

Equal channel angular extrusion for bulk processing of Fe-Co-2V soft magnetic alloy

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Abstract

The soft magnetic alloy Hiperco[®] 50A was subjected to equal channel angular extrusion (ECAE) at elevated temperatures using two different processing routes. The ECAE process dramatically refined the grain size compared to conventional Hiperco[®] bar. Fine grain size in Hiperco[®] has previously only been available in sheet form. Yield strengths of 650-700MPa and ultimate tensile strengths of 900-1400MPa represent a 2 to 3-fold increase in strength compared to conventional bar. The Hall-Petch relationship was summarized alongside previous studies of bar and sheet. ECAE also produced high ductility, due partly to a disordered bcc crystal structure upon quenching after ECAE.

[®]Hiperco is a tradename of Carpenter Technology Corp., Reading, PA.

Hiperco[®] 50A (simply Hiperco hereafter) is a soft magnetic alloy with nominal composition of Fe₄₉Co₄₉V₂ in wt.%. Hiperco has the highest magnetic saturation of any commercial alloy, with high permeability and low coercivity as well. It is used in applications such as solenoids, electric motors, and generators that take advantage of its excellent magnetic behavior. However, Hiperco in bar form has low mechanical strength and is well-known to exhibit low ductility at room temperature [1]. The poor mechanical properties limit its application in high performance components that may encounter mechanical stresses due to shock, vibration, etc. Until now, Hiperco in a high-strength condition has only been available as cold worked sheet, not in bulk products with larger cross-section [2].

The focus of the present work is the role of severe plastic deformation (SPD) of bulk samples via Equal Channel Angular Extrusion (ECAE), also known as Equal Channel Angular Pressing (ECAP), on the resulting mechanical properties, in comparison to conventional bar and cold-worked sheet materials. In ECAE, a bar of material is extruded through a die with an abrupt corner, usually with a 90 degree angle. Large shear deformation, i.e. >100% effective strain, is produced within the bar as it progresses through the die corner but the overall dimensions of the bar do not change. The material undergoes SPD and, depending on the homologous temperature of the process, dynamic recrystallization takes place resulting in a very fine grain size. Grain refinement and work hardening are responsible for the dramatic increases in yield and tensile strength observed with SPD processes, such as ECAE. Increased strength of Hiperco would generally be beneficial for high-performance magnetic components. In most materials, a dramatic increase in strength would come with a concomitant decrease in ductility. However, it has been shown that high levels of cold-work, as obtained in sheet products, can actually improve the

ductility of Hiperco [3-7]. Thus, the possible beneficial effects of ECAE on both strength and ductility provided the motivation for exploring ECAE in the present work.

Zhao and Baker performed *conventional extrusion* on binary Fe-Co alloys and observed moderate reductions in grain size for 7.5:1 area reduction extrusion at 1000°C followed by a 4:1 area reduction extrusion pass at 750°C [8]. Wießner et al. performed ECAE on a ductile FeCo17 alloy at room temperature [9]. They reported an increase in magnetic coercivity as a result of ECAE, but no mechanical properties were reported. No research was found on ECAE of Hiperco 50A or similar alloys with composition near 50/50 FeCo.

In cold-worked sheet, high strength is obtained through grain refinement, i.e. the Hall-Petch effect [1, 10-21]. The improved ductility of sheet could be due to an increase in the disordered bcc phase after rolling as opposed to the more brittle ordered B2 structure. But even with post-rolling heat treatment to induce ordering, relatively high ductility in sheet can be retained. The reasons for this are not well understood and could involve the following: 1) The formation of local concentration-disordered (LCD) zones as proposed by Kawahara [4-7], or 2) Second phase particles, very fine grain size, or partial recrystallization with subgrains acting to prevent dislocation pileups and crack propagation, i.e. “slip partitioning” or “slip dispersion” [19, 22-26]. Note these explanations are separate from the theories put forth to account for the ductility from vanadium additions in FeCo-V compared to (very brittle) binary FeCo [26].

