Stress-Induced Grain Growth in an Ultra-Fine Grained Al Alloy

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This paper reports on a study of the stress-induced grain growth phenomenon in the presence of second-phase particles and solutes segregated at grain boundaries (GBs) during high-temperature deformation of an ultra-fine grained (UFG) Al alloy synthesized *via* the consolidation of mechanically milled powders. Our results show that grain growth was essentially inhibited during annealing at 673 K (400 °C) in the absence of an externally applied stress, whereas in contrast, grain growth was enhanced by a factor of approximately 2.7 during extrusion at 673 K (400 °C). These results suggest that significant grain growth during hot extrusion was attributable to the externally applied stresses stemming from the state of stress imposed during extrusion and that the externally applied stresses can overcome the resistance forces generated by second-phase particles and solutes segregated at GBs. The mechanisms underlying stress-induced grain growth were identified as GB migration and grain rotation, which were accompanied by dynamic recovery and possible geometric dynamic recrystallization, while discontinuous dynamic recrystallization did not appear to be operative.

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I. INTRODUCTION

STRESS-INDUCED grain growth has attracted considerable interest among the scientific community as evidenced by the number of articles published during the past decade.^[1–24] These research efforts have been motivated primarily by two factors: scientific interest and technological importance. From a scientific perspective, it is of interest to provide insight into the fundamental mechanisms that are active during stress-induced grain growth, including the role of any externally applied stresses, as well as the interactions between those mechanisms, for example. From a technological standpoint, understanding these fundamental questions is a prerequisite in order to be able to control grain size which critically influences mechanical behavior, since the manufacturing of the vast majority of materials involves some type of plastic deformation processing and the presence of externally applied stresses which are likely to influence grain growth phenomena. For example, most industrial products are fabricated *via* conventional plastic deformation approaches (*e.g.*, rolling, forging, and extrusion),^[25,26] and more recently, nanostructured (NS) and ultra-fine grained (UFG) metals are frequently synthesized either *via* severe plastic deformation approaches (equal-channel angular pressing,^[27] high-pressure torsion,^[28] accumulative rolling bonding,^[29] *etc.*) or by consolidation of NS powders,^[30] during which the aforementioned conventional plastic deformation approaches are implemented for secondary consolidation.

Recent studies on stress-induced grain growth involve *in situ*^[1–8] and *ex situ*^[9–13] transmission electron microscopy (TEM) analysis, optical microscopy observations^[14,15], and molecular dynamic (MD) simulations.^[16–24] Related published theoretical and experimental results reveal that there are two roles that an externally applied stress plays during grain growth: (1) provides a driving force for grain growth,^[2,16] and (2) promotes the coupling between the two mechanisms responsible for grain growth, including grain boundary (GB) migration and grain rotation.^[2,7,14,18,20,21,24,31] Noteworthy, however, is the fact that most published theoretical and experimental papers related to stress-induced grain growth focus on single-phase materials and as a result, material systems containing second-phase particles have been rarely reported. Moreover, although many materials that contain a high level of solute segregation at GBs are selected as model materials for studies of stress-induced grain growth,^[5,9,11,32,33] the influence of solute atoms segregated at GBs on stress-induced grain growth has been investigated only in very limited cases.^[24,34] For example, in a related study,^[34] the effect of O segregated at GBs in nanocrystalline Al films produced by magnetron sputtering on stress-induced grain

growth during uniaxial tensile testing was investigated *via* TEM and atom-probe tomography (APT). The results of this study revealed that segregated O could inhibit stress-induced grain growth as a consequence of pinning of GBs by segregated O when the concentration of segregated O exceeds a critical level; these results were subsequently validated on the basis of MD simulations.^[24] Interestingly, the MD results^[24] suggest that the critical stress required for triggering coupled GB motion can be expressed as a function of O concentration at the GB.

Furthermore, published experimental studies of stress-induced grain growth predominantly address phenomena that occur at room temperature (RT)^[1,3–7,9,11–13] and cryogenic temperature.^[10] In contrast, stress-induced grain growth at elevated temperatures has been investigated only in few studies,^[2,8] which hinders our basic understanding of the aforementioned commercially important plastic deformation approaches, including extrusion, rolling, forging, *etc.* These plastic deformation processes generally involve elevated temperatures and are widely used, not only in industrial production, but also for the synthesis of NS and UFG materials *via* the consolidation of NS powders, as presented above. These deformation processes result in stress-induced grain growth being accompanied by dynamic recovery (DRV) and/or dynamic recrystallization (DRX), which increase their complexity. Significantly, despite the fact that experimental results^[2,8] as well as MD simulations^[16–18,21] show that stress-induced grain growth is indeed present at elevated temperatures, the interactions between stress-induced grain growth and possible DRV and/or DRX have heretofore not been studied in detail.

On the basis of the above discussion, the present study was motivated by two factors. First, there is a lack of fundamental information on stress-induced grain growth in the presence of both second-phase particles and solute atoms segregated at GBs, which hinders our ability to effectively control grain size evolution in many technically important alloys (*e.g.*, Al alloys, steels, *etc.*). Second, published experimental studies on stress-induced grain growth phenomena during high-temperature plastic deformation processes are very limited,^[2,8] which limit our knowledge of the conditions required to carefully control the behavior of NS and UFG metals during consolidation as well as of engineering materials during thermomechanical processing. In the present study, stress-induced grain growth occurring in an UFG 5083Al (an Al-Mg-Mn-Cr alloy) during hot extrusion at 673 K (400 °C) was first analyzed and discussed by comparing the scenarios of grain growth in the absence and presence of an externally applied stress, and the mechanisms governing stress-induced grain growth, including GB migration and grain rotation, were then identified based on microstructural analysis.

II. EXPERIMENTAL PROCEDURES

The model material 5083Al selected for study was synthesized *via* the consolidation of mechanically milled

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powders, using the following procedure. First, as-atomized 5083Al pre-alloyed powders of less than 40 μm in diameter that contain supersaturated solution of constituent elements Mg, Mn, and Cr were used as starting powders. Second, the gas-atomized powders were mechanically milled using stainless steel balls and a vessel in an argon atmosphere at RT for 4 hours. Third, the mechanically milled powders were then canned, degassed at 673 K (400 °C) for 25 hours, and consolidated *via* hot isostatic pressing (HIPping) under 175 MPa at 648 K (375 °C) for 4 hours to prepare fully dense materials. The composition of the as-hot isostatic pressed (HIPped) 5083Al was measured using chemical analysis conducted at a commercial laboratory (Luvak Inc., Boylston, MA) per ASTM E1097-07, E1019-08, and E1447-09 and is given in Table I. The impurities C, O, N, Fe, Ni, Si, and Cu either originated from the raw materials used or were introduced during milling.

The grain growth phenomenon in the as-HIPped 5083 Al was then investigated for the following two cases: i) annealing at 673 K (400 °C) for 5 hours in the absence of an applied stress, and ii) extrusion at 673 K (400 °C) with an area reduction ratio of 10 (true strain ~ 2.30) under a ram speed of 1.4 mm/s. The extruded material was prepared as follows. First, prior to extrusion, the as-HIPped 5083 Al round billet of 25 mm in diameter and 50 mm in length was held at 673 K (400 °C) for 1 hour. Second, the preheated round billet was extruded using a die of 8.6 mm in the length of the deformation zone. Third, a round rod of 7.9 mm in diameter was produced following extrusion. We estimated that the extrusion process lasted ~ 36 seconds, with an associated deformation time of ~ 2.9 seconds and a strain rate of $\sim 0.8 \text{ s}^{-1}$ on the basis of the extrusion die geometry and the extrusion parameters used in the present study. Following annealing and extrusion, the materials were immediately water quenched to suppress any possible microstructural evolution.

