Strengthening and weakening in the processing of ultrafine-grained metals

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Abstract

It is now well established that ultrafine-grained (UFG) metals, with average grain sizes below 1 μm, may be produced by applying severe plastic deformation to bulk metals using processing procedures such as equal-channel angular pressing (ECAP) and high-pressure torsion (HPT). Generally, these procedures lead to a significant strengthening in the material but there are some examples where processing by ECAP or HPT produces a significant weakening. This report examines examples of both strengthening and weakening in the processing of UFG metals.

K e y w o r d s: equal-channel angular pressing, high-pressure torsion, severe plastic deformation, ultrafine grains

1. Introduction

The grain size of a polycrystalline metal is one of the most important structural parameters. Specifically, the grain size determines the strength of the material such that the strength is increased when the grain size is reduced. This strengthening is a direct consequence of the Hall-Petch relationship in which the yield stress, σ_y, is given by [1, 2]

$$\sigma_y = \sigma_o + k_y d^{-1/2}, \qquad (1)$$

where σ_o is the lattice friction stress, d is the grain size and k_y is a constant of yielding. The production of a small grain size may also provide an opportunity for achieving a superplastic forming capability at elevated temperatures so that the material may be easily formed into a complex shape [3].

The necessity of developing small grains for strengthening is well known in metallurgical industries where thermomechanical processing is used to achieve grain refinement. Nevertheless, the minimum grain sizes achieved by this processing are typically of the order of a few micrometers and it has proven

impossible, using conventional processing techniques, to produce average grain sizes in the submicrometer range. This situation changed about twenty-five years ago when new processing techniques were developed based on the application of severe plastic deformation (SPD) to conventional bulk solids to produce submicrometer or even nanometer grain sizes [4]. A detailed summary was given recently describing the major SPD processing techniques that are now available for the production of ultrafine-grained (UFG) metals [5] where UFG metals are defined as those metals having average grain sizes below 1 μm. To date, most processing is conducted using either equal-channel angular pressing (ECAP) or high-pressure torsion (HPT) [6] where ECAP involves pressing a bar or rod through a die constrained within a channel that is bent through a sharp angle [7] and HPT involves applying a high pressure and concurrent torsional straining to a sample which is generally in the form of a relatively thin disk [8]. Detailed analysis has demonstrated the general validity of Eq. (1) over a very wide range of grain sizes into the nanometer range [9] although at very small grain sizes, below ~ 50 nm, there is some evidence for the

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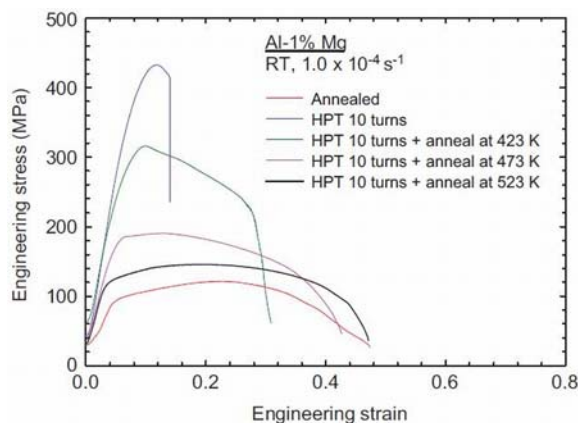
Fig. 1. Stress-strain curves for an Al-1% Mg alloy showing the high strength and lack of ductility after HPT processing through 10 turns and the potential for restoring ductility through short-term (10 minutes) anneals at temperatures from 423 to 523 K [13].

potential of developing super-strength materials [10, 11].

This report is designed to describe the general strengthening that may be achieved using processing by ECAP and HPT. However, it is important to note also that for a limited number of materials there is the possibility that SPD processing may lead to an overall weakening. Accordingly, examples are presented to demonstrate both the strengthening and the weakening that may occur in the processing of UFG metals.

2. Typical strengthening achieved in SPD processing

It is well known that SPD processing generally produces a strong material but this strengthening is accompanied by a loss of ductility. This produces the so-called paradox of strength and ductility in which the strength increases while the elongations to failure decrease [12]. An excellent example of this effect is given by results on an Al-1% Mg solid solution alloy with an initial annealed grain size of $\sim 400\,\mu m$. This alloy was processed by HPT at room temperature (RT) to produce a grain size of $\sim 200\,nm$ and then tested in tension at RT using an initial strain rate of $1.0 \times 10^{-4}\,s^{-1}$. The results are shown in Fig. 1 where the lower curve shows the low strength and relatively high ductility achieved in the annealed material and the upper curve shows the high strength and loss of ductility that is attained immediately after HPT processing [13].