In addition to mechanical properties, the magnetic performance of Hiperco must also be considered when assessing the effects of thermomechanical processing. Well-annealed, soft Hiperco with large grain size and fully-ordered B2 (L₂₀) crystal structure displays the best soft magnetic behavior because there is little impedance to magnetic domain wall movement. Unfortunately, this material in bar form is both mechanically weak and brittle. In materials with fine grain size and/or high levels of cold work (resulting in internal stress and/or high dislocation density), domain wall movement is restricted and the magnetic saturation and permeability generally decrease [27-29]. Although magnetic performance will be mentioned here, it is not the primary focus of this initial study. The goals of the present work are to investigate ECAE of Hiperco following the processes of Ref. 30 and to evaluate the mechanical response of the processed materials in light of the resulting microstructure after ECAE.

As mentioned earlier, Hiperco is relatively brittle at room temperature and would not normally be considered a good candidate for SPD processing. However, since ECAE can be performed at elevated temperatures, it was considered that the high-temperature ductility of Hiperco might be sufficient to survive the ECAE process without catastrophic failure. Therefore, prior to beginning any ECAE trials, the hot ductility of conventional Hiperco bar was investigated using a Gleeble thermo-mechanical simulator. The Gleeble 3500A employs resistive heating to generate high temperatures within a sample while simultaneously applying tensile or compressive forces. Hot ductility tensile tests were performed between 650 and 900°C, as shown in Fig. 1. Hiperco displays substantial necking prior to failure with reduction in area >75% and increasing with test temperatures above 650°C. This behavior contrasts the brittle nature of Hiperco near room temperature where little reduction in area and no necking are observed prior to failure. Based on the hot ductility results, Hiperco was deemed amenable for ECAE trials.

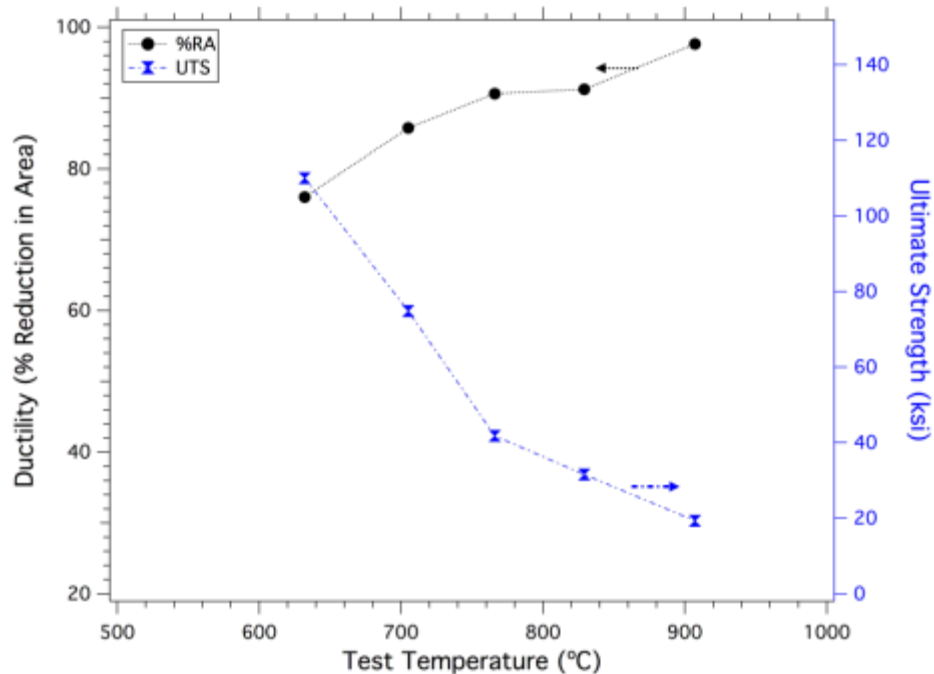


Fig. 1. Elevated temperature mechanical properties (hot ductility) of Hipercó obtained with a Gleeble 3500 thermo-mechanical simulator.

