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Consolidation of Fe-N Magnets Using Equal Channel Angular Extrusion

by SG Sankar and LJ Kecskes

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14. ABSTRACT
We have conducted a series of Equal Channel Angular Extrusion experiments on an Fe16N2 magnetic phase material (containing 3 wt% Mn). Results showed the following: 1) Under the experimental conditions investigated thus far, the best density of the extruded specimens is about 75% of the X-ray bulk density. Extrusions at temperatures up to approximately 150 °C do not have any deteriorating effects on the magnetization values (i.e., compared to the precursor powder). However, extrusions at temperatures approximately 150 °C result in a small change in the intrinsic coercivity. 2) The best saturation induction obtained on the extruded specimen is approximately 12.5 kG. The potential to enhance these properties is very high with $Fe_{16}N_2$ powder, now available at Advanced Materials Corporation, using more optimized extrusion conditions. 3) Only a very limited set of extrusions were performed. As such, we believe that there are many more variables we need to examine to improve the density as well as potentially induce more favorable crystal and magnetic textures.

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1. Introduction

High-strength permanent magnets are crucial components in the construction of lightweight, high-efficiency brushless motors, generators, and magnetic bearings. They play a prominent role in the civilian and military sectors of our economy. Civilian applications include computer peripherals, windmill generators, electric vehicles, and medical devices. Military applications include US Army tanks, helicopters, unmanned aerial vehicles, electric vehicles, electric guns, microelectronic devices, etc. Enhancement of the physical characteristics such as the magnetic remanence, coercivity, and energy product, over a wide operating temperature range are the most desirable goals in permanent magnet development. Furthermore, optimization of processing conditions, examining processing techniques as applicable to the production of permanent magnets, replacing a part of or the entire rare-earth metal component with other less critical materials, and reducing their costs are highly desirable.

At present, magnets based on the $Nd_2Fe_{14}B$ composition are widely used in almost all high-demand commercial applications, while those based on Sm_2Co_{17} are used in a majority of the military applications where thermal stability is a prerequisite.

Many candidate materials for the fabrication of high-strength magnets exist; however, some such as $Fe_{16}N_2$[1–4] and $Sm_2Fe_{17}N_x$, are thermally highly sensitive. As such, powders made from these compositions cannot be consolidated to full density through the use of conventional techniques that are commonly employed in the industry such as powder metallurgy followed by hot deformation. The latter stage requires treatments at high temperatures, typically, around 1,000 °C. Instead, in this work, we have explored the potential use of alternative methods that do not require high temperatures to achieve full densification. Specifically, we have considered Equal Channel Angular Extrusion (ECAE), a form of severe plastic deformation processing, to consolidate the $Fe_{16}N_2$ powders[5] and to prepare dense bodies at significantly lower temperatures of 100 and 150 °C. It is hypothesized that if successful, the combination of low temperatures with ECAE would be novel because it offers unique advantages:

- Ensures the retention of volatile components such as nitrogen in the $Fe_{16}N_2$ or $Sm_2Fe_{17}N_x$ crystal structure and thus retains their unique magnetic properties.

- Prevents or limits the growth of the grains. That is, at such processing temperatures, powders with a nanocrystalline structure do not have the chance to coarsen into larger grains, and therefore, will retain their magnetic, electronic, and mechanical properties, which are unique to nanocrystalline phases such as exchange-coupling.
- Helps in the formation of bulk structures with nearly 100% density.
- By repeatedly passing the sample through the ECAE process, grain refinement, grain boundary character modification, introduction of pinning centers, and changes in defect structures have been shown to develop. Potentially, some—or a selective combination of these modifications—can be successfully accomplished in exchange-coupled permanent magnets to enhance their magnetic properties and usher in new technological breakthroughs. However, this will require a sustained and more focused research effort.
- The ECAE method can be used to impart or induce a specific texture and thus promote a particular crystalline alignment. This feature would be very useful in fabricating uniaxial magnets that would increase the magnetization in one crystalline direction and result in magnets with higher magnetic energy product.

2. Background

The Advanced Materials Corporation (AMC), Pittsburgh, Pennsylvania, has recently developed a low-temperature processing technology to fabricate $Fe_{16}N_2$ powder.[6,7] Briefly, the process consists of first making nanocrystalline α-iron by reducing iron (Fe) oxy-hydroxide at temperatures below approximately 400 °C in a fluidized bed reactor.[8] The resultant α-iron product is then reacted, in-situ, with gaseous ammonia or mixtures of ammonia, hydrogen, nitrogen, and/or helium at temperatures between 120 and 180 °C to form an iron nitride (Fe-N) powder. Figure 1 illustrates the formation and relative ratio of the Fe-N phases ($Fe_{16}N_2$ and Fe_4N) to α-Fe as a function of changes in the processing conditions. A typical illustration of the variation of the magnetization in relation to the nitriding temperature of α-Fe is shown in Fig. 2. Magnetization on a per gram basis of this powder is about 5% higher than that of α-Fe (see Fig. 2). The saturation magnetization of the powder showed that it is nearly 240 emu/g; this is much higher than that of α-Fe. The intrinsic coercivity of this powder is nearly 1,000 Oe.

