Friction Consolidation of Gas-Atomized Fe-Si Powders for Soft Magnetic Applications

X. Jiang^{1,*}, S.A. Whalen¹, J.T Darsell¹, S.N. Mathaudhu^{1,2}, N.R. Overman¹

¹Pacific Northwest National Laboratory, Richland WA, 99352, USA

²University of California, Riverside, Riverside CA, 99521, USA

Abstract. *Soft magnetic materials are often limited in scalability due to conventional processes that do not retain beneficial microstructures, and their associated physical properties, during densification. In this work, friction consolidation (FC) has been studied to fabricate Fe-Si soft magnetic materials from gas-atomized powder precursors. Fe-Si powder is consolidated using variable pressure and tool rotation speed in an effort to evaluate this unique densification approach for potential improvements in magnetic properties. FC, due to the high shear deformation involved, is shown to result in uniform gradual grain structure refinement across the consolidated workpiece from the center nearest the tool to the edge. Magnetic properties along different orientations indicate little, if any, textural orientation in the refined grain structure. The effect of annealing on the magnetic properties is evaluated and shown to decrease coercivity. FC processing was able to retain the magnetization of the original gas-atomized powders but further process optimization is needed to reach the optimal coercivity for the soft magnetic materials applications.*

***Corresponding author:** Tel. 509-372-6861, email: Xiujuan.jiang@pnnl.gov

Keywords: Fe-Si, soft magnet, friction consolidation, coercivity, microstructure

Address: 902 Battelle Blvd, Richland, WA 99352

1.Introduction

The last two decades has witnessed a steadily increasing demand for soft magnetic materials for use in power generation, transmission and conversion applications^{1, 2}. The primary challenge in the design and wider adoption of energy-efficient soft magnets is a limited ability to reduce core loss at different switching frequencies during service. Nanocrystalline and amorphous magnetic materials are promising materials to circumvent this challenge owing to their reduced coercivity and enhanced electrical resistance^{2, 3}, however, a key technical barrier has been the limited ability to industrially scale-up the production of nanostructured magnetic materials and maintain the enhanced performance. Currently, melt-spinning is used to produce commercial nanocrystalline materials⁴, however, broader commercial applications may be realized if nanocrystalline precursor materials could be produced and consolidated more efficiently without degrading magnetic performance.

Friction consolidation (FC) offers unique advantages over conventional soft magnet processing approaches^{5, 6} that can help overcome challenges in the scalable processing of nanostructured soft magnetic materials. FC is a solid-state processing technique that uses a non-consumable rotating tool to generate frictional heat and plastic deformation resulting in particulate bonding, material joining or local microstructural modification. Originally developed for Al- and Mg-based

structural alloys^{7,8}, FC has been extended to ODS steel⁹, and semiconducting Bi_2Te_3 thermoelectrics¹⁰.

In low- to moderate-melting point materials, frictional heat and severe plastic deformation during processing result in continuous dynamic recrystallization in the shearing solid, which can lead to microstructural refinement when appropriate cooling regimes are known and utilized. The extent of densification and microstructural refinement within the processed zone can be affected by adjusting process parameters such as feed force, tool rotation, feed rate, and chamber boundary conditions, such as temperature and cooling rate. FC has also been shown to impart novel crystallographic textures in some materials¹⁰.

This study probes the ability of FC to consolidate fine-grained soft magnetic Fe-Si precursor powders while retaining desirable magnetic properties. The most widely utilized electrical steels are Fe-Si alloys with silicon contents of 3-3.5 wt.% to maintain balance between eddy current loss and ductility.¹¹ Significant improvement in the development of soft magnetic materials has also resulted in preferred microstructural and texture relationships^{12, 13}. For these reasons, this communication reports the fabrication of a Fe- 3.3 wt.% Si alloy system using FC to demonstrate the possibility of producing soft magnetic materials with controllable microstructures and crystallographic texture.

2. Materials and Methods

Commercially obtained gas-atomized Fe-Si (with ~3.3 wt. % Si) powders (Höganäs Corp.) were used in this study. Friction consolidation was performed using a custom friction stir processing apparatus (Transformation Technologies, Inc. Elkhart, IN). Fe-Si powders were loaded into the powder chamber in the top die half, as shown in Fig. 1. During processing, a rotating tool plunges into the chamber while compressive force is applied to the powder charge. As heat and pressure build below the rotating tool face, the powders are consolidated in a state of severe plastic deformation.

Fig. 1 Schematic showing the FC process with feedstock powders

Approximately 20g of powder was loaded into the die for each processing run. Consolidation was performed at several different thermomechanical conditions using a flat-faced tungsten-rhenium-hafnium carbide (W-Re-WC) tool. The FC runs were aborted when the tool temperature reached 950 to 990°C and this dictated the time of the processing. Sample ID and processing conditions are shown in Table I.

