Prediction of Generation of High and Low Angle Grain Boundaries (HAGB and LAGB) during severe plastic deformation

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Abstract

It is well known that a balance between the generation of low angle and high angle grain boundaries (LABG and HAGB) is achieved in materials undergoing a severe plastic deformation (SPD) process. It is also observed that most annealed materials evolve from a substantial fraction of LAGB at the early deformation steps towards a steady state in which an equilibrium between LAGB and HAGB fractions is attained. In the present work, such a balance is analyzed for different alloys, i.e., Cu, Al, and Fe based. A mathematical expression is proposed for the amount of LAGB and HAGB as a function of the strain impaired which in turn can be used to predict the number of passes (especially for ECAP) or the amount of total deformation for any SPD process necessary to attain a steady microstructure. The model seeks to serve as a first approximation of the mechanical and microstructural properties of ultrafine grain metal materials.

Keywords: High angle grain boundaries (HAGB); low angle grain boundaries (LAGB); continuous dynamic recrystallization (CDRX); grain size; dislocations, severe plastic deformation (SPD)

I. Introduction

Recrystallization is a well-known microstructural phenomenon that can happen either during plastic deformation or with annealing heat treatments. For that reason, it is very important to know its evolution because many properties of metallic materials can be modified through this process [1, 2]. The most popular concept of recrystallization involves the occurrence of nucleation and growth. During nucleation, new grains appear in the microstructure whereas during the growing phase the newly formed grains replace the deformed ones [3, 4, 5]. When recrystallization takes place concurrently with the deformation process is called “dynamic”. This process is modernly named Discontinuous Dynamic Recrystallization (DDRX) [6, 7, 8]. During DDRX processes, high and low angle grain boundaries migrate through atomistic processes that occur near grain boundaries. The migration mechanisms depend on several parameters including the structure of the boundary itself (orientation and plane of the boundary), the temperature, the nature and magnitude of the forces at the boundaries, and finally point defects such as solutes and vacancies [1]. Although, the basic process during boundary migration is the transfer of atoms from and to the grains adjacent to the boundary, many aspects of grain boundary migration can also be interpreted in terms of dislocations theory [6].

On the other hand, in the absence of thermal activation, the generation and accumulation of dislocations are the only effective mechanism concerning dislocation motion. However, in the case of severe plastic deformation (SPD) processes such dislocation agglomeration can lead to another recrystallization process named Continuous Dynamic Recrystallization (CDRX) (which is similar but not identical to the recrystallization process observed in high stacking fault energy materials (ferrite, aluminum alloys, and so on) when deformed at high temperature) [9, 10, 11]. In this case, in the early stages of straining, dislocations create walls of high dislocation density. Then, as strain progresses, these walls recombine as low angle grain boundaries (LAGB). Further deformation promotes some rotation of these sub-grains, and the former LAGB becomes high angle grain boundaries (HAGB) [12, 13, 14, 15, 16, 17]. The resulting microstructure is formed by newly refined grains, but without following the classical nucleation and growing steps.

The CDRX processes can promote very fine grain sizes (providing nanostructured metals), in contraposition with DDRX where grains sizes are rarely below 1-5 micrometer. According to various researchers [12, 18], during CDRX, crystallites are sub-divided by emerging new boundaries, without nucleation of new grains. The responsible mechanisms for the new

boundaries have been associated to a) dislocation cell walls which convert from LAGB into HAGB, b) progressive misorientation increments by absorption of dislocations and c) continuous lattice curvature which can be removed by grouping geometrically necessary dislocations into new walls [19, 20].

Accordingly, the extraordinary mechanical properties noticed in nanostructured materials after SPD (as well as functional properties, such as electrical, magnetic, and so on) strongly depend on the amount and balance between LAGB and HAGB [21, 22, 23]. It has been reported that most metallic materials undergoing SPD display a steady grain size, in such a way that no matter to introduce extra strain, as the grain size (and balance between LAGB and HAGB) remains constant [24]. In this moment, no further refinement is observed, even though more strain is added.

