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The significance of self-annealing in two-phase alloys processed by high-pressure torsion

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Abstract. The Zn-22% Al eutectoid alloy and the Pb-62% Sn eutectic alloy were processed by high-pressure torsion (HPT) over a range of experimental conditions. Both alloys exhibit similar characteristics with significant grain refinement after processing by HPT but with a reduction in the hardness values by comparison with the initial unprocessed conditions. After storage at room temperature for a period of time, it is shown that the microhardness of both alloys gradually recovers to close to the initial unprocessed values. Electron backscatter diffraction (EBSD) measurements on the Pb-Sn alloy suggest that the self-recovery behaviour is correlated with the fraction of high-angle grain boundaries (HAGBs) after HPT processing. Thus, high fractions of HAGBs occur immediately after processing and this favours grain boundary migration and sliding which is important in the self-annealing and recovery process. Conversely, the relatively lower fractions of HAGBs occurring after annealing at room temperature are not so conducive to easy migration and sliding.

1. Introduction

Exceptional grain refinement to the submicrometer or even the nanometer scale is achieved most readily in bulk solids through the application of severe plastic deformation (SPD) [1]. In general, most emphasis to date has focused on two techniques termed equal-channel angular pressing (ECAP) where a rod or bar is pressed through a die constrained within a channel [2] and high-pressure torsion (HPT) where a disk is subjected to a high applied pressure and concurrent torsional straining [3].

Since processing by HPT may lead to grain refinement within the nanometer range [3], this technique has attracted much attention over the last few years. There are now numerous reports available describing the microstructural evolution and the enhancement of mechanical properties in both pure metals and simple alloys. For two-phase alloys, for example, several early reports demonstrated the possibility of achieving significant grain refinement in both the Zn-22% Al eutectoid alloy [4,5] and the Pb-62% Sn eutectic alloy [6]. By contrast, no information is available at present on the self-annealing behavior of these two alloys after processing by HPT at room temperature. In practice, the homogeneously dispersed secondary phase particles may hinder any grain growth or grain boundary movements but these two alloys have relatively low melting points (753 K



for the Zn-Al alloy and 465 K for the Pb-Sn alloy) and it is known that self-annealing occurs at room temperature in many pure metals processed by SPD and having much higher melting points such as Cu [7] and Ag [8,9]. Therefore, it is reasonable to anticipate that self-annealing will also take place in the Zn-Al and Pb-Sn alloys after HPT.

Consequently, the present investigation was initiated in order to provide information on the self-annealing behavior of these two representative two-phase alloys after processing by HPT. The experiments were conducted using both the Zn-Al eutectoid alloy and the Pb-Sn eutectic alloy because early reports suggested there was a strain softening in these alloys during HPT [4-6]. This softening is significantly different from other typical two-phase alloys such as the Al-33% Cu eutectic alloy [10] and the Cu-Ag system [11,12] where strain hardening dominates.

2. Experimental material and procedures

The experiments were performed using two different two-phase alloys: a Zn-22% Al eutectoid alloy and a Pb-62% Sn eutectic alloy. The Zn-Al billets were annealed in air at 473 K for 1 h but no annealing treatment was performed on the Pb-Sn alloy because of the very low melting point which leads to self-annealing at ambient temperature. The two materials were machined into disks having thicknesses of ~1.2-1.5 mm and each side of each disk was subjected to careful polishing to give a series of HPT disk samples having thicknesses of 0.80 ± 0.01 mm. The average grain sizes prior to HPT were measured as ~ 1.4 μm for the Zn-Al alloy and ~ 2.9 and ~ 1.9 μm for the Sn grains and the Pb grains in the Pb-Sn alloy. Further information on the initial microstructures of both alloys are available in an earlier report [6]. The processing by HPT was performed at a pressure, P , of 3.0 GPa at room temperature using a quasi-constrained facility [13,14] operating at a rotational speed of 1 rpm. Two disks of each alloy were processed by HPT for a total number, N , of 1 turn. After HPT processing, the disks were stored at room temperature for a period of time in order to examine the characteristics of the self-annealing behaviour.