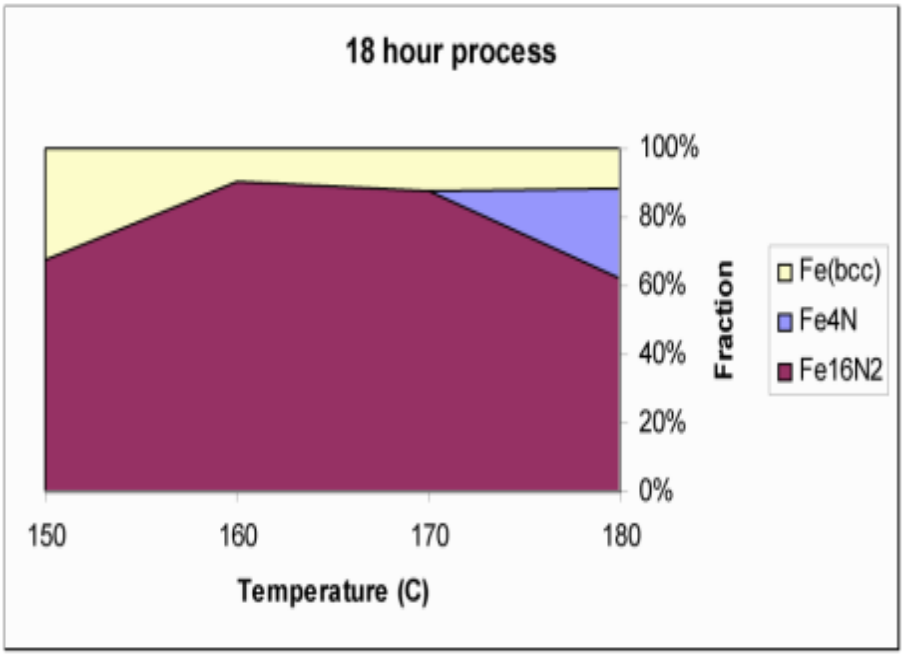

Fig. 1 Fraction of the 3 primary phases in the product: bcc α-Fe, Fe_4N, and $Fe_{16}N_2$. (Note: The relative ratio of the phases are dependent on the process conditions.)

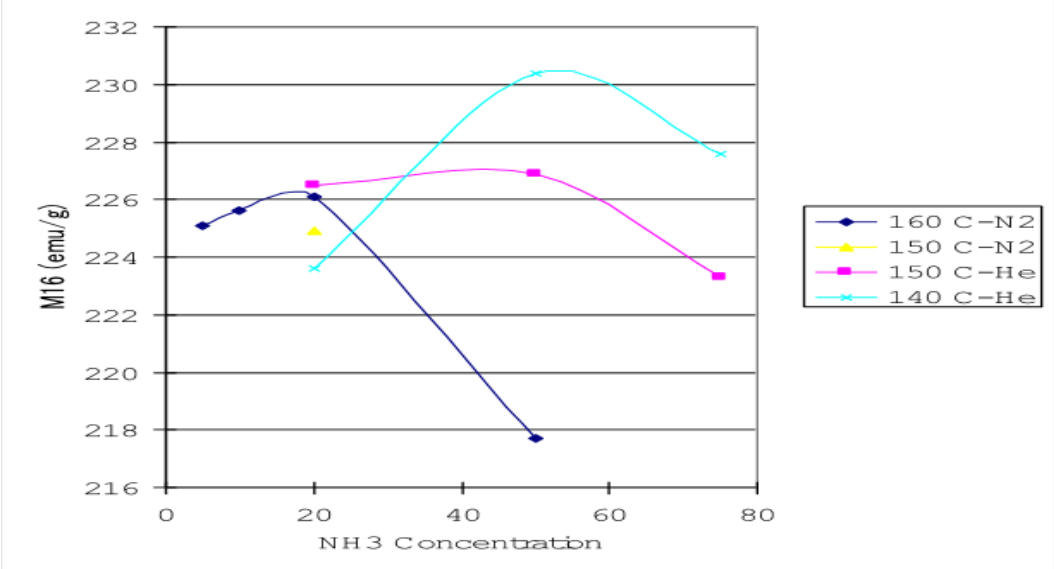

Fig. 2 Magnetization (at an external magnetic field of ~16 kOe) of reaction products as a function of ammonia concentration in a mixture of ammonia and nitrogen at 3 different temperatures

More recently, AMC has successfully synthesized pure nanocrystalline $Fe_{16}N_2$ powder with a magnetization of greater than 200 emu/g, at an external field of approximately 16 kOe, a saturation magnetization of approximately 240 emu/g, and an intrinsic coercivity of approximately 2,300 Oe.

This is the first instance where anyone has produced $Fe_{16}N_2$ powder with both high-saturation magnetization and a high coercivity.[8–11]

Subsequent to this discovery, AMC has developed a reliable and reproducible technique to produce $Fe_{16}N_2$ nanocrystalline powders. In turn, the subsequent challenge, addressed in this work, was to consolidate these powders into dense bodies for use in practical applications. However, the presence of a phase transition at approximately 240 °C in $Fe_{16}N_2$, where the nitrogen atoms begin to reorganize and form a mixture of Fe_4N and α-Fe, imposes a severe restriction on the consolidation method for this material. That is, using conventional high-temperature powder metallurgical techniques that are typically employed in magnetic materials processing, such as those for Sm-Co and Nd-Fe-B are not applicable.

It was hypothesized that ECAE is one of the few techniques that may be applicable to the consolidation of the $Fe_{16}N_2$ powders, due to the fact that this technique could be performed even at near room temperature to densify powders. Published literature on materials using ECAE indicate that it has been used to densify a wide range of powders of iron, copper, or magnesium alloys.

3. Experimental Procedures

3.1 Synthesis of the $Fe_{16}N_2$ Powder

The $Fe_{16}N_2$ powder used in the consolidation experiment was fabricated in a similar manner as described previously. Initially, the prepared iron oxy-hydroxide (FeO-OH) was prepared, decomposed into an oxide, and then reduced in flowing hydrogen to metallic Fe using a fluidized bed reactor at temperatures between 280 and 400 °C, followed by nitriding the resultant metallic Fe powder. The nitride product was formed in a flowing nitrogen (and ammonia) gas mixture at lower temperatures between 140 and 180 °C for several hours. In some experiments, part of the Fe was replaced with manganese (Mn) during the preparation of the hydroxide. Additionally, in more recent experiments, a commercially procured nanocrystalline iron-oxide powder was used as the starting material.