ECAE billets with a 25.4 mm by 25.4 mm cross section and 150 mm long were prepared from Hipercó with composition: 49.1 wt.% Co, 1.93 wt.% V, 0.051 wt.% Nb and balance Fe. ECAE routes C and E were utilized in this work, Table 1. The extrusion rate for all billets was 12.7 mm/sec. Prior to heating, the bars were coated with a high temperature lubricant. Billets were held in a furnace at the specified temperature for 30 minutes, while the die was held at a constant 350°C, thus the extrusions were carried out non-isothermally. All billets were water quenched immediately after the extrusion, except for one sample which was air cooled after the final extrusion pass. Billets were slightly milled between each pass to create flat and level surfaces with accurate dimensions. Samples for microscopy were cut in Extrusion Plane (EP) and Flow Plane (FP) orientations, Fig. 2, at least 3 mm away from the surface, and mechanically polished to 0.06 micron colloidal silica. Microstructural characterization was performed using a FEI Quanta 600 Scanning Electron Microscope (SEM) with 25kv accelerating voltage. For uniaxial tension tests, flat dog-bone samples, 25 mm long and 8 mm gage length with ~1 mm thickness were cut along the Extrusion Direction (ED, EP normal direction) and Flow Direction (FD, FP normal direction) using wire EDM, and tested using an MTS machine with 8mm clip-on extensometer. Two or three samples were tested along the extrusion and flow directions to confirm the repeatability. Conventional Hipercó bar samples, with 6.35 mm diameter, were also tensile tested for comparison.

Table 1. ECAE Route descriptions and those used for Hiperco extrusions

Route name	Min # of passes	Billet rotations about the extrusion axis				Effect on microstructure
		1→	2→	3→	4→ N	
A	1	0°	0°	0°	etc.	elongation (lamellar)
B (B _A)	2	+90°	-90°	+90°	etc.	elongation (filamentary)
C	2	180°	180°	180°	etc.	back/forth shearing
C' (B _C)	4	+90°	+90°	+90°	etc.	back/forth cross-shearing
E	4	180°	90°	180°	etc.	back/forth cross-shearing
Sample	Extrusion experiments in this study					
4E	4 ECAE passes following Route E (4E) at 850°C and water quenching					
6E	4E at 850°C plus 2 more passes following Route E at 750°C and water quenching					
4C	2 ECAE passes following Route C (2C) at 850°C plus 2 more passes following Route C at 750°C and water quenching					
4C-AQ	2C at 850°C plus 2C at 750°C and air quenching (AQ)					

Based on the hot ductility results, an initial extrusion temperature of 850°C was chosen. Successful ECAE results were achieved with four passes of Route E at 850°C, sample 4E. The intent was to keep the temperature as low as possible to attain the greatest microstructural refinement, i.e. by limiting grain growth during extrusion. Initial passes at 750°C or 700°C proved to be unsuccessful presumably due to the formation of the brittle ordered B2 phase; the order/disorder temperature is approximately 720°C in the Fe-Co system [31]. However, a few passes at 850°C allowed for subsequent finishing passes at 750°C as shown in Table 1, suggesting improved ductility from the SPD and grain refinement from the initial passes. Two ECAE routes were investigated in this work, namely C and E, and the microstructure and properties of the extrusions will be discussed below. The reason for the selection of these routes was to obtain near equiaxed grains and a weak crystallographic texture such that texture-induced magnetic anisotropy would not be developed, since these routes are known to lead to the weakest texture among most known ECAE routes. Finally, sample 4C-AQ was a repeat of 4C except after the last pass the sample was air cooled instead of water quenched, in order to approximate the suggested cooling rate for obtaining good magnetic properties [2].

Backscattered scanning electron (BSE) micrographs of sample 6E are shown in Fig. 2 to illustrate the fine grain size obtained with ECAE. The microstructure is generally representative of all extrusions in this study. The top micrographs show the extrusion plane (EP) and the bottom images display the flow plane (FP) structure. Grain size of 1.5 to 3 microns was estimated based on the BSE channeling contrast. No significant differences were observed between the microstructure near the center of the extrusion and that near the outside edge. It is clear that significant grain refinement occurs during ECAE, even at 850°C, which is about $0.64 \times T_m$ homologous temperature for Hiperco. The two extra route C passes and the lower finishing temperature do not appear to significantly alter the microstructure compared to sample 4E (not shown), which was processed at 850°C only. The fine grain size indicates that dynamic recrystallization occurs during ECAE at these temperatures without extensive grain growth, which is likely limited due to the short duration of the extrusion process. In addition, note that 500 ppm Nb was added to this lot of Hiperco. The formation of NbC precipitates could pin grain boundaries to maintain a fine grain size during ECAE, similar to the effect of Nb additions (~3000 ppm) in