The nature of the microstructures was repeatedly investigated during the course of storage using scanning electron microscopy (SEM) for the Zn-Al alloy and electron backscatter diffraction (EBSD) for the Pb-Sn alloy. For sample preparation, the two disks were subjected to careful grinding and polishing with abrasive papers and diamond paste. For the Zn-Al alloy, etching was conducted for ~ 5 s in a solution of 100 ml $\text{C}_2\text{H}_6\text{O}_2$, 20 drops of HNO_3 , four drops of H_2SO_4 and four drops of HCl in order to reveal the grain boundaries. For the Pb-Sn alloy, the polished disk was etched for ~ 5 s in a solution of 25 ml H_2O , 5 ml HCl with a concentration of 37% and 5 g of NH_4HO_3 in order to remove any residual stresses which may be introduced by polishing and to provide an optimum surface for the EBSD measurements. The EBSD patterns were collected with a step size of 0.15 μm and with a clean-up procedure where the total number of modified points was less than 10% of the total points measured.

The Vickers microhardness, H_v , at the edge of each disk was carefully monitored for 50 days in the Zn-Al alloy and 20 days in the Pb-Sn alloy and the process was terminated when it appeared that the values of H_v became saturated at room temperature. The microhardness values across the disk diameters were recorded after 0 day (corresponding to immediately after processing), 15 days and 33 days of storage for the Zn-Al alloy and 0 day, 4 days and 9 days for the Pb-Sn alloy. An FM-300 microhardness tester equipped with a Vickers indenter was used and loads of 100 gf and 10 gf were selected for the Zn-Al alloy and the Pb-Sn alloy, respectively, with dwell times of 15 s for both alloys.

3. Experimental results

3.1 Self-annealing at room temperature in the Zn-Al alloy after HPT

Microstructural observations were undertaken on the Zn-Al disk processed by HPT after 1 turn. Representative microstructures shown in Fig. 1 were taken at the edge of the disk (a) immediately after processing and (b) after storing at room temperature for 33 days. Due to the different sensitivities to the etching solution, Zn grains and Al grains appear to have different depths in Fig. 1. Specifically,

the Zn grains appear to be brighter as a matrix phase and the Al grains appear darker as a convex phase. It is obvious from Fig. 1 that HPT through only 1 turn produces very significant grain refinement in Fig. 1 (a) by comparison with the microstructure in the initial condition where the measured average grain size was $\sim 1.4 \mu\text{m}$. The average grain size immediately after HPT (including both Zn grains and Al grains) was about $\sim 200 \text{ nm}$. It is evident in Fig. 1 (b) that storage for 33 days leads to a growth of both the Zn grains and the Al grains by comparison with the microstructure visible immediately after HPT. The measured average grain size after storage for 33 days was $\sim 450 \text{ nm}$ which is consistent with the results in earlier reports [4,5]. It should be noted that the grains in Fig. 1 are essentially equiaxed in shape and there is no evidence for the presence of any lamellar structure which is common in the Zn-Al alloy in the cast condition.

