Al₂O₃ “self-coated” iron powder composites via mechanical milling

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Abstract

Electrically insulated ferrous powders permit isotropic magnetic flux, lower core losses, and structural freedom for state-of-the-art electromagnetic (EM) core and device designs. Many current coating materials are limited by low melting temperatures, which leads to insufficient insulation of powders, resulting in metal-on-metal contact. Use of a high-temperature coating material, such as alumina, could alleviate these issues. In this work, iron powder was mechanically milled with alumina media, to yield plastically deformed, alumina-coated iron particles with improved magnetic saturation, elastic modulus, and hardness. Various milling times and media ball sizes are investigated to maintain particle size, insulate powders uniformly, and optimize properties after compaction and curing. We found that longer milling times yielded more dense powder coatings and lower magnetic saturation.

Keywords: Composite materials, powder metallurgy, SEM, magnetization

1. Introduction

Soft magnetic composites (SMCs), used in various low power electromagnetic devices, incorporate magnetic, metallic particles (iron-, nickel-, or cobalt-alloys) coated with electrically insulating materials [1–5]. SMCs are advantageous over traditional steel laminates due to their lower core losses, 3D magnetic flux capabilities, and the structural freedom that allow for complex designs. These materials also possess good green strength and formability as well as high saturation magnetization and low coercivity.

The future of SMCs lies in higher frequency applications such as transformers, jet engines, and electric motors; however, the limiting factor for further advancements and implementation of these materials is the stability of the materials currently used as insulating coatings [6–8]. The conventional process of developing SMCs from coated powders requires high compaction pressures (>800 MPa) to maximize density and heat treatment temperatures between 570 °C and 775 °C for proper iron stress relief [9, 10]. Organic coatings such as epoxy resins or silicone polymers cannot withstand post-compaction heating above 450 °C before they begin to degrade, which results in low mechanical strength and metal-to-metal contact points that lead to large eddy currents and reduced magnetic permeability [11–14]. For this reason, inorganic coatings such as phosphates, MgO, Al₂O₃, Y₂O₃, and ZrO₂ are superior high temperature coating alternatives [15–18].

Inorganic, high temperature protective coatings are capable of electrically insulating iron powders even after compaction and heat treatment, to eliminate eddy currents and improve

efficiency of devices. Phosphates and MgO are both limited by reduced resistivity after annealing temperatures below 600 °C [19–21]. Alumina (Al₂O₃), with its high melting point (2050 °C) is a strong candidate for high temperature coatings that confine eddy currents within individual iron particles after compaction and curing steps. The highly stoichiometric structure and large band gap of α -Al₂O₃ make electronic conduction extremely difficult, which is essential for reducing eddy currents in iron-based composites [22].

Several wet chemistry and dry coating techniques have been developed for coating iron-alloy powder surfaces with protective layers of alumina; however, uncontrollable phase transformations often led to degraded magnetic properties. For example, the formation of maghemite (Fe₂O₃) during the thermal decomposition of boehmite (γ -AlOOH), a precursor to alumina, led to decreased volumes of iron, which thus decreased the magnetic permeability [23, 24]. Alumina-coated iron nanocomposites were also synthesized by wet chemistry; however, the magnetization suffered due to the formation of FeAl₂O₄ at reducing temperatures below 800 °C [13]. Both studies had extensive processing steps and led to undesired oxidation of the Fe core, which reduced magnetization. Dry coating methods, based on Van der Waals interactions between large (host) Fe powders and small (guest) Al₂O₃ particles, have also been developed. Jay et al. used a mechanofusion process to coat Fe powders ($d_{ave} = 150 \mu\text{m}$) with alumina particles ($d_{ave} = 9 \mu\text{m}$), then subsequently oxidize and heat treat the powders to study oxidation kinetics [25]. Due to cracks in the Al₂O₃ coating from the processing methods, iron oxide layers formed on Fe surfaces during the oxidation process, which would thus decrease magnetization due to reduced iron content. An additional form of dry coating, is mechanical milling (by high-energy ball milling), conventionally used for deformation and refinement

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purposes, but can also be used for an unique and simple coating method [26–28]. The mechanical forces introduced between the media material and iron powder during milling allow for powders to be internally stressed and externally coated while maintaining particle size improves mechanical strength [29]; but decreases the magnetic permeability by hindering domain wall motion [30]. Therefore, stress-relief temperatures are necessary to improve magnetic properties, but consequently hinder mechanical properties. These temperatures can be reached by using high temperature coatings such as alumina.