3.2 Extrusions

ECAE experiments were performed at the US Army Research Laboratory (ARL), Weapons and Materials Research Directorate, at Aberdeen Proving Ground, Maryland. Unlike conventional clamshell tool designs, the ECAE tool at ARL is equipped with a sliding wall design that allows for reduced friction forces enabling

the extrusion processing of long aspect ratio bars having dimensions of 19.05 × 19.05 × 203.2 mm (3/4 × 3/4 × 8 inches). The die angle is 90°, which imparts the work piece with a strain of roughly 1 during each pass. For process temperatures below 300 °C, the tool is heated to the desired value and the sample after insertion is allowed to equilibrate prior to the start of the extrusion. With this tool geometry, both solid bar and powder extrusions can be made. For the latter case, a suitable diameter and depth hole is bored into a can and back-filled with the powder sample.

Typically, for metallic powders, which are intrinsically hard, the canning materials are made from pure nickel (Ni). This is to match the relative impedance of the extrudate material with that of the can; otherwise, one will flow around the other, without full consolidation. Thus, the nanocrystalline $Fe_{16}N_2$ powder samples were sealed under inert atmosphere into Ni metal containers. They were then loaded into the extrusion press and heated to 100 and 150 °C, respectively. Once the sample stabilized at the target temperature, the sample was extruded. About 18 of such extrusions were performed on the samples. For the extrusions, several routes, namely A, B_C, and C were tried; however, in some cases, especially beyond Pass 3, the canning material failed. Therefore, due to the low ductility of the specimen and the Ni can at the extrusion temperature, imposing a less severe deformation would be preferable. Consequently, hereafter, only Route C was attempted. Using Route C, the extrudate is rotated around its long axis by 180° between successive passes. Two modes of extrusions, videlicet, 1) 4C and 2) 6C gave the most promising results.

After the extrudate containers cooled down to ambient temperature, the $Fe_{16}N_2$ samples were extracted by sectioning using an electric discharge machine. Subsequently, the samples were metallographically polished. In a few cases we have succeeded in obtaining cubes of $Fe_{16}N_2$ samples.

Bulk densities of these specimens were determined using pycnometry in a toluene solution. The best density for an extruded sample is 5.91 g/cm^3, which is approximately 75% of the X-ray density of the $Fe_{16}N_2$ phase.

B-H loop measurements on the extruded samples were performed using a Walker Hysteresisgraph unit (see Section 3.3).

3.3 Evaluation of the Magnetic Properties

Three different types of magnetic properties were determined on the powders as well as extruded specimens prepared in this research. Illustrative examples of these results are described in the following subsections.

3.3.1 Magnetic Properties of the Precursors

Open M-H loops of the powders were determined in a vibrating sample magnetometer (VSM) at room temperature. About 30 mg of the specimen was packed in a specially designed sample capsule. The sample was loaded into this capsule from the reactor without ever exposing it to air after the formation of the nitride in the reactor, which was located inside of an argon glove box—ensuring the specimen did not oxidize. Further, the sample was tightly packed so that the particles did not move physically in the presence of the applied magnetic field. The sample capsule was then loaded into the magnetometer. Measurements were made following the routine procedure prescribed for the VSM.

A 4-quadrant M-H loop was then obtained on all of the samples following this protocol and a standard Ni sample was used as a reference.

A typical M-H loop of a powdered sample is shown in Fig. 3. This sample is made from a co-precipitate of FeO-OH with the addition of 3 wt% Mn. The final composition, after reduction and nitriding contains nearly 83 wt% of the 16:2 phase (as determined from an analysis of the thermo-magnetic analysis [TMA] curve, see Fig. 3). We have succeeded in making a number of powder samples with magnetization of greater than 200 emu/g, at an external field of approximately 16 kOe, a saturation magnetization above 240 emu/g, and coercivities of greater than 1,000 Oe.