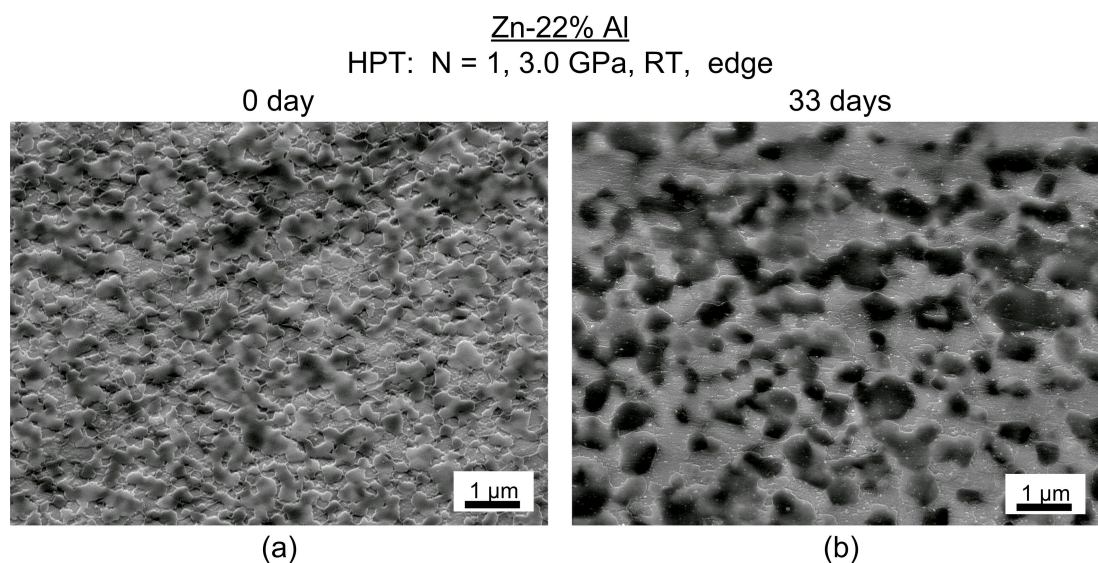


Figure 1. Microstructures of the Zn-Al alloy at the edge of the disk processed by HPT for one turn (a) immediately after processing and (b) after 33 days of storage.

The Vickers microhardness values at the edge of the Zn-Al disk are plotted against the time of storage in Fig. 2 (a). It is apparent that the microhardness increases significantly in the first ~ 30 days of storage but thereafter the values tend to saturate at $H_v \approx 28$. Thus, the saturated value of H_v at the edge of the disk after storage is about 1.6 times higher than the starting value immediately after HPT processing. In addition, values of the microhardness were recorded across the diameters of disks during the course of storage after 0 day (immediately after processing), 15 days and 33 days and these values are plotted against the positions on the disks in Fig. 2 (b). Inspection of Fig. 2 (b) reveals several interesting points. First, each plot exhibits the same general features with higher values at the centres of the disks and lower values at the edges. Second, the value of the microhardness in the centre region immediately after processing was $H_v \approx 77$ which is higher than the initial annealed condition of $H_v \approx 68$. This shows the occurrence of some limited hardening in this central region but nevertheless the hardness values in the central region gradually decrease during storage at room temperature thereby demonstrating a softening through annealing after HPT. Third, in the edge region the microhardness is initially low and subsequently these values gradually increase during storage at room temperature. Thus, it is evident from Fig. 2 (b) that the hardness values change in different ways in the centre region and the edge region during self-annealing and the softening effect in the centre occurs over a diameter of $\sim 4 \text{ mm}$. Therefore, the transition points at $\pm 2 \text{ mm}$ from the centre represent regions where the microhardness values remain essentially unchanged throughout the period of storage. Due to the geometrical features of HPT, the torsional strain introduced is theoretically equal to zero at the

centre of the disk. Therefore, there should be only a limited amount of strain introduced in the centre region and in this region the alloy appears to be hardened rather than softened immediately after HPT processing. Similar strengthening in the early stages of straining was reported in a Zn-10% Al alloy processed by ECAP where the highest ultimate tensile strength was achieved after one pass but decreased through three passes [15].

