Evolution of hardness in ultrafine-grained metals processed by high-pressure torsion

Megumi Kawasaki\textsuperscript{a,b,*}, Han-Joo Lee\textsuperscript{a}, Byungmin Ahn\textsuperscript{c}, Alexander P. Zhilyaev\textsuperscript{d,e}, Terence G. Langdon\textsuperscript{b,d}

\textsuperscript{a} Division of Materials Science and Engineering, Hanyang University, Seoul, South Korea
\textsuperscript{b} Departments of Aerospace & Mechanical Engineering and Materials Science, University of Southern California, Los Angeles, United States
\textsuperscript{c} Department of Energy Systems Research, Ajou University, Suwon, South Korea
\textsuperscript{d} Materials Research Group, Faculty of Engineering and the Environment, University of Southampton, Southampton, United Kingdom
\textsuperscript{e} Institute for Metals Superplasticity Problems, Ufa, Russia

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\textbf{A B S T R A C T}

The processing of metals through the application of high-pressure torsion (HPT) provides the potential for achieving exceptional grain refinement in bulk metals. Numerous reports are now available demonstrating the application of HPT to a range of pure metals and simple alloys. In practice, excellent grain refinement is achieved using this processing technique with the average grain size often reduced to the true nano-scale range. Contrary to the significant grain refinement achieved in metals during HPT, the models of the hardness evolution are very different depending upon the material properties. For a better understanding of the material characteristics after conventional HPT processing, this report demonstrates the hardness evolutions in simple metals including high-purity Al, commercial purity aluminum Al-1050, ZK60A magnesium alloy and Zn-22% Al eutectoid alloy after processing by HPT. Separate models of hardness evolution are described with increasing equivalent strain by HPT. Moreover, a new approach for the use of HPT is demonstrated by synthesizing an Al–Mg metal system by processing two separate commercial metals of Al-1050 and ZK60A through conventional HPT processing at room temperature.

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1. Introduction

The processing of metals through the application of severe plastic deformation (SPD) provides the potential for achieving exceptional grain refinement in bulk metal solids [3]. There are numbers of publications to date demonstrating the significance of SPD techniques but, in general, equal-channel angular pressing (ECAP) [2] and high-pressure torsion (HPT) [3] are accepted as the major SPD methods [4]. For ECAP processing, a sample, in a rod or bar shape, is pressed through a die constrained within a channel so that the microstructure receives shear straining leading to grain refinement to grain sizes in the submicrometer range. For processing through HPT, a sample, in the shape of a disk, is subjected to a high applied pressure and concurrent torsional straining so that the severe deformation leads to submicrometer grains or even true nanometer grains within the metal sample [3]. Accordingly, the present report describes recent published data on simple metals and common alloys processed by HPT.

The essential principles in HPT processing are that the strain introduced within the HPT disk sample is markedly inhomogeneous. Specifically, when a disk is processed by HPT, the equivalent von Mises strain, $\varepsilon_{eq}$, is given by a relationship of the form [5,6]

$$
\varepsilon_{eq} = \frac{2\pi Nr}{h^3}
$$

where $N$ is the number of HPT revolutions and $r$ and $h$ are the radius and height (or thickness) of the disk, respectively. Therefore, it is apparent that the torsional straining imposed within the disk sample is dependent upon the distance from the center of the disk, $r=0$, where there is theoretically no straining by the torsional processing. This implies that there is an inevitable inhomogeneity both in microstructures and hardness in the disk sample processed by HPT. Nevertheless, it is demonstrated experimentally that both sufficiently high numbers of HPT turns and high applied pressures lead to homogeneous microstructures and hardness throughout the disk [7,8].

