

Net Shape Processing of Alnico Magnets by Additive Manufacturing

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Alternatives to rare earth permanent magnets, such as alnico, will reduce supply instability, increase sustainability, and could decrease the cost of permanent magnets, especially for high temperature applications, such as traction drive motors. Alnico magnets with moderate coercivity, high remanence, and relatively high energy product are conventionally processed by directional solidification and (significant) final machining, contributing to increased costs and additional material waste. Additive manufacturing (AM) is developing as a cost effective method to build net-shape three-dimensional parts with minimal final machining and properties comparable to wrought parts. This work describes initial studies of net-shape fabrication of alnico magnets by AM using a laser engineered net shaping (LENS) system. High pressure gas atomized (HPGA) pre-alloyed powders of two different modified alnico “8” compositions, with high purity and sphericity, were built into cylinders using the LENS process, followed by heat treatment. The magnetic properties showed improvement over their cast and sintered counterparts. The resulting alnico permanent magnets were characterized using scanning electron microscopy (SEM), energy dispersive spectroscopy (EDS), electron backscatter diffraction (EBSD), and hysteresisgraph measurements. These results display the potential for net-shape processing of alnico permanent magnets for use in next generation traction drive motors and other applications requiring high temperatures and/or complex engineered part geometries.

Index Terms—Additive manufacturing, alnico, permanent magnets.

I. INTRODUCTION

Continuing supply uncertainty and extreme price fluctuations in rare earth oxides and metals, stemming from a near monopoly position (by China) for production of such critical elements as Dy, Nd, and Pr, which are key components in high energy product ($BH_{\max}$) rare earth (RE) permanent magnets with higher temperature stability, have motivated research into viable alternative magnet solutions [1], [2]. Improvement of permanent magnet alternatives, such as alnico, could present the opportunity to effectively reduce supply instability and drastically decrease manufacturing costs associated with permanent magnets for certain applications such as traction drive motors [1], [2]. Diversifying the supply of magnets for medium energy product applications and high temperature conditions ($\sim 200^\circ\text{C}$) could create new opportunities for revitalization of domestic magnet production and enhanced national security by removing reliance on foreign production of critically important products. Alnico, in particular, is a promising near-term non-RE permanent magnet because of its impressive saturation magnetization and remanence, improved mechanical properties, and excellent thermal stability, featuring a nearly flat temperature dependence of magnetic properties up to 400°C [1].

Co is not considered a resource critical element and has a diversified supply, unlike RE materials [1]. Additionally, work within this group has been able to reduce the Co content of alnico (patent US20170121783 A1), while producing coercivities approaching 3kOe, in light that Co is the most expensive element within alnico magnets. New next generation motor designs with appropriate power outputs for automotive applications currently exist to utilize alnico magnets and some larger motor manufacturers are even incorporating hard ferrites in their secondary drive motors – all before considering energy product at elevated temperatures where RE magnets have a significant disadvantage [1].

Alnico is a functional nanostructured alloy due to the spinodal decomposition of the solid solution into a magnetic Fe-Co-rich (α_1) phase and an Al-Ni-rich (α_2) phase during cooling [3], [4]. This class of permanent magnets was developed in the 1930s and the complex processing, heat treatment, and compositions were optimized empirically over the following decades [5]–[11]. Alnico magnets exhibit coercivity from shape anisotropy after thermal annealing in a magnetic field, resulting in the spinodal nanoscale structure elongated along the applied field direction [3], [4]. More recent investigations of different alnico grades have shown how changes in chemistry and processing produce distinctly different micro- and nanoscale structures, and result in differences in magnetic properties, especially remanence and coercivity [3].

Directionally solidified castings are currently required to process alnico magnets, alnico 9, with the highest energy product ($BH_{\max} \sim 10.5$ MGOe) from grains aligned primarily along the [001] direction, increasing costs and complexity considerably [3], [12]. These directionally solidified castings must undergo significant amounts of machining for final net

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shape, removing exterior surfaces that are not textured in order to obtain the best properties. Also many of these castings have defects that must either be accommodated or must be re-processed or scrapped, further increasing costs and material waste. However, as an added benefit, alnico can be re-processed and recycled into the material stream with less effort than RE based magnets, which currently cannot be effectively recycled on a commercial basis for reuse.