The microstructures of as-HIPped, annealed, and extruded 5083 Al were investigated using TEM Philips CM 12 and high-resolution TEM (HRTEM) JEOL JEM 2500 SE operated at voltages of 120 and 200 kV, respectively. The grain size (d) and the second-phase particle size (l) are represented by an area-equivalent circle diameter; in TEM bright-field (BF) images, cross-sectional areas of the grains (S_g) and of the second-phase particles (S_p) were measured and converted to area-equivalent circle diameters using the following equations: $d = (4S_g/\pi)^{1/2}$ and $l = (4S_p/\pi)^{1/2}$. On the basis of a large number of randomly selected TEM BF images, the statistical distributions of grain sizes and second-phase particle sizes were determined and their average values were calculated by fitting the statistical distributions using lognormal probability function.^[34–36] The preparation of TEM and HRTEM specimens is described as follows. First, the materials were sectioned,

mechanically thinned to $<30 \mu\text{m}$, and dimpled (VCR D500i dimpler) to 10 to 20 μm in thickness. For the extruded 5083 Al, the materials were taken along the extrusion direction and thinned perpendicular to the extrusion direction. Then, the materials were further thinned to an electron-transparent thickness using a Gatan PIPS 691 precision ion milling system under an Ar^+ accelerating voltage of 4 kV. X-ray diffraction (XRD) analysis was also conducted on as-milled 5083 Al powders, as-HIPped, and extruded 5083 Al bulk materials using Scintag XDS2000 X-ray diffractometer that is equipped with a Cu target operated at 1.8 kW.

For the extruded 5083 Al, the solute concentrations at GBs and at grain interiors (GIs) were measured *via* a Cameca local-electrode atom-probe (LEAP) 4000X-Si tomograph with a pulsed ultra-violet laser (355 nm wavelength), a pulse repetition rate of 500 kHz, and a pulse energy of 10 to 15 pJ. Steady-state direct-current voltages between 2.0 and 8.0 kV were applied, with the specimen base temperature being 30 K (-243 °C). Nine datasets were collected and analyzed by three-dimensional (3D) reconstruction and visualization using Cameca's software program IVAS version 3.6.4. The specimens for APT analysis were prepared by sectioning rectangular cuboids with dimensions of $\sim 10 \text{ mm} \times \sim 0.3 \text{ mm} \times \sim 0.3 \text{ mm}$ from the bulk materials and then electropolishing the rectangular cuboids to needle-shaped tips with a diameter of $\sim 50 \text{ nm}$ at the apex using 10 pct perchloric acid in acetic acid and 2 pct perchloric acid in butoxyethanol as the electrolytes.

III. RESULTS

A. Microstructure of the as-HIPped 5083 Al

The typical TEM microstructure of the as-HIPped 5083 Al is shown in Figure 1(a), and it shows reasonably equiaxed ultra-fine grains. HRTEM analysis revealed that more than 85 pct of these grains are separated by HAGBs (misorientation $>15 \text{ deg}$). A representative HRTEM image containing a GB is displayed in Figure 1(b), where grain A shows a clear two-dimensional atomic image as a result of being on $\langle 110 \rangle$ zone axis, while in contrast, the neighboring grain B exhibits an amorphous-like characteristics, indicative of an orientation far away from the zone axes. The difference in the HRTEM feature between grains A and B suggests that their GB is of high-angle nature. The above result is consistent with the conclusion in the published studies on UFG Al alloys synthesized *via* consolidation of mechanically milled NS powders.^[37,38] The grain size distribution was obtained based on measurements on 302 grains randomly selected, as shown in Figure 1(c); the average grain size was determined to be $\bar{d} = 244 \text{ nm}$.

Table I. Chemical Composition of the as-HIPped 5083Al (Wt Pct)

C	O	N	Mg	Mn	Cr	Fe	Ni	Si	Cu	Al
0.019	0.45	0.004	4.5	0.70	0.16	0.20	0.019	0.13	0.014	balance

Fig. 1—The microstructure of the as-HIPped 5083 Al: (a) TEM BF micrograph showing grains and second-phase particles (indicated by red and white arrowheads), (b) HRTEM micrograph showing the GB between two neighboring grains, (c) statistical distribution of grain size, and d) TEM BF micrograph showing the finer second-phase particles at GIs (Color figure online).

Careful inspection of Figure 1(a) shows the presence of dispersed second-phase particles. The second-phase particles can be divided into two categories: i) coarser particles (marked by red arrowheads in Figure 1(a)) of 20 to 100 nm in size despite the occasional presence of particles >100 nm, which are distributed at both GBs and GIs; and ii) finer particles (marked by white arrowheads in Figure 1(a)) at GIs, which range from 5 to 20 nm in size as revealed by a TEM image of higher magnification (Figure 1(d)). The statistical analysis based on a large number of TEM view fields revealed ~ 30 nm of the average second-phase particle size ($\bar{l}$) in the as-HIPped 5083 Al. Inspection of the published literature^[39] indicates that the second phases in the 5083 Al synthesized by consolidation of mechanically milled powders have been identified as $\text{Al}_{12}\text{Mg}_2(\text{CrMnFe})$, $\text{Al}_{12}(\text{FeMn})_3\text{Si}$, $\text{Al}_6(\text{CrMnFe})$, MgO , SiO_2 , and CuMgSiAl . Given that mechanically milled 5083 Al powders underwent a prolonged period during degassing and HIPping at high temperatures [degassing at 673 K (400 °C) for 25 hours and HIPping at 648 K (375 °C) for 4 hours], the as-HIPped 5083Al can be considered to achieve the equilibrium state and consequently, the volume fractions of the various second-phase particles were determined primarily

by the equilibrium thermodynamic conditions [*i.e.*, the equilibrium solubility of elements in the matrix Al at 648 K (375 °C)]. Given the extremely low equilibrium solubility of Mn, Cr, Fe, and O in Al^[40] and relatively high contents of these elements (Mn 0.7 wt pct, Cr 0.16 wt pct, Fe 0.20 wt pct, and O 0.45 wt pct) and of Si (0.13 wt pct), appreciable amounts of $\text{Al}_{12}\text{Mg}_2(\text{CrMnFe})$, $\text{Al}_{12}(\text{FeMn})_3\text{Si}$, $\text{Al}_6(\text{CrMnFe})$, and MgO should be observed. In the present 5083 Al, Cu-containing particles (CuMgSiAl), as well as carbide, nitride, and Ni-containing particles, were rarely found due to the very low contents of Cu (0.014 wt pct), C (0.019 wt pct), N (0.004 wt pct), and Ni (0.019 wt pct). Based on the above discussion, the second-phase particles in the as-HIPped 5083 Al should primarily comprise the following phases $\text{Al}_{12}\text{Mg}_2(\text{CrMnFe})$, $\text{Al}_{12}(\text{FeMn})_3\text{Si}$, $\text{Al}_6(\text{CrMnFe})$, and MgO .