Hiperco 50HS sheet [2,20]. Furthermore, a subgrain structure might exist (with low angle grain boundaries), which could be an important factor for both strength and ductility and may not be apparent in SEM/BSE images. Grain refinement and partial recovery can lead to increases in both strength and ductility as shown in a few recent ECAE studies, such as for a Nb-1%Zr alloy [32]. Detailed sub-micron characterization will be pursued in subsequent work. For comparison to Fig. 2, conventional Hiperco bar exhibits grain sizes in the range of ~20-75 microns, depending on the heat treat temperature [15-17]. Conventional bar is typically used in the heat treated condition. However, as-received bar possesses a grain structure on a similar scale resulting in similar yield strength [16]. Post-ECAE annealing, which may be performed to induce ordering and increase magnetic performance, will likely result in coarsening of the ECAE microstructure. Nevertheless, an important aspect here is that there is significant heat treat processing space to be explored between the as-ECAE structure and conventional bar microstructure. That is, intermediate heat treatment times and temperatures may be investigated to potentially improve magnetic performance without excessive grain growth, thereby maintaining a high strength level.

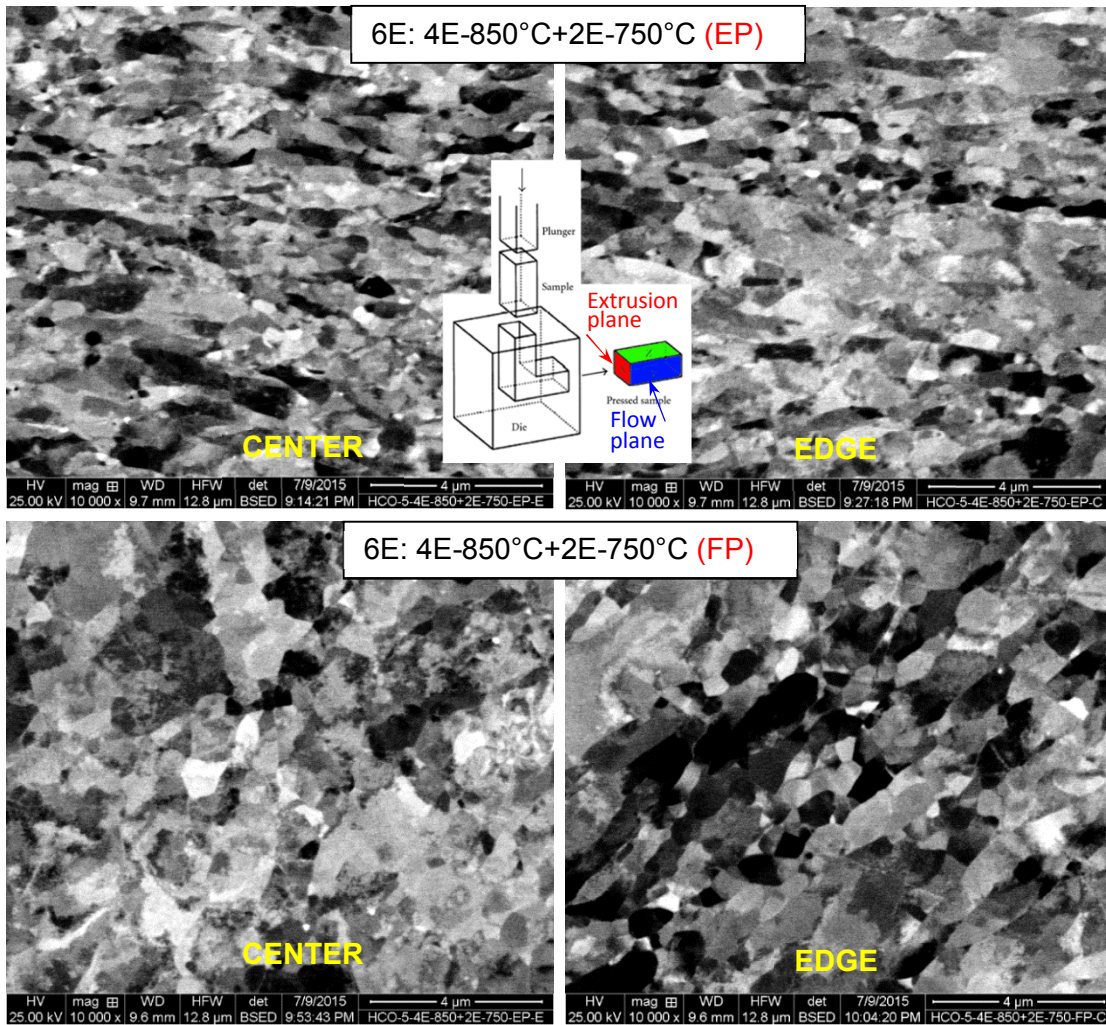


Fig. 2. Backscattered scanning electron micrographs of ECAE 6E samples showing the extrusion plane (top) and the flow plane (bottom) microstructures.

After ECAE processing, Hiperco bar exhibits very high strength as shown in Fig. 3. The yield and tensile strengths, in the range of 676-862MPa (98-125ksi) and 869-1103MPa (126-160ksi), respectively, are comparable to strengths found in highly cold-worked sheet. This result correlates well with the small grain size discussed previously, i.e. Hall-Petch considerations. High strengths have not been previously achievable in bulk sections of Hiperco. The as-ECAE condition represents more than a two-fold increase in strength compared to conventional Hiperco bar (200-400MPa, 29-58 ksi) yield strength, Fig. 3. The conventional bar data are from three different heat treatments from a previous study [16].

The failure strains between 15 and 20% for the flow direction in the 6E sample and for both directions in the 4C sample are among the highest reported for Hiperco in any condition [4-7,19,26]. Due to water quenching after ECAE, it is likely that the as-ECAE material is in the disordered (bcc) or only slightly ordered condition. In general, disordered material displays higher ductility than the alloy in the ordered state, but some observed differences, at this time, are attributed to the extrusion finishing