In this paper, we discuss the use of alumina media balls during mechanical milling to coat iron powder surfaces in the absence of additional powder. We show, through a combination of structural and chemical analyses along with magnetic and mechanical testing, that these “self-coated” powders possess uniform alumina coatings capable of minimizing eddy current losses, and have relatively strong magnetization and mechanical strength.

2. Experimental

High purity, Ancorsteel 1000C iron powders were provided by Hoeganaes Corp. (Cinnamonsin, NJ). Powders were sieved to produce large particle sizes of 250 - 400 μm . A high-energy SPEX 8000M shaker mill was used to ball mill iron powder in an alumina ceramic vial, with one of four alumina media ball sizes (0.5 mm, 1 mm, 2 mm, and 3 mm). Trials varied from 2 to 24 h with ball-to-powder weight ratios (BPRs) of 2:3. Powders were uniaxially compacted with 3 GPa pressure and cured for 1 h at 500 °C or 900 °C in an argon and hydrogen (3%) atmosphere. The experimental procedure is depicted in Figure 1.

The phase identification and structural analysis of powder samples were characterized by x-ray diffraction (XRD) (Rigaku, Japan) with Cu-K α radiation ($\lambda=1.541 \text{ \AA}$). Williamson-Hall plots were used to analyze the crystallite size and microstrain of milled powders [31]. Magnetic properties were measured from $\pm 1.5 \text{ T}$ at room temperatures using a Physical Property Measurement System (PPMS®) with a vibrating sample magnetometry (VSM) extension (Quantum Design, USA). The surface morphology and porosity of powder samples were observed by scanning electron microscopy (SEM) (FEI, USA). Additionally, powder compacts were traditionally polished and imaged via SEM coupled with energy-dispersive spectroscopy (EDS) for chemical analysis of powders and powder compacts. Mechanical properties were investigated by nanoindentation (MTS, USA) at 200 nm depth. Alumina-coated iron powders were analyzed for magnetic, mechanical, and structural property dependence of milling time and media ball sizes.

3. Results

3.1. Effect of Milling Time

Iron powders ball milled at room temperature with 2 mm Al_2O_3 media balls in an alumina vial were imaged using SEM for particle and surface morphologies. Figure 2 shows a transition of particle shape from irregular for as-received powders to

Figure 1: Procedure for producing alumina coated iron powders via mechanical milling.

comparatively spherical for powders milled for 24 h. Extended milling times resulted in reduced particle surface areas creating rounded edges and spherical morphologies.

Ball milled powders became coated directly from the alumina media and media balls; with no additional powder added. A “self-coated” surface was characterized by a textured morphology and sub-micron sized particles as seen in Figure 3, for powder milled for 24 h. Powders milled for longer periods of time ($> 12 \text{ h}$) exhibited an excessive amount of agglomerations of finer sized particles in outermost regions, which is an indication of coated surfaces.

X-ray diffraction patterns confirmed increased microstrain and alumina content on iron powders milled for 2 to 24 h (Figure 4). Initial, unmilled iron powder displayed minimal peak broadening and no trace of alumina content as expected. As milling time increased, Fe peak broadening increased and peak intensity decreased as evident of reduced crystallite size and increased microstrain. Calculations using the Williamson-Hall technique confirmed the decrease in crystallite size and increase in microstrain. Al_2O_3 peaks began to emerge for powders milled for 4 h or longer.

Figure 2: Iron powders milled with 2 mm media balls for (a) 0 h, (b) 2 h, (c) 8 h, and (d) 24 h in alumina vial.

Less deformed powders allowed for improved mechanical interlocking and higher densities (7.3 - 7.4 g/cm^3), compared to more deformed powders ($\sim 6.8 \text{ g/cm}^3$) using 3 mm balls or times greater than 16 h. Nanoindentation was performed on polished powder compacts to measure elastic modulus and

Figure 3: Iron powder milled for 24 h with 2 mm Al_2O_3 media balls in an alumina vial. Inset image shows lower magnification of zoomed in surface.

hardness of iron after being milled, compacted, and cured. Figure 5 illustrates as milling time increased to 16 h, the hardness of Fe increased slightly, indication of more stressed regions, and decreased at 24 h. Higher curing temperatures, nominally 900 °C compared to 500 °C led to decreased hardness. Elastic modulus remained relatively constant throughout the experiments.