Alnico 5-7 is also a grain-aligned alloy and exhibits the highest remanence (13.5 kG) of any of the commercial alnico grades [3]. Alnico 8 is crystallographically isotropic and has the highest coercivity (1.9 kOe) of any of the commercially available alnico compositions [3]. However the coercivities and energy products of all these grades are ~2-3 times below the value theoretically possible from the spinodally decomposed nanostructure [3]. Thus it is theorized that further processing and composition optimization could produce alnico permanent magnets that achieve an energy product of ~20 MGOe, a value comparable to both Dy enriched Nd-Fe-B and Sm-Co-based permanent magnets operating at elevated temperatures (150-180°C) [3].

Additive manufacturing (AM) is emerging as a method to build net-shape parts with final properties that can meet and exceed cast and wrought processed materials, with great potential to be applied in the permanent magnet industry [13]. In most AM processes, parts are built up layer-by-layer using an electron beam or laser as a heat source to melt the new material onto the layer below. The molten pool is then solidified through heat conduction through the part and substrate on the order of 10^2 - 10^3 K/s for laser engineered net shaping (LENS) [14]. The strength of AM lies both in its ability to eliminate the need for costly dies and molds, but also to drastically reduce material waste with net-shape parts and minimal final machining [13]. Additionally AM opens the processing space and enables complex geometries not possible by current manufacturing methods, as well as interesting prospects in creation of functionally graded permanent magnet materials and control of grain texture to produce tailored isotropic or anisotropic properties [13], [15].

This work describes enhanced alnico magnets produced by AM using experimental high-pressure gas atomized powders (HPGA) and a LENS system. The resulting alnico magnets have been characterized using scanning electron microscopy (SEM) and closed loop hysteresisgraph measurements. The improved properties demonstrate the promise for “printing” alnico magnets into net-shapes for use in next generation motors and other applications, especially those requiring high temperatures and possibly complex engineered part geometries.

II. EXPERIMENTAL PROCEDURES

A. High Pressure Gas Atomization

Elemental and pre-alloyed charge materials were batched to form powders of two modified alnico-8 alloys after melting and superheating each alloy using an induction furnace to 1625°C (for LENS-1-37) and 1750°C (for LENS-1-262) in a

zirconia crucible before atomization. Atmosphere control was maintained with high purity Ar at 1 psi of over-pressure during atomization. Each alloy melt was atomized with high purity Ar gas at 425 psi (for LENS-1-37) and 500 psi (for LENS-1-262) and the molten droplets rapidly solidified into pre-alloyed alnico powders [16]-[19]. The resulting HPGA powders were screen size classified for the laser AM build experiments.

B. Additive Manufacturing

Each batch of screened HPGA powders was built into ≥ 3 mm diameter x ~20mm tall cylinders using an Optomec MR-7 LENS system. The powder was fed into a melt pool created by a Yb-fiber laser (1070nm wavelength), while the deposition head was continuously translated upwards (z-direction) at a rate of 0.203 mm/s. No rastering in the x- or y-directions was used for the builds. A melt pool sensor, which measures the melt pool size based on pixel intensities collected by an infrared (IR) camera, was utilized to maintain a constant size via automatic adjustment of laser power (50-200W).

The LENS-1-37 samples were built on stainless steel substrates, whereas the LENS-1-262 samples were built on both stainless steel (SS) and alnico 9 substrates (A9). Each sample was centerless ground, cut into 3mm x 8mm tall cylinders, and heat treated as listed in Table 1.

TABLE 1 HERE

C. Characterization

The full hysteresis loops were measured using a closed loop Laboratorio Elettrofisico AMH-500 hysteresisgraph with a maximum applied field of 12 kOe and the resulting cross-sectional microstructures were analyzed with an FEI Quanta FEG SEM and an FEI Teneo FEG SEM. The properties of the LENS samples were compared with sintered samples of the same compositions from previous work [17]-[19].

III. RESULTS AND DISCUSSION

A. HPGA Powder Characterization

The resulting powders created during the HPGA runs were highly spherical with few satellite particles and very low trace impurities (O, S, N, and C). A significant portion of each powder batch contained fine powders ($<45\mu\text{m}$), shown in Fig. 1, which were utilized for other net shape compression molding and sintering experiments [17]-[19]. The LENS-1-262 composition included deliberate Ti and Nb substitutions for Fe with the intent to further increase the coercivity of the alloy, as shown in Table 2. The LENS-1-37 powder maintained the original charge composition, whereas the LENS-1-262 alloy was near the desired target alloy, but exhibited anomalous Zr pickup in the powder, which appears to have been transferred into the subsequent builds. The origin

of the Zr pickup is at this point unknown, but may have originated from the melt system ceramics used in the HPGA process.