temperatures. Only the ECAE samples with finishing temperatures of 750°C are displayed in Fig. 3. Extrusion 4E, processed only at 850°C, also had high strength but its ductility was lower, ~ 4-6%, similar to conventional bar. The reasons for relatively low ductility in extrusion 4E are not fully understood but, by comparison to extrusions 6E and 4C, it appears that the finishing temperature of 850°C is high enough for recovery and recrystallization (but with minimal grain growth). Higher failure strains for extrusions 6E and 4C, especially in the flow direction, suggest that a cold worked structure, not simply fine grain size, is required to sustain deformation without failure. It is possible that extrusions 6E and 4C contain a higher dislocation density and/or a subgrain structure contributing to ductility, but further microstructural characterization must be performed to determine the exact mechanism responsible. Overall, the higher ductility obtained with high amounts of process deformation is consistent with the results for cold-worked sheet obtained by Kawahara and others [4-7,18,25].

Extrusion 6E shows considerable anisotropy in ductility between the flow direction and extrusion direction, although the ~7% strain in the extrusion direction is still quite high for Hiperco. The six passes of extrusion route E, therefore, produce similar anisotropy to what might be expected in sheet material (although with lower ductility in the “long” dimension of the product). Conversely, the tensile response of extrusion 4C is more isotropic and reproducible. Importantly, extrusion 4C was produced with only four passes, thereby reducing the processing time as well. The reasons for differences in the degree of anisotropy among the samples is not known and requires further microstructural characterization.

Typical ECAE processing involves water quenching in between extrusion passes and after the final pass. Extrusion 4C-AQ utilized the same extrusion parameters as extrusion 4C, except that air-cooling was employed after the final pass. This experiment was an attempt to produce a partially or fully ordered B2 crystal structure by slowly cooling through the ordering range, which may impart better magnetic performance. It would be highly desirable to produce both high strength (and good ductility) and good magnetic performance without the need for a separate heat treatment step. As shown in Fig. 3, high yield and ultimate strengths of sample 4C-AQ are retained with air-cooling. Some ductility is lost, however, and more anisotropy is indicated for extrusion 4C-AQ. Even so, the failure strains of ~7-8% and ~14% in the ED and FD orientations, respectively, compare favorably to conventional bar material.