Given the importance of the high and low angle grain boundaries fractions on the mechanical response, especially in pure materials where there is no other contribution to their hardening than interactions between dislocations and dislocations with grain boundaries, it would be advantageous to predict the microstructural and mechanical properties achieved after applying a SPD process to establish the amount of strain imposed on the material to attain the proposed mechanical resistance. As well, this prediction would indicate the practical limit beyond which there is no sense to continue applying deformation as the material has attained a steady state. For this purpose, an expression predicting the amount of high and low angle grain boundaries become necessary. Therefore, this work is focused on deriving such expression, first from a theoretically point of view, then fitting the model with own experimental results, and finally extending it to results reported in the literature. Since the grain boundaries evolution follows different mechanisms depending on the stacking fault energy, alloying elements, and processing conditions, the model presented in this manuscript serves as a qualitative tool for the microstructural properties evolution of metallic materials processed using SPD techniques.

II. Experimental procedure

An Armco iron was selected as based material for this study [25], and Equal Channel Angular Pressing (ECAP) was chosen as the technique to apply SPD. Samples having lengths of ~60mm and diameter of 8mm were deformed by ECAP at room temperature following route Bc. Previous to the ECAP processing, samples were annealed at 930 °C for 20 min. The die used in this study was fabricated from an insert tool steel with two channels intersecting at an

inner angle of $\Phi = 90^\circ$ and an outer angle of $\psi = 37^\circ$ (Figure 1 (a)), resulting in a strain of ~1 per pass. Molybdenum disulfide was used as the lubricant. Samples were subjected up to 16 ECAP passes.

The microstructural characterization of the samples was performed by Electron Back-Scattered Diffraction (EBSD). For this purpose, samples from the center of the billets (TD plane, Figure 1 (b)) processed by ECAP were conventionally polished until 0.02 μm colloidal silica suspension.

The HKL CHANNEL 5 software was used to analyze EBSD results by measuring grain sizes and fractions of HAGB and LAGB. Regions separated by boundaries having misorientation angles (θ) greater than 15° were defined as grains, while subgrains had boundaries with misorientation angles within the range of 2° and 15° . The EBSD system was coupled to a scanning electron microscope (SEM) JEOL JSM-7001F operating at an accelerating voltage of 20 kV, using a step size of 0.03 μm .

Fig. 1- (a) ECAP configuration. (b) TEM and EBSD samples.

For the observation of finer microstructural features, a transmission electron microscope (TEM) FEI-TECNAI G2 S-TWIN was used. It operated at 200 kV, to which it was coupled an orientation-phase mapping precession unit NanoMEGAS ASTAR that makes use of the electron-beam together with recognition of electron diffraction spot patterns, offering the opportunity to perform Automated Crystal Orientation Mapping (ACOM).

TEM samples were initially obtained by cutting billets in the extrusion direction (TD plane, Figure 1 (b)). Then, thin foils were produced by electropolishing using a STRUERS Tenupol-3 machine operating at a voltage of 20 V, applying a current of 0.05 A during 1.5 min. In this study, a mixture of 6% perchloric acid (HClO_4) + and 94% glacial acetic acid (CH_3COOH) was used as an electrolyte at room temperature.

III. Results and discussion

A. Microstructural characterization

Figure 2(a)-2(f) shows the microstructural evolution for the different ECAP passes together with the Grain Orientation Spread (GOS) evolution, highlighting the reduction in grain size from the initial condition (coarse grains) to the one corresponding to sixteen passes (ultrafine grains). The initial annealed microstructure consists mainly of equiaxed grains, with an average grain size of $\sim 72\mu\text{m}$. It is evident that grain refinement took place, i.e., from $72\mu\text{m}$ in the annealed condition to $\sim 360\text{ nm}$ after sixteen passes. On the other hand, other authors have reported even smaller grain sizes for iron processed by different SPD techniques, e.g. Tejedor et al. [26] and Casas et al. [27] obtained grain size values of $\sim 200\text{ nm}$. In those cases, the greater grain size reduction can be attributed to the deformation techniques used by the authors (High Pressure Torsion (HPT) and mechanical milling respectively), which are well known by their capacity to generate more dislocations at high deformations.

