H_{ci} remain almost unchanged with increasing loading fraction of Nd-Fe-B powders (Fig. 4c). The inductive coercivity H_{cb} increases at higher contents of Nd-Fe-B powders, which is ascribed to the higher remanence for the bonded magnet with larger filling fraction of Nd-Fe-B powders. If the demagnetization curve $M-H$ has ideal squareness, i.e. the value of H_{cb} is same at that of B_r , the maximum energy product $(BH)_{max}$ equals to $\frac{1}{4}(B_r)^2$. It is

expected that the value of $(BH)_{max}$ increases parabolically with Br or filling fraction volume. However, the volume fraction dependence of $(BH)_{max}$ deviates slightly from a parabolic curve since the real magnet lacks the perfect squareness of $M-H$ curve. (Fig. 4(d)).

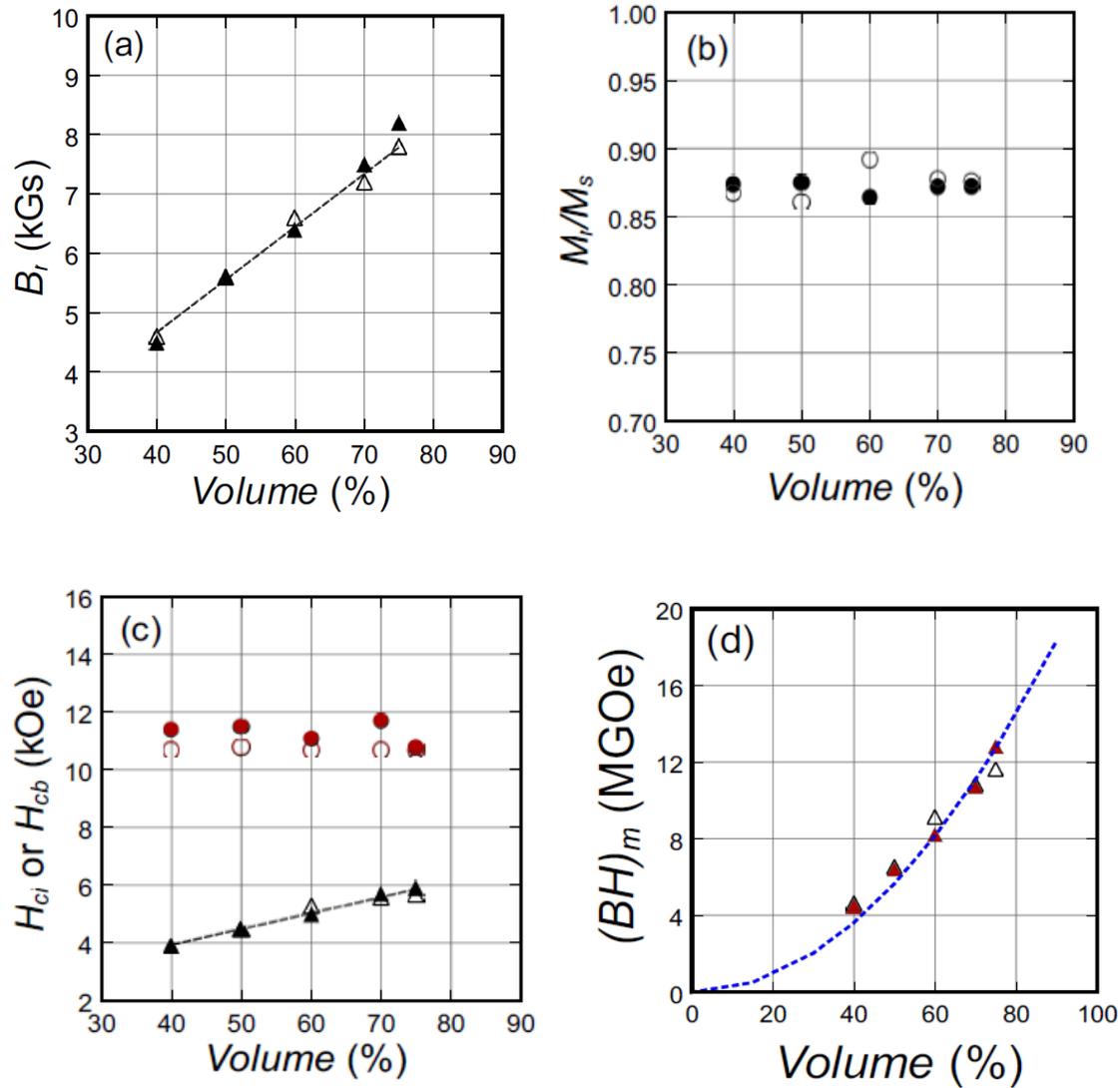


Figure 4. Dependence of magnetic properties on filling fraction for bonded magnets made with MQA Nd-Fe-B powders in Nylon (solid symbol)- and PPS (open symbol): (a) Magnetic remanence; (b) Magnetic remanence ratio; (c) Intrinsic (circle) and inductive (triangle) coercivity; and (d) Maximum energy product.