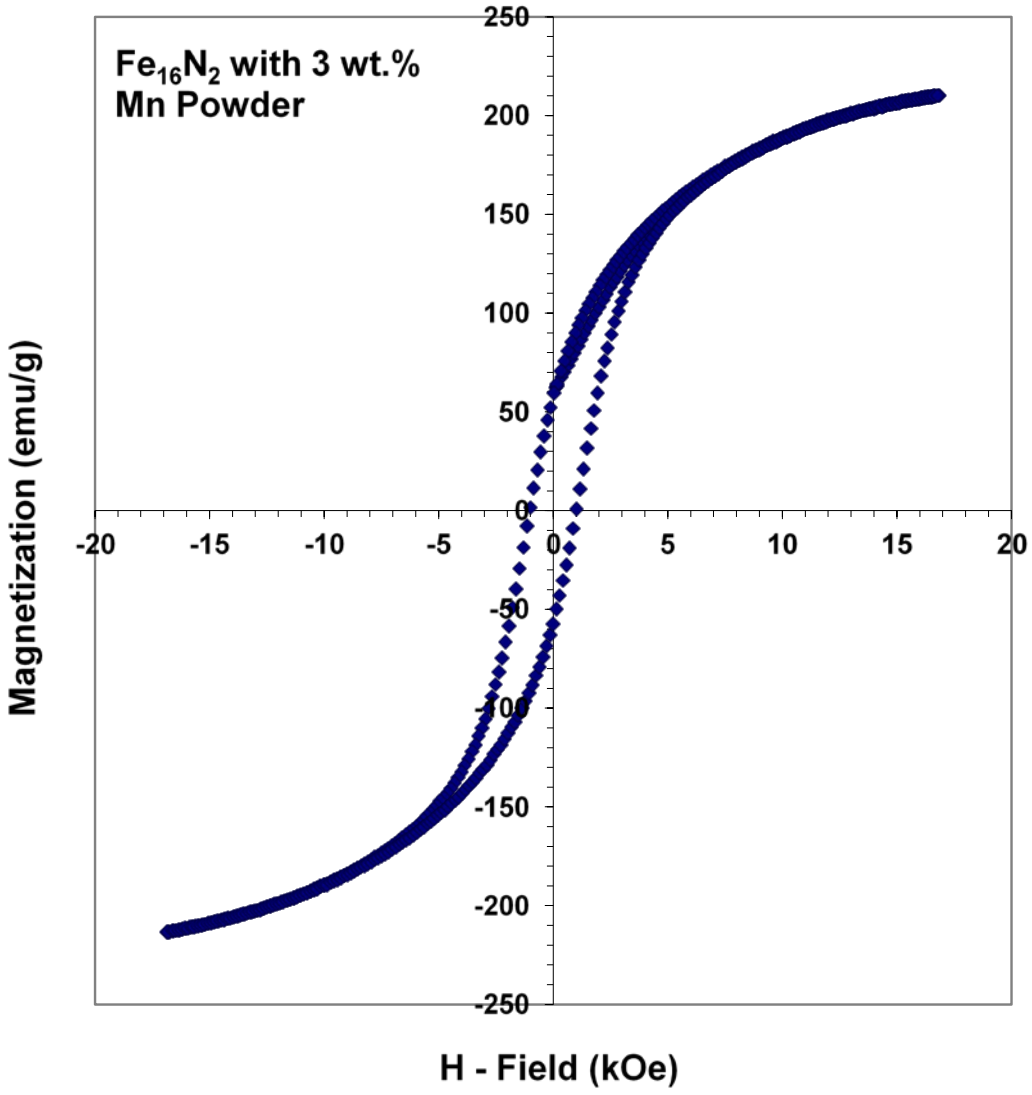

Fig. 3 M-H loop of an $Fe_{16}N_2$ (with 3 wt% Mn) powder. (Note: The magnetization at 16 kOe is ~208 emu/g. The saturation magnetization is 240 emu/g and the coercivity of this powder is ~1,000 Oe.)

In more recent experiments, nanocrystalline powder of $Fe_{16}N_2$ from commercially obtained nanocrystalline α-Fe powder was prepared. These specimens showed coercivities of approximately 2,300 Oe. An M-H loop of such a powder sample is shown in Fig. 4. Typical high-resolution transmission electron micrographs (TEMs) of this powder are shown in Fig. 5; this was obtained on the specimen supplied by AMC to Dr Larry Allard at Oak Ridge National Laboratory (ORNL), Oak Ridge, Tennessee, under a program supported by Advanced Research Projects Agency-Energy (ARPA-E).

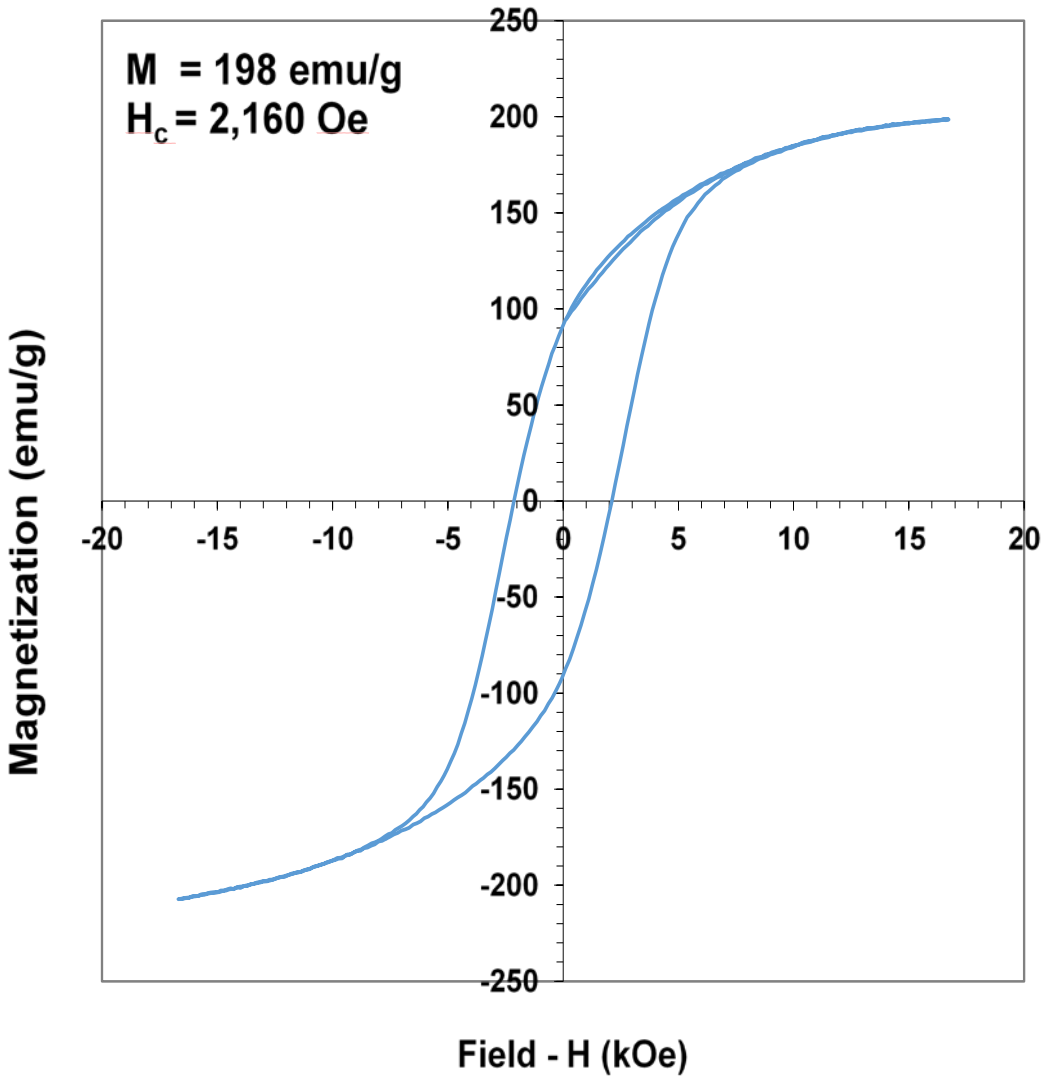

Fig. 4 M-H loop of an $Fe_{16}N_2$ powder prepared from nanocrystalline commercially procured Fe_2O_3. (Note: This sample exhibits a magnetization of 198 emu/g at an external field of 16 kOe, a saturation magnetization of 245 emu/g, and a coercivity of 2,160 Oe. High-resolution TEM of this powder is shown in Fig. 5.)