A few other features of interest are found in Fig. 3. First, unlike conventional bar, material in the as-ECAE condition does not show an upper/lower yield point (discontinuous yielding portion of the stress-strain curve), although a slight plateau is evident just after yielding. Upper/lower yield point with 1-2% discontinuous strain and the propagation of Luders bands are typical of large-grain annealed Hipercor bar [4,16,24,28]. Second, the material fails without appreciable necking (with the possible exception of extrusion 4C) which is also characteristic of Hipercor alloys. And third, visual examination of the fracture surface (not shown here) confirms a brittle, transgranular, quasi-cleavage fracture mode even after significant tensile straining is incurred. It should be noted that conventional Hipercor occasionally failed in the threaded grip region of the round tensile samples. This behavior was observed in previous studies of Hipercor bar and illustrates the notch sensitivity of this material.

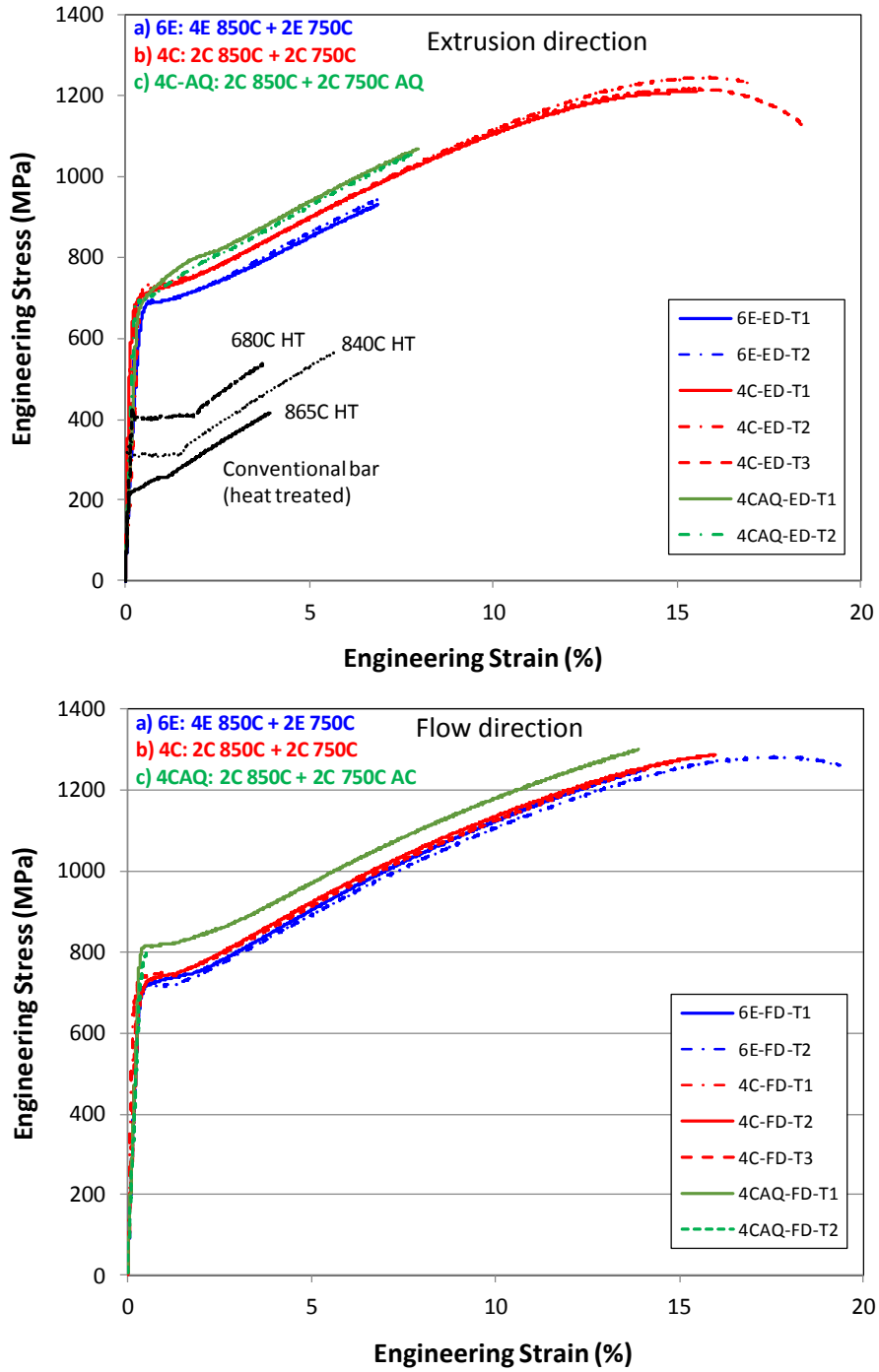


Fig. 3. Stress-strain curves from ECAE processed Hipercro in the a) extrusion direction and b) flow direction. Results also shown for conventional bar heat treated at 680, 840, and 865°C.

To summarize the tensile behavior in this work, Hall-Petch plots of yield strength vs. (grain size)^{-1/2} were constructed in Fig. 4 according to:

$$\sigma_y = \sigma_0 + kd^{-1/2},$$

where σ_y is yield strength, σ_0 is intrinsic friction stress (Peierls stress), k is a constant (Hall-Petch coefficient) and d is grain size. The results for ECAE bar and a few studies on conventional bar are shown in Fig. 4a. The Hiperc conventional bar material (Sandia bar) was heat treated in the range of 680-870°C. The ECAE data points were produced from the approximate grain size measurements obtained with backscattered SEM channeling contrast, while grain size of conventional bar was acquired from optical micrographs. Other results from the literature have similar Hall-Petch slopes (k -values) with some degree of scatter [15,17]. Note the data from Zhao and Baker includes only their results for material in the ordered condition (slowly cooled from high temperature). Figure 4b also comprises Hiperc sheet material from Nabi et al., Ren and Yu, George, Shang, and