Figure 5 show the magnetic hysteresis loop of Magfine (HDDR) Nd-Fe-B bonded magnets. The remanence increases with magnetic alignment and results in a maximum energy product of about

13 MGOe. Different from the MQA bonded magnets (Fig. 3), the intrinsic coercivity remains unchanged for non-aligned and aligned samples. The result confirms that the domain wall pinning plays an important role in the magnetization reversal process[27]. It should be noted that the magnetic alignment occurs at a temperature range between 475 to 485 K (Fig. 5b), less than that for MQA Nd-Fe-B Nylon bonded magnets by about 15 K (Fig. 3c). This is partially ascribed to the different morphology of Magfine and MQA Nd-Fe-B powder (supplementary material, Fig. S2). The Magfine particle has an irregular sphere-like shape while MQA powder has a plate-like morphology. For the same viscosity of melted binders, the sphere-like particles are easier to move or rotate than plate-like ones. The correlation between the magnetic alignment temperature and types of magnetic powder will be discussed in section 3.2.

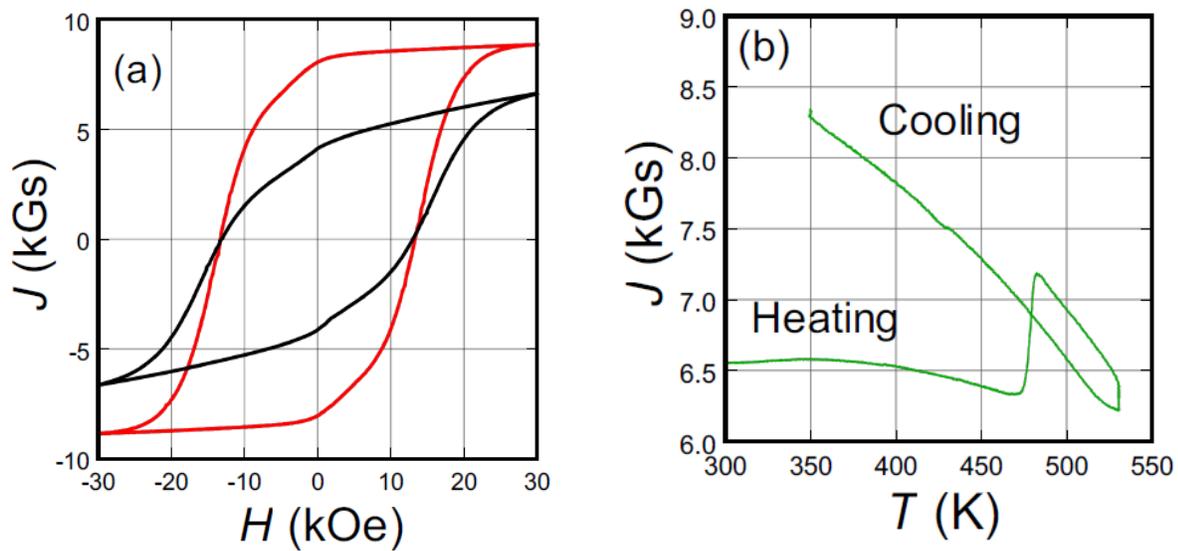


Figure 5. (a) Magnetic hysteresis plots, J - H (black and red lines are for non-aligned and aligned samples, respectively); and (b) Magnetization vs. temperature plot, J - T for the heating and cooling profiles. All data are for 75 vol% Magfine Nd-Fe-B Nylon bonded magnets.

Figure 6 displays the effect of loading fraction of Magfine powders on magnetic properties of the bonded magnets. The magnetic remanence B_r increases linearly with filling fraction of Nd-Fe-B powder for both Nylon- and PPS- bonded magnets. For all the Magfine bonded magnets, the

remanence ratio M_r/M_s is about 0.89 and comparable to that of the MQA bonded magnets (0.88). The magnetic alignment of the particles is very similar for both types of anisotropic Nd-Fe-B powders under processing conditions and achieves a particle DOA $\Phi_p=0.97$. Similar to MQA bonded magnets, the inductive coercivity and energy product of Magfine bonded magnets increases almost linearly and parabolically with loading fraction of magnetic powders, respectively.

