The characteristics of two-phase Al-Cu and Zn-Al alloys processed by high-pressure torsion

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Abstract. Experiments were conducted on two different two-phase alloys, the Al-33% Cu eutectic and the Zn-22% Al eutectoid. These alloys were processed by high-pressure torsion (HPT) and then measurements were taken to determine the distributions of hardness values across the disk diameters and tensile tests were conducted to examine the potential for achieving superplastic elongations. Both alloys showed grain refinement through the HPT processing but the Al-Cu alloy exhibited a conventional work-hardening with torsional straining whereas the Zn-Al alloy exhibited a work-softening due to the loss of Zn-rich precipitates under the high imposed pressure. Excellent superplastic elongations were achieved in both alloys when pulled in tension at elevated temperatures with a maximum elongation of 1800% in the Zn-Al alloy.

1. Introduction
Processing through the application of severe plastic deformation (SPD) provides the opportunity for achieving exceptional grain refinement in bulk solids [1,2]. Typically, metals processed using SPD techniques have grain sizes within the submicrometer range or even in the true nanometer range where the grain size is <100 nm. These grain sizes cannot be attained using conventional thermo-mechanical processing. An important characteristic of SPD processing is that a very high strain is imposed without incurring any significant change in the overall dimensions of the sample. This contrasts with other processing methods, such as rolling and extrusion, where at least one of the dimensions is reduced during the processing operation. Several different SPD processing techniques are now available [3] but most attention has been centered to date on the procedures of equal-channel angular pressing (ECAP) [4] and high-pressure torsion (HPT) [5]. Experiments show that HPT is generally preferable to ECAP because it produces materials with smaller grain sizes and with a higher fraction of grain boundaries having high-angles of misorientation [6-8]. However, there tends to be a disadvantage because the samples are often in the form of thin disks and there have been only limited attempts to extend the process to larger cylindrical samples [9-12]. Due to the very small grain sizes that may be attained in HPT processing, the present experiments were conducted using HPT with two different two-phase alloys.
There are two major advantages in reducing the grain size of polycrystalline materials [13]. First, the Hall-Petch relationship shows that the strength increases when the grain size is reduced [14,15]. Therefore, it is reasonable to anticipate that ultrafine-grained materials will exhibit exceptional strength at ambient and low temperatures. Second, at high temperatures, in the regime where diffusion becomes sufficiently rapid, the fundamental relationship for the flow process shows that the strain rate varies inversely with the grain size raised to a power of $p$ [16] and this provides an opportunity for achieving superplastic ductilities at rapid strain rates in tensile tests conducted at elevated temperatures. Superplasticity is defined formally as the ability of a polycrystalline metal to exhibit an elongation of at least 400% when pulled in tension and with an associated value for the strain rate sensitivity, $m = \frac{\partial \ln \sigma}{\partial \ln \dot{\varepsilon}}$, of ~0.5 where $\sigma$ is the flow stress and $\dot{\varepsilon}$ is the strain rate [17]. The ability of some metals to exhibit superplastic flow forms the basis for the superplastic forming industry in which sheet metals are formed into complex shapes and curved parts for use in a wide range of applications from aerospace to automotive and architectural [18]. For conventional metals not processed by SPD, it is now well established that superplasticity may be achieved in tensile testing at temperatures above ~0.5 $T_m$, where $T_m$ is the absolute melting temperature, provided the grains remain small at values less than ~10 $\mu m$ [19]. Generally, in the superplastic forming industry the sheet metals are processed to give initial grain sizes of ~3-5 $\mu m$. However, the advent of SPD processing provides an opportunity for attaining grain sizes that are about one order of magnitude smaller than in the conventional metals and this suggests these materials may exhibit excellent superplastic properties and, because the strain rate varies inversely with grain size, it is probable that these superplastic elongations will occur at very rapid strain rates [20]. The ability to achieve superplasticity at high strain rates after processing by ECAP was first demonstrated using commercial aluminum-based alloys where the elongations to failure were superplastic and occurred at strain rates at and above $10^{2} \ \text{s}^{-1}$ [21]. These experimental results directly confirmed the occurrence of high strain rate superplasticity which is defined formally as superplastic elongations occurring at strain rates faster than $10^{2} \ \text{s}^{-1}$ [22].