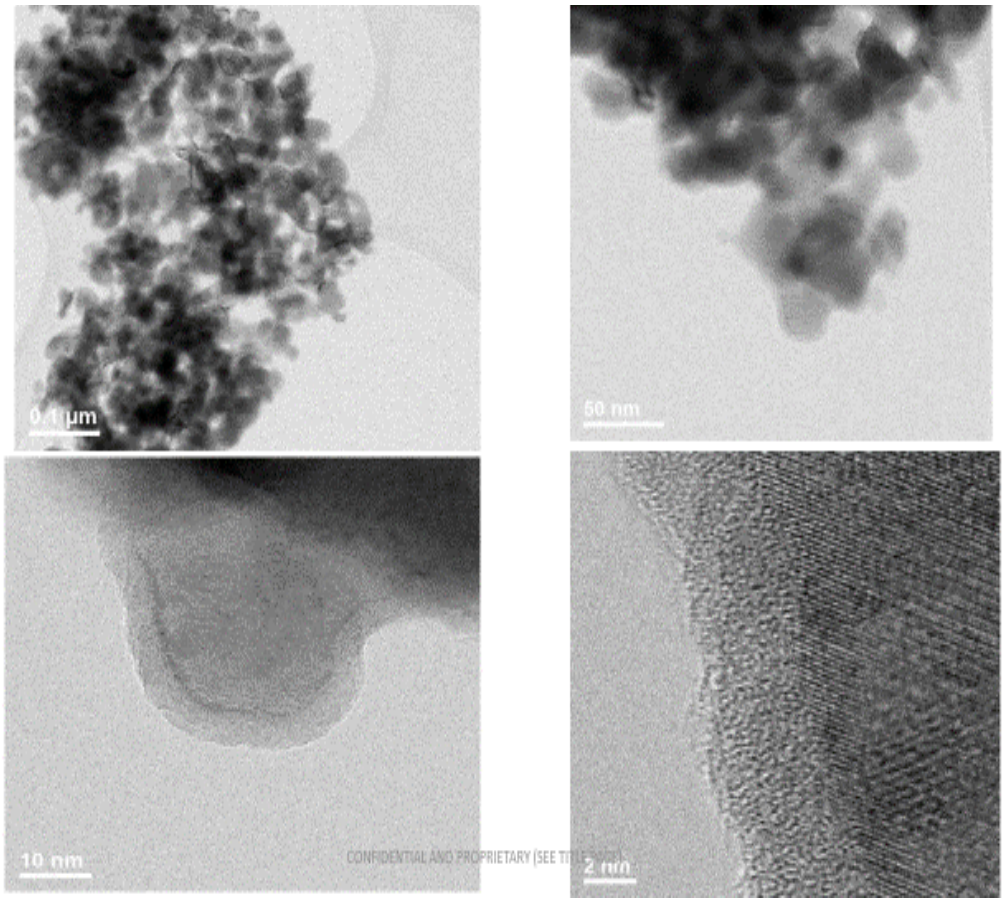

Fig. 5 High-resolution TEM images of the $Fe_{16}N_2$ powder made from commercial nanocrystalline Fe_2O_3 powder. (Note: Top left image shows a cluster of a large number of particles. Top right image shows that the size of the particles are ~20 nm. Bottom left image shows that the particle surface is coated with an iron-oxide layer of ~2-nm thickness (this layer is created for passivation purposes). The bottom right image shows the interior of the particle, having an ordered crystalline lattice; the spacing of the lattice planes confirms that the particles consist of the $Fe_{16}N_2$ composition. (TEM images are used with permission courtesy of Dr Larry Allard, ORNL).

3.3.2 TMA of the Precursors

TMA was performed on the powders; this method employs a combination of VSM and the use of a Bitter magnet. The external magnetic field was approximately 300 Oe and the temperature of the specimen was increased from room temperature to about 800 °C. The sample was maintained under a pure-argon atmosphere to prevent oxidation of the specimen. For instance, Fig. 6 shows the variation of magnetization versus temperature at an applied field of approximately 300 Oe.