B. Microstructure of the Annealed 5083 Al

Figure 2(a) shows the typical TEM microstructure of the annealed 5083 Al. Based on statistical analysis of 421 grains observed in a series of randomly selected TEM images, the grain size distribution was reported in

Figure 2(b), and the average grain size was evaluated as $\bar{d} = 262$ nm. Comparison of the average grain size of the annealed 5083 Al (262 nm) with that of the as-HIPped 5083 Al (244 nm) shows that very slight grain growth occurred during annealing.

In addition, by statistical analysis of the second-phase particles in the as-HIPped 5083 Al (e.g., as shown in Figures 1(a) and (d)) and those in the annealed 5083 Al (e.g., as displayed in Figures 2(a) and (c), the latter being the higher-magnification TEM image of finer particles), it is revealed that, although subjected to annealing at 673 K (400 °C) for 5 hours, the volume fraction, size, and distribution of both coarser and finer particles remain almost unchanged. Based on the discussion in Section III-A, the as-HIPped 5083 Al was in an equilibrium state. Thus, during annealing of the as-HIPped 5083 Al, the volume fraction of the second-phase particles was also determined by the equilibrium solubility of the elements. Due to the very low equilibrium solubility of Mn, Cr, Fe, and O in Al,^[40] the effect of the difference in equilibrium solubility of these elements at 648 K and 673 K (375 °C and 400 °C) on the amounts of second-phase particles containing these elements, including $\text{Al}_{12}\text{Mg}_2(\text{CrMnFe})$, $\text{Al}_{12}(\text{FeMn})_3\text{Si}$, $\text{Al}_6(\text{CrMnFe})$, and MgO , can be neglected. Again, as a result of very low contents of Cu, C, N, and Ni, it is highly unlikely that CuMgSiAl , carbide, nitride, and Ni-containing particles are present in the annealed 5083 Al. Accordingly, it is appropriate to assume that the volume fraction of the second-phase particles remains essentially unchanged. The unchanged size and distribution of the second-phase particles suggest a high thermal stability of the four types of second-phase particles. This phenomenon can be attributed to the extremely low equilibrium solubility^[40] and/or diffusivity^[41] of some constituent elements of the four types of particles (i.e., Mn, Cr, Fe, and O) in the matrix Al, which inhibits the dissolution-precipitation process responsible for coarsening of the second-phase particles.^[42,43]

C. Microstructure of the Extruded 5083 Al

The TEM BF image in Figure 3(a) shows the typical microstructure of the extruded 5083 Al. In the extruded materials, the grain size distribution was statistically analyzed by measuring grains that exhibit strong diffraction conditions (i.e., a low-index zone axis is parallel or close to the direction of the incoming electron beam) and thus appear dark and have clear boundaries with the neighboring grains in the TEM BF images. Such grains are normally characterized by high misorientations (i.e., HAGBs) with the neighboring grains,^[44,45] which can be confirmed *via* HRTEM analysis (Figure 3(b)). Based on the measurements of 412 grains, the grain size distribution exhibited an average grain size of $\bar{d} = 647$ nm, as shown in Figure 3(c). By comparing the average grain sizes of the as-HIPped (244 nm) and the extruded (647 nm) 5083 Al, it is evident that significant grain growth, by a factor of ~ 2.7 , occurred during extrusion.

Fig. 2—The microstructure of the annealed 5083 Al: (a) TEM BF micrograph showing grains and second-phase particles, (b) statistical distribution of grain size, and (c) TEM BF micrograph showing the finer second-phase particles at GIs (Color figure online).

In addition, *via* quantitative analysis of the second-phase particles in the as-HIPped 5083 Al (as shown in Figures 1(a) and (d)) as well as those in the extruded 5083 Al (as displayed in Figures 3(a) and (d), the latter being the higher-magnification TEM image of finer particles in extruded 5083 Al), the volume fraction, size, and distribution of both coarser and finer particles remain almost unchanged. The unchanged volume fraction of the second-phase particles in the extruded 5083 Al can be rationalized as follows. As analyzed in Section III-A, the as-HIPped 5083 Al was characterized by an equilibrium state. In the present study, extrusion

Fig. 3—The microstructure of the extruded 5083 Al: (a) TEM BF micrograph showing grains with three different features (marked by I to III) and second-phase particles, (b) HRTEM micrograph showing the GB between two neighboring grains, (c) statistical distribution of grain size, and (d) TEM BF micrograph showing the finer second-phase particles at GIs (Color figure online).

of 5083 Al was performed at a temperature as high as 673 K (400 °C) (homologous temperature of ~ 0.80) with a moderate straining magnitude of 2.3 under a relatively low strain rate of 0.8 s^{-1} . Thus, 5083 Al can be presumably considered to undergo an equilibrium process during hot extrusion. The volume fraction of the second-phase particles during hot extrusion of as-HIPped 5083 Al was also primarily determined by the equilibrium solubility of the elements, as discussed above. Given the fact the elements Mn, Cr, Fe, and O in Al exhibit an extremely low equilibrium solubility in Al,^[40] we can presumably neglect any effects associated with differences in equilibrium solubility of these elements at 648 K and 673 K (375 °C and 400 °C) on the volume fractions of the second-phase particles containing these elements, *i.e.*, $\text{Al}_{12}\text{Mg}_2(\text{CrMnFe})$, $\text{Al}_{12}(\text{FeMn})_3\text{Si}$, $\text{Al}_6(\text{CrMnFe})$, and MgO. As discussed in Section III-B, the very low contents of Cu, C, N, and Ni lead to negligible volume fractions of CuMgSiAl , carbide, nitride, and Ni-containing particles. The unchanged size and distribution of the second-phase particles are attributable to two factors: (1) high thermal stability of the second-phase particles in 5083 Al as discussed in Section III-B, and (2) negligible fragmentation of the nano-scale second-phase particles during

plastic deformation, consistent with experimental observations in the published literature.^[35,46]

Careful inspection of the microstructure at GIs in the extruded 5083 Al (Figure 3(a)) reveals three types of grains based on microstructural features at GIs. First, most grains (as marked by “I” in Figure 3(a) as examples) contain well-defined smaller subgrains with size ranging from ~ 100 to $\sim 300 \text{ nm}$. The subgrain structure inside such grains can be more clearly demonstrated in a high-magnification TEM micrograph (Figure 4(a)), where the GBs are delineated by dots and subgrains are marked by “SG.” The HRTEM micrograph (the zone axis $\langle 110 \rangle$ parallel to the incoming electron beam direction) as shown in Figure 4(b) confirms the presence of the low-angle subgrain boundaries (SGBs) with misorientation of 2 to 3 deg between the subgrains. It is noted that, in addition to the subgrains (marked by “SG” in Figure 4(a)), there exists a region (marked by “D”) where subgrain structure was not well developed and consequently, SGBs cannot be observed. HRTEM observation indicates an almost identical orientation inside the region (not shown herein). Second, TEM examination also reveals some grains (marked by “II” in Figure 3(a) as examples) that consist of only larger well-defined subgrains with size falling in the range of ~ 450 to $\sim 1000 \text{ nm}$.

Fig. 4—Higher-magnification micrograph of grains with microstructural features marked by “I” in Fig. 3(a): (a) microstructure at the GIs, where the HAGBs are delineated by dotted lines, the subgrains are marked by “SG,” and the regions where SGBs are not developed are marked by “D,” and (b) HRTEM micrograph of the region marked by a black square in (a), showing the low-angle SGB, where the SGB is delineated by the asterisks and the low-misorientation angle (<15 deg) is revealed by the two straight black lines.