Fig. 2- GOS evolution maps for Armco iron after (a) 0, (b) 1, (c) 2, (d) 4, (e) 8, (f) 16 ECAP passes and (g) GOS values plot.

The GOS values correspond with the average deviation orientation between each point in a grain and the average orientation of the grain [28]. Figure 2(g) shows that the highest GOS value of 27° was found in the material with one ECAP pass, indicating the substantial amount of defects introduced in the material, which creates significant orientation differences (more than 76% of the grains present GOS values higher than 6°), taking into account that the initial material (annealed condition with very low defects density) presented a maximum GOS value of 2.3° (99% of the grains showed GOS values lower than 6°).

The GOS maps in Figure 2(a)-2(f) seem to indicate that the homogenization of microstructure (more grains with GOS values lower than 6°-blue color) proceeds regularly with an increasing

number of ECAP passes following a tendency to increase until the first ECAP pass. While with the subsequent ECAP passes the GOS values decrease continuously until the sixth pass where a tendency to level out around 7° is observed (see Figure 2(g)). This behavior in the material, trying to return to a steady state like the initial microstructure, can be interpreted as a microstructural regeneration with a smaller grain size due to the occurrence of CDRX.

The results obtained for the present Armco iron subjected to ECAP show that in the initial five ECAP passes (Figure 3) the LAGB fraction is more predominant. This behavior is in accordance with the literature [29]. The increase in the fraction of LAGBs or the reduction of HAGBs in stage I can be attributed to the development of dense dislocation walls (DDWs), dislocation tangles (DTs) in the original grains and the refined cells followed by the transformation of DDWs and DTs into sub-boundaries with small misorientations separating individual sub-grains (Figure 4).

It can also be seen in Figure 4(a) that the fraction of HAGB begins to saturate with the increase in the number of passes. As further deformation is introduced in the material, more DDWs and DTs are created increasing the misorientation in the grains due to the evolution of LAGB into HAGB (Figure 4(b)-4(e)), allowing the grain refinement. However, by increasing the strain, the rate of dislocation generation is reduced, reaching a saturation value because the grain size starts to limit the free path for the movement of the dislocations [25].

The microstructural characteristics resulting from ECAP processing was also studied by transmission electron microscopy (TEM). During the first ECAP passes (less than 4 passes), a morphology of elongated grains and several regions in the material with significant dislocation density were observed (see Figure 3 and Figure 4(b)-4(e)). After sixteen ECAP passes it occurs a shift from elongated grains and subgrains to more equiaxed ones. The final state highlights the gradual transition from the initially predominant arrangement of low angle grain boundaries to a dominant state of high-angle grain boundaries, as shown in Figure 3.

Fig. 3- HAGB evolution calculated by EBSD and TEM for all ECAP passes.

On the other hand, LAGB and HAGB fraction values calculated by TEM (Figure 3) follow the same trend than when calculated by EBSD, with some minor differences for the initial stages. These differences can be explained by the dimensions of the grains, which are of the order of few microns, being almost the size of the thin area, thus reducing the number of observable grains and not allowing obtaining statistically sound data for analyses.

As mentioned before, the characteristic of CDRX is that nucleation and growth stages of DDRX do not occur. Instead of that, deformation bands, where the number of sub-boundaries (2° - 5° misorientation) is higher than in the surrounding material, are developed during deformation, as shown in the sketch of Figure 4(a). Along with the continuous deformation, sub-boundaries turn into LAGB (5° - 15° misorientation) via dislocation motion and develop into high angle boundaries by merging of lower angle boundaries, as shown in Figure 4(a) (green arrow).

Therefore, it can be understood that larger areas with fractions of high angle boundaries are obtained by increasing the number of dislocations stored in cells, which will finally form subgrain boundaries. Thus, an increment in the level of deformation increases the probability of activation of mechanical annihilation processes, which according with Muñoz et al. [30] results in larger orientation differences between angles of the boundaries.

