Microstructural Evolution in Pure Aluminum in the Early Stages of Processing by High-Pressure Torsion

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Measurements were taken to evaluate the evolution of homogeneity in disks of high-purity aluminum in the early stages of processing by high-pressure torsion. The results demonstrate that samples processed through 1/4 or more whole revolutions exhibit microhardness values which are generally higher in the centers of the disks than at the edges whereas after 1/8 turn the hardness is higher at the edge than in the center. It is shown that all of the hardness measurements are mutually consistent and they scatter around a unique curve when plotted against the equivalent strain. The measurements of hardness are supplemented by microstructural observations which provide evidence of a gradual evolution in the microstructure with increasing strain from an initial formation of subgrains to an array of ultrafine grains separated by boundaries having high angles of misorientation. [doi:10.2320/matertrans.MB200910]

(Received August 3, 2009; Accepted October 2, 2009; Published November 26, 2009)

Keywords: hardness, high-pressure torsion, homogeneity, severe plastic deformation, ultrafine grains

1. Introduction

Processing through the application of severe plastic deformation is now an accepted procedure for producing polycrystalline metals with exceptionally small grain sizes.¹ An example of this approach is high-pressure torsion (HPT) where a sample, generally in the form of a thin disk, is subjected to an applied pressure and concurrent torsional straining.² An important advantage of HPT is that it is capable of introducing exceptional grain refinement and producing materials with grain sizes in the lower submicrometer range or even in the nanometer range.

When a disk is processed by HPT, the equivalent strain imposed by torsional straining, \( \varepsilon_{eq} \), varies across the disk through a simple relationship of the form³

\[
\varepsilon_{eq} = \frac{r \theta}{\sqrt{3} h}
\]

where \( r \) is the distance from the center of the disk, \( \theta \) is the total rotational angle imposed on the disk in radians and \( h \) is the thickness of the disk. It follows from eq. (1) that the strain is a maximum at the periphery of the disk and reduces to zero at the disk center. This means that, in principle at least, it is reasonable to anticipate that the strain imposed in processing by HPT is very inhomogeneous with little or no strain introduced in the central region.

Early experiments on polycrystalline Ni confirmed that there were higher values of the microhardness, indicative of larger strains and more refined microstructures, around the edges of the HPT disks but with lower microhardness values in the disk centers. However, detailed experiments showed that the hardness values across the disks became more homogeneous when the pressure imposed during torsional straining was increased.⁴ The gradual evolution towards a more homogeneous structure in disks of Ni was later demonstrated by constructing plots of the hardness values over the disk surfaces after processing through different numbers of torsional revolutions and different applied pressures and these plots demonstrated greater homogeneity at larger numbers of revolutions and higher applied pressures.⁵ To date, several reports are now available on the variations of hardness across HPT disks for a range of metals including Al and aluminum alloys,⁶-⁸ Cu,⁹-¹¹ Ni¹² and steel.¹³

It is possible to achieve an excellent pictorial representation of the hardness distributions introduced in HPT by taking large numbers of individual hardness measurements following rectilinear grid patterns on the disk surfaces and then plotting these data in the form of color-coded contour maps. This approach was first introduced for high-purity Al¹⁴ and subsequently the same approach was applied to an aluminum Al-6061 alloy,¹⁵ Cu and Cu-Zn alloys¹⁶ and high-purity Al processed by HPT using strain reversals.¹⁷

The results from these experiments revealed a clear dichotomy between different materials. First, and as anticipated from application of eq. (1), the hardness values tended to be higher at the disk peripheries and lower at the disk centers in the Al-6061 alloy¹⁵ and in pure Cu and the Cu-Zn alloys.¹⁶ Second, and contrary to eq. (1), the hardness values in high-purity Al were initially higher in the centers of the disks and lower at the edges.¹⁴ However, both sets of materials showed a gradual evolution towards a reasonably homogeneous distribution of hardness values when the HPT processing was continued through larger numbers of turns.