The additional three curves shown in Fig. 1 delineate the effect of taking the HPT-processed samples and annealing for the very short time of 10 min at

temperatures from 423 to 523 K. These short-term anneals lead to some minor grain growth which reduces the overall strength of the material but at the same time the annealing introduces reasonable ductilities with elongations up to > 0.2. It follows from these results that short-term annealing after HPT processing may be an effective and simple procedure for achieving reasonable ductility and overcoming the strength-ductility paradox. Very recently, experiments on an Al-7%Si alloy showed there is also an excellent potential for achieving high strength and high ductility in HPT processing by increasing the number of turns in HPT [14].

3. Examples of strengthening and weakening in metals subjected to SPD processing

3.1. Processing by ECAP

Numerous experiments have demonstrated the use of ECAP for achieving significant strengthening in a very wide range of metals. For example, detailed experiments on a number of commercial aluminum alloys showed that each alloy was strengthened by factors of about two or three after processing by ECAP [15]. Nevertheless, experiments on a spray-cast Al-7034 alloy with an initial grain size of $\sim 2.1\,\mu m$ showed that processing by ECAP at a temperature of 473 K refined the grain size to $\sim 0.3\,\mu m$ but produced a weakening in subsequent tensile testing at room temperature due to the partial loss during ECAP of the hardening metastable η' phase [16]. Accordingly, comprehensive experiments were conducted in compression by cutting small samples from ECAP billets of an Al-6061 alloy which is representative of a strengthening material [15] and the Al-7034 alloy which is representative of a weakening material [16]. These samples were oriented in three mutually perpendicular directions and then tested at RT (298 K) using an initial strain rate for all samples of $5.5 \times 10^{-4}\,s^{-1}$ [17]: each test was terminated at an engineering strain of 30 % and, for comparison purposes, samples were tested in the as-received and unprocessed condition and after processing by ECAP through 1, 2 and 6 passes (p).

The results are shown in Figs. 2 and 3 for the Al-6061 and Al-7034 alloy, respectively [17], where, for simplicity, results are presented only for samples cut in the Z direction where the Z plane is defined as the longitudinal plane parallel to the top surface at the point of exit from the ECAP die [18, 19]. In practice, the results for both alloys showed that samples cut in the three mutually perpendicular directions give essentially the same stress-strain curves thereby confirming a lack of any significant anisotropy.

In Fig. 2, the as-received condition of the Al-6061 alloy shows a gradual yielding and then a significant
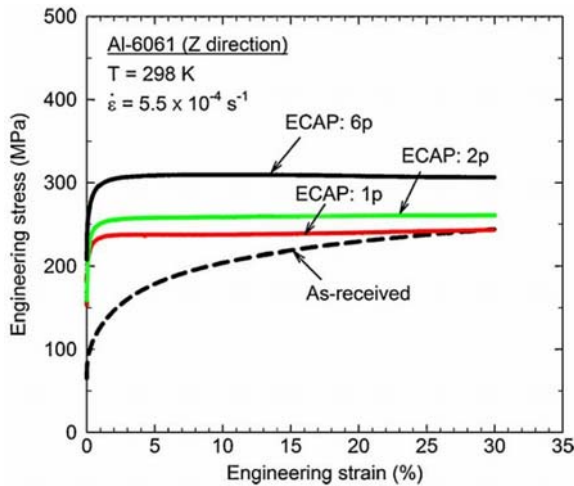
Fig. 2. Stress-strain curves for an Al-6061 alloy showing the strengthening introduced through processing by ECAP from 1 to 6 passes [17].