Although there is significant grain refinement through HPT processing, the models of hardness evolution into homogeneity are not consistent between the different metals and alloys [9,10]. The present report is initiated to provide recent experimental results on the hardness evolution observed in a series of representative metals and alloys including high-purity Al, commercial purity aluminum Al-1050, ZK60A magnesium alloy and Zn-22% Al eutectoid alloy after they are processed through HPT. These materials are selected because, as also shown in a recent report summarizing the hardness evolution models for metals after HPT [9], high-purity Al [10–18] and Zn–Al alloy [10,19–22] demonstrate unique softening behaviors after HPT compared with most common metals which generally exhibit strain hardening during deformation.

The present report demonstrates two separate contents. In the following two sections, the three separate models of hardness evolution are displayed showing recent experimental results in the metals processed through HPT processing. In the last section, as a new approach for the use of HPT, a potential of improving the upper limit of hardness is demonstrated in the Al–Mg system when processed from two separate commercial metal disks of Al-1050 and ZK60A through conventional HPT processing.

2. Separate models of hardness evolution through HPT

2.1. Commercially pure Al-1050 aluminum alloy and ZK60A magnesium alloy

In this section, two different materials of commercial purity aluminum Al-1050 and ZK60A magnesium alloy are selected for demonstrating the general model of hardness evolution through HPT where it is observed in most metals showing strain hardening.

A separate billet was machined from an as-received bar of a commercial purity (99.5%) aluminum Al-1050 containing 0.30% Fe and 0.25% Si [23,24] and an as-extruded bar of ZK60A, respectively. These billets of the alloys were sliced and polished to have final disk shapes with a diameter of 10 mm and a thickness of ~0.8 mm. All disks were processed at room temperature in quasi-constrained HPT conditions [25,26] with the procedure described earlier [12]. A series of the disks was processed for both alloys under a compressive pressure of $P = 6.0$ GPa and a rotation speed of 1 rpm for total revolutions of $N = 1/4$, 1 and 5 turns.

After processing, Vickers microhardness measurements were conducted on the polished surfaces of both the Al-1050 and ZK60A disks. For the Al-1050 disks, the measurements were taken over the total surface of each disk with an increment of 0.6 mm between the datum points on both axes, X and Y. The measured microhardness values, $H_v$, were used to construct a three-dimensional color-coded contour map for each disk in order to show the distributions of the local microhardness on the surfaces of the HPT disks after 1/4, 1 and 5 turns. For the ZK60A alloy, the measurements were conducted along the diameters on the polished surfaces of the processed disks. As described earlier [12], each $H_v$ value at the selected representative position along the disk diameter was calculated from four separate cruciform positions lying at a distance of 0.15 mm so that the individual positions were separated by incremental distances of 0.3 mm. For both metals, in order to remove the influence of the possible hardness gradient through the disk thickness direction [18,27], all measurements were conducted on the surfaces at the mid-sections through the disk heights.

Fig. 1 shows the microhardness variation in the three-dimensional color-coded contour maps for the Al-1050 disks processed by HPT for (a) 1/4 turn, (b) 1 turn and (c) 5 turns where the $H_v$ values are presented by unique colors given at the right in the figure [23,24]. For references, the average $H_v$ value of ~25 was measured as the initial hardness in the as-received condition before HPT processing [28]. It is apparent that high hardness with $H_v \approx 60–65$ was recorded at the peripheral regions of $r \approx 3–5$ mm in the disk after processing for 1/4 turn as shown in Fig. 1(a) and there is a sharp drop in hardness toward the center of the disk with the minimum $H_v$ value of ~45 at $r = 0$. After HPT for 1 turn as shown in Fig. 1(b), the
Fig. 1 – Three-dimensional color-coded contour maps of the hardness distributions for commercial purity Al-1050 alloy after HPT for (a) 1/4, (b) 1 and (c) 5 turns under 6.0 GPa at room temperature [23,24].

central region demonstrating lower hardness is reduced and the minimum Hv value at the center of the disk increased to ~50–55. Additional HPT through 5 turns showed the saturated high hardness of Hv ≈65 throughout the total disk surface as shown in Fig. 1(c), thereby demonstrating hardness homogeneity in the Al-1050 through HPT for 5 turns. Earlier study using a commercial purity (99.7%) Al reported a similar hardness evolution toward homogeneity with increasing numbers of HPT turns for up to 8 under 1.0 GPa [29].