Cross-sectional SEM images of powder compacts revealed a highly dense, alumina coating on iron powders. Powders milled for 24 h, compacted, then cured at 500 °C, are seen in Figure 6, in which light grey regions represent Fe particles, while dark areas bordering iron particles portray Al_2O_3 coating as confirmed by XRD. The inset image of Figure 6 represents the highlighted region depicting a Fe- Al_2O_3 -Fe junction. Finer, sub-micron sized particles of alumina were found between iron powders with a layer thickness of roughly 10 μm .

Complimentary elemental mapping from a magnified region in Figure 6 reveals aluminum and oxygen elements uniformly distributed on iron powder surfaces (Figure 7a). The oxygen content found within the iron powder regions was most reasonably due to exposure to air after mounting and polishing. EDS images exhibit clear isolation of iron particles with oxygen and aluminum, which reduce metal-on-metal contact points. Similar results were found for powders milled for shorter periods of time and higher curing temperatures up to 900 °C. Alumina coating thickness determined by cross-sectional analysis ranged from 1 to 20 μm , depending on processing parameters. Therefore, alumina coating of iron powders was effective by means of mechanical milling.

Magnetization saturation and coercivity were evaluated using VSM measurements of cured powder compacts. Figure 8 indicates that magnetization decreased with increased mill time. Powders milled for 2 h possessed the highest magnetization, comparable to that of pure iron (~217 emu/g [32]). Figure 9 shows hysteresis loops of powder compacts milled for 2, 4, and

Figure 4: X-ray diffraction patterns of Fe/ Al_2O_3 powders milled with 2 mm media balls for various amounts of time.

Figure 5: Nanoindentation results for hardness and elastic modulus for powders milled with 2 mm media balls for various amounts of time.

24 h, representing soft magnetic properties and low coercivities for all samples. For as-compacted powder composites, magnetization decreased with increased mill time. Powder compacts cured at 500 °C had higher saturation magnetization than 900 °C cured compacts. All samples had relatively low coercivities below 15 Oe.

3.2. Effect of Media Ball Size

With a constant milling time, structural, mechanical, and magnetic properties were found to depend on media ball sizes, ranged from 0.5 to 3 mm in diameter. SEM images presented in Figure 10 show powders milled with (a) 1 mm or (b) 3 mm media balls for 4 h. Spherical particles were obtained when milled with large (3 mm) Al_2O_3 media balls, but were not seen in powders milled with smaller (0.5 or 1 mm) media balls.

EDS, performed in conjunction with SEM images, allowed for chemical analysis of exterior regions of powder milled for 4 h (shown in Figure 11). Finer particles of aluminum-oxide

Figure 6: SEM image of polished powder compact from powder milled for 24 h with 2 mm Al_2O_3 media balls, compacted, and cured at 500 °C. Inset shows zoomed in view of boxed area of SEM image.

sufficiently coated iron powder surfaces from ball milling with alumina media. Powders milled with 3 mm Al_2O_3 media had increased amounts of agglomerations of finer particles on outermost regions as seen in Figure 11b, similar to that of powders milled for longer periods of time shown previously. The dominant elemental components of the surfaces of milled powder were Al, O, and Fe (Figure 11c). All powders milled for 4 h with 0.5 to 3 mm media had evidence of Al_2O_3 coating according to XRD data. Fe peaks broadened and peak intensity decreased as media ball sizes increased indication of increased microstrain and decreased crystallite size, similar to previously reported XRD patterns for increased mill times.

Figure 7: SEM images and EDS maps of a compact from powders milled for 24 h with 2 mm Al_2O_3 media balls, compacted, and cured at 500 °C.

We found the hardness of powder milled with 0.5 or 1 mm balls to be slightly lower than the hardness of powder milled with larger media balls, as presented in Figure 12. Less deformation and lower defect densities were present when smaller

media balls were used, therefore reduced hardness was expected. As before, compacts cured at higher temperatures had lower hardness. Elastic moduli remained relatively constant throughout, as expected.