FIGURE 1 HERE

TABLE 2 HERE

B. Additive Manufacturing Builds

The lower power appeared qualitatively to produce improved sample builds, whereas other parameters did not have a significant influence on the builds that were produced. Build parameters and optimization was not a primary goal of this work, only initial process verification and demonstration.

The different compositions affected the build microstructures as shown in Fig. 2 in the as-built condition. The LENS-1-37 samples transitioned from equiaxed fine grains near the substrate and exterior of the build into elongated columnar-appearing larger grains with aspect ratios of approximately 2:1 in the center, mostly oriented along the length of the build. The LENS-1-262 samples maintained fine equiaxed grains throughout the build with some compositional segregation around grain boundaries as shown in Fig. 3 in the as-built condition. In particular significant Zr segregation on the grain boundaries was evident, whereas Ti showed slight enrichment and Al slight depletion in the same zones. Zr has been used in the past to improve magnetic properties in a similar way to Ti in sintered alnico samples, and it would appear that this mechanism is still operating in the AM builds [20]. The grains in the LENS-1-262 samples were randomly oriented as demonstrated by the representative electron backscatter diffraction (EBSD) orientation image micrograph (OIM) map included in Fig. 3 with the corresponding orientation key.

FIGURE 2 HERE

FIGURE 3 HERE

C. Magnetic Characterization

The magnetic properties of the LENS-1-37 samples are included in Table III with the full hysteresis loops included in Fig. 4. The remanence of the LENS-1-37 samples averaged 9 kG, the intrinsic coercivity 1.8 kOe, and the energy product 6 MGOe. When these properties were compared with the corresponding sintered samples (Table V), produced using the same HPGA powder, they showed a marked improvement of 0.5 kG, ~0.150 kOe, and 1.4 MGOe for remanence, coercivity, and energy product, respectively. From the microstructural analysis (Fig. 1a) the improvement in properties was partially

attributed to the evolution of columnar grains throughout the majority of the sample length and cross section. This is supported by the high remanence ratios of 0.76-0.78, indicating an underlying texture. Additionally the rapid solidification inherent to the LENS process likely caused finer compositional segregation than what occurs during casting or after sintering for 4 hours at 1250°C, as is typical for the sintered samples [14], [17]-[19].

TABLE 3 HERE

FIGURE 4 HERE

The impressive results from the initial LENS-1-37 experiments prompted further investigation into this method for alnico permanent magnet manufacturing. The magnetic properties of the second round of samples, LENS-1-262, are included in Table IV, with full hysteresis loops included in Fig. 5. Since the initial deposit of material in the LENS process requires a small melt pool to be formed in the substrate, two different substrates – stainless steel, a typical material, and alnico 9 – were used to study the influence of possible substrate epitaxy on the microstructures of the built rods. In particular, it was theorized the alnico 9 substrate could epitaxially “seed” the growth of [100] aligned grains in the LENS printed samples, if the aligned grains could be maintained through the substrate to sample interface.

For the samples built on the stainless steel substrate, the remanence averaged 7.5 kG, intrinsic coercivity averaged 2 kOe, and energy product averaged 4.8 MGOe. The samples built on the alnico 9 substrate averaged 7.7 kG for remanence, 1.9 kOe for coercivity, and 4.4 MGOe for energy product. It is evident from the magnetic properties, and the randomly oriented microstructure in Fig. 3, that the alnico 9 substrate provided trivial benefit because the remanence ratio of 0.73 was not considerably higher than the 0.72 of the stainless steel substrates. Although the alnico 9 substrate samples exhibited higher remanence, the coercivity and energy product were lower.