The high-magnification TEM microstructure of this type of grains is shown in Figure 5(a). Again, the presence of low-angle SGBs inside the grain was confirmed by the HRTEM analysis as shown in Figure 5(b). Third, in addition to the grains containing a subgrain structure (both smaller and larger subgrain sizes as mentioned above), grains, inside which subgrain structure was not well developed, can also be observed, as marked by “III” in Figure 3(a). The high-magnification TEM microstructure of this type of grains is shown in Figure 6.

D. XRD Results

The XRD patterns of gas-atomized 5083 Al powders, the as-HIPped, and the extruded bulk 5083 Al are shown in

Fig. 5—Higher-magnification micrograph of a grain with microstructural features marked by “II” in Fig. 3(a): (a) microstructure at the GIs, where the HAGBs are delineated by dotted lines and the subgrains are marked by “SG,” and (b) HRTEM micrograph of the region marked by a black square in (a), showing the low-angle SGBs, where the SGBs are delineated by the asterisks and each of the low-misorientation angles (<15 deg) between two neighboring subgrains is revealed by the two straight black lines.

Figure 7. Each pattern exhibits five peaks corresponding to $\{111\}$, $\{200\}$, $\{220\}$, $\{311\}$, and $\{222\}$. The intensities of the diffraction peaks were taken from these XRD patterns, as given in Table II. The crystallographic textures in the aforementioned materials were determined based on the intensities using a model originally developed by Lotgering.^[47] In the model, the degree of orientation of a crystal plane $\{hkl\}$, O_{hkl} , is defined as:

$$O_{hkl} = \frac{V_{hkl} - V_{hkl,0}}{1 - V_{hkl,0}}, \quad [1]$$

where $V_{hkl} = I_{hkl} / \sum I_{hkl}$ (I_{hkl} is the intensity of the peak corresponding to $\{hkl\}$ and $\sum I_{hkl}$ is the sum of intensity of all peaks for the investigated bulk material) and $V_{hkl,0} = I_{hkl,0} / \sum I_{hkl,0}$ ($I_{hkl,0}$ is the intensity of the peak

corresponding to $\{hkl\}$ and $\sum I_{hkl,0}$ is the sum of intensity of all peaks for the corresponding powder diffraction). Based on the intensities provided in Table II, the degrees of orientation of various $\{hkl\}$ were then calculated, as reported in Table III. It is noted that, when the calculations were performed, only the intensities corresponding to the five peaks in Figure 7 were used to obtain the sums of intensities. Since the sum of intensities corresponding to the five peaks accounts for a predominant percentage of the total sum of intensities of all peaks,^[48] those corresponding to other peaks can be safely neglected. The

Fig. 6—Higher-magnification TEM micrograph of an on-axis dark grain, showing the microstructural features marked by “III” in Fig. 3a.

Fig. 7—XRD patterns of gas-atomized 5083 Al powders, as-HIPped, and extruded 5083 Al (Color figure online).

Table II. The Intensities of Peaks in the XRD Patterns of Gas-Atomized 5083 Al Powders, as-HIPped, and Extruded 5083 Al

	{111}	{200}	{220}	{311}	{222}
Powders	6529	2652	1142	956	287
As-HIPped	6221	2539	1136	1012	294
Extruded	9148	2382	594	1215	656

model proposed that $O_{hkl} = 0$ and 1 correspond to completely random (e.g., powders) and preferential (e.g., a single crystal) orientations, respectively, and accordingly, $0 < O_{hkl} < 1$ represents the presence of $\{hkl\}$ texture to a certain extent. Based on the aforementioned discussion, the calculated degrees of orientation in Table III suggest that the as-HIPped 5083 Al are randomly oriented and no textures are present, and that only {111} texture was developed in the extruded 5083 Al, which is consistent with the previously reported texture in the UFG Al-7.5 wt pct Mg synthesized by HIPping of mechanically milled powders followed by hot extrusion.^[49] In Reference 49, it is observed that two spots in the {111} ring of the selected area electron diffraction pattern, which represent the crystallographic direction parallel to the extrusion direction, exhibit much higher intensity than that of other locations in the ring, suggesting a $\langle 111 \rangle$ fiber texture parallel to the extrusion direction; in contrast, other rings in the diffraction pattern show fairly uniform intensity, indicating that other types of texture were not developed.

Based on XRD peak broadening after subtraction of instrumental broadening, the average dimension of coherent domains ($\bar{d}_{CD}$) and microstrain ($\langle \epsilon^2 \rangle^{1/2}$) can be determined using the following equation^[50]:

$$\frac{\Delta_{\alpha}^2}{\tan^2 \alpha} = \frac{\lambda}{\bar{d}_{CD}} \frac{\Delta_{\alpha}}{\tan \alpha \sin \alpha} + 25 \langle \epsilon^2 \rangle \tag{2}$$

where λ is the X-ray wave-length, α is the angle corresponding to the peak maximum position, and Δ_{α} is the integral breadth corresponding to α after subtraction of instrumental broadening effect. By performing a least-square fit to $\frac{\Delta_{\alpha}^2}{\tan^2 \alpha}$ against $\frac{\Delta_{\alpha}}{\tan \alpha \sin \alpha}$ for all measured peaks, $\bar{d}_{CD}$ and $\langle \epsilon^2 \rangle^{1/2}$ can be obtained. $\langle \epsilon^2 \rangle^{1/2}$ can be used to evaluate the dislocation density ρ by the following equation^[51,52]:

$$\rho = \frac{2\sqrt{3} \langle \epsilon^2 \rangle^{1/2}}{b \bar{d}_{CD}} \tag{3}$$

where b is Burgers vector. Using Eq. [3], the calculated dislocation densities are $\sim 2.4 \times 10^{14}$ and $\sim 7.7 \times 10^{13} \text{ m}^{-2}$, corresponding to the as-HIPped and the extruded 5083 Al, respectively.

Table III. The Calculated Degrees of Orientation in the as-HIPped and Extruded 5083 Al

	{111} + {222}	{200}	{220}	{311}
As-HIPped	~0	~0	~0	~0
Extruded	~0.27	~0	~0	~0

E. APT Results

Figure 8(a) shows a typical 3D APT reconstruction for the 5083 Al extruded at 673 K (400 °C) with atoms of all elements present in the material displayed. Careful inspection reveals the presence of a GB in the reconstructed volume, which is clearly delineated by the 6 at. pct Mg isoconcentration surface (Figure 8(b)) due to strong Mg segregation at the GB. A selection box with the Z-axis parallel to the GB plane normal was implemented to measure the concentrations of Al and various solutes across the GB, and the concentration profiles are shown in Figures 8(c) and (d). In addition, the solute concentrations at GBs and at GIs were measured using rectangular or cylindrical selection boxes, whose sizes were selected such that only the matrix solid solutions were included within the analysis boxes. The measurements of solute concentrations were performed for multiple analysis volumes corresponding to multiple APT datasets. The measurements revealed the GB segregation layers of 5 to 10 nm in thickness. The averaged concentration values from the multiple APT datasets for GIs and GBs are reported in Table IV. It is clearly shown that, for all solutes, the concentrations at GBs are higher than those at GIs, indicative of

solute segregation at GBs. However, it is noted that, despite solute segregation at GBs, the absolute values of solute concentrations at GBs are still very low except for that of Mg as a result of very low overall contents of these elements (Table I). Accordingly, GB segregation of these elements is not evident from the atom maps, and the GBs cannot be delineated by isoconcentration surfaces of elements other than Mg. As discussed in Section III-C, 5083 Al may undergo an equilibrium process during hot extrusion. Hence, GB segregation occurring during hot extrusion can be considered to be of an equilibrium nature, which is primarily related to temperature.^[53] Based on this discussion, it is considered appropriate to assume that the annealed and the extruded 5083 Al exhibit the same general solute segregation at GBs.