Table I. Processing conditions of different samples

Sample ID	Tool Speed (rpm)	Max. Pressure (MPa)	Max. Tool Temp. (°C)
A	125	14	960
B	125	34	990
C	300	122	950

Each consolidation run produced a puck nominally 6 mm thick and 25.4 mm in diameter. Following sample fabrication, microstructural and magnetic characterization was performed on each of the puck specimens. Samples were epoxy mounted and polished to 0.05 μm using colloidal silica. A JEOL 7600 field emission scanning electron microscope (SEM) was used to perform imaging and electron backscatter diffraction (EBSD) mapping. All EBSD analyses were carried out using Oxford's AZtec™ software package, an accelerating voltage of 20 keV, a probe current setting of 16 A and a working distance of 22 mm. A body centered cubic (b.c.c.) crystal structure was utilized for the analysis. Phase indexing was performed using lattice parameters for b.c.c. Iron, Laue group 11(m-3m) and space group 229 with a lattice parameter of 2.866 Å¹⁴. EBSD results confirmed the b.c.c. structure following all processing and annealing treatments. Local misorientation mapping was performed using a 9x9 filter and a defined subgrain angle of 5 degrees.

In order to test the microstructural homogeneity, a nominally 2 mm x 2 mm x 2 mm cube was extracted from two different locations of the consolidated piece (illustrated in Fig. 2) for magnetic measurements.

Fig.2 Cutting regions from half consolidated puck (top) with direction indications and an actual typical cut piece for VSM (bottom)

The magnetic properties were tested using a Microsense Vibrational Sample Magnetometer (VSM), with a maximum magnetic field of 2.3 Tesla. To study the effects of residual stress on magnetic properties after consolidation, samples B and C were annealed at 700 and 900 °C for 2 h and furnace cooled using a vacuum furnace held at 10^{-5} Torr.

3. Results and Discussion

3.1 Fe-Si Powder Characterization

In order to assess the effect of FC on the starting gas-atomized Fe-Si powder, baseline morphology, microstructure and magnetic properties are investigated. SEM images of the as-received powder are shown in Fig. 3. Size and morphology of the powder is highly irregular compared to typical spherical powder produced using gas atomization¹⁵. Larger particles are shown to be on the order of $\sim 120\mu\text{m}$ in length.

Fig. 3 20kV SEM backscatter images, showing morphology and structure of as received Fe 3.3 wt.%Si powder.

EBSD results shown in Fig. 4 illustrate the grain size from several particles. Average grain diameter of the as-received powder is measured and found to be $6.9 \pm 4.9\mu\text{m}$, with multiple grains exhibiting diameters $>20\mu\text{m}$.

Fig. 4 EBSD maps showing grayscale pattern quality images (left) along with the respective out-of-plane grain orientation maps with inverse pole figure coloring.

Magnetic evaluation of the Fe-Si powder shows a magnetization of 200 emu/g and a coercivity of 0.6 Oe. The magnetization is close to the reported value of Fe-3.3 wt % Si, 206 emu/g¹⁶.

3.2 Friction Consolidated Specimen Characterization

As shown in Table I, powders are processed with increasing tool speed and pressure, both of which contribute to the temperature change on the tool. Fig. 5 shows the tool force and temperature profiles of these samples during processing.

Fig. 5 Time-dependent profiles of FC load (top), tool position (middle) and tool temperature (bottom), during the friction consolidation of Fe-Si powder for samples A, B, and C.

Fig. 5 shows that sample A experiences 8kN (34MPa) with a tool displacement of 3.3mm. Sample B is loaded at approximately 18kN (14MPa) with a tool displacement of 3.5mm. A load of 10-20kN is applied to sample C for the first 30 s. Then, a final load of 60kN (122MPa) is applied for approximately 10s at a displacement of 4.2mm. The tool temperature profiles are shown in Fig. 5. Here, we see that the time at which the peak tool temperature is reached generally follows a trend

with process rotational velocity and applied force. That is, sample C reaches peak tool temperature first, followed by sample B, and finally sample A.

Cross-sectional images of the produced pucks are shown in Fig. 6. The red rectangular highlighted regions indicate the areas where magnetic properties were measured and EBSD analysis is performed.

Fig.6 Cross sections of consolidated Fe-Si specimens where material flow lines are visible. Increased densification is shown as a result of higher applied pressure during consolidation.