Fig. 4- CDRX occurrence, (a) HAGB and LAGB evolution for Armco iron processed by ECAP, (b) microstructure evolution after 1pass indicating the formation of DDWs (yellow arrows), DTs (red arrows) and grain boundaries (Light blue arrows), (c) after 4 passes, (d) after 6 passes and (e) after 16 passes.

B. Model description.

As already stated, the dependence of the HAGB and LAGB fraction with the number of ECAP passes (i.e., strain) displays two stages as illustrated in Figure 4(a) for the present Armco iron. The first and shorter stage is characterized by a sharp increment in the LAGB

fraction (or heavy diminution of the HAGB fraction). The second stage however displays a gradual diminution of the LAGB until attaining a sort of steady state. These results agree with the literature [31, 32], although, depending on the initial microstructural state and the SPD method, stage I can be very short or even absent [33].

This shape reminds the flow curves of materials displaying DDRX. Typical DDRX flow curves exhibit a single peak stress followed by a gradual fall towards a steady state stress. In this curve, and at low strains, the deformation controlling mechanism are strain hardening and softening by dynamic recovery (DRV). When DRX is activated, an evident additional softening after the peak stress in the flow curve is observed. From the first studies of Ludwik [34], Hollomon [35] and Bergström [36] until the most recent studies of Estrin et al. [12, 37, 38] among others, the different plasticity models have used exponential type functions to describe the dislocation evolution as well as the flow stress. Various models [39, 40, 41] have been proposed to describe metals undergoing DDRX exclusively. The model proposed by Estrin and Mecking [38] and Bergström [36] to predict the flow curve σ - ε states the following relation between stress and strain:

$$\sigma^2 = [\sigma_s^2 + (\sigma_o^2 - \sigma_s^2) \cdot e^{-\Omega\varepsilon}] \quad (1)$$

where

$$\sigma_o = \alpha' \mu b \sqrt{\rho_o} \quad (2)$$

and

$$\sigma_s = \alpha' \mu b \sqrt{U/\Omega} \quad (3)$$

Assuming that yield stress σ_o at high-temperature conditions can be neglected, equation (1) becomes:

$$\sigma^2 = \sigma_s^2 \cdot (1 - e^{-\Omega\varepsilon}) \quad (4)$$

here α' is a geometric constant, μ the shear modulus, b the burgers vector, U and Ω the characteristic parameters describing the work hardening and DRV behavior respectively, and σ_s the extrapolated saturation stress in the absence of DDRX.

The modeling of DDRX can be accomplished by treating it as a solid-state transformation. In this case, the kinetics of DDRX can be represented by an Avrami equation [41]. Assuming that the mechanical softening is proportional to the recrystallized volume fraction X the constitutive equation that applies after the onset of DDRX is given by:

$$\sigma = \sigma_s - (\sigma_s - \sigma_{ss})X \quad (5)$$

The recrystallization volume fraction, in turns, follows an Avrami kinetics:

$$X = 1 - e^{(-Bt^k)} \quad (6)$$

where B is a constant, t the time and k the Avrami exponent, where, at a constant strain rate, the time becomes equal to the ratio strain/strain rate.

Equations (1) to (8) represent the flow behavior of the material, although some rate controlling equations to quantify and σ_{ss} , X , U and Ω must be provided. When these are available, the flow stress can be expressed as an explicit function of temperature T , strain rate $\dot{\epsilon}$, and strain ϵ .