TABLE 4 HERE

FIGURE 5 HERE

When these LENS-1-262 samples were compared with their sintered counterparts they had ~0.3 kG lower remanence, ~0.5 kOe higher coercivity, and ~1 MGOe higher energy product, as given in Table V. When compared with their cast counterparts, that had no Zr contamination, the LENS-1-262 samples had lower coercivity, lower energy product, and similar remanence. The lower coercivity, and therefore energy

product, can be attributed to the influence of grain boundary Zr, whereas the similar remanence is a combination of alloy chemistry and underlying microstructure. As the trace Zr was found primarily on grain boundaries, but also in minor amounts intergranularly, the influences to the alloy chemistry is likely minimal but still non-zero, suggesting the AM process effect in improving remanence was again established, though perhaps less clearly.

Overall comparison of both the LENS-1-37 samples and the LENS-1-262 samples to textured alnico magnets of the same composition, as processed by Anderson et al., showed the remanence ratios to lie within the same range of 0.70-0.79 [17]. The trend appears to continue to suggest that the LENS AM process can be utilized in alnico to enhance the magnetic properties of these compositions.

TABLE 5 HERE

With further composition and processing optimization, AM processing of alnico has the potential to provide the highest coercivity and remanence of any alnico grade currently commercially available, perhaps even exceeding them in magnetic properties. Additionally AM processing provides the added benefits of net-shape processing without expensive molds and dies and of greatly reduced material waste.

IV. CONCLUSION

HPGA pre-alloyed alnico powders have been LENS processed into near net-shapes and their microstructures and magnetic properties analyzed. These AM alnico permanent magnets have higher coercivities, remanences, and energy products than their cast and sintered counterparts of the same compositions.

The improvement in alnico with AM processing showed remanence values up to 9.0 kG which nears the 10.6 kG of the directionally solidified, highly textured, anisotropic cast alnico 9. This is indicative that the AM processing is very promising as a method to not only improve properties by squaring the hysteresis loop, but also for potentially reducing processing costs by eliminating the need for expensive molds for directional solidification, extensive final machining, and material waste.

The LENS built samples also had coercivities as high as 2.03 kOe, which matches the 2.02 kOe of sintered alnico 8H, the highest coercivity available from any current commercial alnico grade. Again this indicates that AM processing is a promising method to obtain high coercivity alnico permanent magnets.

Thus AM processing has been shown to be a promising method for net-shape permanent magnet processing in order to improve magnetic properties and reduce processing costs and wastes. These findings are important steps towards near-term alternatives to RE permanent magnets. Transmission electron microscopy (TEM) analysis of the fully heat-treated

microstructures is currently underway to isolate the nanostructured and compositional mechanism(s) of magnetic property improvement from AM processing. Future work will explore composition and processing optimization and also other AM processing methods, such as electron beam melting of a powder bed process to investigate further property and process improvements.

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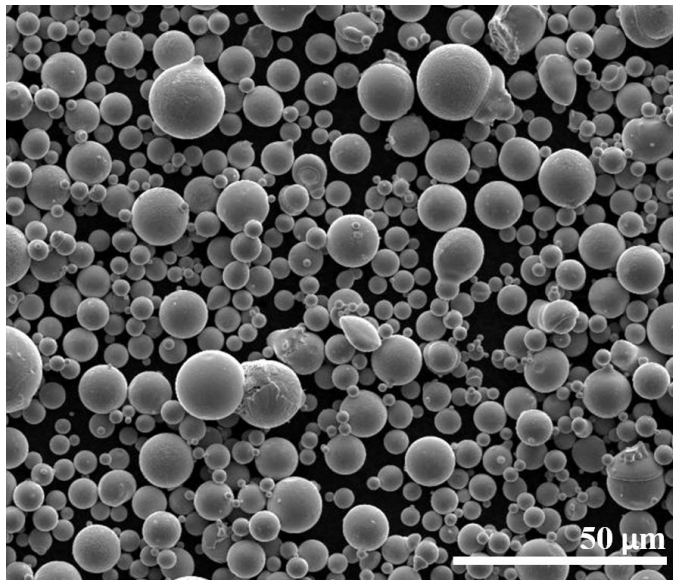


Fig. 1. SEM micrograph of LENS-1-37 composition powder screened as diameter $<20\mu\text{m}$, demonstrating the highly spherical morphology of the gas atomized powder.