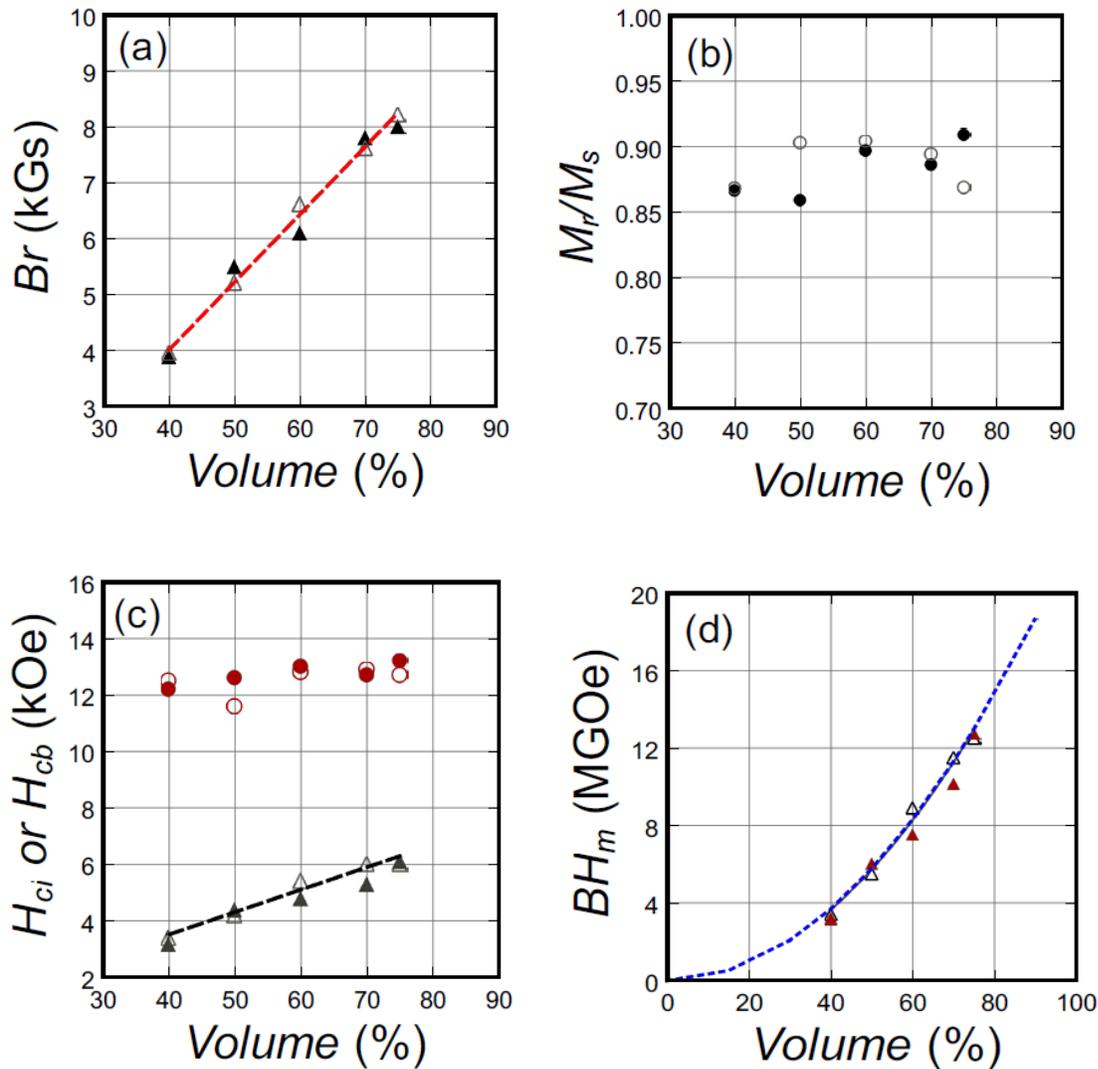


Figure 6. Dependence of magnetic properties on filling fraction for bonded magnets made with Magfine Nd-Fe-B powders in Nylon (solid symbol)- and PPS (open symbol): (a) Magnetic remanence; (b) Magnetic remanence ratio; (c) Intrinsic (circle) and inductive (triangle) coercivity; and (d) Maximum energy product.