When analyzing the evolution of LAGB and HAGB in Figure 4(a), it is clear that the function correctly describing the evolution of LAGBs is similar to the one that predicts flow stresses. This similarity, in turns, sounds logic assuming that HAGBs and LAGBs are proportional to the dislocation density involved, which in turns, according to equation (2) is proportional to σ^2 . Consequently, the dependence of LAGB on strain must be similar to equations (1) to (6). Accordingly, the following relation is proposed:

$$LAGB = 1 - HAGB = 1 - [HAGB_0 e^{-\lambda^* \epsilon} + HAGB_{ss} (1 - e^{-\varphi^* \epsilon^n})] \quad (7)$$

where $HAGB_0$ corresponds to the fraction of HAGB for the deformation-free material, while $HAGB_{ss}$ is the saturation value of HAGB reached by means of the deformation process. Here, n corresponds to the growth exponent of the HAGB fraction. This exponent shows how easy it is for a material to reach a saturation value of the HAGB fraction after SPD, i.e., the grade of the material reaching full CDRX. The term λ is denominated as the exponent for LAGB generation, whereas φ is referred to as the generation exponent of HAGB or the probability of converting LAGBs into HAGB due to the CDRX process by which the sub-grains become

grains (see Figure 5). As can be seen in equation (7), the second term is quite similar to the Avrami equation where the B and k constants were related with nucleation and growth rates, whereas in this model the constants φ and n explain the ease and the capacity to transform LAGB into HAGB.

Fig. 5- Model description.

It is worth mentioning here that although some authors have proposed in the literature that when large plastic strains are involved, the Hencky definition of strain should be used instead of the equivalent Von Mises values, here, the later will be used for comparison purposes because most of the experimental results reported in the literature consider equivalent Von Mises expressions.

In the same way that the model to describe DDRX establishes a direct relation between the recrystallized fraction and the flow stress, equivalents relations between CDRX and the flow stress have been established by Hughes et al. [42]. These authors proposed that the hardening

effect or the flow stress after significant deformations can be obtained as the contribution of both LAGB and HAGB fractions (equation (8)).

$$\sigma = \sigma_0 + \sigma_{GB} + \sigma_{SGB} \quad (8)$$

where σ_{GB} and σ_{SGB} are the strengthening contributions from HAGB and LAGB, respectively and can be calculated by the following equations:

$$\sigma_{SGB} = \alpha' \mu M b \sqrt{\rho_0 + \rho_{LAGB}} \quad (9)$$

$$\sigma_{GB} = k' \left(\frac{HAGB}{d} \right)^{1/2} \quad (10)$$

with $\rho_{LAGB} = \frac{3(LAGB)\bar{\theta}_{LAGB}}{bL}$, where L represents the sub-grain size, $\bar{\theta}_{LAGB}$ the average misorientation of the LAGB, M the Taylor factor (3.02 for Armco iron), ρ_0 the dislocations density for the annealed condition, d the grain size, k' the Hall-Petch constant (0.36 MPa m^{1/2} for Armco iron [25]) μ the shear module (80 GPa for Armco iron), and α' a material constant (0.3 for Armco iron). The model proposed by Hughes was applied before in a previous study [43] indicating a good correlation with the experimental data.

Thus, considering that the values of σ_0 and ρ_0 are small with respect to the σ_{SGB} and ρ_{LAGB} magnitudes, the flow stress can be approximated by mixing equations (8), (9) and (10) as follows:

$$\sigma \approx \sqrt{1 - LAGB} \frac{k'}{\sqrt{d}} + \sqrt{LAGB} \cdot \alpha' \mu M b \sqrt{\frac{3\bar{\theta}_{LAGB}}{bL}} \quad (11)$$

C. Model validation

Taking the HAGB values calculated through the EBSD measurements, the model fitting presents a good correlation with the present experimental values (Figure 6(a)). In Figure 6(a)-6(c) the application of the model using different data of materials processed by severe plastic deformation has also been adjusted. The model fitting procedure uses the highest experimental value of HAGB as first approximation, and after some iterations, it calculates

the n , λ , and φ parameters. Then, the model recalculates the $HAGB_{ss}$ value according to the new n , λ , and φ values to obtain the best fitting and the convergence criteria. In the case that convergency is not reached, a new initial value for $HAGB_{ss}$ is used, and the fitting process starts again. The final value of the exponents is defined by fixing each one and looking at how the other parameters change after some iterations, so the final values are chosen when after fixing each exponent, the other values do not change more than 10%.