A similar trend of hardness evolution was observed in the ZK60A magnesium alloy when processed through HPT under 6.0 GPa at room temperature. Fig. 2 shows the variation of microhardness versus the distance from the center for the ZK60A disks processed by HPT for 1/4, 1 and 5 turns at 6.0 GPa. The dashed line in the figure denotes the average Hv value of ~72 in the as-received condition. After HPT for 1/4 and 1 turn, the hardness evolution showed a similar variation of hardness to the Al-1050 disks where higher hardness was shown at the peripheries and the hardness values decreased toward the centers of the disks. Consistent minimum Hv values of ~75 were observed at r = 0 for both processed disks after 1/4 and 1 turn and there was no significant difference between these two disks except for the slightly higher Hv values at some measurement points in the peripheral region in the disk after 1 turn. Increasing the numbers of turns to 5, the hardness values are saturated to Hv ≈105–110 through the total disk diameter and thus the ZK60A demonstrated a reasonable hardness homogeneity after HPT through 5 turns. A similar hardness evolution toward homogeneity was reported in a recent report on a ZK60 after HPT at room temperature under 2.0 GPa and a homogeneous distribution of high hardness was demonstrated after 5 revolutions [30].

The hardness variations demonstrated in both the Al-1050 and ZK60A disks during HPT represent the general model of hardness behavior with strain hardening. In practice, the Hv values in the central region are lower compared with those in the outer region and it is consistent with the imposed equivalent strain as calculated in Eq. (1). This general model of hardness evolution toward a uniformly distributed high saturated hardness was predicted with a theoretical approach using strain gradient plasticity modeling [31]. Moreover, a very recent analysis by three-dimensional FEM confirmed the predictions of increasing strength with increasing distance from the centers of the disks and with increasing numbers of HPT turns in strain-hardening materials [32].

2.2. High-purity Al

Processing by HPT was conducted using a polycrystalline high-purity (99.99%) Al after annealing for 1 h at 773 K. The annealed billet of the high-purity Al was sliced and then polished to have disks with a diameter of 10 mm and a thickness of ~0.8 mm. All disks were processed by HPT at room temperature under the same conditions as mentioned in the previous section for both the Al-1050 and ZK60A alloy. A series of the disks was processed at F = 6.0 GPa for total revolutions of N = 1/2, 1, 5 and 20 turns [18,33,34].

Fig. 2 – Distribution of the Vickers microhardness values versus distance from the centers of ZK60A disks processed by HPT for 1/4, 1 and 5 turns under 6.0 GPa at room temperature where the lower dashed line shows an average hardness of Hv ≈72 in the as-received condition without HPT.
The Vickers microhardness measurements were conducted on the high-purity Al after HPT by using the same procedure as noted for the ZK60A alloy in the earlier section. Thus, the Hv values were recorded along the diameter on each polished surface of the processed disk. All measurements were conducted at equally polished sections which were ~200 μm from the lower (or bottom) surfaces.

Fig. 3 shows the variation of the Hv values against the distance from the centers of the disks after processing by HPT for 1/2 to 20 turns where the lower dashed line denotes the average hardness of Hv ~ 20 measured in the annealed condition prior to processing and the error bars denote the 95% confidence limit [9,18]. It is apparent for all disks that the hardness values across the diameters increased significantly after HPT to ~35 which is higher than in the annealed condition without HPT by a factor of ~2. Although the increasing hardness after HPT is a consistent result as seen in the Al-1050 and the ZK60A alloy in the earlier section, the unique behavior of the high-purity Al is that there is high hardness at the central regions compared with the hardness at the edges of all the processed disks. In practice, the Hv values at the peripheral regions remain essentially constant at Hv ~ 37 through 1/2 turn to 20 turns. Detailed investigation shows the central regions with high hardness are widely spread with wide scatter shown by the large error bars when in the early stage of HPT for 1/2 turn. Thereafter, the central regions in the disks tend to decrease with increasing numbers of HPT turns from 1 to 5 so that the central region becomes almost a point with Hv ~ 42 recorded with short error bars after 5 turns. Additional HPT turns to 20 demonstrated an essential disappearance of the high hardness at the central region, thus the experiments show the occurrence of a high level of hardness homogeneity across the diameter of the high-purity Al disks after HPT for more than 5 turns.