3.2 Effect of temperature on magnetic alignment

In Nylon- and PPS- bonded magnets, the magnetic powder was bonded to achieve the near net shape and mechanical integrity. To align the magnetic particles with externally applied magnetic field, the binder should be at low viscosity state to facilitate the rotation of magnetic particles. The magnetic alignment must be done at temperatures slightly above the melting point of the binder. To understand the effect of temperature on magnetic alignments, we have measured the melting point of binder in bonded magnets with different loading fraction of magnetic powders using DSC (see supplementary materials, Fig. S3 and S4 for the DSC curves of Nylon- and PPS-bonded magnets, respectively). The starting-temperature and ending-temperature for magnetic alignment were derived from thermal magnetization curves in bonded magnets during post-compaction magnetic alignment process.

Figure 7 displays the binder melting temperature and magnetic alignment temperature in anisotropic Nd-Fe-B Nylon- and PPS- bonded magnets. The binder melting temperature is same in bonded magnet with MQA- and Magfine magnetic powders and remains almost unchanged with different loading fractions of Nd-Fe-B magnetic powders. However, the magnetic alignment temperature is different for the different types of magnetic powders. As shown in Fig. 7a, the starting- and ending- magnetic alignment temperatures of MQA is higher than that of Magfine in Nylon bonded magnet by about 10-20 K. It means that the MQA powder is more difficult to align than Magfine powders in Nylon bonded magnets, i.e. MQA powder needs higher temperature to facilitate magnetic alignment. This is contrary to expectation because the MQA magnetic powder has a slightly lower coercivity than Magfine powder (Fig.1), indicating that the response to magnetic alignment should be less in MQA at a fixed applied magnetic field and viscous state of

the binder. Further, comparing Fig. 3c and Fig. 5b, the magnetization of MQA powder is higher than that of Magfine powder by about 15% around 450-500 K, under the applied alignment magnetic field of 30 kOe. In other words, the driving force for magnetic alignment of MQA powder is slightly higher than that for Magfine powders which, again, indicates that MQA should be easier to align. One reason for the difficulty in magnetic alignment of MQA powder is related to the different morphology of Magfine and MQA powders, as discussed in section 3.1. The irregular sphere-like Magfine particle is easier to rotate than the plate-like MQA powder in binder with similar viscosity. This morphology difference is likely a reflection of the methods via which each of the powders was made.

In contrast, the difference between starting- and ending- magnetic alignment temperatures is only about 3-5 K both for the MQA- and Magfine powders in PPS bonded magnets (Fig. 7b). The possible reason is that the temperature dependence of viscosity for PPS is different from that of Nylon-12.[28,29] As the temperature reaches the critical point, the viscosity of PPS is relatively low so that both MQA and Magfine powder can be easily rotated in a narrow temperature range (3-5 K). On the other hand, the relatively wider temperature range (10-15 K) needed to complete magnetic alignment of magnetic powder in Nylon-12 indicates a slower change in viscosity, compared to PPS. Further work is needed to clarify the correlation between magnetic alignment and the viscosity of binders.