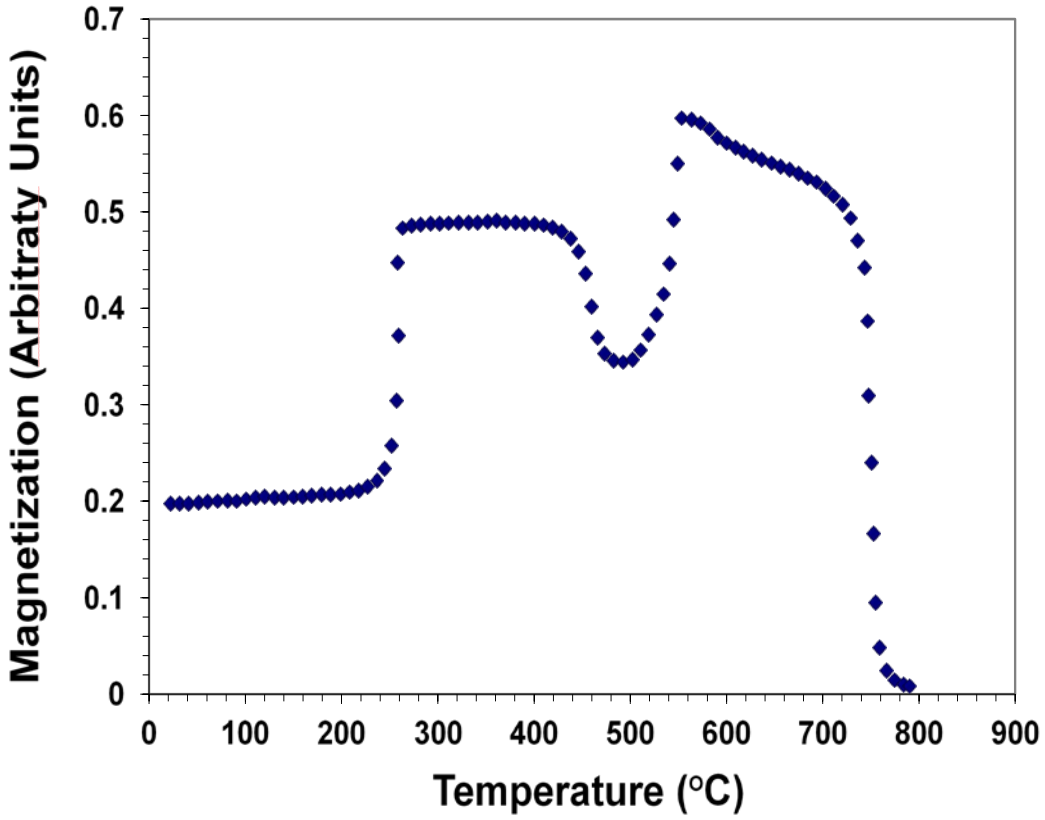

Fig. 6 TMA of $Fe_{16}N_2$ powder performed at a low-applied magnetic field of ~300 Oe. (Note: The structure of $Fe_{16}N_2$ rearranges into Fe_4N + α-Fe at a temperature of ~240 °C.)

At room temperature the measured magnetization is very small. This is due to the fact that the applied field is much lower (i.e., ~300 Oe) than the anisotropy field (16,000 Oe) of the 16:2 phase. At temperatures slightly above 220 °C (see Fig. 6), the magnetization increases sharply. This transition shows up as a typical first-order phase transformation. This change is attributed to a transformation from the $Fe_{16}N_2$ phase to a mixture of $Fe_{14}N$ and α-Fe phases. Both latter phases are cubic and thus the sample no longer has magnetic anisotropy. In earlier studies, we have verified the X-ray diffraction patterns of the powder before and after this phase transformation. This has also been confirmed by several other researchers, for instance, see Widenmeyer et al.[8]

The relative height of this change in the magnetization (i.e., relative to the total height in the plot of M vs. T) gives a reasonably good, semiquantitative percentage concentration of the $Fe_{16}N_2$ phase.

Thus, in summary, it has been successful in determining whether or not the $Fe_{16}N_2$ phase is present in a specimen (from the presence of the phase transition at ~240 °C) and, if so, to what degree (from the relative magnitude of this change). Such an analysis was carried out for all of the powder samples as well as the extruded specimens in this study.

3.3.3 Magnetic Measurements of the Extrudates

Results obtained on the B-H loop measurements from the ECAE sample, determined with a Walker Hysteresis-graph unit are shown in Figs. 7 and 8. Again, a standard Ni sample was used as a reference. These measurements are obtained in a closed-loop mode and, therefore, are not subject to any uncertainty (as would be the case with open-loop M-H measurements made in a VSM that is subject to demagnetization corrections).

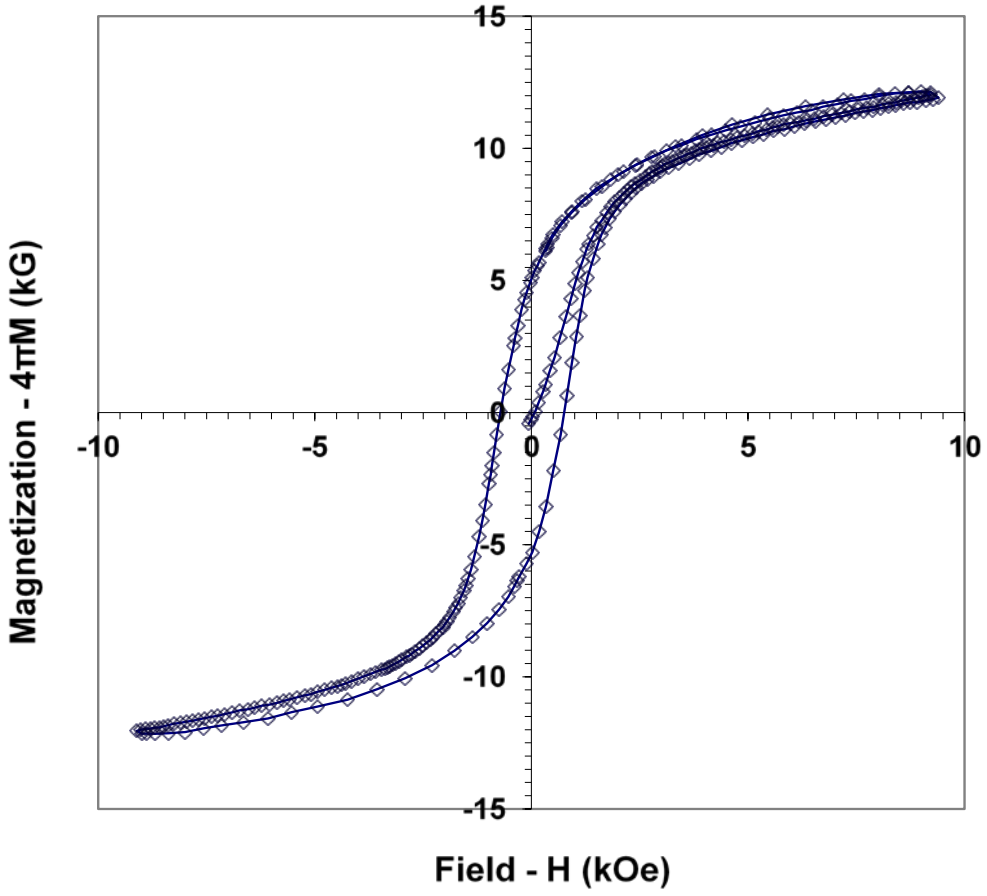

Fig. 7 B-H loop of an extruded sample (Extrusion 8) from the $Fe_{16}N_2$ (with 3 wt% Mn) powder. (Note: This sample exhibits the highest value of maximum induction, 12 kG. The remanence of this sample is ~5 kG and its coercivity is ~800 Oe. The density of this sample measured using pycnometry is 5.41 g/cm^3—about 73% of the X-ray density of the material.)