A similar hardness evolution with increasing numbers of HPT turns was shown in a high-purity Al in several earlier reports [10–18] and the special emphasis on the large hardness deviation at the central regions was observed in an early stage of HPT through 1 turn with significant refinement of microstructure in a high-purity Al in recent reports [14,17]. Moreover, this unique hardness behavior was observed in a high-purity Mg [35] and Zn [36] after HPT at room temperature. Nevertheless, an important conclusion in this section is that the hardness evolution in the high-purity Al is different from commercial Al alloys that show the general strain hardening model. Thus, the hardness behavior of this type shown in the high-purity Al includes a significant strain softening after a rapid strain hardening in the very early stage of HPT processing.

2.3. Zn-22% Al eutectoid alloy

The experiments used a commercial Zn-22% Al (in wt.%) eutectoid alloy which contains a binary microstructure with about equal amounts of an Al-rich α phase and a Zn-rich β phase. These phases are homogeneously distributed and there are areas of both equiaxed grains and of a lamellar structure [19–22]. The disks of the Zn–Al alloy were prepared from a billet in the same way as noted in the earlier Sections 2.1 and 2.2 to have final disk dimensions with a diameter of ~10 mm and a thickness of ~0.83 mm. Before processing by HPT, all disks were annealed in air at 473 K for 1 h to remove any residual stresses. Processing by HPT was conducted on the Zn–Al disks at room temperature under P = 6.0 GPa for N = 1, 2, 4, 5, and 20 turns.

Following the procedure noted in the earlier sections for both the ZK60A and high-purity Al, the measured Hv values were recorded across the diameters of the Zn–Al disks after HPT for 1, 2, 4, 5 and 20 turns. Fig. 4 shows the average values of Hv against the distance from the centers of the disks where
the upper dashed line denotes the measured hardness value of Hv ≈ 68 for the annealed and unprocessed condition [22].

There are several critical conclusions from an inspection of Fig. 4. First, all of the disks after HPT demonstrated significantly lower hardness than in the unprocessed condition, thereby demonstrating the occurrence of significant softening during HPT. Second, there is a consistent trend of higher hardness at the disk centers and the high Hv values decrease toward the peripheries where the minimum Hv value is recorded for each disk after HPT. Thus, the hardness behavior involves strain softening with increasing equivalent strain as given in Eq. (1). Third, there is a decrease in the Hv values over the disk surfaces after processing with increasing numbers of turns. In practice, the hardness values of Hv ≈ 60 at the center toward ~ 30 at the edge after 1 turn changes to ~ 30 at the center to ~ 23 at the peripheries through 20 turns. Thus, with increasing HPT revolutions, the difference in local hardness between the center and edge in each processed disk tends to decrease and this demonstrates a gradual hardness evolution toward homogeneity throughout the disk surfaces in the Zn–Al alloy.

This hardness behavior in the Zn–Al alloy is, in practice, the opposite of the behavior observed in the strain hardening metals such as the Al-1050 and ZK60A where, as shown in the earlier Section 2.1, these metals show significant strengthening with higher hardness at the edges of the processed disks. Therefore, HPT processing introduced a strain softening behavior accompanied with significant weakening. This strain softening behavior was also reported in binary microstructures of Al-10%, ~ 20% and ~ 30% Zn alloys after HPT at 5.0 GPa up to 5 turns [37–39] and the Pb-62% Sn eutectic alloy after HPT for 1 turn at 3.0 GPa [21].