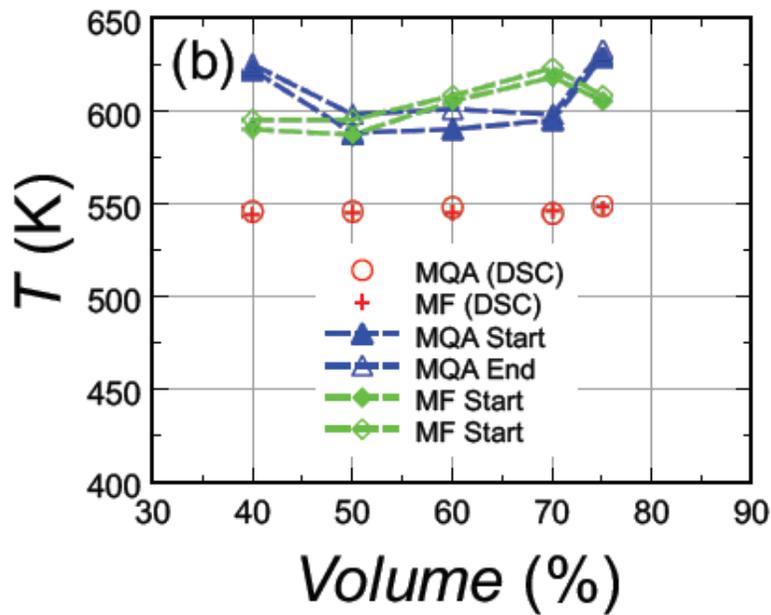
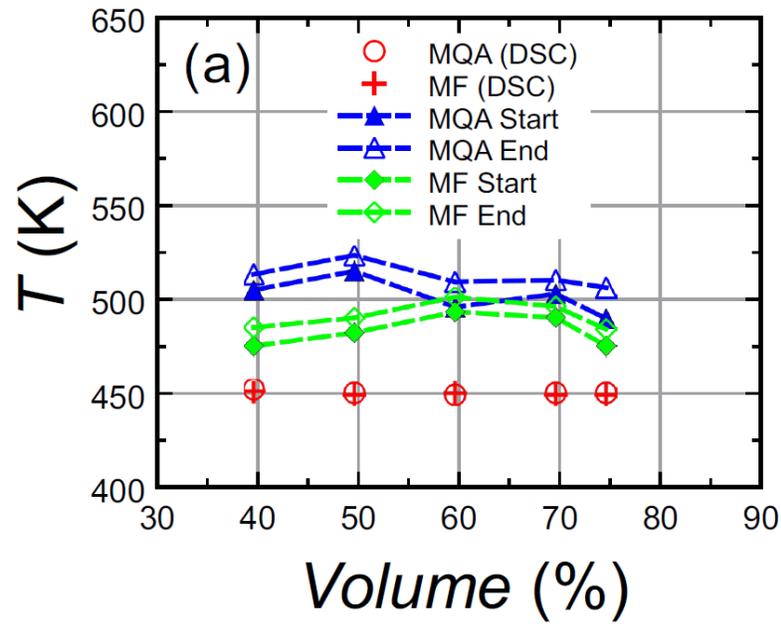


Figure 7. The binder melting temperature (symbols of circle and plus are for MQA- and Magfine-bonded magnets, respectively); starting magnetic alignment temperature (solid symbol) and ending magnetic alignment temperature (open symbol), as a function of loading fraction of bonded magnets made with Nd-Fe-B magnetic powders in Nylon- (a) and PPS- (b). The symbols of triangle and diamond represent MQA- and Magfine bonded magnet samples, respectively.

3.3 Magnetic energy variation during post-compaction magnetic alignment process

During magnetic alignment process, there are mainly three types of competing interactions.[30,31] Firstly, the Zeeman energy due to the interaction between external applied magnetic field and magnetic powder ($E_{app.}$) is the driving force to align magnetic particles. Secondly, the inter-particle magneto-static energy ($E_{stat.}$) promotes the random distribution of the magnetization direction of different magnetic powder and discourage the magnetic alignment. Thirdly, the interaction between magnetic particle and binder (E_{bind}) prevents the particles from moving or rotation. However, E_{bind} depends on the temperature. When processing temperature is higher than melting point of the binder, the interaction between particle and binder is very weak. The spatial distribution of the direction of the magnetic easy axes for the magnetic particles (i.e. DOA) is determined by the balance between the Zeeman energy ($E_{app.}$) and the inter-particle static energy ($E_{stat.}$) in bonded magnets, when E_{bind} is negligible (e.g. near the melting temperature of the binder). We considered these two contributions to simulate the relationship between DOA and magnetic alignment field H_{app} during magnetic alignment process in anisotropic Nd-Fe-B bonded magnets. The achieved DOA results from minimization of total magnetic energy (E_{tot}).

$$E_{tot} = E_{stat.} + E_{app.} \quad (3)$$

During magnetic alignment process, the coercive Nd-Fe-B magnetic powders are magnetized and display net magnetization. E_{stat} increase while E_{app} reduces with increasing net magnetization under a fixed DOA and an external applied field. If the increased part of E_{stat} are compensated by the reduction in E_{app} (i.e. ignoring the particle-binder interaction at alignment temperature), the magnetic particle will rotate to reduce the angle between its effective easy magnetization axis (EMA) and the direction of alignment field and continue until the distribution of magnetic particle reaches a new balance. Further increase in alignment magnetic field reduces the angle between the EMA of magnetic powder and the direction of applied field, i.e. enhancement of DOA. The DOA

has been calculated as a function of alignment field. In the numerical implementation, we fix the particle, but rotate the direction of its EMA in response to externally applied alignment magnetic field, which are mathematically equivalent. The room temperature magnetic properties of anisotropic Nd-Fe-B powder are selected as the starting point. The coercivity and remanence are 15 kOe and 12.5 kGs, respectively (See Figure 1).