The values of the different parameters obtained by adjusting the HAGB model are shown in Figure 7 and Table 1 for each one of the materials analyzed in Figure 6. The evolution of model parameters presented in Figure 7(a)-7(b) as a function of transition deformation (i.e., the deformation at the lowest LAGB fraction), indicates that values of λ and φ exponents are higher for materials processed by ARB, followed by materials deformed by ECAP, while, materials processed by HPT show lower values. On the other hand, in Figure 7(c) the opposite happens with the evolution of the n exponent. The highest values were obtained for materials processed by HPT followed by ECAP and ARB. This explains why the materials processed by ARB and ECAP can transform LAGB into HAGB faster than HPT, but neither ECAP nor ARB can reach a complete saturation in the final fraction of HAGB as HPT. This is coherent with the fact that combining deformation processes (i.e., ECAP + HPT) promotes the increase in the fraction of HAGB and the reduction of the grain size.

Fig. 6- Model fittings, (a) ECAP processed (Cu-0.2Mg [32], Pure Cu [31], AA8079 [48], Al-0.13Mg [48], AA6060 [50], pure Al [51]), (b) ARB processed (Armco iron [44], IF Steel [49], Trip steel [46], Stainless steel [47], AA1050 [52], Al/SiC composite [52]) and (c) HPT (AA6061 [53], Cu-0.17Zr [54], Cu-Ni-Si [55], pure Cu [45], Pure Mg [56]) processed materials.

Comparing the same material processed by different techniques (Armco iron processed by ECAP (this study) and Armco processed by ARB [44]), it can be observed that the λ and φ values are higher for the ARB processing, while the value of the n exponent is similar for both cases. This shows that the ARB process can convert LAGB into HAGB faster than ECAP, but both processes get similar values of saturation of the HAGBs. Similar behavior was observed with pure copper processed by ECAP [31] and copper processed by HPT [45]: higher values of λ and φ were obtained for copper processed by ECAP, while copper processed by HPT presented a higher value of n . In other words, materials with higher values of λ and φ could indicate the faster and easier occurrence of CDRX, and the higher values of n would indicate an efficiency of the CDRX giving rise to smaller grain sizes.

Fig. 7- Model parameters evolution, (a) λ , (b) ϕ and (c) n exponents for different materials and SPD techniques.

One important characteristic of λ and ϕ exponents is that the values of the λ exponent tend to be higher than ϕ values. Such characteristic is not only coherent with the fact that the highest amount of LAGBs are present at lower values of deformations but also with the feature that the multiplication rate of the dislocations is also high during the initial stages of deformation [25].

Figure 7(a)-Figure 7(b) demonstrate that high λ and ϕ values are related to deformations values smaller for the ECAP and ARB than the HPT process, which corroborates the faster transition from stage I to stage II after ECAP and ARB processing. On the other hand, Figure 7 (c) plots the n exponent as a function of the maximum deformation applied in each process. In this way, the n exponent for the HPT process shows higher values than ECAP and HPT as a consequence of the significant deformation differences introduced in each process.

Analyzing materials with low Stacking Fault Energy (SFE), TRIP steel [46] and stainless steel [47] processed by ARB, they present similar values in all three exponents (λ , ϕ and n). Besides, the values of the n exponent were the lowest, together with the one for Cu-0.2Mg

[32] processed by ECAP between the others materials. Such behavior is coherent with the one observed for materials presented in Figure 6(a) and 6(b) from which one may perceive no significant increments in the HAGB fraction in stage II, which is also related to the fact that CDRX occurs more readily in materials with high SFE, as Gourdet et al. [19] indicated in their CDRX model. On the other hand, materials like Al-0.13Mg [48], IF steel [49] and pure Cu [45] processed by ECAP, ARB and HPT respectively, displayed the highest values of the exponent n . However, the grain boundaries evolution in materials with low SFE follows a more complicated pathway due to the alloy elements that give rise to additional hardening mechanisms. Therefore, the present model results can be interpreted qualitatively.