A significant limitation of the earlier work on high-purity Al was that the hardness distributions were recorded only after processing from 1 to 5 turns using applied pressures from 1.25 to 6.0 GPa.¹⁴ No measurements were taken when disks were rotated through fractional numbers of whole revolutions although there are some recent reports of hard-
ness measurements taken after less than one whole revolution in Al\(^{18-20}\) and Ti\(^{21}\). Accordingly, the present investigation was initiated to extend the earlier work on high-purity Al to cover samples tested through fractional numbers of turns.

2. Experimental Material and Procedures

The experiments were conducted using polycrystalline aluminum of 99.99% purity which matches the material used in the earlier investigation\(^{14}\). Full details of the preparation of the material were given earlier\(^{4}\) but briefly an aluminum ingot was rolled, swaged into 10 mm diameter rods, sliced into disks having thicknesses of \(~0.85\) mm and the disks were annealed at 773 K for 1 h to give an initial grain size of \(~1\) mm. Disks with the same size and the same purity were also prepared from extruded rods and were annealed at 773 K for 1 h to produce an initial grain size of \(~250\) µm. The latter disks were especially used for electron back-scatter diffraction (EBSD) analysis after processing by HPT.

The processing by HPT was conducted at room temperature \((~298\) K) under an applied pressure, \(P\), of 1.25 GPa with a constant rotational speed of 1 rpm using a quasi-constrained processing condition\(^{8}\) and the processing facility described earlier.\(^{14}\) Because of the low pressure used in these experiments and the requirement to rotate through relatively small fractions of whole revolutions, no lubricant was applied around the edges of the samples prior to HPT\(^{22}\) in order to reduce any slippage between the sample and the die. As demonstrated in earlier experiments,\(^{22}\) very little slippage generally occurs in aluminum when processing by HPT. Measurements showed a small reduction in the thickness of each disk during processing; typically, the thicknesses of the disks were reduced from \(~0.85\) to \(~0.76\) mm after processing through one whole revolution. The HPT was conducted by applying the pressure of 1.25 GPa, carefully straining through selected amounts corresponding to 1/8, 1/4, 1/2, 3/4 and 7/8 of a whole revolution, respectively, and then discontinuing the rotation, removing the applied pressure and taking the disk from the HPT facility.

Following HPT, the disks were mounted and polished mechanically using polishing papers with 600, 800 and 1200 grits and then polishing on cloth to a mirror-like finish. All of the microhardness measurements were taken using an FM-1e microhardness tester equipped with a Vickers indenter. In order to ensure consistency with the earlier experiments, two different procedures were used for taking measurements of the Vickers microhardness, \(H_v\). First, individual microhardness values were recorded across the diameter of each disk in incremental steps of 0.3 mm up to a distance of 1.2 mm on either side of the center of each disk and then by spacings of 0.6 mm at greater distances from the center, where each separate value is the average of four measurements uniformly positioned around the selected point at a distance of 0.15 mm. Special attention was directed to the central region of each disk because of the uncertainties associated with the validity of eq. (1) in this region and the special need to ensure a very high degree of accuracy in this central area. Second, individual measurements of \(H_v\) were recorded on the surface of each disk following a regular rectilinear grid pattern with spacings of 0.3 mm between every separate point. These latter measurements provided a complete set of datum points covering each disk and the points were then plotted in the form of color-coded contour maps where separate colors denote different hardnesses and the maps give a pictorial representation of the variations in local hardness over the entire surface of each sample.

Microstructural analysis was conducted after processing by HPT using an Hitachi S-4300SE scanning electron microscope (SEM) equipped with a field emission gun. Electron back-scatter diffraction (EBSD) analysis was performed at an accelerating voltage of 20 kV and the individual crystal orientations were determined using an automatic beam scanning with step sizes of 0.08–0.3 µm. This analytical treatment was used to provide both the sizes of the individual grains and the misorientation angles across the various grain boundaries. All data acquisition and subsequent analysis was performed using TSL orientation image microscopy software (v.3.5) and a cleaning-up procedure was applied to all EBSD images to adjust the points with a confidence index (CI) of less than 0.1. Angular misorientations of less than 2\(^\circ\) were excluded from the analysis because of the limitations inherent in the angular resolution of the EBSD procedure.\(^{23}\)