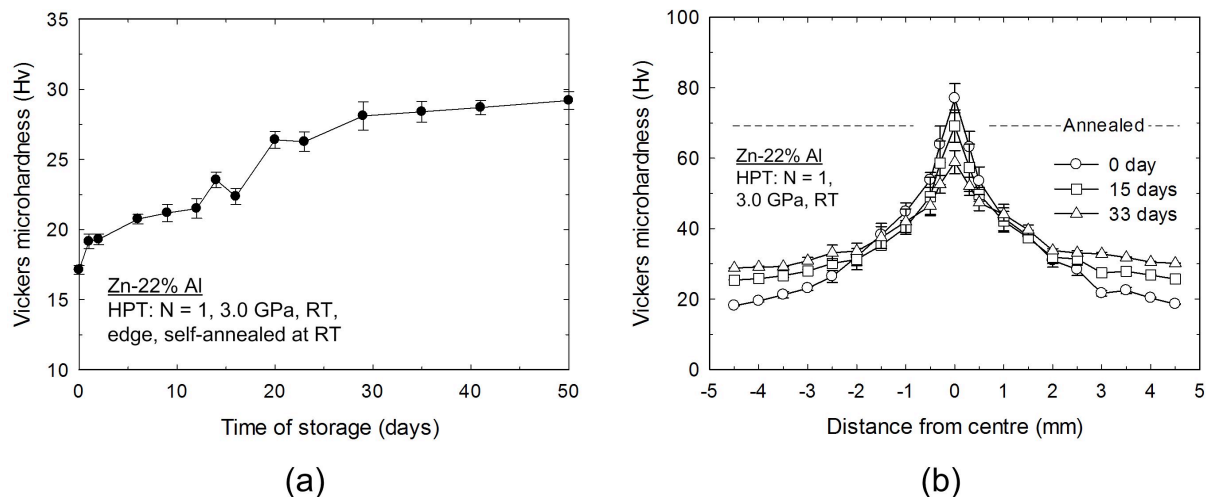


Figure 2. Variation of microhardness in the Zn-Al alloy during storage (a) at edge of disk for up to 50 days and (b) across the diameter immediately after processing and for 15 and 33 days of storage.

3.2 Self-annealing at room temperature in the Pb-Sn alloy after HPT

Figure 3 shows images of the Pb-Sn alloy taken at the edge of the disk after processing by HPT for 1 turn immediately after processing (a, c and e) and after 11 days of storage at room temperature (b, d and f). Figures 3 (a) and (b) show two reconstructed phase maps with the Sn-rich phase appearing blue and the Pb-rich phase appearing red. Grain reconstruction maps for individual phases are shown in (c) and (d) for the Sn-rich phase and (e) and (f) for the Pb-rich phase. In these maps, high-angle grain boundaries (HAGBs) with misorientations greater than 15° are shown in thicker black lines and the thinner black lines represent low-angle grain boundaries (LAGBs). It is evident that, as noted earlier for the Zn-Al alloy, there is significant grain growth in the Pb-Sn alloy after storage at room temperature. The measured initial average grain sizes prior to HPT were ~ 2.9 and ~ 1.9 μm for the Sn and Pb grains, respectively. The measured average grain sizes immediately after processing were ~ 1.4 μm for the Sn grains and ~ 800 nm for the Pb grains whereas these values increased to ~ 2.0 and ~ 1.3 μm after 11 days of storage. The Pb-rich phase particles are broken and distributed essentially homogeneously by the very large strain introduced by HPT and these particles rejoin to form larger particles during the self-annealing process. There is a high fraction of HAGBs in both phases immediately after processing but the fraction decreases when the disk is stored for a long time. Thus, there are more thin black lines in Fig. 3(d) than in Fig. 3(c) and in Fig. 3(f) than in Fig. 3(e).

In Fig. 4 (a), microhardness measurements are shown at the edges of the disks for the Pb-Sn alloy and again there is rapid initial growth during the first 4 days and a saturation at $H_v \approx 7$. In Fig. 4 (b) all microhardness values are lower than the initial condition where $H_v \approx 10$ and all values are higher in the centre and lower at the edge. However, there is essentially a plateau in the values of H_v in the central region of the disk over a diameter of ~ 1 mm after 4 and 9 days of storage. There is also no evidence of strengthening in the early stage of HPT in the Pb-Sn alloy.