Inspection of the published literature reveals that the solutes segregated at GBs generally reduce the GB energy.^[54,55] Based on the changes of enthalpy and configurational entropy during grain coarsening, Kirchheim^[56] developed a formula that describes the GB energy per unit area (γ) as a function of GB segregation of the solute in an ideal dilute binary solution. Given the negligible interactions between different solute elements in an ideal dilute solution containing multiple

Fig. 8—The results of APT analysis of the extruded 5083 Al: (a) a typical tomographic 3D reconstruction (9 M atoms in reconstruction) with atoms of all elements displayed in different colors, where a GB is included, (b) Mg atoms in the reconstruction with 6 at. pct Mg isoconcentration surface superimposed, which delineates the GB, (c, d) Al and solutes' concentration profiles across the GB in (a), measured using a selection box with the Z-axis parallel to the GB plane normal (Color figure online).

Table IV. The Concentrations of Al and Solutes at GBs and GIs Measured by APT and the Calculated Solute Excess at GBs

Element	Concentration at GBs (At. Pct)	Concentration at GIs (At. Pct)	Solute Excess (mol/m ²)
Al	94.1142 ± 0.2657	96.4831 ± 0.1083	—
Mg	5.3283 ± 0.0347	3.1550 ± 0.0140	4.43 × 10 ⁻⁶
Mn	0.1150 ± 0.0077	0.0408 ± 0.0018	1.48 × 10 ⁻⁷
Cr	0.0293 ± 0.0034	0.0177 ± 0.0010	2.37 × 10 ⁻⁸
Fe	0.1054 ± 0.0062	0.0699 ± 0.0019	7.33 × 10 ⁻⁸
Ni	0.0266 ± 0.0032	0.0138 ± 0.0009	2.59 × 10 ⁻⁸
Si	0.0702 ± 0.0051	0.0651 ± 0.0020	1.32 × 10 ⁻⁸
Cu	0.0440 ± 0.0040	0.0265 ± 0.0013	3.58 × 10 ⁻⁸
C	0.0334 ± 0.0036	0.0273 ± 0.0014	1.33 × 10 ⁻⁸
O	0.1110 ± 0.0088	0.0843 ± 0.0038	5.67 × 10 ⁻⁸
N	0.0226 ± 0.0030	0.0165 ± 0.0010	1.28 × 10 ⁻⁸

Table V. The Calculated Enthalpy of Segregation at GBs ($H_{\text{seg},i}$) for the Constituent and Impurity Elements in 5083 Al Used in the Present Study (kJ mol⁻¹)

Mg	Mn	Cr	Fe	Si	Cu	Ni	C	O	N
34.7	52.0	25.1	37.6	36.6	20.2	89.6	27.0	71.3	70.3

components, using the same arguments as those that Kirchheim used for solute atoms of each element, an equation that correlates γ with GB segregation of multiple elements can be derived as follows:

$$\gamma = \gamma_0 - \sum_i^m \Gamma_i (RT \ln c_i^{\text{GI}} + H_{\text{seg},i}) \quad [4]$$

where γ_0 is the GB energy of pure solvent per unit area, m is the number of the solute type, Γ_i is the excess of solute i at GBs, c_i^{GI} is the concentration of solute i at GIs, $H_{\text{seg},i}$ is the enthalpy of segregation of solute i , R is universal gas constant, and T is the temperature. By selecting a symmetrical tilt $\Sigma 5$ (012) [100] GB to represent the general HAGBs in the present study, $H_{\text{seg},i}$ is determined *via* calculations based on first-principles simulations, as shown in Table V; the details can be found in Appendix. Based on the measured solute concentrations at GBs and at GIs, Γ_i can be assessed by the following equation^[56]:

$$\Gamma_i = \frac{N^{\text{GB}}}{N_A} \left(c_i^{\text{GB}} - c_{\text{sv}}^{\text{GB}} \frac{c_i^{\text{GI}}}{c_{\text{sv}}^{\text{GI}}} \right) \chi \quad [5]$$

where N^{GB} is the total atomic number per unit volume at GBs, N_A is the Avogadro's constant, c_i^{GB} is the concentration of solute i at GBs, $c_{\text{sv}}^{\text{GB}}$ and $c_{\text{sv}}^{\text{GI}}$ are the solvent concentrations at GBs and GIs, respectively, and χ is the GB width (taking $\chi = 2$ nm herein^[57,58]). The calculated values of the solute excess are shown in Table IV.

Using the modified Read-Shockley expression, which is valid for both low- and high-angle GBs,^[59] γ_0 can be calculated by the equation:

$$\gamma_0 = \sin \theta [E_c - E_s \ln(\sin \theta)] / b, \quad [6]$$

where θ is the GB misorientation (θ is taken as 36.9° for the symmetrical tilt $\Sigma 5$ (012) [100] GB^[64] herein), and E_c and E_s are dislocation-core and strain-field energies. As

a result of the unavailability of the data corresponding to E_c/b and E_s/b for pure Al to calculate γ_0 using Eq. [6], we rely on those for metal Pd that can be found in the published studies^[60]: $E_c/b = 1.01$ Jm⁻² and $E_s/b = 0.70$ Jm⁻² at 0 K. With E_c/b and E_s/b being proportional to Gb (where G is the shear modulus),^[61] the values of E_c/b and E_s/b for pure Al at 673 K (400 °C) can be calculated as $E_c/b = 0.28$ Jm⁻² and $E_s/b = 0.20$ Jm⁻², on the basis of the following data^[62]: $G = 72.5$ GPa and $b = 0.275$ nm for Pd at 0 K, and $G = 19.5$ GPa ($G = 30.22 - 0.016T$ (GPa)^[63]) and $b = 0.286$ nm for pure Al at 673 K (400 °C). The GB energy per unit area for pure Al is then calculated to be $\gamma_0 = 0.229$ Jm⁻².

Given the relatively low solute concentrations of constituent and impurity elements in the 5083 Al studied (as shown in Table I), it may be reasonable to assume the solution of the constituent and impurity elements in the matrix Al to be an ideal dilute solution. Based on the assumption, γ in the 5083 Al can be evaluated using Eq. [4]. The calculated result indicates that GB segregation reduces the GB energy per unit area at 400 °C from $\gamma_0 = 0.229$ Jm⁻² for pure Al to $\gamma = 0.144$ Jm⁻² for 5083 Al in the present study.