Fingers [10-14, 18-21]. Here it is confirmed that the ECAE material lies in a similar strength/grain size regime as cold-worked sheet. The dashed trendline appears to be a reasonable fit to the sheet results as well, with the exception of the higher strength values from Shang. Shang performed Knoop hardness measurements, from which yield strengths were estimated in a review by Sourmail [1,10]. The Hall-Petch slope is about the same as that shown from the other studies and the shift to higher strengths may be due to uncertainty in the hardness to strength conversion. With regard to order/disorder, Sourmail discussed the fact that details were not given in the Shang paper on quenching vs. slow-cooling; the degree of order could have significant influence on the Hall-Petch behavior as well. Based on the dashed trendline in Fig. 4, the estimated k -value (Hall-Petch slope) and σ_0 (Peierls stress) are 871MPa- $\mu\text{m}^{-1/2}$ and 104MPa, respectively. These parameters are presented along with values from other studies in Table 2. In general, for both Hiperc bar and sheet, the relationship between yield strength and grain size can be described by a Hall-Petch slope in the range 700-875MPa- $\mu\text{m}^{-1/2}$ and estimated σ_0 of 100-200MPa at room temperature. Again, the σ_0 estimated from Knoop hardness (Shang/Sourmail) is higher than the others. Regardless, there is a notable difference between Peierls stresses reported in different studies, the reason for which is not clear.

In summary, in the present work, the significant grain refinement through ECAE at high temperatures were demonstrated in bulk Hiperc soft magnetic alloys for the first time. The resulting samples exhibited high strength levels comparable to Hiperc sheets and the ductility levels much higher than the heat treated conventional bars with large grain sizes. Hall-Petch slope in the present work was found to fit well with those from large grain sized bars and rolled sheets. The increase in ductility was attributed to the disordered phase that may co-exist with the ordered phase and the small grain sizes. Magnetic properties of the ECAE processed of bulk ultrafine grained Hiperc bars should be evaluated thoroughly to evaluate the applicability of these materials as superior bulk soft magnets.