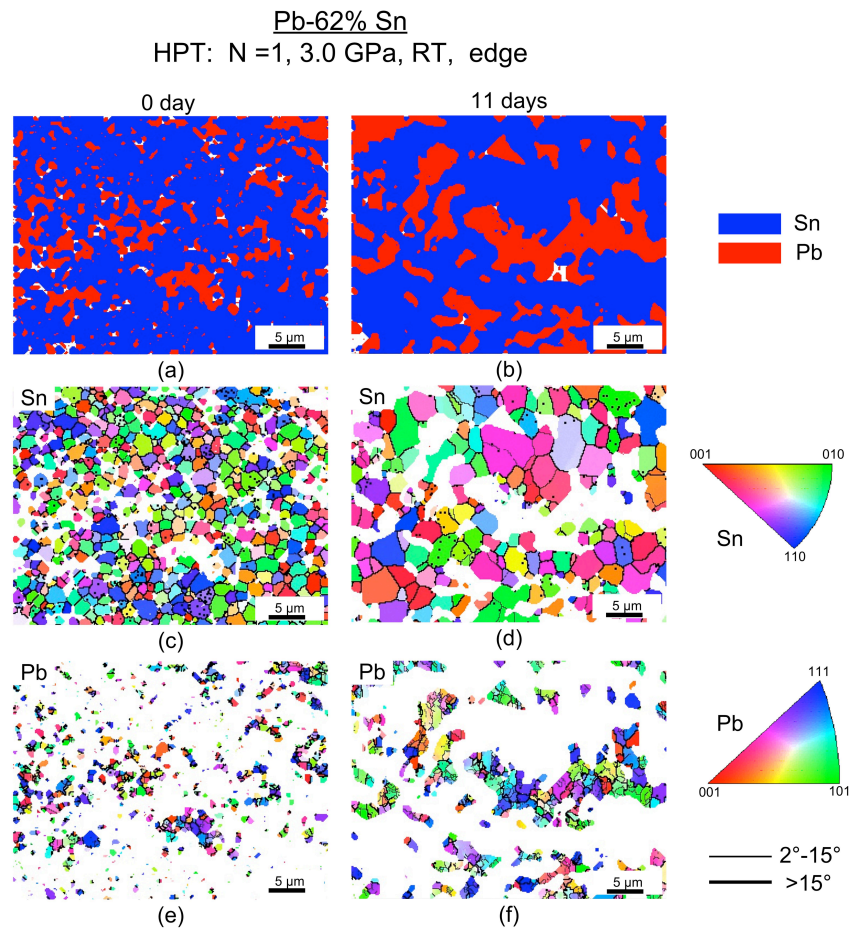


Figure 3. Images of the Pb-Sn alloy at the edge of the disk processed by HPT for one turn immediately after processing (a, c and e) and after 11 days of storage (b, d and f): the colours correspond to different orientations depicted in the unit triangles on the right.

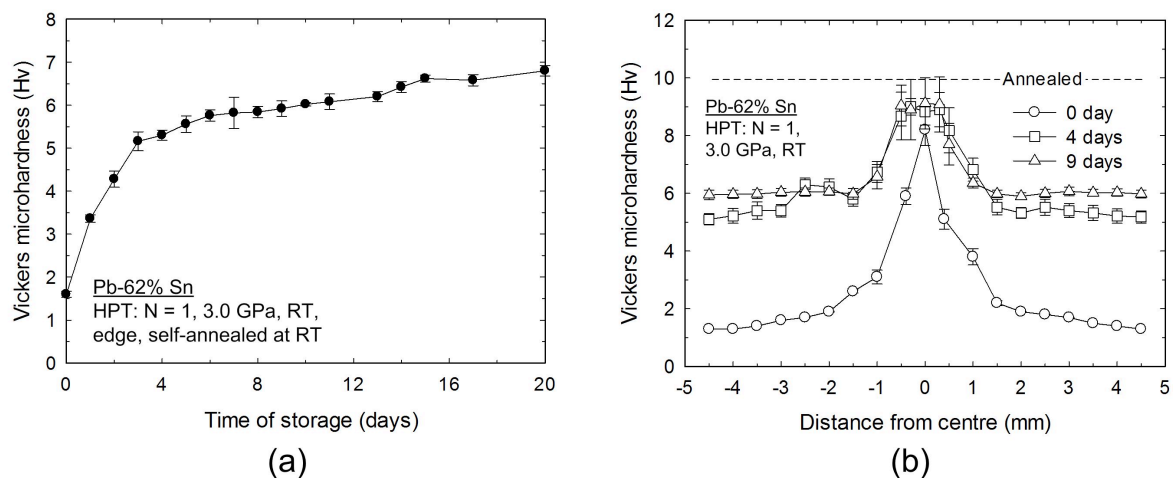


Figure 4. Variation of microhardness in the Pb-Sn alloy (a) at edge of disk after storage up to 20 days and (b) across the diameter immediately after processing and after 4 and 9 days of storage.