Based on the measured solute concentrations of Mn, Cr, Fe, Si, and O at GBs (the average thickness of the segregation layers is taken as 7.5 nm as estimated by averaging 5 to 10 nm of the segregation layer thickness) and at GIs (average grain size $\bar{d} = 647$ nm), as well as the overall contents of these elements in the 5083 Al (Table I), the amount of these elements in precipitates can be evaluated, and accordingly the volume fractions of Al₁₂Mg₂(CrMnFe), Al₁₂(FeMn)₃Si, Al₆(CrMnFe), and MgO can be estimated as 1.45, 0.84, 0.65, and 0.74 pct, respectively. Thus, the total volume fraction of the second-phase particles is $f = 3.68$ pct in the resultant extruded 5083 Al. Based on the discussion in Sections III-B and III-C, the total volume fraction of second-phase particles remained essentially unchanged during the annealing and extrusion processes.

IV. DISCUSSION

A. Stress-Induced Grain Growth During Hot Extrusion

As presented in Section III-B, very limited grain growth occurred during annealing of the as-HIPed 5083 Al at 673 K (400 °C) for 5 hours. This observation suggests that, in the absence of an applied stress, the two possible mechanisms responsible for grain growth, *i.e.*, GB migration and grain rotation, as discussed in Section I, were almost inhibited by second-phase particles and GB segregation, which is described in detail as follows. First, solute segregation at GBs reduced the GB energy per unit area and thus the driving force for grain growth provided by GB curvature. Second, the thermal stability of second-phase particles, in combination with solute segregation at GBs, promoted the retention of high resistance forces to GB migration and grain rotation. Both of these factors facilitated the inhibition of grain growth. In contrast, significant grain growth was observed during extrusion of the as-HIPed 5083 Al at 673 K (400 °C) (the extrusion process lasted for only ~36 seconds as mentioned in Section II) plus preceding holding at 673 K (400 °C) for 1 hour. Comparing the processing conditions of annealing and extrusion (*i.e.*, much longer time of exposure to 673 K (400 °C) for the former than that for the latter, which facilitates thermally activated grain growth for the former) and the grain sizes after annealing and extrusion, it then follows that grain growth during hot extrusion is predominantly influenced by the type and magnitude of externally applied stresses. Although the thermal stability of second-phase particles in combination with solute segregation at GBs tended to maintain high dragging stresses and solute segregation at GBs lowered the driving force for grain growth, our results suggest that the externally applied stresses overcame the resistance forces generated by second-phase particles and GB segregation, enabling GB migration and grain rotation. The mechanisms underlying stress-induced grain growth during extrusion are discussed in the section that follows.

B. Mechanisms Underlying Stress-Induced Grain Growth During Hot Extrusion

As discussed in Section I, two mechanisms, *i.e.*, GB migration and grain rotation (the latter being caused by GB translation or sliding for the curved GBs^[22,31]), are responsible for grain growth. It has been well documented^[1,2,64,65] that GB migration is a ubiquitous mechanism underlying grain growth. Recently, the contribution of grain rotation and subsequent grain coalescence to grain growth has been extensively reported for NS materials.^[3–5,66] Whether grain rotation was operative or not in the UFG 5083 Al in the present study will be analyzed and discussed as follows. In addition, grain growth involves HAGB motion and consequently, the following three processes may affect grain growth: (1) discontinuous dynamic recrystallization (dDRX),^[64] operated by nucleation of new recrystallized grains and migration of new HAGBs of these

grains, consuming the surrounding deformed microstructures, (2) geometric dynamic recrystallization (gDRX),^[67–69] characterized by relatively localized movement of the existent GBs that have become corrugated, giving rise to impingement of these GBs and production of new equiaxed grains, and (3) transformation from low-angle SGBs to HAGBs. Inspection of published scientific literature^[64,70] reveals that the mechanism of transformation from low-angle SGBs to HAGBs generally plays a very limited role during microstructural evolution in most Al alloys subjected to plastic deformation, especially during high-temperature deformation, where the rate of dislocation accumulation is minimized due to the enhanced rate of dislocation annihilation by the elevated temperatures. Actually, during hot extrusion in the present study, the dislocation density decreased from $2.4 \times 10^{14} \text{ m}^{-2}$ in the beginning (in the as-HIPed 5083 Al) to $7.7 \times 10^{13} \text{ m}^{-2}$ at the end of extrusion. The likelihood for dDRX and gDRX to operate in the present study will be also analyzed and discussed as follows.

The possibility that dDRX occurred during hot extrusion is first examined based on the analysis of crystallographic texture. The results of crystallographic texture analysis presented in Section III-D indicate that the strong $\langle 111 \rangle$ fiber texture was developed while the $\langle 100 \rangle$ fiber texture was undetectable following hot extrusion of the almost randomly oriented as-HIPed 5083 Al. Inspection of the published literature indicates that, for Al alloys (*e.g.*, Al-Mg alloys) of high stacking fault energy (SFE) subjected to uniaxial deformation, such as extrusion, the $\langle 111 \rangle$ fiber texture component is predominant in the deformed state,^[71,72] and an appreciable concentration of the $\langle 100 \rangle$ fiber texture component can always be observed in the Al-Mg alloys that undergo dDRX.^[72–75] Based on the preceding discussion, it is suggested that, in the present study, the presence of the $\langle 111 \rangle$ fiber texture component represents the occurrence of dislocation slip that provides a mechanism to accommodate the plastic strain during extrusion, and the absence of the $\langle 100 \rangle$ fiber texture component indicates inhibition of dDRX during hot extrusion. Inspection of the published literature reveals that, in NS and UFG materials, when the HAGB area fraction exceeds 70 pct, the HAGBs form a stable network that can prevent the occurrence of dDRX.^[76] In related work, Humphreys suggested that, in Al alloys, more than 64 pct of the HAGB area fraction can completely inhibit dDRX by stabilizing the deformed microstructure against heterogeneous microstructural evolution (*e.g.*, dDRX^[67,69]). Suppression of dDRX in the present study can be presumably attributed to more than 70 pct of the HAGB area fraction throughout hot extrusion.

The possibility that gDRX occurred during hot extrusion is then analyzed as follows. References 68 and 77 describe the fundamental mechanisms that underlie gDRX of an Al alloy. During high-temperature deformation, the HAGB spacing decreases along a certain direction; simultaneously, inside the grains subgrains with low-angle boundaries (misorientation <15 deg) are progressively developed *via* DRV due to a

high SFE of the Al alloy that facilitates motion and rearrangement of dislocations by minimizing dissociation of unit dislocations^[64] until a steady state is reached. In the present study (5083 Al deformed at temperature of 673 K (400 °C) with strain rate of 0.8 s⁻¹ as presented in Section II), the strain value corresponding to onset of the steady state is estimated as 0.3 to 0.5.^[78,79] Under steady-state conditions, the subgrain size remains almost unchanged with increasing the magnitude of the strain. When the spacing decreases, the HAGBs always become corrugated due to the surface tension of low-angle SGBs. Pinned by SGBs, the adjacent corrugated HAGBs cannot impinge each other when the HAGB spacing is larger than subgrain size. When the HAGB spacing decreases to a value that is close to the subgrain size, the HAGBs pinch off due to minimized pinning of SGBs and gDRX then occurs, leading to the fragmentation of the original grains and the generation of new equiaxed grains. Based on the preceding discussion, in an effort to determine the possibility for gDRX to occur, the steady-state subgrain size produced by DRV (δ) during hot extrusion in the UFG 5083 Al is required. There are no data available for the subgrain sizes in 5083 Al containing nanoscale second-phase particles (e.g., the average size ~30 nm) that underwent high-temperature deformation. In order to render the problem tractable, we will estimate the subgrain size herein. Inspection of the available scientific literature reveals that, during high-temperature deformation of an Al alloy, the δ value is a sole function of Zener-Hollomon parameter (Z) by the following empirical correlation^[79,80]:

$$\delta^{-1} = A + B \ln Z \quad [7]$$

where A and B are the material constants, and Z is the combination of strain rate ($\dot{\epsilon}$) and the deformation temperature (T) as defined by^[81]:

$$Z = \dot{\epsilon} \exp(Q/RT) \quad [8]$$

where Q is the apparent activation energy of high-temperature deformation. The values of Q , A , and B of the UFG 5083 Al in the present study will be determined *via* extrapolation based on the published data, which is presented as follows.