TABLE I
FULL HEAT TREATMENTS FOR MODIFIED ALNICO-8 MAGNETS

STEP	LENS-1-37- x	LENS-1-262-x
Solutionize (Vacuum & Oil Quench)	1250°C, 30 min.	1200°C, 30 min.
Magnetic Anneal (1T, Air Cool)	840°C, 10 min.	840°C, 9 min.
Draw (Furnace Cool)	650°C, 5hrs 580°C, 15hrs	670°C, 1.5hrs 650°C, 4hrs 620°C, 4hrs 580°C, 4hrs 550°C, 4hrs 520°C, 4hrs

Full heat treatment details for the magnets produced from the modified alnico-8 type alloy HPGA powders, as optimized using corresponding cast samples.

TABLE II
COMPOSITIONS OF MODIFIED ALNICO-8 HPGA POWDERS

ELEMENT	LENS-1-37 (at%)	LENS-1-262 (at%)	DIFFERENCE
Fe	30.93	28.33	-2.6
Al	14.01	13.45	-0.56
Ni	11.90	12.39	+0.49
Co	33.69	34.62	+0.93
Ti	6.99	8.28	+1.29
Nb	<0.001	0.29	+0.29
Cu	2.48	2.49	+0.01
Zr	<0.001	0.15	+0.15

Compositions of the modified alnico-8 type alloy HPGA powders, as analyzed by inductively coupled mass spectroscopy (ICP-MS).

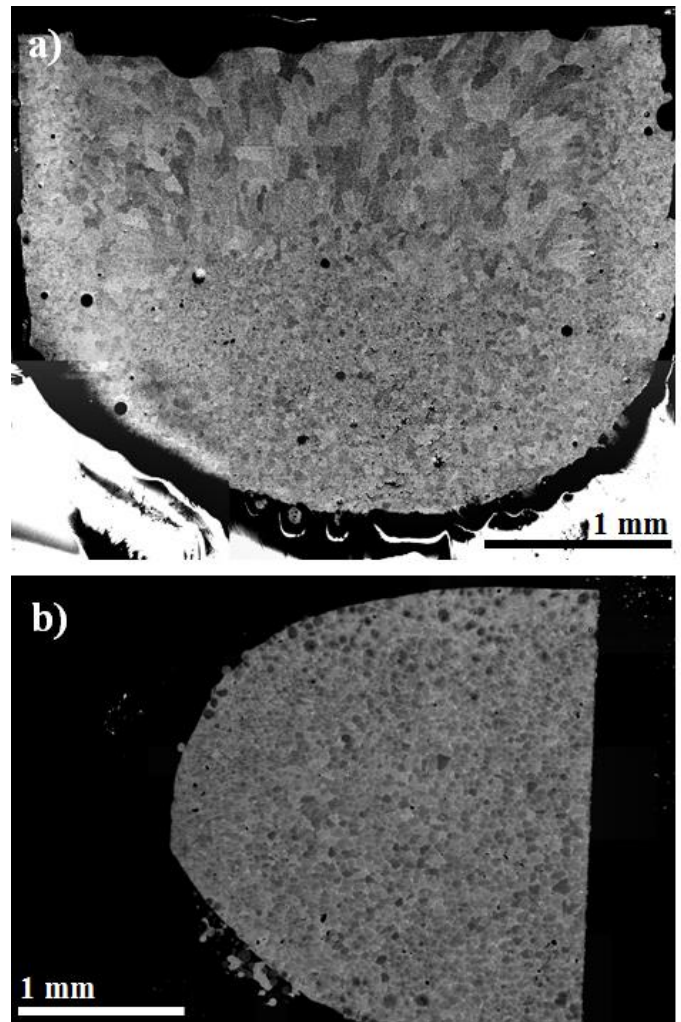


Fig. 2. SEM montage micrographs demonstrating the longitudinal representative microstructures of the a) LENS-1-37 and b) LENS-1-262 rod tips. Note the difference in grain size and orientation.