Fig. 4. Hardness measurements showing strengthening in an Al-7075 alloy through processing by HPT for up to 10 turns [22].
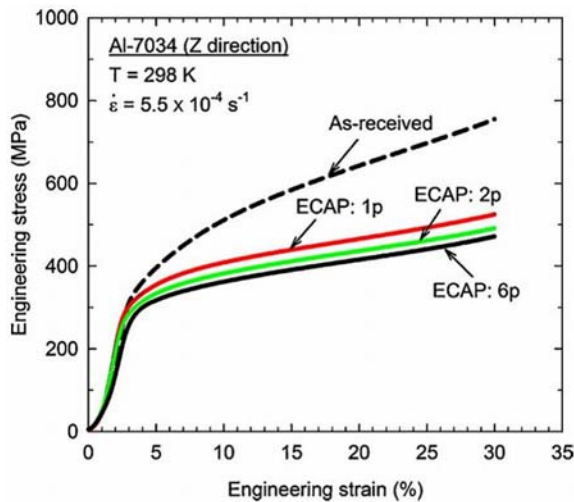

Fig. 3. Stress-strain curves for an Al-7034 alloy showing the weakening introduced through processing by ECAP from 1 to 6 passes [17].

rate of strain hardening whereas after processing by ECAP the strength is increased, there is an abrupt and well-defined yield point and thereafter there is no significant strain hardening up to the termination of the test at an engineering strain of 30 %. It is also apparent that the overall strength of the Al-6061 alloy increases with increasing numbers of ECAP passes. By contrast, for the as-received Al-7034 alloy shown in Fig. 3 there is a gradual yielding and significant strain hardening but after processing by ECAP there is a lower yield stress which tends to decrease with increasing numbers of ECAP passes. It is apparent also that the strain hardening after yielding occurs at a slower rate after ECAP than for the as-received alloy.
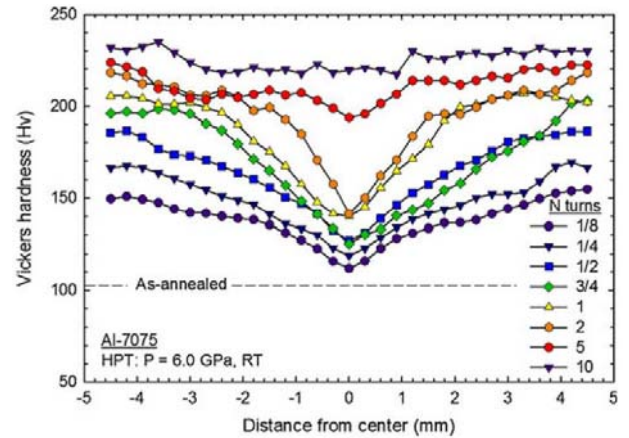
Thus, the results for these two alloys provide a clear demonstration of the occurrence of both strengthening and weakening in alloys processed by ECAP.

3.2. Processing by HPT

3.2.1. Strengthening by HPT

Processing by HPT generally involves the use of a thin disk which is subjected to a high pressure and torsional straining. In practice, the equivalent von Mises strain, $\varepsilon_{\mathrm{eq}}$, imposed on the disk in HPT is given by a relationship of the form [20]:

$$\varepsilon_{\mathrm{eq}} = \frac{2\pi N r}{h\sqrt{3}}, \tag{2}$$

where N is the number of turns in HPT, r is the radial distance from the center of the disk and h is the height (or thickness) of the disk. It follows from Eq. (2) that the strain varies across the disk from zero strain at the disk center where $r = 0$ to a maximum at the edge of the disk. This suggests there should be significant inhomogeneities in the measured hardness values across the disk but in practice microstructural evolution occurs during processing and after large numbers of turns it is often possible to achieve a reasonable level of homogeneity. Furthermore, this evolution to homogeneity is consistent with the predictions of a theoretical analysis based on the application of strain gradient plasticity modeling [21].

The strength imposed in HPT processing may be measured in a very simple way by taking hardness measurements across diameters of the disks after processing. An example is shown in Fig. 4 for an Al-7075 alloy processed by HPT at RT under a pressure, P,
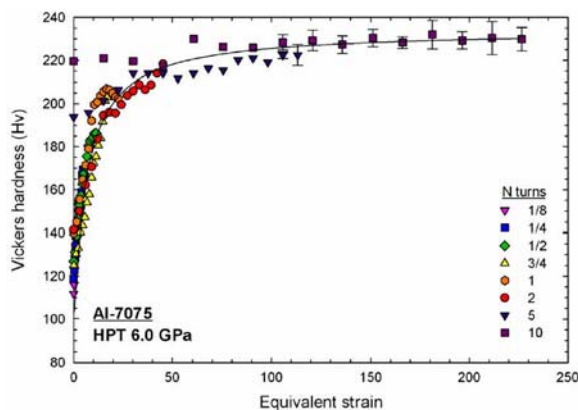
Fig. 5. Values of the Vickers hardness plotted against equivalent strain for an Al-7075 alloy processed by HPT for up to 10 turns [24].