3. Experimental Results

3.1 Microhardness measurements after HPT

The values of the Vickers microhardness across the diameter of each disk are plotted against the distance from the center in Fig. 1 where the lower solid points correspond to the as-received and unprocessed condition and the remaining points denote torsional straining through total numbers of turns, \(N\), from 1/8 to 1 whole revolution. It is apparent that the hardness increases significantly at the onset of straining. Specifically, after one eighth turn of HPT the hardness increases by a factor which is slightly lower than 2 where this increase is comparable to the increase in strength typically recorded in a range of commercial aluminum-based alloys after processing through 1 pass in ECAP.\(^{24}\)

A critical and important feature after \(N = 1/8\) in Fig. 1 is that, similar to the results reported for several different materials,\(^{4,13,15,16,25,26}\) the measured hardness in the center of the disk is lower than in the surrounding areas. However, this result differs from earlier measurements on aluminum of 99.99% purity where the measured hardness was consistently higher in the centers of the disks after small numbers of whole revolutions.\(^{14,17,26}\) Thus, these measurements provide a direct demonstration of the importance of recording the values of hardness after fractional numbers of turns in order to obtain a complete understanding of the microstructural evolution occurring during processing by HPT.

Figure 1 shows also that the variation in the microhardness values changes after 1/4 turn of HPT where the hardness decreases at the periphery but increases in regions close to the center. Nevertheless, after 1/4 turn the hardness at the central point remains relatively low and it is comparable to the hardness after 1/8 turn. With increasing fractional turns, the peaks in hardness in the central region of the disk build up and after 1/2, 3/4 or 7/8 turns the hardness is higher in the center than at the edges. These results show that at least
1/2 turn is needed with high-purity Al in order to attain the conventional result of an increased hardness at the center and lower values of hardness around the edge.

All of the microhardness measurements taken from the rectilinear grid patterns, with incremental spacings of 0.3 mm, were plotted in the form of color-coded contour maps to provide pictorial images of the distributions of the individual hardness values over the disk surfaces. These plots are shown in Fig. 2 for disks torsionally strained through 1/8, 1/4, 1/2, 3/4 and 7/8 turns where the X and Y axes denote two arbitrarily-selected orthogonal axes marked in mm such that the central position is at (0,0). All measurements of Hv are shown within incremental values of Hv = 5 with a total range in Hv from 30 to 60 and with the scale shown as the inset at the lower right. An earlier report presented similar color-coded representations for the same material processed by HPT at a pressure of 1.25 GPa when torsionally strained through 1, 3 and 5 turns. In the earlier report it was shown that the disks become reasonably homogeneous in hardness values when strained to \( N = 5 \) turns with a hardness of \( \sim 40 \) whereas after \( N = 1 \) turn the hardness in the central region is higher than around the edge of the disk by \( Hv \approx 10 \). The present results are consistent with these general trends for all measurements taken at and above \( N = 1/4 \) turn. However, in the earliest stages of processing, at \( N = 1/8 \) turn, the situation is reversed and the highest hardness values occur around the edge. It is also apparent from Fig. 2 that the total extent of the region of higher hardness decreases with increasing amounts of torsional straining.