The application of the model for each deformation process is shown in Figure 8(a)-8(c). It can be seen a similar trend, where, for the HPT process, the n exponent presents the highest value (Figure 8c), whereas after ARB deformation the highest values were obtained for the λ and φ exponents (Figure 8(b)). The ARB process also showed less data scattering and presented a smaller action zone (shaded areas in Figure 8(d)) as compared to ECAP and HPT. Such an observation is coherent with the fact that ECAP and HPT involve higher hydrostatic pressures that allow the introduction of further deformation. For that reason, HPT showed a larger action zone during material processing. Additionally, Figure 9 and Table 2 display the model application following equations (9) to (11) and the comparison with the experimental yield stress values for the Armco iron processed by ECAP. The experimental and theoretical values keep a good correlation, demonstrating the model is also a useful tool to predict the mechanical behavior of metallic materials after the SPD process.

Based on the results of the model, it can be demonstrated that the most important parameter during the evolution of the HAGB is the mode of deformation (i.e., the used SPD technique). Processes like ECAP and ARB showed faster transitions from LAGB (stage I) to HAGB (stage II) (higher values of the λ and φ exponents) while the HPT processing proved to be the most efficient method to obtain HAGB (i.e., more continuous dynamic recrystallization) showing the highest values of the n exponent.

Fig. 8- Overall model fitting, (a) ECAP process, (b) ARB process, (c) HPT process and (d) approximated action zones for each process.

Fig. 9- Comparison of the model yield stress prediction and the comparison with the experimental values for Armco iron processed by ECAP.

IV. Conclusions

Armco iron with grains ranging in micrometer size was processed by ECAP to obtain a nanostructured material. Different techniques, such as EBSD and TEM, confirmed the grain refinement. The occurrence of CDRX was corroborated by GOS maps that showed how the microstructure was regenerated with smaller grain size, leading to GOS values quite similar to the annealed material (0 passes).

Armco iron processed by ECAP at a different number of passes was used as reference material to develop a new model which could predict the HAGB evolution as a function of the deformation introduced in the material during severe plastic deformation processing at room temperature. The model showed good correlation with the experimental data obtained from the EBSD measures as well as with different experimental data of materials deformed by different SPD techniques.

The model pretends to serve as a first **qualitative** approximation of the CDRX. The model can establish which method can produce a faster conversion from a microstructure dominated by the LAGB fraction towards one dominated by HAGB (larger λ and φ exponents) and which

processes are more efficient to reach higher HAGB fractions during plastic deformation (larger n exponent).

The model also gave an approximation of the action zones for the ARB, ECAP and HPT processes, indicating that HPT presents a larger range followed by ECAP and ARB.

The new model indicates a good correlation between the theoretical and experimental yield stress and works as a connection between the microstructural characteristics (mainly, grain size and LAGB fractions) and mechanical properties of metallic materials after severe plastic deformation.

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Data availability

The raw/processed data required to reproduce these findings can be provided upon request

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Figure captions:

Figure 1. (a) ECAP configuration. (b) TEM and EBSD samples.

Figure 2. GOS evolution maps for Armco iron after (a) 0, (b) 1, (c) 2, (d) 4, (e) 8, (f) 16 ECAP passes and (g) GOS values plot.

Figure 3. HAGB evolution calculated by EBSD and TEM for all ECAP passes.

Figure 4. CDRX occurrence, (a) HAGB and LAGB evolution for Armco iron processed by ECAP, (b) microstructure evolution after 1pass indicating the formation of DDWs (yellow arrows), DTs (red arrows) and grain boundaries (Light blue arrows), (c) after 4 passes, (d) after 6 passes and (e) after 16 passes.

Figure 5. Model description.

Figure 6. Model fittings, (a) ECAP processed (Cu-0.2Mg [32], Pure Cu [31], AA8079 [48], Al-0.13Mg [48], AA6060 [50], pure Al [51]), (b) ARB processed (Armco iron [44], IF Steel [49], Trip steel [46], Stainless steel [47], AA1050 [52], Al/SiC composite [52]) and (c) HPT (AA6061 [53], Cu-0.17Zr [54], Cu-Ni-Si [55], pure Cu [45], Pure Mg [56]) processed materials.