Fig.7 shows light microscopy images taken at various locations across specimen B. Consolidation of Fe-Si results in mostly equiaxed grains in the processed zone (center middle of the puck) with visible material flow lines typical of friction consolidation, radiating to the puck edges. The density in the processed zone achieved within the targeted thickness is 7.29 g/cm^3 , approximately 95 % of the bulk value.¹⁶ All samples, despite different processing conditions, exhibit material flow lines along the specimen cross-section.

Fig. 7 Light microscopy images showing grain structure of specimen B at various locations.

The shearing action during the friction consolidation process likely results in a high density of dislocations leading to recrystallization at elevated processing temperatures and ultimately

significant grain size reduction¹⁷. The crystal orientation as well as the grain size distribution of each of the samples is evaluated in the homogenous regions identified in Figs 6& 7. The results of this analysis are shown in Figs. 8& 9. Differences in the crystallographic orientation are shown using grain orientation maps with inverse pole figure coloring. A higher degree of texturing is visible as increased load and rotational speed are applied during processing. Maximum multiple of uniform distribution (MUD) values for samples A, B and C are measured at 2.36, 3.79 and 3.41 respectively, indicating that specimen A shows the weakest texturing.

Fig. 8 Grain orientation maps showing the out-of-plane (Z texture) of the consolidated samples using inverse pole figure coloring. A higher fraction of {111} is visible in samples B and C.

Fig. 9 shows average grain size distributions, calculated from the EBSD results shown in Fig. 8. This illustrates the ability of friction consolidation to dramatically reduce grain size in Fe-Si when compared with the as-received powder (Fig. 4). Despite the high temperatures reached during processing¹⁸, Sample A showed significantly weaker texture than the higher load samples indicating the applied force during consolidation plays a key role in texture development with all other variables constant. It can also be seen when comparing specimens B and C that increasing both the load force and rotational speed together did not result in any further change in either the average grain size or texture development. The maximum grain size, however, did increase by several micrometers. It is hypothesized that the larger load force and rotational speed promote

some grain growth as a result of increased heat generation, suggesting that ideal processing parameters for this alloy system fall between the processing conditions used for samples B and C.

Fig. 9 Grain size analysis of the processed zone of sample A, B and C.

3.3 Magnetic Properties of the Consolidated and Consolidated + Annealed Fe-Si

The FC processed Fe-Si materials are characterized to reveal the effects of the various process parameters on consolidated specimen magnetic properties. Magnetic measurements of specimen A were not performed as a result of the poor consolidation indicated by the optical and EBSD assessments. Fig 10 shows a measured typical hysteresis loop of the consolidated Fe-Si material.

Fig. 10 Hysteresis loop of the consolidated material measured by VSM.

The magnetization (Table II) of the as-consolidated samples B and C remains unchanged (within experimental error) compared to that of the starting powders indicating that the phase constituents likely remain relatively constant. The apparent minor increase in magnetization of sample B along the transverse direction in the midway radial location when compared to the center piece is much higher than the same comparison made for sample C. This result corroborates the EBSD measurements and may be due to the weakened texture and slightly elevated maximum grain size of sample C.

Table II. Magnetization and coercivity of the consolidated specimens at different locations (magnetization within ± 10 emu/g and coercivity is within ± 0.5 Oe)

A pronounced increase in coercivity is seen when comparing the consolidated samples with the starting powders (0.6 Oe). A reduction in grain size of ~40% was achieved through friction consolidation processing, the increased coercivity agrees with the well-known grain size versus coercivity trend demonstrated by G. Herzer¹⁹.

From Table II, we can also see that coercivity values from sample Coverall are slightly lower than those from sample B; this is possibly a synergic effect of the grain morphology, grain size distribution, texture, grain boundary conditions as well as the local stress field. Coercivity is highly sensitive to variations in the local stress field^{20, 21}. To address this possibility, annealing at 700 °C and 900 °C for 2h, respectively, was performed for sample B and C on the midway radial specimens (see Fig.2). Grain size of the annealed samples was obtained from EBSD and was compared to the as-processed sample (see Fig. 11 inset). No significant grain growth is observed. However, a significant reduction in coercivity (a maximum of 30%) is observed (see Fig.11). It should be noted that despite the minor differences in the magnetic behavior seen between the transverse direction and the radial direction, both directions should be equivalent, as a result of puck symmetry.

Fig.11 Coercivity as a function of annealing temperature for sample B and sample C. Inset shows that grain size for both samples B and C after annealing at 900 °C is comparable to those of the as-processed, respectively.

With little grain growth observed following annealing, the measured decrease in coercivity is believed to be primarily due to stress relief occurring as a result of annealing. To verify this hypothesis, EBSD local misorientation maps were generated to visualize and compare low angle lattice distortion in the specimen before and after annealing, as shown in Figure 12.