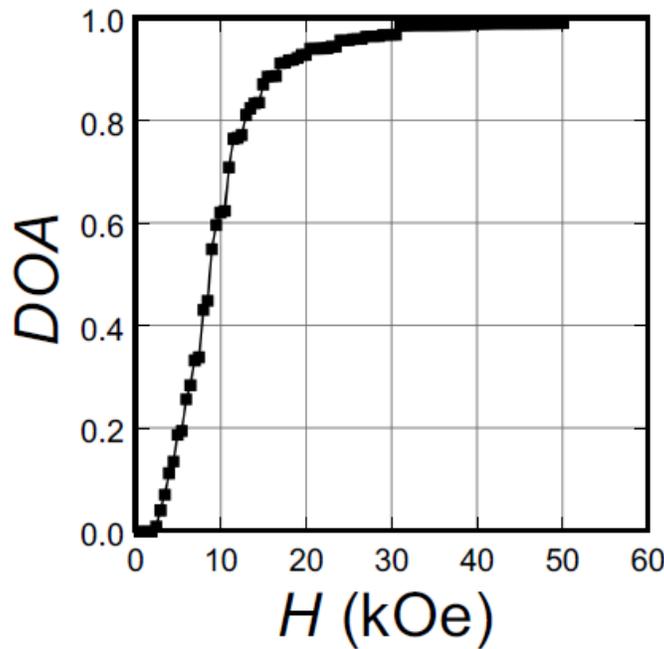


Figure 8. Effect of applied field on DOA of anisotropic MQA Nd-Fe-B bonded magnets.

As shown in Fig. 8, DOA increases slowly at low external field (<5 kOe), then increases rapidly and reaches a value of 0.9 at the external field of 15 kOe (the same as the coercivity). When the external applied field reaches twice that of the coercivity (~30 kOe), the perfect alignment is achieved (DOA=1.0). Further, the coercivity decreases almost linearly with temperature, which will reduce the required magnetic field strength for alignment. For example, the coercivity of magfine powder is around 4.5 kOe at ~425 K (Fig. 9). In other words, an external field of 10 kOe is enough to fully align Magfine or MQA Nd-Fe-B powder in bonded magnets above 425 K. The

numerical calculations confirm that DOA is 0.99 under an alignment field of 10 kOe for magnetic powder with a coercivity of 4.5 kOe and a remanence of 12 kGs. In our previous work [12], an alignment field of 10 kOe can achieve a complete alignment of MQA Nd-Fe-B powder in nylon-12 bonded magnets (30 vol% binder) with an alignment temperature of 550 K, in qualitative agreement with the modeling results.

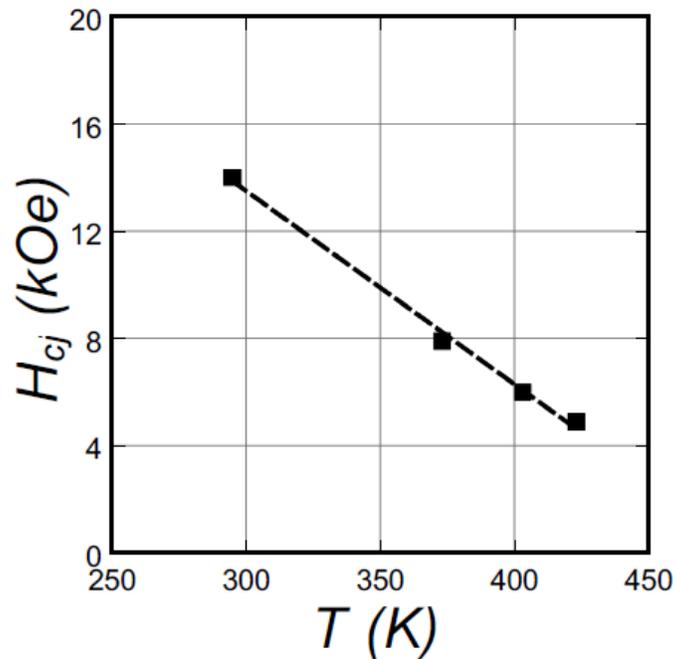


Figure 9. Temperature dependence of coercivity in anisotropic Magfine Nd-Fe-B bonded magnets.