Figure 7. Model parameters evolution, (a) λ , (b) ϕ and (c) n exponents for different materials and SPD techniques.

Figure 8. Overall model fitting, (a) ECAP process, (b) ARB process, (c) HPT process and (d) approximated action zones for each process.

Figure 9. Comparison of the model yield stress prediction and the comparison with the experimental values for Armco iron processed by ECAP.

Table captions:

Table 1. Model parameters and grain size reported before and after deformation.

Table 2. Model validation parameters

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Table 1. Model parameters and grain size reported before and after deformation.

Material	HAGB ₀	λ	φ	n	HAGB _{ss}	Grain size (initial-final) [μm]	Reference
ECAP Processed materials							
This study	95.40	3.83	0.09	1.54	72.30	(70-0,35)	
Cu-0.2Mg	88.00	3.51	0.14	0.56	67.00	(670-0,31)	[32]
Pure Cu	94.00	1.17	0.12	1.57	67.00	(5-0,45)	[31]
AA8079	87.00	7.60	0.37	1.31	80.00	(1000-0,51)	[48]
Al-0.13Mg	88.00	1.40	0.02	2.46	80.00	(1000-0,68)	[48]
AA6060	62.00	1.12	0.18	1.32	59.00	(52-0,47)	[50]
Pure Al *	90.00	3.30	0.15	1.01	80.00	(1000-1,2)	[51]
ARB Processed materials							
Armco iron	80.00	4.47	0.27	1.61	63.00	(15-0,58)	[44]
IF steel	80.00	1.68	0.12	1.94	90.00	(33-1)	[49]
Trip Steel	97.00	3.17	0.87	0.67	50.00	(35-0,3)	[46]
Stainless steel	80.00	3.50	0.84	0.70	75.00	(10-not specified)	[47]
AA1050	84.00	4.27	0.42	1.36	73.00	(14-0,3)	[52]
Al/SiC composite	86.00	4.77	0.45	0.97	85.00	(12-0,19)	[52]
HPT Processed materials							
AA6061	95.00	0.30	0.14	0.97	65.00	(150-0,52)	[53]
Cu-Ni-Si	80.00	0.90	0.02	1.71	70.00	(20-0,2)	[55]
Cu-0.17Zr *	90.00	0.27	0.05	1.55	82.00	(not specified-0,2)	[54]
Pure Cu *	90.00	0.70	0.01	3.02	69.00	(not specified-0,47)	[45]
Pure Mg *	90.00	0.56	0.14	1.48	84.00	(not specified-<0,6)	[56]

*The value of HAGB₀ was assumed to be ~90% since it was not specified in the paper, but the authors indicated a previous heat treatment before the respective SPD process.

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Table 2. Model validation parameters

ECAP passes	Subgrain size [μm]	Grain size [μm]	LAGB	$\bar{\theta}$ [rad]	ρ_{LAGB} [m^{-2}]	σ_{SGB} [MPa]	σ_{GB} [MPa]	σ_y experimental [25] [MPa]
0	72.42	90.12	0.046	0.011	8.53E+10	19.2	37.0	204
1	1.08	2.71	0.912	0.045	4.54E+14	392.9	62.9	441
2	1.13	2.55	0.82	0.059	5.28E+14	423.9	92.2	506
3	0.34	0.8	0.67	0.029	7.47E+14	503.9	212.1	579
4	0.27	0.66	0.577	0.026	7.26E+14	496.9	274.3	703
5	0.23	0.77	0.41	0.026	7.28E+14	497.3	283.4	715
6	0.27	0.51	0.42	0.027	5.57E+14	435.1	371.7	752
7	0.25	0.49	0.327	0.025	4.81E+14	404.4	396.8	774
8	0.26	0.51	0.405	0.026	4.25E+14	380.3	404.8	747
16	0.29	0.42	0.246	0.021	2.43E+14	287.7	471.2	844

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