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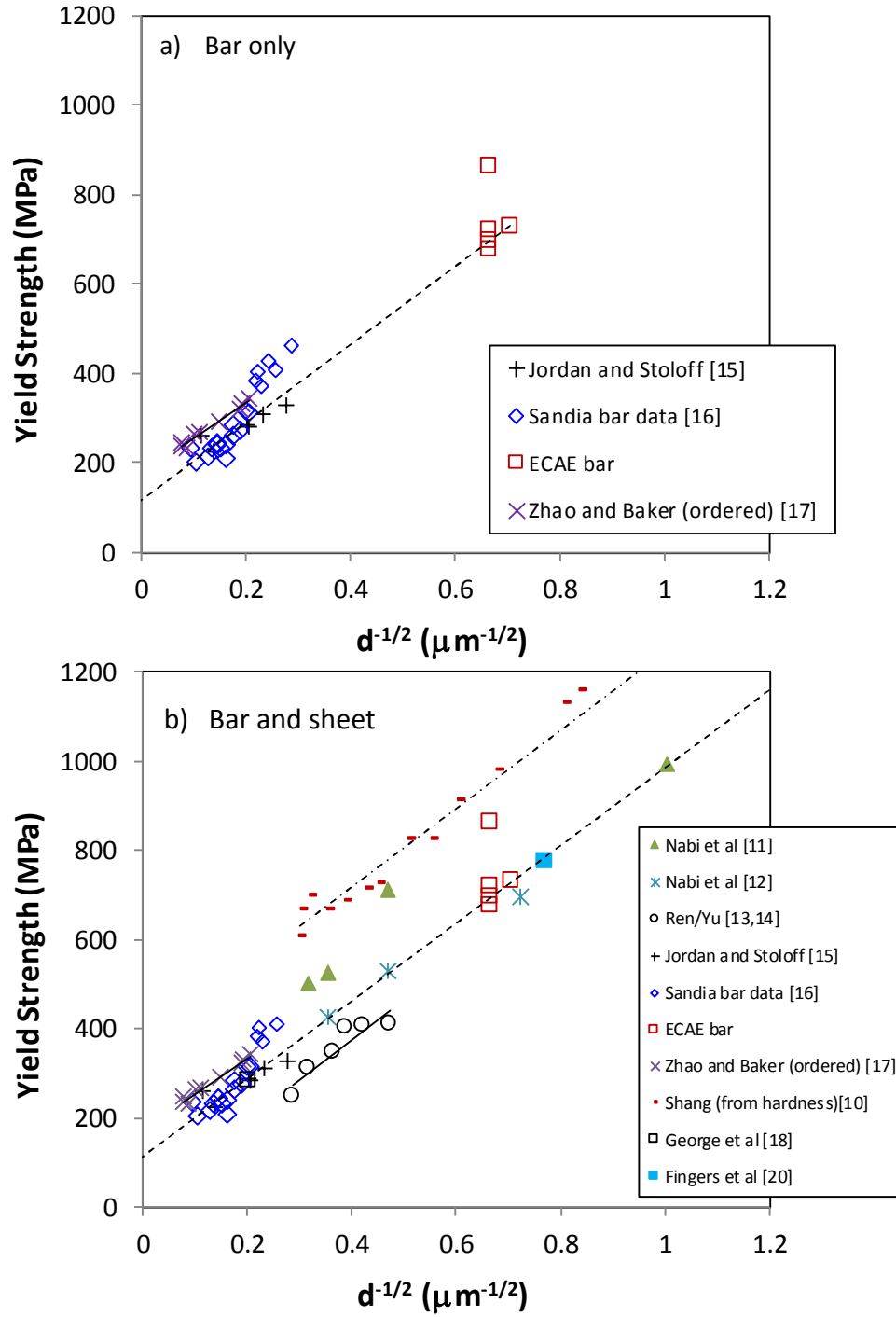


Fig. 4. The Hall-Petch grain size dependence of yield strength for a) FeCo-based alloys and ECAE Hipercor bar, and b) bar and cold-worked sheet.

Table 2. Hall-Petch parameters, σ_0 and k for FeCo-based alloys.

Material	Peierls Stress (MPa)	k (MPa- mm ^{-1/2})	Method	Reference
FeCo (ordered)	172	806	compression	Zhao and Baker [17]
FeCo-2V (ordered)	143	714	tensile	Jordan and Stoloff [15]
FeCo-2V (ordered)	26	848	tensile	Yu et al. [13,14]
Fe-27Co, H50, H50HS	404	837	Knoop hardness	Shang, Sourmail [1,10]
FeCo-2V (600ppm Nb)	104	871	tensile	Current/previous study [16]

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