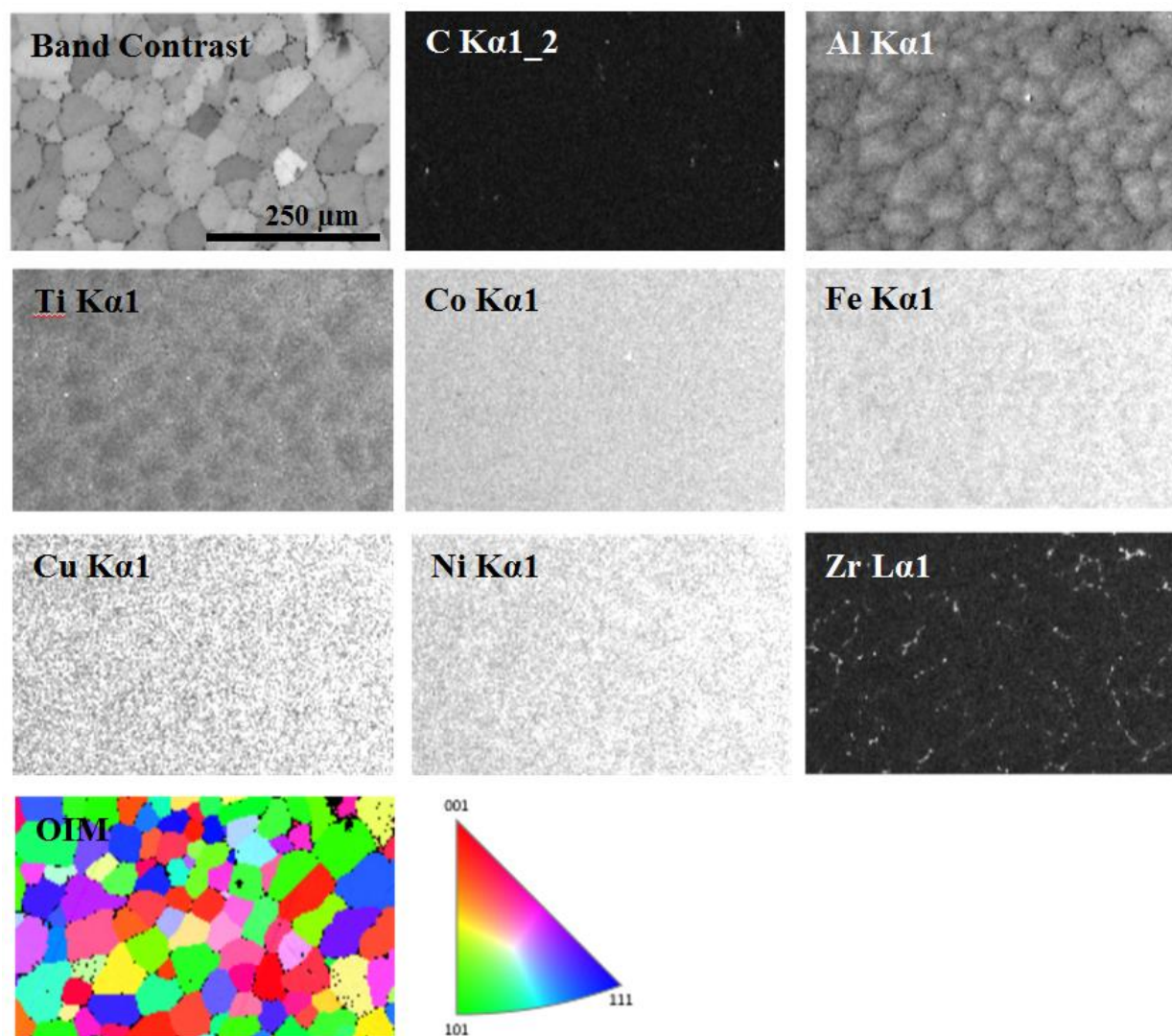


Fig. 3. SEM/EDS micrographs demonstrating the grain structure and compositional segregation within the LENS-1-262 samples. Note the significant presence of Zr isolated to the grain boundaries. Also note the random grain orientation of the same area as demonstrated by the EBSD OIM map with orientation key.

TABLE III
MAGNETIC PROPERTIES OF LENS-1-37 BUILT ALNICO MAGNETS

SAMPLE (SUBSTRATE)	B_r (kG)	H_{CB} (kOe)	H_{CJ} (kOe)	BH_{MAX} (MGOE)	REMANENCE RATIO (B_r/M_s)
LENS-1-37-2a (SS)	8.9	1.73	1.85	6.0	0.76
LENS-1-37-2b (SS)	9.0	1.74	1.85	6.1	0.78
LENS-1-37-3a (SS)	9.2	1.68	1.79	6.0	0.78

Magnetic properties of the modified alnico-8 type alloy HPGA powders built by LENS into rods, as determined using a closed loop hysteresisgraph. The (SS) samples were built on a stainless steel substrate.