This finding is important for integrating alignment magnetic field source into additive manufacturing systems in order to print and align at the same time, especially considering that 90% of alignment can be achieved with <5 kOe external magnetic field at 425 K. The reason is that it is much easier to incorporate an alignment field source if the required magnetic field strength is low. It means that sufficient magnetic field for alignment during additive manufacturing could be supplied with sintered magnets, even with the sintered magnet positioned far enough to shield it from thermal demagnetization.

4 Conclusion

In summary, the magnetic alignment of anisotropic Nd-Fe-B bonded magnets depends on the binder type, thermo-magnetic properties of magnetic powder and alignment magnetic field. When the binder is at molten state, the magnetic powder can be aligned in bonded magnets by an external field. The degree of alignment is determined by the balance between the Zeeman energy ($E_{app.}$) and the inter-particle static energy ($E_{stat.}$) in bonded magnets around the melting temperature of binders. Nearly complete alignment can be achieved by applying magnetic field which is twice the coercivity in anisotropic Nd-Fe-B bonded magnets. It is expected that similar behavior can be achieved with other anisotropic bonded magnets. To achieve good alignment in anisotropic bonded magnets with selective binders, the temperature dependence of coercivity of the selected magnetic powder determines the minimum value of the necessary alignment magnetic field.

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References

- [1] Coey J M D and O'Donnell K 1997 New bonded magnet materials (invited) *J. Appl. Phys.* **81** 4810–5
- [2] Ormerod J and Constantinides S 1997 Bonded permanent magnets: Current status and future opportunities (invited) *J. Appl. Phys.* **81** 4818–20
- [3] Sugimoto S 2011 Current status and recent topics of rare-earth permanent magnets *J. Phys. D: Appl. Phys.* **44**
- [4] Ma B M, Herchenroeder J W, Smith B, Suda M, Brown D and Chen Z 2002 Recent development in bonded NdFeB magnets *J. Magn. Magn. Mater.* **239** 418–23
- [5] Paranthaman M P, Shafer C S, Elliott A M, Siddel D H, McGuire M A, Springfield R M, Martin J, Fredette R and Ormerod J 2016 Binder Jetting: A Novel NdFeB Bonded Magnet Fabrication Process *Jom* **68** 1978–82
- [6] Li L, Tirado A, Conner B S, Chi M, Elliott A M, Rios O, Zhou H and Paranthaman M P 2017 A novel method combining additive manufacturing and alloy infiltration for NdFeB bonded magnet fabrication *J. Magn. Magn. Mater.* **438** 163–7
- [7] Li L, Tirado A, Nlebedim I C, Rios O, Post B, Kunc V, Lowden R R, Lara-Curzio E, Fredette R, Ormerod J, Lograsso T A and Paranthaman M P 2016 Big Area Additive Manufacturing of High Performance Bonded NdFeB Magnets *Sci. Rep.* **6** 36212
- [8] Huber C, Abert C, Bruckner F, Groenefeld M, Schuschnigg S, Teliban I, Vogler C, Wautischer G, Windl R and Suess D 2017 3D Printing of Polymer-Bonded Rare-Earth Magnets with a Variable Magnetic Compound Fraction for a Predefined Stray Field *Sci. Rep.* **7**
- [9] von Petersdorff-Campen K, Hauswirth Y, Carpenter J, Hagmann A, Boës S, Daners M S, Penner D and Meboldt M 2018 3D printing of functional assemblies with integrated polymer-bonded magnets demonstrated with a prototype of a rotary blood pump *Appl. Sci.* **8**
- [10] Jaćimović J, Binda F, Herrmann L G, Greuter F, Genta J, Calvo M, Tomše T and Simon R A 2017 Net Shape 3D Printed NdFeB Permanent Magnet *Adv. Eng. Mater.* **19**
- [11] Baldissera A B, Pavez P, Wendhausen P A P, Ahrens C H and Mascheroni J M 2017 Additive Manufacturing of Bonded Nd-Fe-B - Effect of Process Parameters on Magnetic Properties *IEEE Trans. Magn.* **53**
- [12] Wendhausen P P, Ahrens C H, Baldissera A B, Pavez P D and Mascheroni J M 2017 Additive manufacturing of bonded NdFeB, process parameters evaluation on magnetic properties 2017 *IEEE International Magnetism Conference, INTERMAG 2017*
- [13] Coey J M D 2020 Perspective and Prospects for Rare Earth Permanent Magnets *Engineering* **6**