Relative to pure Al, the Q value of an Al alloy is usually higher as a result of the retardation of DRV by solutes and second-phase particles.^[79,82] For 5083 Al where most Mg is dissolved in the Al matrix and other alloying elements than Mg, including Mn, Cr, Fe, *etc.*, are precipitated in the form of second-phase particles,^[25,38] an increase in Q is primarily attributable to the combined contributions of both the Mg solute and the second-phase particles, which are determined as follows. For a single-phase Al-Mg binary alloy, the increase in Q represents the influence of the solute Mg concentration. Based on available data in published studies,^[83–88] the increase in Q of a series of single-phase Al-Mg alloys, where Mg is the predominant solute, relative to that of pure Al as a function of Mg concentration is plotted in Figure 9(a), with a linear fitting coefficient in excess of 0.99. For an Al-Mg alloy that contains Mg as the

predominant solute and the second-phase particles, the contribution of the second-phase particles to the increase in Q can be obtained by subtraction of the contribution of the solute Mg from the total increased amount. Moreover, inspection of the published literature^[64] reveals that the influence of the second-phase particles on DRV and thereby Q can be reflected by a parameter, namely “particle dispersion level” defined as $f/\bar{l}$. Based on available data in the published studies,^[68,79,86,89,90] the increase in Q of Al alloys, where Mg is the predominant solute, due to the presence of second-phase particles as a function of $f/\bar{l}$ is demonstrated in Figure 9(b), where the linear fitting coefficient is ~0.99. In terms of Figures 9(a) and (b), the Q value of 5083 Al in the present study can be estimated as 193 kJ/mol *via* extrapolation (solute Mg concentration 2.93 wt pct in terms of APT measurement in Section III–E, and $f/\bar{l} = 1.23 \mu\text{m}^{-1}$ based on the values of $f = 3.68$ vol pct and $\bar{l} = 30$ nm as presented in Sections III–E and III–A, respectively).

Inspection of the published literature reveals that, during high-temperature deformation, the δ value of a single-phase Al alloy is primarily determined by deformation conditions (*i.e.*, Zener-Hollomon parameter) and insensitive to the particular alloy compositions^[68,91],

Fig. 9—(a) the increase in Q as a function of Mg solute concentration in single-phase Al-Mg alloys, and (b) the increase in Q as a function of “particle dispersion level” ($f/\bar{l}$) in Al alloys.

however, the δ value can be effectively reduced in an Al alloy containing the dispersion of fine second-phase particles.^[68] Based on the discussion, it can be presumably suggested that the presence of second-phase particles changes the values of A and B in Eq. [7]. Given that the influence of second-phase particles on DRV and thereby on δ can be represented by $f/\bar{l}$ as discussed above, A and B can be considered as a function of $f/\bar{l}$. Based on the data of δ and Q available in the published literature,^[68,77,78,90] δ as a function of $\ln Z$ under a series of $f/\bar{l}$ values is plotted and fitted based on Eq. [7] (*i.e.*, linear fitting) in Figure 10(a), where the plots for a series of $f/\bar{l}$ exhibit an excellent linear fitting coefficient larger than 0.92. Then, the A and B values obtained based on the fitted lines in Figure 10(a) as a function of $f/\bar{l}$ are demonstrated and linearly fitted in Figure 10(b) with the linear fitting coefficients of 0.91 and 0.90 for A and B , respectively. Based on the fitted lines in Figure 10(b), the A and B values corresponding to the 5083 Al in the present study are determined as $A = -7.07273 \mu\text{m}^{-1}$ and $B = 0.35857 \mu\text{m}^{-1}$.

Based on $Q = 193 \text{ kJ/mol}$, $A = -7.07273 \mu\text{m}^{-1}$, and $B = 0.35857 \mu\text{m}^{-1}$ as determined above, as well as the processing parameters $\dot{\epsilon} = 0.8 \text{ s}^{-1}$ and $T = 673 \text{ K}$, the δ value of the 5083 Al in the present study is estimated as 157 nm. We then divide the grains in the as-HIPped 5083 Al into two groups: <157 nm, and 157 to 450 nm (very few grains >450 nm as shown in Figure 1(c)). Purely from a geometrical perspective, the GB spacing of the grains of 157 to 450 nm can be reduced to less than $\delta = 157 \text{ nm}$ during extrusion (the reduction ratio of ~ 3.2 in the directions perpendicular to the extrusion direction). However, the GB spacing was simultaneously increased by GB migration. Hence, the actual status for the change of GB spacing, *i.e.*, the likelihood for gDRX to be operative, is determined by the relative magnitude of the rate of lowering GB spacing by extrusion and the rate of widening GB spacing by GB migration. For grains <157 nm (less than δ), our results suggest that it is unlikely that subgrains are produced inside 5083 Al grains during extrusion. As a consequence of the absence of SGBs inside grains, the pinning pressure to local movement and impingement of HAGBs should be minimized. In a more recent MD investigation of plastic deformation of NS Al at high temperature,^[17] it was demonstrated that, during compression to the strain of 21 pct, the one serrated GB segment of a grain that contains no subgrains meets another GB segment on the opposite side of the grain, triggering gDRX. In the present study, however, the possibility for gDRX to be active in the grains <157 nm is still determined by the relative magnitude of the rate of reducing GB spacing by extrusion and the rate of increasing GB spacing by GB migration, as discussed above.

Next, the possibility for grain rotation to occur during hot extrusion is examined as follows. Generally, two mechanisms are responsible for the formation of a subgrain structure during plastic deformation: (1) DRV and (2) rotation and incomplete coalescence of grains with HAGBs, *i.e.*, leaving low-angle SGBs between the prior grains with HAGBs. In the extruded 5083 Al,

Fig. 10—(a) δ^{-1} as a function of $\ln Z$ in a series of Al alloys with different $f/\bar{l}$, where the data points correspond to the line with the same colors, and (b) A and B as a function of $f/\bar{l}$, where the red and black data points represent values for A and B , respectively (Color figure online).

subgrains of ~ 450 to $\sim 1000 \text{ nm}$ in size have been observed (Figure 5(a)), which are much larger than those formed *via* DRV ($\delta = 157 \text{ nm}$). It is then argued that such a grain comprising subgrains of ~ 450 to $\sim 1000 \text{ nm}$ in size may originate from rotation and incomplete coalescence of a few prior grains that were initially separated by HAGBs. These grains with HAGBs come from either the original grains in the as-HIPped 5083 Al or the grains after GB migration of the original grains. For these grains, plastic deformation was primarily accommodated by grain sliding and rotation, whereas intragranular dislocation activity may have been weakened and consequently, the magnitude of strain used to induce intragranular dislocation activity is insufficient to develop subgrain structure inside these grains. It is noted that the dark grain in Figure 6 is characterized by the absence of subgrain structure at the entire GI. One of the possible mechanisms responsible for the formation of such a grain may be the rotation and complete coalescence of the prior grains with HAGBs.