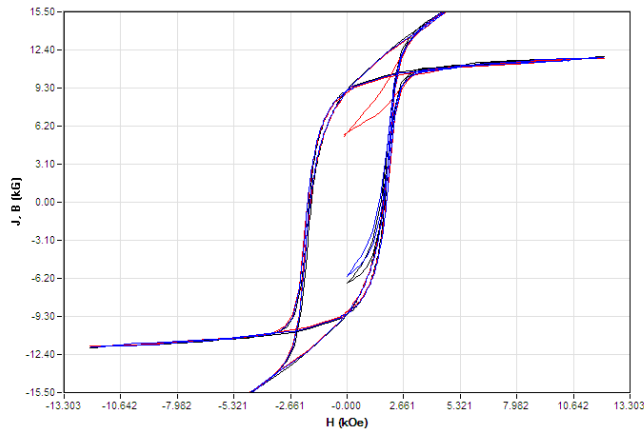


Fig. 4. Full hysteresis loops for the LENS-1-37 samples. Note the low deviation between samples.

TABLE IV
MAGNETIC PROPERTIES OF LENS-1-262 BUILT ALNICO MAGNETS

SAMPLE (SUBSTRATE)	B_r (kG)	H_{CB} (kOe)	H_{CJ} (kOe)	BH_{MAX} (MGOE)	REMANENCE RATIO (B_r/M_s)
LENS-1-262-1b (SS)	7.59	1.84	2.02	4.9	0.72
LENS-1-262-2 (SS)	7.47	1.84	2.04	4.8	0.72
LENS-1-262-3 (A9)	7.89	1.71	1.92	4.5	0.73
LENS-1-262-4 (A9)	7.65	1.72	1.92	4.4	0.73

Magnetic properties of the modified alnico-8 type alloy HPGA powders built by LENS into rods, as determined using a closed loop hysteresisgraph. The (SS) samples were built on a stainless steel substrate, whereas the (A9) samples were built on alnico 9 substrates.

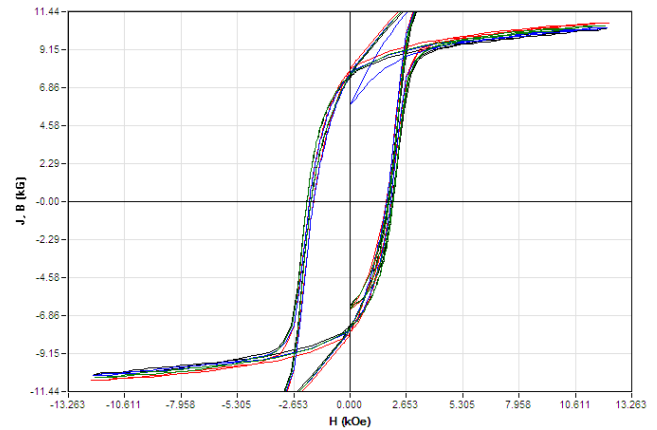


Fig. 5. Full hysteresis loops for the LENS-1-262 samples. Note the low deviation between samples.

TABLE V
COMPARISON OF MAGNETIC PROPERTIES BETWEEN VARIOUS ALNICO MAGNETS

SAMPLE	B_r (kG)	H_{CJ} (kOe)	BH_{MAX} (MGOE)
Sintered 37	8.8	1.70	4.9
LENS-1-37	9.0	1.83	6.0
Cast 262 (No Zr)	7.8	2.32	5.9
Sintered 262	8.1	1.53	3.7
LENS-1-262 (SS)	7.5	2.03	4.8
LENS-1-262 (A9)	7.8	1.92	4.4
AMT sintered alnico 8H	6.7	2.02	4.5
AMT cast alnico 9	10.6	1.50	9.0

Magnetic properties of the modified alnico-8 type alloy HPGA powders built by LENS into rods, as determined using a closed loop hysteresisgraph. The (SS) samples were built on a stainless steel substrate, whereas the (A9) samples were built on alnico 9 substrates. These are compared with sintered and cast samples of the same compositions as well as Arnold Magnetic Technologies, Inc. (AMT) commercial alnico 8H and alnico 9 grades. The cast 262 samples did not have contamination by Zr.