4. Discussion

The present results with the Zn-Al and Pb-Sn alloys demonstrate the unusual situation of metals processed by HPT where exceptionally low values of hardness were recorded after HPT processing. This result is not due to the use of a two-phase eutectic or eutectoid alloy because it was shown recently that the hardness increased after HPT processing for the Al-33% Cu eutectic alloy [10] and Cu-28% Ag alloys [11,16]. It is now well established that the variations of hardness with equivalent strain introduced by HPT depend upon the nature of the recovery in the material [5,6,17,18]. In general, three types of behaviour are available in terms of the hardness after HPT processing. First, the hardness may increase initially with an increase of equivalent strain and then saturate at a reasonably high strain. This type of hardness variation is found in many metals of commercial purity and in metallic materials such as Cu and Al alloys [16,19-22] where there is strain hardening with little or no recovery during straining. Second, the hardness variation follows an initial increase, a subsequent decrease and thereafter the hardness reaches essentially a saturation value with increasing straining. For this condition, the stacking fault energy is very high so that cross-slip occurs easily to give rapid microstructural recovery. This type of behavior is observed in materials such as high-purity Al [17,18,23,24] and high-purity Mg [25]. Finally, as demonstrated in this investigation with the Zn-Al and Pb-Sn alloys, the hardness values after HPT processing are significantly lower than in the initial condition despite the fact that the grain sizes in both materials are reduced in the processing. Recent reports summarize these different models of hardness evolution for a range of metals and alloys processed by HPT [26,27].

In most metallic alloys processed by HPT the grain size is reduced and the material becomes harder while a common annealing process invariably produces grain growth and a general softening. By contrast, the Zn-22% Al and Pb-62% Sn alloys exhibit a significant work-softening behaviour with a reduction in grain size when processed by HPT and then an anneal-hardening behaviour with an increase in grain size when annealed at room temperature. Similar trends were reported for Pb (99%), Sn (99.9%) and In (99.999%) after processing by HPT [28]. A recent report also showed a similar work-softening and anneal-hardening effect in the Zn-22% Al alloy but the alloy was processed by hot rolling and annealed at room temperature as well as several elevated temperatures [29]. In this latter work, a dynamic recovery model was employed to explain the work-softening effect where dislocation pile-ups are absorbed by grain boundaries during deformation and this leads to a decrease in the dislocation density and hardness and an increase in the grain boundary misorientations. However, this theory fails to explain the higher hardness measured in an initial cast or annealed condition by comparison with the deformed condition because it is reasonable to anticipate there will be an initial low dislocation density. An alternative explanation for the occurrence of work-softening appears to lie in microstructural characteristics that were recorded earlier for the Zn-Al alloy [30,31] where HPT processing led to a significant reduction in the numbers of rod-shaped precipitates of Zn within the Al-rich grains and thereafter to an absorption of the Zn precipitates by the Zn-rich grains.

In the present investigation, grain boundary information was obtained from EBSD measurements on the Pb-Sn alloy. These data are analyzed in plots of the number fractions of grain boundary misorientation angles in the Pb-Sn alloy and the results are shown in Fig. 5 where (a and b) are for the initial annealed condition prior to HPT, (c and d) are immediately after HPT processing and (e and f) are for samples after 11 days of storage. It is readily apparent that there is a significant increase in the fractions of HAGBs after HPT (c and d) compared to the initial cast or annealed condition (a and b) and, in addition, the fractions of HAGBs gradually decrease as the alloy is annealed at room temperature for both phases (e and f). In view of the low melting points for both alloys, grain boundary migration and sliding probably occurs at room temperature and it is well known that sliding requires boundaries having high angles of misorientation [32] and migration occurs more rapidly in high-angle boundaries [33,34]. Thus, the high fractions of HAGBs immediately after HPT processing favour the occurrence of migration and sliding and this can produce anneal-hardening during storage at room temperature.