- [14] Croat J J, Herbst J F, Lee R W and Pinkerton F E 1984 Pr-Fe and Nd-Fe-based materials: A new class of high-performance permanent magnets (invited) *J. Appl. Phys.* **55** 2078–82
- [15] Nozawa Y, Iwasaki K, Tanigawa S, Tokunaga M and Harada H 1988 Nd-Fe-B die-upset and anisotropic bonded magnets (invited) *J. Appl. Phys.* **64** 5285–9
- [16] Takeshita T 1993 Present status of the hydrogenation-decomposition-desorption-recombination process as applied to the production of magnets *J. Alloys Compd.* **193** 231–4
- [17] Horikawa T, Matsuura M, Sugimoto S, Yamazaki M and Mishima C 2016 Crystallographic alignment in the recombination stage in d-HDDR process of Nd-Fe-B-Ga-Nb powders *AIP Adv.* **6** 10–6
- [18] Gutfleisch O, Gebel B and Mattern N 2000 Texture in a ternary Nd_{16.2}Fe_{78.2}B_{5.6} powder using a modified hydrogenation-disproportionation-desorption-recombination process *J. Magn. Magn. Mater.* **210** 5–9
- [19] Qin W, He J and Meng J 2012 Research on composite powder and magnet properties of bonded NdFeB magnets *Adv. Mater. Res.* **535–537** 1314–8
- [20] Rowlinson N, Ashraf M M and Harris I R 1989 New developments in bonded Nd-Fe-B magnets *J. Magn. Magn. Mater.* **80** 93–6
- [21] Gandha K, Li L, Nlebedim I C, Post B K, Kunc V, Sales B C, Bell J and Paranthaman M P 2018 Additive manufacturing of anisotropic hybrid NdFeB-SmFeN nylon composite bonded magnets *J. Magn. Magn. Mater.* **467** 8–13
- [22] Jung I D, Park J M, Yu J H, Kang T G, Kim S J and Park S J 2014 Particle size effect on the magneto-rheological behavior of powder injection molding feedstock *Mater. Charact.* **94** 19–25
- [23] Parans Paranthaman M, Yildirim V, Lamichhane T N, Begley B A, Post B K, Hassen A A, Sales B C, Gandha K and Nlebedim I C 2020 Additive manufacturing of isotropic NdFeB PPS bonded permanent magnets *Materials (Basel)*. **13** 1–10
- [24] Gandha K, Nlebedim I C, Kunc V, Lara-Curzio E, Fredette R and Paranthaman M P 2020 Additive manufacturing of highly dense anisotropic Nd-Fe-B bonded magnets *Scr. Mater.* **183** 91–5
- [25] Huber C, Abert C, Bruckner F, Pfaff C, Kriwet J, Groenefeld M, Teliban I, Vogler C and Suess D 2017 Topology optimized and 3D printed polymer-bonded permanent magnets for a predefined external field *J. Appl. Phys.* **122**
- [26] Khlopkov K, Gutfleisch O, Hinz D, Müller K-H and Schultz L 2007 Evolution of interaction domains in textured fine-grained Nd₂Fe₁₄B magnets *J. Appl. Phys.* **102** 23912
- [27] Li W F, Ohkubo T, Hono K, Nishiuchi T and Hirosawa S 2009 The role of grain boundaries in the coercivity of hydrogenation disproportionation desorption recombination processed Nd-Fe-B powders *J. Appl. Phys.* **105** 07A706
- [28] Bai J, Goodridge R D, Hague R J M, Song M and Okamoto M 2014 Influence of carbon nanotubes on the rheology and dynamic mechanical properties of polyamide-12 for laser sintering *Polym. Test.* **36** 95–100
- [29] Mwanja F M, Maringa M and van der Walt K 2020 A Review of Methods Used to Reduce the Effects of High Temperature Associated with Polyamide 12 and Polypropylene Laser Sintering ed B Xu *Adv. Polym. Technol.* **2020** 9497158

- [30] Gao R W, Zhang J C, Zhang D H, Dai Y Y, Meng X H, Wang Z M, Zhang Y J and Liu H Q 1999 Dependence of the magnetic properties on the alignment magnetic field for NdFeB bonded magnets made from anisotropic HDDR powders *J. Magn. Magn. Mater.* **191** 97–100
- [31] Liu J and Walmer M 2008 Process and Magnetic Properties of Rare-Earth Bonded Magnets *Handbook of Advanced Magnetic Materials* pp 1008–44