3.2 Microstructural evaluation after HPT

Detailed microstructural observations were undertaken and representative examples obtained by EBSD are shown in Fig. 3 where the upper line corresponds to \( N = 1/8 \) turn in the center of the disk, the second line corresponds to \( N = 1/4 \) turn at the edge of the disk, the third line corresponds to \( N = 1/2 \) turn at the edge of the disk and the bottom line corresponds to \( N = 1 \) turn at the edge of the disk: the images in the left column depict the grain configurations and the relative orientations of the individual grains and the images in the right column show the individual grain boundaries within each area where the high-angle boundaries having misorientations \( \geq 15^\circ \) are marked in red and the low-angle boundaries having misorientations from \( 2^\circ \) to \( 15^\circ \) are marked in blue. The grain size is defined for the area surrounded by the former red lines and the subgrain size by the latter blue lines as in the conventional definitions. It should be noted also that the magnifications increase with increasing fractional turns because of the gradual development of an ultrafine-grained structure.
Inspection of Fig. 3(a) shows there is only a relatively small change in the microstructure in the central region of the disk after 1/8 turn. The grain size was measured as \( \sim 250 \mu m \) but subgrains with low-angle boundaries have formed within these larger grains. However, there is a large average spacing between the subgrain boundaries. It should be noted that the presence of these subgrain boundaries and/or the presence of dislocations within the grains leads to the gradual changes in color or color gradients which are clearly visible in the EBSD image. The grain size was also \( \sim 250 \mu m \) at \( N = 1/8 \) turn near the edge of the disk as shown in Fig. 3(b) and again there are subgrains formed with the larger grains. However, in this condition the subgrains were relatively densely developed and the average spacing between the low-angle boundaries was very small.

Close inspection shows there is also some evidence at this early stage of processing for a gradual evolution into high-angle boundaries. An earlier investigation using transmission electron microscopy showed the dislocation density was very high within the subgrains at this early stage of processing.27)

Figures 3(c) and (d) show the edges of the disks after straining through 1/4 and 1 turn, respectively. After 1/4 turn in Fig. 3(c) the grain size is reduced to \( \sim 5 \mu m \), the subgrains are well developed and the fraction of grains having high-angle boundaries has increased. In this condition the microstructure is heterogeneous with some regions having subgrains and a high density of dislocations and other regions having ultrafine grains with relatively few dislocations. Finally, the situation after \( N = 1 \) turn is shown in Fig. 3(d) where the grain size is \( \sim 2 \mu m \) and there are very few subgrains but there is a large fraction of grains having high-angle grain boundaries. For this condition there is no measurable difference between the sizes of the grains and the subgrains, thereby confirming the ultrafine-grained microstructures produced in HPT are due to an evolution in the boundaries from predominantly low-angle misorientations to predominantly high-angle misorientations. This is consistent with recent results for aluminum of the same purity processed by equal-channel angular pressing (ECAP) where the fractions of high-angle boundaries were recorded up to 12 separate passes through the ECAP die.28) In this condition the dislocation density within the individual grains is relatively low27) which leads to the measured decrease in hardness by comparison with the initial stages of HPT.

4. Discussion

The present results provide a very useful addition to the data presented earlier showing the variations of hardness in high-purity Al processed by HPT through 1, 3 and 5 turns under an applied pressure of 1.25 GPa.14) The experimental data presented in Figs. 1 and 2 give additional information for samples of the same material processed through fractional...
numbers of whole turns and this is supplemented by the comprehensive microstructural information obtained by EBSD and shown in Fig. 3.

At first sight, the experimental datum points shown in Fig. 1 appear to scatter randomly with no clear and consistent variation with increasing number of turns and therefore with increasing amounts of applied strain. However, there is a critical difference between processing in ECAP and HPT because in ECAP the strain remains constant throughout the billet whereas in HPT the strain varies across the disk as represented by eq. (1). This means in practice that, although it is meaningful to plot the values of the Vickers microhardness directly against the number of imposed passes in processing by ECAP,29) it is not possible to construct a similar simple plot for samples processed by HPT because of the continuous variations in strain within each sample. This problem was first recognized in very early work where HPT was applied to ring samples and this led to the introduction of the equivalent strain.30) In a comprehensive recent study of the processing of an austenitic steel by HPT, it was shown that a plot of the individual values of the microhardness against equivalent strain gave a unique plot with all datum points falling on or about a single line and with similar results also for copper and chromium.15)

More recently this same approach was used to interpret hardness data obtained in the processing by HPT of both disk and ring samples.27,31–35)

In order to make use of this analytical approach in the present investigation, it is first noted that the thicknesses of the HPT disks are reduced in HPT processing and this reduction generally increases with increasing numbers of turns.25) In this investigation, the sample had an initial thickness of \( \sim 0.85 \text{ mm} \) and this was reduced to \( \sim 0.80 \text{ mm} \) on application of the applied pressure and it was further reduced to \( \sim 0.76 \text{ mm} \) after one turn of HPT. Since the thickness reductions for different fractional revolutions are all within 5% of these values, a thickness of \( h = 0.8 \text{ mm} \) was used to estimate the equivalent strain using eq. (1). The results of these calculations are shown in Fig. 4 where all of the datum points from Fig. 1 are replotted as a function of the estimated equivalent strain.