The microstructural evolution inside the grain as shown in Figure 4(a) may be primarily attributable to GB migration. As a site for relaxation of dislocation strain energy, GBs behave as sinks for dislocation

absorption.^[92,93] When GB migration occurred, transgranular movement of HAGBs induced an extensive interaction between these HAGBs and the dislocations, leading to a significant decrease in dislocation density; however, subsequent straining would increase the dislocation density again. The regions that were swept by HAGBs in the earlier stage may undergo a magnitude of subsequent strain, which is higher than the critical strain corresponding to the onset of the steady state, leading to the formation of well-defined subgrain structure *via* DRV in the regions as marked by “SG” in Figure 4(a). The sizes of these subgrains fall in the range from 100 to 300 nm as analyzed in Section III–C, similar to the subgrain size produced by DRV (*i.e.*, $\delta = 157$ nm). In contrast, the regions that were swept by HAGBs in the later stage may be subjected to a relatively low magnitude of subsequent strain, which is insufficiently high to develop subgrain structure in the regions as marked by “D” in Figure 4(a).

On the basis of the aforementioned discussion, the mechanisms underlying the microstructural evolution associated with HAGB motion during hot extrusion include GB migration, grain rotation, and possible gDRX. GB migration and grain rotation were responsible for grain growth, while gDRX led to possible grain refinement. The final grain size in the extruded 5083 Al indicates that GB migration and grain rotation played a dominant role, leading to overall grain growth. Despite the strong resistance forces generated by the second-phase particles and the solutes segregated at GBs, which even almost inhibited GB migration and grain rotation during annealing at 673 K (400 °C) as reported in Section III–B, the introduction of externally applied stresses during hot extrusion may overcome these resistance forces, enabling the operation of GB migration and grain rotation. Given that both stress-induced GB migration and grain rotation were operative during extrusion, it is difficult to reveal the effect of second-phase particles and solute segregation at GBs on the two mechanisms separately *via* experiments. In order to solve this problem, we have formulated a theoretical framework that can be implemented to quantify the effect of second-phase particles and solute segregation at GBs on the two mechanisms. This analytical model is described in detail elsewhere.^[94]

V. CONCLUSIONS

1. The results in the present study indicate that, in an UFG 5083Al containing second-phase particles and solutes segregated at GBs, grain growth was essentially inhibited during annealing at 673 K (400 °C) for a prolonged period of 5 hours in absence of an externally applied stress; in contrast, grain growth by a factor of ~ 2.7 occurred during extrusion at 673 K (400 °C). The significant grain growth during hot extrusion can be attributed to the externally applied stresses coming from the state of stress imposed during extrusion.
2. The mechanisms underlying the microstructural evolution related to HAGBs during hot extrusion

involved GB migration, grain rotation, and possible gDRX while dDRX did not appear to occur. GB migration and grain rotation contributed to stress-induced grain growth, and simultaneously possible gDRX led to grain refinement. Our results suggests that GB migration and grain rotation dominate, resulting in overall grain growth. Although the strong resistance forces were generated by the second-phase particles and the solutes segregated at GBs, the exertion of externally applied stresses can overcome these resistance forces, enabling the operation of GB migration and grain rotation.

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APPENDIX

The calculation of enthalpy of segregation using first-principles simulations

Enthalpy of segregation ($H_{\text{seg},i}$) is defined as the difference in the total enthalpies of a system with solute atoms of a specific element i at GIs (H_i^{GI}) and at GBs (H_i^{GB}), *i.e.*, $H_{\text{seg},i} = H_i^{\text{GI}} - H_i^{\text{GB}}$. Thus, $H_{\text{seg},i}$ can be considered to be temperature independent if neglecting the difference in specific heat between the solute atoms at GIs and at GBs. Given that $H_i^{\text{GI}} = E_i^{\text{GI}} - PV_i^{\text{GI}}$ and $H_i^{\text{GB}} = E_i^{\text{GB}} - PV_i^{\text{GB}}$ (where E_i^{GI} and E_i^{GB} are the total energies of a system with solute atoms of a specific element i at GIs and GBs, respectively, V_i^{GI} and V_i^{GB} are the volumes of a system with solute atoms of a specific element i at GIs and GBs, respectively, and P is the pressure of the system), if one neglects the difference in the volumes between the solute atoms at GIs and at GBs (*i.e.*, $V_i^{\text{GI}} = V_i^{\text{GB}}$), then $E_{\text{seg},i} = E_i^{\text{GI}} - E_i^{\text{GB}} = H_{\text{seg},i}$, where $E_{\text{seg},i}$ is defined as the energy of segregation. In the present study, $H_{\text{seg},i}$ is determined by calculating $E_{\text{seg},i}$ at 0 K *via* first-principles simulations. The details of calculations are described as follows.

As a typical HAGB in Al, symmetrical tilt $\Sigma 5$ (012) [100] GB has been extensively studied by experimental^[95,96] and theoretical^[97,98] investigations. Hence, it was selected to represent the general HAGBs in the present study. The GB structure was simulated using a supercell that was repeated periodically in the three-dimensions to reproduce the entire crystal lattice studied

Fig. A1—The 40-atom supercell that contains a symmetrical tilt $\Sigma 5$ (012) [100] GB, together with the crystallography in the supercell, where the GB is marked by a solid line and the atoms located in different planes along [100] direction are denoted by balls with different colors (Color figure online).

(namely “periodical boundary conditions”). Figure A1 shows the supercell of 40 atoms used in the present study, as well as the crystallography involved. The supercell includes two identical grains, and each of the two grains contains eleven (012) atomic layers with one layer shared by the two grains, which represents the GB as indicated by the solid line in Figure A1. When the total energies of the supercell with solute atoms at the GIs and at the GBs were calculated to acquire the energy of segregation, the solute atoms replaced the positions of Al atoms in the lattice at the GIs and at the GB for Mg, Mn, Cr, Fe, Ni, Si, and Cu, but were inserted in the interstitial sites of the Al lattice for C, O, and N.

The first-principles simulations were conducted within the framework of density functional theory (DFT), as implemented in the CASTEP code (Cambridge Serial Total Energy Package).^[99] The ion-electron interactions were modeled by the ultrasoft pseudopotential method.^[100] The exchange and correlation interaction was described by the generalized gradient approximation (GGA) with the Perdew-Burke-Ernzerhof (PBE) parameterization.^[101,102] Plane wave cutoff energies of 310, 250, 290, 280, 270, 180, 270, 240, 300, and 220 eV were set for Mg, Mn, Cr, Fe, Ni, Si, Cu, C, O, and N, respectively. The Brillouin zone integrations were performed using a Monkhorst-Pack k -point,^[103] and the k -point mesh was sampled by $5 \times 2 \times 1$ for the supercell used herein. The optimized lattice constant for bulk Al at 0 K (4.05 Å) is well consistent with the experimental data at 20 K (4.03 Å)^[104] and other simulated result (4.05 Å).^[97] The total energy was converged numerically to 2×10^{-6} eV/atom with respect to electronic, ionic, and unit cell degrees of freedom.

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