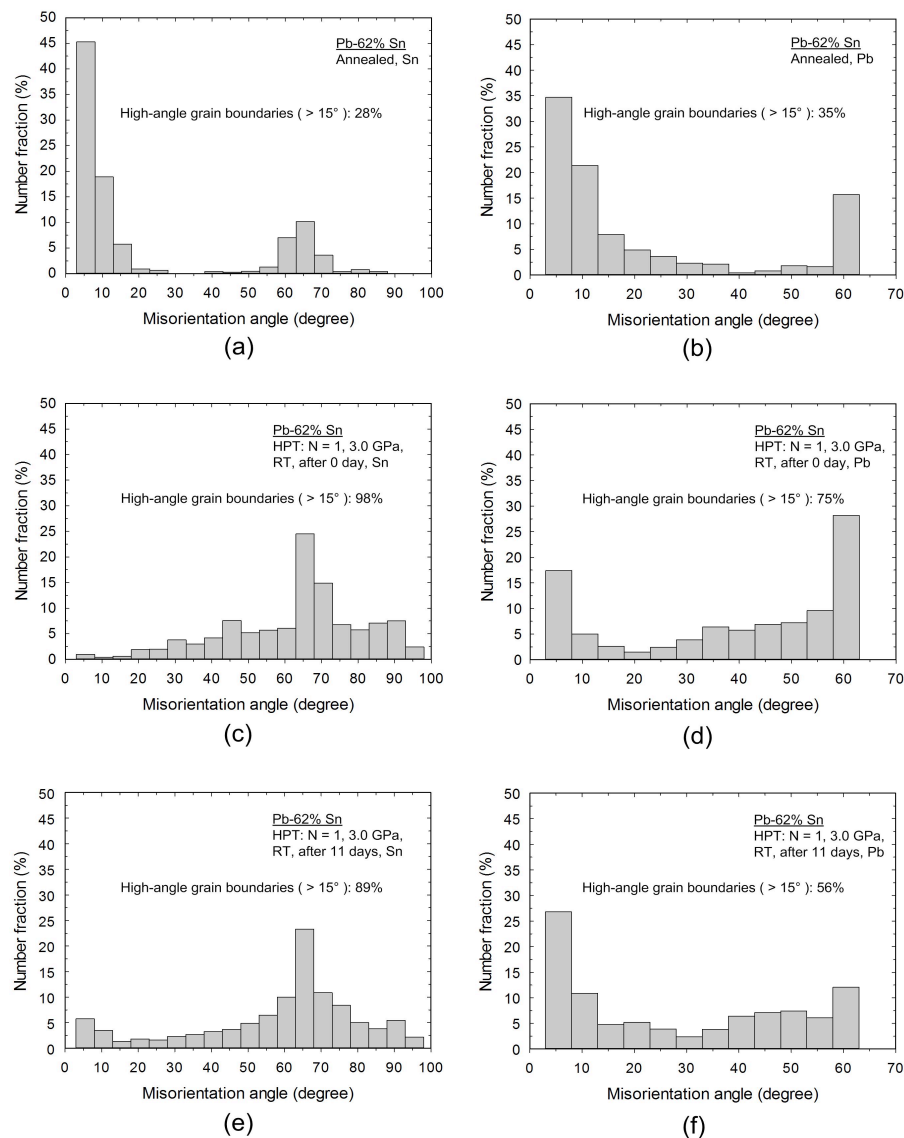


Figure 5. The distributions of the boundary misorientation angles in the Pb-Sn alloy for (a and b) initial annealed condition prior to HPT, (c and d) immediately after processing and (e and f) after 11 days of storage.

Very recent experiments using nanoindentation demonstrated the occurrence of grain boundary sliding as the dominant deformation mechanism at room temperature in the Zn-22% Al alloy after HPT through two turns by observing the relatively high values of the strain rate sensitivity, m , of ~ 0.25 and the associated low value for the activation volume [35]. These results are consistent with the present investigation but additional experiments are now needed to provide information on the grain boundary effects associated with the occurrence of work-softening and anneal-hardening in both of these two-phase alloys.

5. Summary and conclusions

1. Disks of the Zn-22% Al eutectoid alloy and the Pb-62% Sn eutectic alloy were processed by HPT for 1 turn. Microstructural examination showed significant grain refinement but with a corresponding decrease in the measured hardness by comparison with the initial unprocessed conditions.

2. This strain softening effect is accompanied by a significant increase in the fraction of high-angle grain boundaries (HAGBs) after HPT processing. The presence of a high fraction of HAGBs is consistent with the occurrence of grain boundary migration and sliding.

3. A significant self-annealing effect occurs in both alloys after storage of the HPT-processed disks at room temperature. This self-annealing leads to the occurrence of some limited grain growth and an increase in the values of the microhardness.

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