It is apparent from Fig. 4 that the datum points for different fractional revolutions and for different positions on the HPT disks all tend to lie around a single line showing a peak at an equivalent strain of \( \sim 2.0 \) and at higher strains, above \( \sim 7 \), the hardness values attain a steady-state value which remains essentially constant to even higher strains. This result matches earlier results plotted for aluminum of the same purity where the datum points were extended to strains of \( >40 \) using ring samples.31,34) The scatter in the points visible in Fig. 4 at equivalent strains in the range of \( \sim 2–7 \) is attributed to the inhomogeneous development of the microstructure when processing thin disks by HPT.

It is also instructive to delineate the EBSD images shown in Fig. 3 in terms of the estimated equivalent strain within the disk at the point used for microstructural examination. For Fig. 3(a), the sample was examined near the center of the disk so that the equivalent strain is probably \( <0.3 \) which corresponds to the initial increase in hardness before the maximum. For Fig. 3(b), the equivalent strain near the edge of the disk is \( \sim 2 \) and this corresponds to the hardness maximum. For Fig. 3(c), the equivalent strain near the edge of the disk after 1/4 turn is \( \sim 4 \) which correspond to the region where the hardness decreases prior to attaining a steady-state condition. Finally, for Fig. 3(d) the equivalent strain near the edge of the disk after 1 turn is \( \sim 16 \) which is within the steady state region.

Three conclusions may be drawn from Fig. 4. First, hardness values vary directly with the estimated equivalent strain and this variation is directly comparable to the variation in hardness with the imposed strain when processing by ECAP because earlier measurements on the same material showed a peak in hardness after 2 ECAP passes which, for an ECAP die with a channel angle of 90°, corresponds to an applied strain of \( \sim 2.36) \) Second, the hardness increases at small equivalent strains up to \( \sim 2 \), decreases thereafter, and reaches a steady-state condition at equivalent strains above \( \sim 7 \). These changes are consistent with the very high stacking fault energy in high purity aluminum and the consequent rapid microstructural recovery that was documented in an earlier study.14) Third, the hardness values saturate at equivalent strains above \( \sim 7 \) as revealed in this investigation and in experiments with ring samples.31–35)

5. Summary and Conclusions

(1) Samples of high-purity aluminum were processed by high-pressure torsion under a pressure of 1.25 GPa at room temperature. The torsional straining was terminated after fractional numbers of whole revolutions between 1/8 and 7/8 turns. Microhardness measurements were taken on polished surfaces of the disks and the microstructures were examined using electron back-scatter diffraction.

(2) After 1/4 or more fractional revolutions the values of the microhardness are higher in the centers of the disks than at the edges whereas after 1/8 turn the hardness is higher at the edge than in the center of the disk. All hardness measurements scatter around a unique curve when plotted against the equivalent strain, with the hardness initially increasing up to a strain of \( \sim 2 \), thereafter decreasing gradually to a strain of \( \sim 7 \) and then remaining constant at higher strains.

(3) Microstructural observations demonstrate a gradual evolution of the microstructure with subgrains evolving into an array of ultrafine grains separated by boundaries having high angles of misorientation.

Acknowledgements

We thank Mr. Yuki Ito of Kyushu University for assistance in obtaining the EBSD data. This work was supported in part by a Grant-in-Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science and Technology, Japan, in Priority Areas “Giant Straining Process for Advanced Materials Containing Ultra-High Density Lattice Defects”, in part by Kyushu University Interdisciplinary Programs in Education and Projects in Research Development (P&P) and in part by the National Science Foundation of the United States under Grant No. DMR-0855009.
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