The effect of Cu-based core-sheath configurations on the processing of Nd-Fe-B-based permanent magnets via equal-channel angular pressing

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Abstract. Equal channel angular pressing (ECAP) has been used as an alternative manufacturing route for preparation of Nd\textsubscript{2}Fe\textsubscript{14}B-based anisotropic magnets, facilitating processing temperatures much lower than conventional die upsetting. While this method can produce a suitable texture and microstructure in permanent magnetic materials, it still remains novel; involving extremely high pressures which present a high risk of both process failure and die and tooling damage. Powder metallurgical processes frequently incorporate an external layer of secondary material (commonly an outer foil layer or can) for separation between the primary material and die as well as the control of surface effects such as friction through appropriate choice of secondary material. This work implements such modifications to this manufacturing route by incorporation of an outer layer of Cu foil, the addition of which negatively affected both the powder compaction and strength of texture produced via ECAP. Also investigated was the incorporation of solid Cu bar as part of the sample cross section. This modification facilitated processing without any compromise on observed magnetic properties, whilst also reducing damage to both the die and tooling. This type of methodology may aid in improving the reliability of producing bulk anisotropic permanent magnets via ECAP.

1. Introduction

Equal channel angular pressing (ECAP) is a thermo-mechanical processing method capable of developing suitable texture in Nd\textsubscript{2}Fe\textsubscript{14}B-based permanent magnets at temperatures low enough to avoid grain coarsening [1]. However, extremely high pressures during pressing [2] of permanent magnet materials, which are highly brittle [3][4], frequently lead to failure of the process and tooling damage. Such a risk of failure presents an obstacle to the upscaling and reliability of this manufacturing process.

It is common practice for powder metallurgical processes to incorporate a secondary material to isolate the primary material from the die and control properties arising from material-die contact such as friction [5]. With a view towards reducing pressure during ECAP and increasing the chance of a successful procedure, a suitably low friction material, Cu, was chosen for incorporation into the ECAP process. Two arrangements of Cu incorporation were investigated: a thin outer layer of foil acting as a sheath insulating the powder from the die wall, and a solid Cu bar at the leading end of the sample.
2. Methods

Commercial melt-spun Nd$_{13.5}$Fe$_{73.8}$Co$_{6.7}$B$_{5.6}$Ga$_{0.4}$ powders (Magnequench MQU-F1, referred to herein as ‘F1’) were placed into an ECAP die with channel angle of 90° and channel diameter of 10mm. The sample was then processed via ECAP at a temperature ($T$) equal to 500°C with a backing pressure ($\sigma$) approximately equal to 0.5 GPa and a pressing speed of approximately 3 mm/s. This process was then modified by first placing a sheath of Cu foil with a thickness of approximately 76 µm into the die channel and then placing F1 powders into this foil. The foil was folded at each end to prevent powder escaping, and processed via ECAP under the same conditions.

Figure 1 illustrates the experimental setup for the ECAP process modified by incorporating a Cu bar. A solid cylindrical Cu bar with a diameter slightly under 10 mm was first placed into an ECAP die and deformed at room temperature to fill the channel. F1 powder was then inserted above this bar. The top of the bar remains above the channel corner, meaning all powder is confined entirely in the pressing channel. The sample was then processed via ECAP under the same conditions.

Figure 2 shows schematically the sample after ECAP and three principle manufacturing directions, the pressing direction (PD), exit direction (ED) and transverse direction (TD). Following ECAP, cubes with a length of 3 mm were cut from the center of the sample for magnetic measurements via vibrating sample magnetometer along the PD, TD and ED directions. Prior to each directional measurement, the sample was magnetised in a pulsed field of $\mu_0H = 7$ T using a pulse magnetiser. Disc-shaped samples were cut in the plane normal to ED for texture analysis on a GBC X-ray diffractometer. Finally, samples were cut along the plane normal to TD for optical microscopy.

In the addition to the experimental work, modelling of ECAP process utilising a finite element package QForm VX [6] was carried out. This package is designed to simulate metal forming processes and uses an automated re-meshing procedure which is advantageous in the case of severe plastic deformation processes. The set-up of the simulation included the drawing of the geometry of the ECAP die, forward and backward punches and the workpieces. The values for the pressing speed, temperature, back-punch pressure were adopted from the experiments. In order to simulate the material behaviour of the F1 alloy and Cu, experimental stress-strain curves of both materials were imported into the database of QForm VX as flow stress curves, where the stress is represented as a function of strain, strain rate and temperature. This information was compiled from other published data describing the mechanical behaviour of these materials [7]. The interaction between two workpieces was modelled using high value of friction factor $\mu = 1$. 
3. Results

3.1. Pressure output during ECAP

Figure 3 shows the relationship between the pressure applied to the forward punch (forward pressure) and sample displacement during ECAP for trials without Cu. Trials marked 1, 2, and 3 are trials where the ECAP process failed. In these trials, the forward pressure increase rapidly with little subsequent change in sample displacement. The initial static friction is not overcome, and the sample does not begin to flow. At a critical forward pressure of approximately 4 GPa, punch failure occurs. Trial 4 shows the forward pressure behaviour of a successful passing without any Cu. This successful Cu-free trial was selected for further testing, and forms the basis for measurements in Figs. 6, and 7.