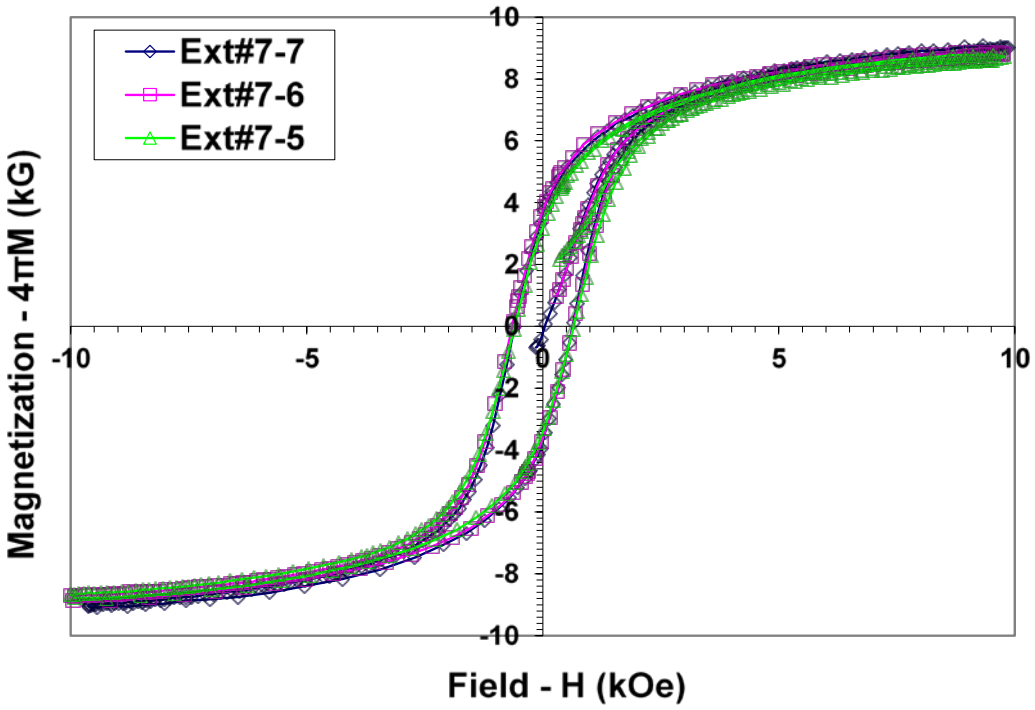

Fig. 8 B-H loops of the extruded sample (Extrusion 7) from the $Fe_{16}N_2$ (with 3 wt% Mn) powder. (Note: The sample was tested in 3 different orientations relative to the extrusion direction. The almost identical loops for the 3 directions indicate a likely lack of strong magnetic texture.)

For some extrusions, we were successful in being able to section and obtain cube-shaped specimens from the extruded samples using a diamond wheel. As such, we have measured the B-H loops of the specimens along the 3 orthogonal directions, relative to the extrusion direction. An illustrative result is shown in Fig. 8. The extrusion conditions, temperature, route, and number of passes, including the results are summarized in the following table. It is clear from the results that under the extrusion conditions employed in this study, we were unable to obtain samples with magnetic texture. However, it is possible that under different extrusion conditions, we may be able to prepare higher density samples that may show magnetic texture. This is an area of research worth pursuing in the future.

Table Extrusion conditions, bulk density, estimated concentration of the $Fe_{16}N_2$ phase, induction, remanence, and coercivity of the ECAE Samples 7, 8, 9, and 10

Sample	Extrusion conditions	Density (g/cm^3)	Fraction of $Fe_{16}N_2$ phase (%)	$4\pi M_{max}$ (kG)	B_r (kG)	H_c (kOe)
Ext 7-5	4C – 150 °C	4.85	65.3	8.78	3.18	0.62
Ext 7-6	4C – 150 °C	…	…	8.87	3.51	0.64
Ext 7-7	4C – 100 °C	…	…	9.09	3.55	0.61
Ext 8	4C – 150 °C	5.41	72.8	12.16	5.04	0.72
Ext 9-7	6C – 150 °C	5.27	70.9	10.37	3.18	0.41
Ext 10	8C – 100 °C	5.08	68.4	11.14	4.93	0.62
Precursor powder	…	…	83.0	210 emu/g	…	~1.0

Note: Properties for Sample 7 were measured along the 3 orthogonal directions, relative to the extrusion direction. For comparison, results on the presursor powder sample are also included. All of the extrusions were performed using the same batch of presursor powders.

An examination of the samples consolidated by other means is illustrated in Fig. 9. The M-H loops reveal that the shock-compacted specimen at the Georgia Institute of Technology resulted in the formation of a pure-Fe specimen; this is reflected in the low coercivity and high magnetization as well as the shape of the loop.[9] The magnetically compacted magnets at IAP Research, Inc., Dayton, Ohio, had low-bulk densities. This is clear from its low magnetization value. However, the coercivity remained almost intact. In contrast, ECAE resulted in a 75% dense specimen that showed high magnetization and reasonably good coercivity.