3. Three models of hardness evolution with equivalent strain by HPT

In the earlier sections, three different hardness behaviors toward homogeneity were discussed demonstrating the hardness development in four representative metals and alloys with increasing numbers of HPT revolutions. The experimental results presented imply that the tendency of increasing/decreasing local hardness is closely correlated with the imposed equivalent strain at the measured position as given in Eq. (1) and thus it is indispensable to evaluate the hardness evolution in terms of the estimated equivalent strain.

The approach of using equivalent strain for evaluating the hardness evolution was first applied on an austenitic steel where a series of steel disks was processed by HPT for a range of turns from 0.17 to 16 turns under $P = 5.3$ GPa [40]. In the examination, the processed steel demonstrated the general strain hardening behavior in the hardness evolution during HPT where the behavior is consistent with those in the Al-1050 and ZK60A as shown in Figs. 1 and 2. The same approach was applied for many different alloy systems [9] and it is generally understood that the hardness behaviors in these metals fall into three different models which are all dependent upon the nature of recovery within the material [9,11,19]. These three possible models of hardness evolution toward homogeneity with increasing equivalent strain are depicted schematically in Fig. 5 for the metals processed by HPT where the initial annealed conditions are marked as horizontal bars on the vertical axes [19].

Fig. 5(a) represents the general hardness model for the materials demonstrating the hardness increases initially with equivalent strain and then saturates to the maximum hardness at a reasonably high strain. The conventional variation of hardness with strain is for materials having little or no recovery and most commercial purity metals and simple alloys exhibit this type of relationship [9]. Fig. 5(b) illustrates the hardness evolution with increasing equivalent strain for the high-purity Al. Although the material shows a similarity with the strain hardening metals exhibiting overall high hardness after HPT, it demonstrates higher hardness values at the central regions of the disks, which received limited imposed strain. Thereafter, there is a subsequent softening with microstructural recovery so as to achieve saturation at high hardness in the high-purity Al after reasonably high numbers of HPT turns. Thus, this hardness behavior shows a bell-shape curve as shown in Fig. 5(b).

By contrast, for a material such as the Zn-22% Al eutectoid alloy, the hardness values after HPT are lower than in the unprocessed condition even though there is a significant grain refinement by processing [19–22]. The weakening in a Zn-22% Al alloy is due to the change in the precipitation kinetics by the severe deformation during HPT leading to a loss of hard Zn precipitates in the Al-rich phase [41]. Similarly, in Al-Zn alloys, the significant weakening besides grain refinement is due to the decomposition of a supersaturated solid solution of
Zn in an Al-rich phase during HPT [37–39]. Moreover, a recent report demonstrates that strain softening within a Zn–Al alloy disk after HPT is due to the transformation of lamellar structure into an ultrafine-grained equiaxed structure [22]. Thus, this type of metal exhibiting strain softening demonstrates a decrease in hardness followed by an ultimate saturation with increasing equivalent strain as shown in Fig. 5(c). As was noted earlier, this hardness behavior is essentially the opposite of the conventional strain hardening behavior given in Fig. 5(a).

4. Potentials of the HPT technique for synthesizing hybrid metals

Although there are unique hardness behaviors with strain softening and additional weakening in a limited number of metals after HPT, processing by HPT generally provides the potential for achieving high hardness in most metallic systems due to significant grain refinement. In recent years, because of the introduction of intense plastic strain during processing, HPT has also been applied for the consolidation of metallic powders [42–50] and bonding of machining chips [51,52]. However, there is almost no report examining the validity of HPT for synthesis of bulk hybrid metal from simple commercial metals. Accordingly, in this last section a new approach for the formation of an Al-Mg hybrid system is demonstrated by processing two commercial metal disks of the Al-1050 and ZK60A alloy through HPT.