These very early results demonstrating superplasticity in alloys processed by ECAP were subsequently followed by many other investigations where superplasticity was reported after processing by ECAP. A comprehensive summary and tabulation of all of the results available to date was published in 2007 and at that time there were more than sixty different publications describing true superplastic flow after ECAP [23]. However, more recently there are also examples of superplasticity occurring in metals processed by HPT. This type of processing provides a bigger challenge because the samples are generally in the form of thin disks and it is well known that the strain introduced in HPT processing varies across the disk from zero strain at the center to a maximum strain at the edge. Although this suggests the development of gross inhomogeneities in HPT, experiments show there is a gradual evolution towards a reasonable level of homogeneity in most materials processed by HPT [24]. Therefore, to avoid any problems associated with variations in homogeneity in the center of the disks, it has become a standard practice to cut two similar tensile specimens from each disk where these two specimens are cut from off-center positions on either side of the mid-point. An example of this procedure, and the associated measurements in mm, is shown in Fig. 1 where the centers of each tensile specimen lie at a distance of 2 mm from the mid-point of the disk [25]. These types of tensile specimens are relatively easy to obtain using electro-discharge machining and they provide an opportunity to evaluate the ability to achieve superplastic elongations in HPT samples while avoiding the center of the disk.

Two-phase alloys tend to be ideal candidate materials for obtaining superplasticity because the presence of two different phases means that grain growth is restricted and it is usually relatively easy to retain an ultrafine-grained microstructure. Accordingly, the present report brings together results on two different two-phase alloys, the Al-33% Cu eutectic alloy and the Zn-22% Al eutectoid alloy. Both alloys were processed by HPT and this report describes the values of the Vickers microhardness that were attained after HPT processing and the results of tensile testing at elevated temperatures. As will be demonstrated, both alloys exhibit superplastic elongations of >1000%.
2. Experimental materials and procedures
The experimental results described in this report were obtained using an Al-33% Cu eutectic alloy and a Zn-22% Al eutectoid alloy. Full details of these experiments were given in earlier reports for the Al-Cu alloy [26] and the Zn-Al alloy [27-31], respectively.

In brief, the Al-Cu alloy was received as a plate and then machined into disks with 10 mm diameters which were homogenized at a temperature of 673 K for 1 hour [26]. These disks were sliced and polished to a final thickness of ~0.83 mm for processing by HPT. The Zn-Al alloy was also received as a plate and it was prepared for HPT in essentially a similar way except that it was given an annealing treatment for 1 hour at 473 K [27]. All processing by HPT was conducted at room temperature for both alloys by placing the disk in a depression on the lower anvil and then bringing the upper and lower anvils together. The processing was conducted using an applied pressure, $P$, of 6.0 GPa and a rotation speed for the lower anvil of 1 rpm. The principle of processing by HPT was described earlier [32] and the same approach was followed in the present experiments except that a lubricant was not placed around the edges of the depressions on the upper and lower anvils. All processing was performed under quasi-constrained conditions in which there is a small outflow of material between the two anvils during the pressing operation [33-35]. Several disks of each alloy were processed by HPT using different numbers of turns from 1/4 to 10.

The values of the Vickers microhardness, $H_v$, were measured along diameters on each disk after processing by HPT. Each disk was carefully polished to a mirror-like finish and the hardness measurements were recorded using an FM-1e microhardness tester equipped with a Vickers indenter. These measurements used loads of either 200 gf for the Al-Cu alloy or 100 gf for the Zn-Al alloy with a dwell time for each measurement of 15 s. Measurements were taken along diameters at points separated by distances of 0.3 mm and at each point the average hardness was determined from four separate measurements located in a cruciform configuration and separated from the selected position by a distance of 0.15 mm. These average values were plotted against the distance from the center of each disk. To evaluate the extent of superplasticity, tensile specimens were machined as shown in Fig. 1 and then each specimen was pulled to failure at initial strain rates in the range from $10^{-5}$ to 1.0 s$^{-1}$. 

![Figure 1. The process adopted for cutting two tensile specimens from off-center positions in HPT disks using electro-discharge machining: all measurements are in mm [25].](image)
3. Experimental results

3.1 Al-33% Cu alloy

Figure 2 shows the microstructure in the Al-Cu alloy after annealing [26]. This eutectic alloy initially contained approximately equal amounts of an Al-Cu solid solution phase, $\alpha$, and an intermetallic CuAl$_2$ phase, $\theta$. In Fig. 2, the $\alpha$-phase is dark, the $\theta$-phase is almost white and the equiaxed linear intercept grain size was measured as $\sim 8.0 \mu m$ for both phases.