Figure 4 shows forward pressure vs. displacement after addition involving Cu. Trials 1 and 2 included Cu foil, while trials 3 and 4 involved Cu bar. With the addition of Cu foil, the pressure vs. displacement curves behave similarly to trial 4 in Fig. 3 (a successful pass without Cu) where the build up of forward pressure reaches approximately 3 GPa before flow can begin. With the addition of a Cu bar, the build up of forward pressure before the sample overcomes the initial static friction and begins to pass is significantly reduced, allowing deformation to proceed at a forward pressure of around 2 to 2.5 GPa. Trials 1 and 4 from Fig. 4 were chosen for further testing.

3.2. Simulation of deformation behaviour

Figure 5 displays a comparison of the shape of the extruded sample after one passing of ECAP with the predicted simulation. It can be seen that a good agreement between the simulation and the experiment is obtained. The result obtained by finite element simulation indicates that the equivalent strain in the central region of the sample is close to the analytically predicted value of 1.15 [8].

3.3. Magnetic measurements

Figure 6 shows demagnetisation curves taken in manufacturing directions PD, TD and ED (which are defined in Fig. 2). The highest remanent magnetisation value ($M_r$, i.e. $M(H = 0)$) in the pressing direction suggests a high degree of alignment of the magnetically easy (001) axes parallel to PD; a sign of developed texture [9]. This is the case for F1 processed both without
3.4. Texture analysis

Figures 7 and 8 are (001) pole figures taken from the plane normal to the exit direction (ED) after ECAP for F1 processed without Cu and with Cu bar respectively. The center of each pole figure is parallel to ED, with each ring marking a 5° increase in altitude away from ED. The peak m.r.d (multiple of random distribution) intensity is observed at an altitude of between 50-70° away from ED for both trials. The azimuthal range and maximum m.r.d value of approximately 4.5 remain consistent between F1 without Cu and F1 with Cu bar. This similarity is also consistent with magnetic measurements in Fig. 6, further supporting the notion that an equivalent texture has been produced in these two trials.
3.5. Optical microscopy

Figure 9 shows the transverse face of samples after ECAP. Without Cu (Fig. 9(a)), the sample becomes quite dense and particles tend to both elongate and realign at an inclination angle of $\theta \approx 26^\circ$ (consistent with the angle produced via simple shear in ECAP with a die angle of 90° [10]) away from the exit direction, ED. When processed with Cu foil, the microstructure retains a significant amount of porosity (as visible in Fig. 9(b)) and particle alignment and elongation is less prominent. In 9(c), when a Cu bar is incorporated, a dense microstructure with aligned and elongated particles similar to Fig. 9(a) is observed.

4. Discussion and Conclusions

The incorporation of Cu foil successfully reduced the risk of process failure during ECAP and the initial build up of forward pressure before significant deformation occurs is reduced. However, the degree of texture produced via ECAP is significantly reduced. The microstructure retains an appreciable degree of residual porosity. Effects stemming from the interaction between the die wall and primary material, such as friction, may play a key role in successful development of a well-textured, dense microstructure for Nd$_2$Fe$_{14}$B-based materials via ECAP.

The incorporation of a Cu bar also successfully reduced the risk of process failure during ECAP. The build up of forward pressure before deformation occurs is significantly reduced. Unlike modifications based on Cu foil, the interaction between the sample and die wall is not affected by the Cu bar, and an equivalent degree of texturing and densification as in the case without Cu is achievable, which is supported by results showing an equivalent texture (Figs. 7 and 8), magnetic performance (Fig. 6) and microstructure (Fig. 9). This modification presents
a promising technique for improving the reliability of the ECAP process for brittle materials requiring high forces without compromise on material and functional properties.

References
[1] Onal E, Lapovok R, Kishimoto H, Kato A, Davies C H J and Suzuki K 2015 J. Appl. Phys. 117
[2] Segal V M 2004 Mat. Sci. Eng. A 386 269
[3] Horton A and Wright J L 1996 IEEE Trans. Magn. 32 4314–4316
[4] Grünberger W, Hinz D, Kirchner A, Müller K H and Schultz L 1997 IEEE Trans. Magn. 33 3889–3891 ISSN 00189464
[5] Roberts P R and Ferguson B L 1991 International Materials Reviews 36 62–79 ISSN 09506608
[6] Qform simulation software URL http://www.qform3d.com/
[7] Yasuda H Y, Kumano M, Nagase T, Kato R and Shimizu H 2011 Scr. Mater. 65 743–746 ISSN 13596462
[8] Segal V 1995 Mat. Sci. Eng. A 197 157–164 ISSN 09215093
[9] Mishra R K 1987 J. Appl. Phys. 62 967–971 ISSN 00218979
[10] Beyerlein I J and Tóth L S 2009 Progress in Materials Science 54 427–510 ISSN 00796425