Magnetization was found to depend greatly on media ball sizes. Higher saturation magnetization was measured for powder milled with 0.5 or 1 mm media balls for 4 h, and was found to be similar to that of powder milled for 2 h with 2 mm balls. Powders milled with 3 mm balls for 4 h resulted in lower magnetization results, similar to that of powders milled for 8 or 16 h with 2 mm media balls. As previously reported, higher curing temperatures resulted in lower magnetic saturation of powder compacts.

4. Discussion

Ball-to-powder ratios are critical in determining deformation and size reduction outcomes using high-energy ball milling. Low BPRs allow for minimal amounts of particle size reduction while continuing to deform powders to obtain the desired mechanical strength. In the following two sections, we outline our results and how they compare to studies with varying parameter space in media ball sizes, milling times, and particle sizes.

Figure 8: Magnetization results (M_s) using VSM as a function of mill time (h) for alumina-coated iron powder compacts cured for 1 h at the specified temperature.

4.1. Effect of Milling Time

We found powders had less deformation and maintained rough morphologies when milled for shorter periods of time, nominally 2 to 4 h, due to less interactions with the medium, directly related to the small BPRs and media ball sizes. High-energy mixers are predominately used for the reduction of particle sizes, often from mm down to nm. High ball-to-powder ratios (BPRs) (8:1 or higher) and large media balls (6 mm to 12 mm) reduce particle sizes via plastic deformation and fracturing processes. Liu et al. [26] reduced iron powder by high-energy

milling of particle sizes from 25 to 2 μm after 200 h of milling using BPRs of 10:1 with stainless steel balls of 10 mm and 6 mm. Our methods of utilizing low BPRs of 2:3 and small media balls (2 mm) allow for numerous contact sites leading to minimal size reduction due to reduced kinetic energy and collision velocities [33], however, irregularly shaped particles were still deformed and often fractured to form spherical-like shapes when milled for 12 to 24 h.

Fe particle sizes were not reduced in order to improve magnetic properties, due to more ferromagnetic content increasing magnetic saturation and less defects resulting in pinning sites for domain walls hindering the magnetization process [34]. Larger particle sizes would allow higher densities as found by Taghvaei et al. [24] for iron powders of $d_{ave} = 150\mu\text{m}$ compacted at 1 GPa with 0.7 wt% phenolic resin. High uniaxial compaction pressures (>2 GPa) allowed for high green densities and minimal amounts of internal pores, thus improving mechanical strength and magnetization.

Figure 9: Hysteresis loops for alumina-coated iron powders milled for 2 h (red), 4 h (blue), and 24 h (black), then compacted and cured at 900 °C.

As work hardening intensified due to longer mill times, spherical-like shapes and Al_2O_3 coated surfaces were observed, nominally between 12 to 24 h of mill time. XRD peak broadening indicated increased defect densities, most notably dislocations [35], and decreased crystallite sizes as milling time increased. The largest amount of defects were present for powders milled for 24 h, determined from microstrain and crystallite size calculations based on the Williamson-Hall technique [31]. Results were confirmed with iron powders ball milled using BPRs of 10:1, in which powders milled for the longest amount of time, 20 h, had the largest dislocation densities and smallest particle sizes [35].

Higher defect densities and more alumina content led to a decrease in the magnetization due to the hinderance of domain wall movement. Powders milled for 24 h were thus thought to have the highest defect density and the thickest coating layers for this study due to the low M_s . Magnetic saturation

for our studies were reported as being higher than Fe- Al_2O_3 nanocomposites studied by Liu et al. [13]. Fe nanoparticles ($d_{ave}=35\text{nm}$) coated with Al_2O_3 (20%) exhibit low magnetic saturation (~ 175 emu/g, much lower than our “self-coated” powder compacts. Thicker Al_2O_3 coatings increase the overall resistivity, therefore potentially lowering eddy current losses. However, more Al_2O_3 coating leads to lower magnetization and more regions of discontinuity between ferromagnetic volume. The lowest magnetization of about 202 emu/g was found for powder milled for 24 h, compacted, and cured at 900 °C, which is much greater than results from literature.