Fig.12 Local misorientation maps (top) showing low angle lattice distortions before annealing (left) and after annealing (right). The respective local misorientation distributions (bottom) illustrate a reduced degree of low angle distortion in the annealed sample.

Results of the EBSD local misorientation mapping show a subtle reduction of the low angle lattice strains following the annealing heat treatment. This effect is most easily identified in the misorientation histogram and supports the hypothesis that decreased coercivity following annealing was a result of reduced internal stress fields in the material. No significant changes in texture were observed in the annealed samples.

4. Conclusion

Gas-atomized Fe-Si powders consolidated using FC were evaluated to study the effect of processing conditions on the grain size, texture and magnetic properties. The viability of this consolidation method to significantly homogenize and densify powdered soft magnetic Fe-Si precursors is demonstrated. The FC process successfully refines the grain size and has demonstrated the ability to impart texture development. Annealing of the processed material was shown to decrease the coercivity due to residual stress relief. These results point to the possibility of practical scale-up of nanostructured materials, however further research is needed to unravel and understand the complex processing conditions needed to control and homogenize the microstructure and thereby the magnetic behavior.

5. Acknowledgements

This research was supported by the MS³ (Materials Synthesis and Simulation Across Scales) Initiative at Pacific Northwest National Laboratory, a multi-program national laboratory operated by Battelle for the U.S. Department of Energy.

References

- ¹O. Gutfleisch, M. A. Willard, E. Brück, C. H. Chen, S. Sankar and J. P. Liu, *Adv. Mater.* **23** (7), 821-842 (2011).
- ²G. Herzer, *Acta Materialia* **61** (3), 718-734 (2013).
- ³M. Huang, W. Wallace, M. McHenry, Q. Chen and B. Ma, *J. Appl. Phys.* **83** (11), 6718-6720 (1998).

- ⁴K. Takenaka, A. D. Setyawan, Y. Zhang, P. Sharma, N. Nishiyama and A. Makino, *Materials Transactions* **56** (3), 372-376 (2015).
- ⁵R. Hasegawa, *Journal of non-crystalline solids* **329** (1), 1-7 (2003).
- ⁶J. Sort, D. Ile, A. Zhilyaev, A. Concustell, T. Czeppe, M. Stoica, S. Surinach, J. Eckert and M. Baró, *Scripta materialia* **50** (9), 1221-1225 (2004).
- ⁷W. Tang and A. P. Reynolds, *Journal of Materials Processing Technology* **210** (15), 2231-2237 (2010).
- ⁸G. Buffa, D. Campanella, L. Fratini and F. Micari, *International Journal of Material Forming*, 1-6 (2015).
- ⁹D. Catalini, D. Kaoumi, A. P. Reynolds and G. J. Grant, *Journal of Nuclear Materials* **442** (1), S112-S118 (2013).
- ¹⁰S. Whalen, S. Jana, D. Catalini, N. Overman and J. Sharp, *Journal of Electronic Materials*, 1-10 (2016).
- ¹¹A. J. Moses, *Scripta materialia* **67** (6), 560-565 (2012).
- ¹²H.-Z. Li, H.-T. Liu, Z.-Y. Liu, H.-H. Lu, H.-Y. Song and G.-D. Wang, *Materials Characterization* **88**, 1-6 (2014).
- ¹³H.-T. Liu, Z.-Y. Liu, Y. Sun, F. Gao and G.-D. Wang, *Materials Letters* **91**, 150-153 (2013).
- ¹⁴P.M. Giles, M.H. Longenbach, and A.R. Marder, *Journal of Applied Physics* **42** (11), 4290-4295 (1970).
- ¹⁵R. P. Koseski, P. Suri, N. B. Earhardt, R. M. German and Y.-S. Kwon, *Materials Science and Engineering: A* **390** (1), 171-177 (2005).
- ¹⁶R. C. O'handley, *Modern magnetic materials: principles and applications*. (Wiley New York, 2000).

- ¹⁷R. S. Mishra, P. S. De and N. Kumar, *Fundamentals of the Friction Stir Process*. (Springer, 2014).
- ¹⁸C. Chang, C. Lee and J. Huang, *Scripta materialia* **51** (6), 509-514 (2004).
- ¹⁹G. Herzer, *Magnetics*, *IEEE Transactions on* **26** (5), 1397-1402 (1990).
- ²⁰P. Zou, W. Yu and J. A. Bain, *Magnetics*, *IEEE Transactions on* **38** (5), 3501-3520 (2002).
- ²¹I. Tabakovic, V. Inturi and S. Riemer, *Journal of The Electrochemical Society* **149** (1), C18-C22 (2002).

Fig1

Fig2

Half of the consolidated puck

Fig3

Fig4

Fig5

Fig6

Fig7

Fig8

Fig10

Fig11

Fig12