The measured values of the Vickers microhardness are shown in Fig. 3 for disks processed through 1/4, 1/2, 1, 2, 5 and 10 turns, respectively, where the lower broken line corresponds to the initial value of $H_v \approx 100$ for the alloy in the annealed condition without any HPT processing [26]. It is readily apparent that the hardness increases very rapidly even in the earliest stages of HPT processing. Thus, after 1/4 turn the hardness increases to $H_v \approx 180$ at the edges of the disk but in the central region the hardness values are lower at $H_v \approx 120$. With further straining, the hardness values at the edges remain reasonably constant but the values increase in the central region until ultimately, after 10 turns of HPT, the hardness in the center is almost as high as at the edge. These results therefore provide a direct example of the increasing homogenization that may be achieved with increasing numbers of torsional revolutions. Measurements showed the grain size was $\sim 3.0 \mu m$ after 10 turns [26].

Tensile tests were conducted to failure at a temperature of 723 K using examples of the off-center samples depicted schematically in Fig. 1. Emphasis was placed primarily on testing samples taken through 10 turns at different initial strain rates and the results are shown in Fig. 4 where the top sample is untested and the remaining specimens were pulled to failure at strain rates from $3.3 \times 10^{-5}$ s$^{-1}$ (bottom sample) to $1.0 \times 10^{-1}$ s$^{-1}$ (top sample below the untested sample). It is apparent that excellent superplastic flow was achieved at the two slowest strain rates where the elongations to failure were 1220% and 1200%, respectively. Inspection shows also that both of these specimens pull out in a uniform manner without evidence for any necking within the gauge lengths. An absence of necking is another requirement for true superplasticity [36]. At faster strain rates, such as $10^{-2}$ and $10^{-1}$ s$^{-1}$, there is clear evidence for necking within the gauge lengths and the relatively lower elongations of $\sim 200-300\%$ show that this alloy does not exhibit high strain rate superplasticity at least at a testing temperature of 723 K.

3.2 Zn-22% Al eutectoid alloy

A typical initial microstructure in the Zn-Al alloy is shown in Fig. 5 where the Zn-rich phase appears white and the Al-rich phase appears black [27]. This microstructure in the Zn-Al alloy was significantly different from the Al-Cu alloy because there was a duplex structure consisting of reasonably equiaxed grains interspersed with a lamellar structure. Measurements showed that the average grain size was $\sim 1.4 \mu m$ in the region of equiaxed grains whereas the average thickness of the thin layers in the lamellar structure was measured as $\sim 100$ nm.
**Figure 3.** Variation of hardness across disk diameters for the Al-Cu alloy after processing by HPT through different numbers of turns [26].

**Figure 4.** Tensile testing to failure of the Al-Cu alloy at 723 K showing examples of superplasticity with elongations up to >1200%.

**Figure 5.** The initial annealed microstructure in the Zn-Al alloy [27].
Figure 6 shows the variation in hardness with distance from the center of the disk after processing through 1, 2 and 4 turns with the broken line again corresponding to the initial annealed condition [27,30]. These results are unusual because all datum points for the processed material now lie below the line for the annealed condition thereby showing a work-softening effect. This loss in hardness is not generally observed in metallic alloys because of the reduction in grain size. For example, the average grain size was measured as ~350 nm at the edge of the disk and ~700 nm at the center of the disk after 4 turns with an average lamellar width of ~200 nm [27]. It should be noted that there is a report of a similar work-softening behavior in the Pb-62% Sn eutectic alloy [31], there are also reports for Pb of 99% purity, Sn of 99.9% purity and In of 99.999% purity [37] and work-softening was documented in the Zn-22% Al alloy after processing by hot rolling [38]. Inspection of Fig. 6 shows that the hardness is reduced significantly around the edges of the disk after 1 turn but the hardness remains high, and almost at the annealed level, in the center of the disk. With increasing numbers of turns, there is a small reduction in hardness at the edges but a significantly greater reduction in hardness near the center of the disk. Thus, all hardness values are again evolving towards a saturation level but now the saturation condition is at a lower level, rather than a higher level, by comparison with the annealed material.