The present experiments used the commercial alloys of Al-1050 and ZK60A where these alloys demonstrate the general strain hardening behavior as shown earlier in Figs. 1–2. The disks of these alloys were prepared in the same way as demonstrated in the earlier sections and conventional HPT processing was conducted at room temperature under quasi-constrained conditions. In practice, two separate disks of the Al and Mg alloys were piled up with the order of Al/Mg/Al where the Mg disk was placed between two Al disks without placing any glue or using a metal brushing treatment. Then, a set of three piled up disks was processed by HPT at 6.0 GPa for a total of 5 revolutions at a constant rotational speed of 1 rpm.

After processing, an overview of the microstructure was examined by an optical microscope at the vertical cross-section of the processed disk after polishing and chemically etching. Subsequently, the distribution of Vickers microhardness, Hv, was examined over the vertical cross-section of the processed disk. The measurements were conducted to record the individual Hv values following a rectangular grid pattern with an incremental spacing of 0.2 mm. This data set was used to construct a color-coded contour map to visualize the hardness distribution within the disk.

An optical micrograph taken at the vertical cross-section of the Al-Mg disk after HPT for 5 turns is shown in Fig. 6(a) in which the dark area denotes a Mg-rich phase and the brighter area denotes an Al-rich phase. The micrograph shows very unique microstructures including a thick multi-layered structure at the central region within \( r \approx 2.5 \) and there is a very fine dispersion of the Mg-rich phase in the Al-rich matrix phase at the peripheral region. The multi-layers observed at the central region have thicknesses of \( \approx 200 \mu m \) whereas the peripheral region displays a fine dispersion of the Mg-rich phase with thicknesses of \( \approx 5-10 \mu m \) to even a true nano-scale of \( \approx 100-500 \text{ nm} \).

The color-coded contour maps are displayed in Fig. 6(b) where the upper map is for the Al-Mg multi-layered disk after HPT for 5 turns. For comparison purposes, a similar color-coded map is also constructed for the ZK60A alloy after HPT for 5 turns and it is shown in the lower row in Fig. 6(b) where the Hv values on the mid-section plane are equivalent to those shown in Fig. 2 for the same sample after 5 turns. It should be recalled that the saturated hardness value after HPT for 5 turns is \( \approx 65 \) for the Al-1050 as shown in Fig. 1 and \( \approx 110 \) for the ZK60A alloy as shown in Figs. 2 and 6(b).

The hardness values are indicated by the color key on the right and it is apparent that the central region containing the thick layered structure demonstrates low Hv values of \( \approx 70 \) in the Al-Mg disk after HPT for 5 turns. This hardness value is
reasonably similar to the saturated hardness of the Al-1050 disk after HPT for 5 turns. However, high hardness with a maximum of Hv = 130 was achieved in the peripheral region where the fine Mg phase is homogeneously distributed in the processed Al–Mg disk. This high hardness is even higher than the hardness values observed in the ZK60A disk after HPT for 5 turns and the formation of a new Al–Mg system with intermetallic compounds may provide the possibility of improving the upper limit on the maximum hardness value in the Al–Mg system through HPT processing.

Multi-layered microstructures of the Al–Mg systems from separate plates of Al and Mg alloys were constructed through hot/cold-rolling [53,54] and accumulative-roll bonding [55–57]. These experiments demonstrated the successful construction of intermetallic compounds at the interfaces of the metal layers but these techniques are not feasible to introduce fine dispersions of dissimilar phases as shown in Fig. 6(a) by HPT processing. Therefore, this report demonstrates that there is a considerable potential for the use of HPT to synthesize new alloy systems from simple metals. Further investigations are now necessary to fully understand the effect of this approach on the detailed microstructural changes in the newly produced metal systems.

5. Summary and conclusions

The hardness evolution toward homogeneity was demonstrated using four representative metals of Al-1050, ZK60A, high-purity Al and Zn–22% Al alloy.

Three separate models of hardness homogeneity were discussed for metals with increasing equivalent strain through HPT. The nature of the variation depends upon the rate of recovery and the microstructural evolution during HPT processing.

There is a great potential for HPT processing to introduce new alloy systems from different metals and further investigations are necessary to fully make use of this approach.

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Conflicts of interest

The authors declare no conflicts of interest.

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