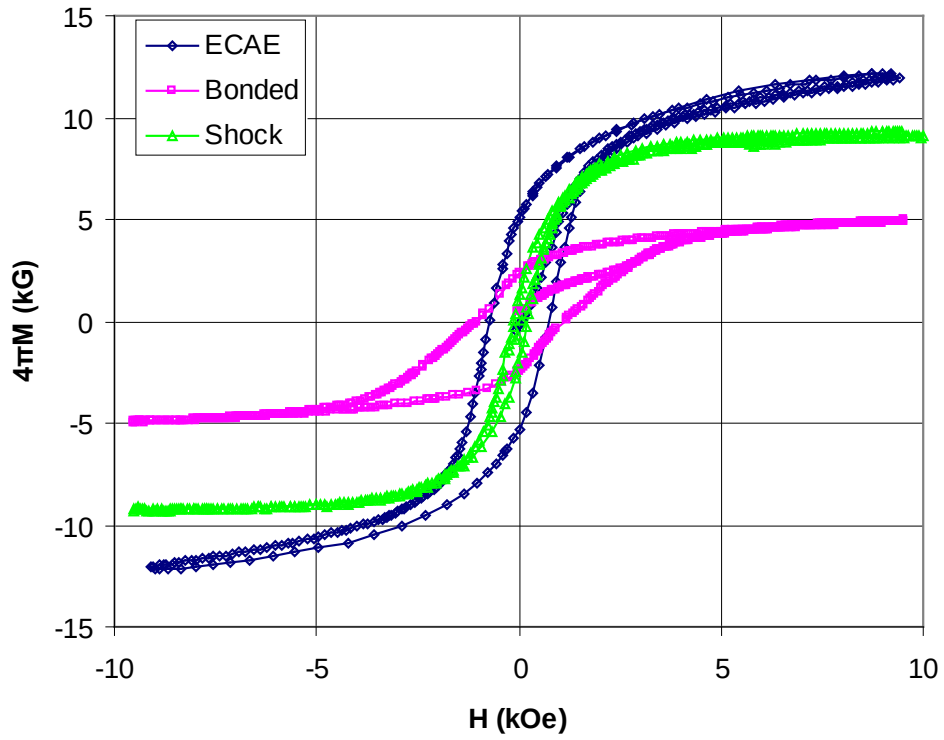

Fig. 9 M-H hysteresis loops obtained for the $Fe_{16}N_2$-based permanent magnets prepared by 3 different consolidation methods. (Note: Comparatively, the ECAE sample has the best performance with the highest energy product and saturation magnetization. The other 2 methods either lack the coercivity, or a large energy product, or high magnetization.)

4. Results and Conclusions

The experiments evaluating the extrudability via ECAE of the $Fe_{16}N_2$ phase (containing 3 wt% Mn) showed several important observations:

- Under the limited experimental conditions investigated thus far, the best density of the extruded specimens is about 75% of the X-ray density.
- Extrusions at temperatures up to approximately 150 °C do not result in appreciable changes in the magnetization (i.e., compared to the powder).
- Extrusions at a temperature of approximately 150 °C result in a decrease of approximately 200 Oe out of approximately 1,000 Oe in the intrinsic coercivity. However, we believe that if we start the extrusions with powders that exhibit higher coercivities (i.e., the powder now synthesized, with a coercivity of ~2,300 Oe), it is conceivable to compromise, accept some losses in the coercivity, and yet still succeed in making fairly strong magnets.

The best saturation induction obtained on the extruded specimen is approximately 12.5 kG. It is believed that this value will improve considerably if we can succeed in fully densifying the material.

The number of extrusions was limited. Furthermore, options in possible or alternate ECAE routes have not been explored in this feasibility study. There is reason to believe that there are several more tangible process variables that can be adjusted to improve the density and induce crystallographic and magnetic texture in the material.

Recent more optimized experiments have succeeded in obtaining nanocrystalline $Fe_{16}N_2$ powder. High-resolution TEM shows that uniform particles of approximately 20 nm are formed under these experimental conditions. The results are highly reproducible.

It is believed that further optimization of these extrusion experiments, especially, on nanocrystalline powders, may yield bulk magnets with energy products of approximately 20 MG·Oe in isotropic magnets and greater than 35 MG·Oe in anisotropic magnets—provided that we are able to induce texture (i.e., in the latter case).

Finally, as shown in Fig. 9, in a comparison of B-H loops (i.e., a shock-compacted magnet prepared at the Georgia Institute of Technology, magnetically compacted bulk magnet prepared at IAP Research, Inc., Dayton, Ohio, and an extruded bulk magnet prepared via ECAE at ARL from $Fe_{16}N_2$ powder specimens supplied by AMC), it may be concluded that, despite an incomplete densification, the ECAE extrusion method provides the best combination of coercivity retention and greatest energy product in obtaining bulk magnets based on this composition.

5. Prognosis

Based on our research, it is desirable to continue the extrusion activity to explore the following:

- Move forward the basic fundamental research in Fe-based magnetic materials. For example, the study of the nature of the magnetic coupling between Fe and Mn atoms in the 16:2 phase, how to improve the thermal stability of the 16:2 phase (see Fig. 6), and the mechanism of coercivity in these types of magnets all need better understanding and explanation.
- To explore additional experimental conditions in the ECAE extrusion process to fabricate low-cost, high-energy magnets with both isotropic and anisotropic characteristics. These include the introduction of a

preconsolidation step, higher number of extrusion passes, the use of confinement pressures, and alternative route geometries.

- To retain US leadership in this emerging area of magnetic materials—without containing a rare-earth metal. This is extremely critical for the scientific, technical, and economic development of the United States of America. Although, there is a temporary relief from the “rare-earth crisis”, it is better and more scientifically sound in the long term to pursue this type of research and development. For instance, extruded Fe-N magnets could play a significant role in fulfilling the demand for isotropic magnets with higher energy products of about 15–20 MG·Oe, which could serve as a bridge between the existing inexpensive ceramic ferrite magnets (i.e., maximum energy product of 3–5 MG·Oe) and the expensive bonded Neo magnets (i.e., Nd-Fe-B-based) with energy products of about 15–20 MG·Oe.

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List of Symbols, Abbreviations, and Acronyms

AMC	Advanced Materials Corporation
ARL	US Army Research Laboratory
ARPA-E	Advanced Research Projects Agency-Energy
B	boron
B-H	Magnetic Induction – Magnetizing Field Intensity
bcc	body centered cubic
C	degree Celsius
cm	centimeter
Co	cobalt
DOE	Department of Energy
ECAE	Equal Channel Angular Extrusion
emu	electromagnetic unit
Fe	iron
Fe-N	iron nitride
FeO-OH	iron oxy-hydroxide
g	gram
kG	kilogauss
mg	milligram
MG	megaGauss
M-H	Magnetization – Magnetizing Field Intensity
mm	millimeter
Mn	manganese
Nd	neodymium
nm	nanometer
Ni	nickel

Oe	Oersted
ORNL	Oak Ridge National Laboratory
Sm	samarium
TEM	transmission electron micrograph
TMA	thermo-magnetic analysis
VSM	vibrating sample magnetometer
weight-percent	wt%

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