Figure 10: Iron powders milled for 4 h with (a) 1 mm and (b) 3 mm media in alumina vial.

As the amount of crystal imperfections increased, microstrain and thus hardness also increased. Nanoindentation confirmed higher hardness for more deformed powders. Increased domain volume leads to increased hardness, as apparent for composites of Fe/ Al_2O_3 (50/50) with hardness of 4.2 GPa [36]. These matrices have lower densities (~ 4.9 g/cm³) compared to that of our compacts with greater than 6.8 g/cm³ densities. Higher curing temperatures resulted in reduced hardness, due to significantly more stress relief and recovery. Elastic moduli remained constant throughout the milling process. Variance in measurements was due to possible indentation from grain boundaries or alumina coating. Further testing is necessary and will be compared with backscattered SEM images to identify grain boundaries and specific grain orientations.

4.2. Effect of Media Ball Size

Reduced amounts of Al_2O_3 coating were present for smaller media balls when milling time was held constant, due to reduced surface areas and collision velocities. Shin et al. [33] found that milling efficiency for particle size reduction is directly correlated to ball diameter and rotational speeds. Smaller diameters greatly reduce the media ball speeds due to lower mass and therefore have reduced kinetic energy resulting in less damaging effects. Iron powders milled for 4 h with 3 mm media balls had larger amounts of alumina coating, reducing the overall ferromagnetic volume, and higher amounts of defect densities, increasing the hardness, as compared to powders milled with 0.5 to 2 mm balls. The data shows that powders milled for 4 h with 3 mm media balls, compacted, and cured at 500 °C produced the highest hardness (~ 3.5 GPa) and high magnetic saturation (~ 213 emu/g). Similar results were found for powders milled for 8 h with 2 mm media balls, indicating smaller media balls can obtain comparable, but require longer milling times. Magnetic saturation was found to be comparable

Figure 11: Surface of iron powders milled for 4 h with (a) 1 mm and (b) 3 mm Al_2O_3 media, with (c) EDS scan of particle in (b).

for both curing temperatures, for these two sample sets. Equivalent deformation may have occurred, creating similar defect densities and coating layer thicknesses, which supports similar magnetic properties. Additionally, powders milled for 4 h with 0.5 or 1 mm media balls had comparable magnetic and mechanical results to powders milled for 2 h with 2 mm media balls. These similarities are directly relatable to their structural defects and alumina coating content. Our results permit a tunability of magnetic saturation and hardness, dependent on milling time and media ball sizes.

Figure 12: Nanoindentation results of hardness and elastic modulus for powders milled for 4 h with different media ball sizes.

Figure 13: Changes in the M_s value as a function of media ball sizes.

5. Conclusions

Soft magnetic composites comprised of electrically insulated magnetic powders are ideal for more efficient, high frequency applications. Ferrous powders coated with nonmagnetic ceramic material will reduce metal-on-metal contacts to minimize eddy current losses and improve overall electrical efficiency. Ball milling methods allow for highly dense, “self-coated” powders to be synthesized directly through the use of a preferred media.

The experimental technique used in this work successfully coated Fe powders with Al_2O_3 and allowed for improved magnetic and mechanical properties compared to literature. Low BPRs allow for more contact points to emerge between powders thus decreasing collision velocities of media balls and minimizing particle size reduction. Small media balls (≤ 3 mm), permit uniform coatings and stressed regions that result in high magnetic saturation (~ 210 emu/g) and good mechanical hardness (~ 3.0 GPa) even after curing temperatures of 900 °C. Alumina coating allows for proper isolation of individual iron particles to reduce eddy current losses and be used for high frequency applications.

Various media types such as zirconia, readily available and with an even higher melting point than alumina, are predicted to have similar trends in structural and coating layer dependence of milled powders. Our “self-coating” technique has little potential of being utilized in industrial manufacturing because of the costly medium necessary for coating powder. Alumina as a coating material does have great potential for SMC applications due to proven magnetic saturation and hardness results shown here, however, higher-than-normal compaction pressures are required in order to obtain high density compacts. Overall, alumina “self-coated” iron powders were synthesized and studied for their improved magnetic and mechanical properties.

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7. References

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