Disks were processed to 5 turns and then tensile specimens were machined from the disks and pulled to failure at 473 K over a range of strain rates. The results are shown in Fig. 7 [29]. Thus, these specimens exhibit the conventional transition through regions I, II and III which is a characteristic feature of conventional superplastic alloys that have not been processed by SPD techniques [39]. For conventional materials, true superplasticity occurs over a range of intermediate strain rates covering about two orders of magnitude in region II and there are decreases in the measured elongations to failure both at more rapid strain rates in region III where there is a transition to conventional dislocation creep and at lower strain rates in region I where flow is restricted by the presence of impurities [40]. Thus, the conventional interpretation of region I is that it occurs because of the presence of an impurity-dominated threshold stress [41,42] and, consistent with this interpretation, there are experimental results showing that region I may be eliminated by using alloys of exceptionally high purity [43,44]. Inspection of Fig. 7 shows that the Zn-Al alloy achieves excellent elongations up to a maximum of 1800% at a strain rate of $1.0 \times 10^{-1}$ s$^{-1}$ which is within the region of high strain rate superplasticity.

4. Discussion
The two alloys subjected to HPT processing in this report exhibit both similarities and differences. The similarity arises because of the ability to achieve excellent superplastic elongations in Figs 4 and 7 and the difference arises in the nature of the hardness distributions as shown in Figs 3 and 6.

Superplasticity is achieved in these alloys because the small grains are easily retained in the two-phase structures at elevated temperatures and a small grain size is an important prerequisite for superplastic flow. For the Al-Cu alloy, elongations were attained of up to 1220% at a strain rate of $1.0 \times 10^{-4}$ s$^{-1}$ whereas for the Zn-Al alloy the maximum elongation was 1800% at the faster rate of $1.0 \times 10^{-1}$ s$^{-1}$. The improved superplasticity achieved in the Zn-Al alloy is associated with the smaller grain size in this alloy because the grain size at the mid-radius position within the disk was ~500 nm whereas in the Al-Cu alloy the grain size was ~3.0 μm. In conventional superplasticity, the high elongations occur through grain boundary sliding [45] and the theoretical model for superplasticity is based on the accommodation of sliding through intragranular slip and the climb of these dislocations into the opposing grain boundaries [46]. This mechanism is consistent with experimental evidence showing the occurrence of intragranular slip during superplasticity [47] and the model predicts that the strain rate varies inversely with the grain size raised to a power of two so that, as in Fig. 7, smaller grain sizes produce superplastic elongations at faster strain rates. It is important to note that the elongation of 1800% in Fig. 7 appears to represent the largest tensile elongation achieved to date in any material processed by HPT.
The distributions of the hardness values shown in Fig. 3 are consistent with a large number of metals including Al alloys [48,49] and Cu alloys [50,51]. In these materials, the imposed strain due to the torsional straining is higher at the edge of the disk but there is also an imposed pressure which leads to a gradual evolution towards homogeneity throughout the disk. This evolution has been accurately predicted using a solid mechanics approach and strain gradient plasticity modeling [52]. Nevertheless, the hardness distributions in Fig. 6 for the Zn-Al alloy are different because there is a work-softening rather than the usual increase in hardness with additional straining. This loss of strength is consistent with earlier observations showing that HPT processing at room temperature leads to a significant reduction in the distribution of rod-shaped Zn precipitates within the aluminum-rich grains and the high pressure applied in HPT produces an absorption of these Zn precipitates by the zinc-rich grains [53,54].

**Figure 6.** Variation of hardness across disk diameters for the Zn-Al alloy after processing by HPT through different numbers of turns [27,30].

**Figure 7.** Tensile testing to failure of the Zn-Al alloy at 473 K showing examples of superplasticity with elongations up to a maximum of 1800% [29].
5. Summary and conclusions
Two different two-phase alloys, the Al-33% Cu eutectic and the Zn-22% Al eutectoid, were processed by high-pressure torsion at room temperature through different numbers of turns. Measurements were taken of the hardness distributions after processing and tensile tests were conducted to evaluate the potential for achieving superplasticity.

The hardness distributions were different because the Al-Cu alloy exhibited conventional hardening with increasing numbers of HPT turns whereas the Zn-Al alloy exhibited a work-softening effect due to the loss of Zn precipitates during the HPT processing.

Both alloys exhibited superplastic elongations when pulled in tension at elevated temperatures. A maximum elongation of 1800% in the Zn-Al alloy represents the largest elongation recorded to date in an alloy processed by HPT.

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