Fig. 6. Hardness measurements showing weakening in a Zn-22%Al alloy through processing by HPT for up to 20 turns [28].

of 6.0 GPa [22]. Measurements are shown for various numbers of turns from 1/8 to 10 and the individual datum points are plotted as a function of the position on the disk with the lower broken line denoting the initial hardness in the annealed condition of Hv ≈ 102. It is readily apparent that the hardness increases rapidly around the edge of the disk in the early stages of processing but after 10 turns there is a reasonable level of homogeneity across the whole disk.

It was suggested in early experiments on HPT processing that the individual hardness measurements after different numbers of revolutions may be conveniently correlated by plotting the hardness values against the predicted equivalent strain calculated using Eq. (2) [23]. This type of plot is shown in Fig. 5 using the experimental points from Fig. 4 and with the 95 % error bars shown at the higher equivalent strains [24]. In Fig. 5 it is apparent that all of the datum points now scatter around a single line and there is a saturation hardness of Hv ≈ 230 at equivalent strains higher than ∼ 180. Similar sets of results are also now available where samples are processed initially by ECAP and then by HPT [24, 25].

3.2.2. Weakening by HPT

The strengthening associated with HPT processing, as depicted in Figs. 4 and 5, is representative of a very wide range of metals. Nevertheless, it has been reported that some metals exhibit a weakening on processing by HPT. This weakening effect was first described in detail when processing a Zn-22%Al eutectoid alloy [26, 27] and subsequently the effect was widely documented. An example of weakening is shown in Fig. 6 where the individual hardness measurements are plotted for a Zn-22%Al alloy processed by HPT at RT using a pressure of 6.0 GPa and an anvil rotation speed of 1 rpm [28]. The
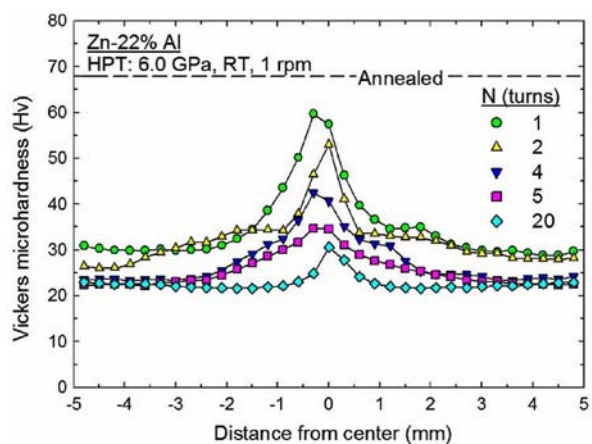
results are shown after 1, 2, 4, 5 and 20 turns and they demonstrate that initially there is a sharp drop in hardness at the edges of the disk with much less weakening in the center but gradually, with increasing numbers of turns, the hardness decreases in the center and, except in the immediate center of the disk, all hardness measurements become essentially identical after 20 turns.

The weakening in the Zn-Al alloy on processing by HPT has similarities to the processing by ECAP of the spray-cast Al-7034 alloy shown in Fig. 3 where the weakening was due to a partial loss of the metastable η' phase. For the Zn-Al alloy, it was shown in early observations using transmission electron microscopy that HPT processing at RT leads to a significant reduction in the distribution of rod-shaped precipitates of stable hexagonal close-packed Zn which are contained within the Al-rich grains in the annealed condition but become absorbed by the Zn-rich grains during processing [29, 30].

Weakening by HPT processing has now been reported in the Pb-62%Sn eutectic alloy [31, 32], the Bi-42%Sn eutectic alloy [33], an Al-30%Zn alloy [34, 35] and in samples of pure Pb, Sn and In when processing by HPT at RT [36]. However, the weakening is not associated especially with eutectic or eutectoid alloys because a hardening effect, similar to most metals, was recorded in the Al-33%Cu eutectic alloy [37].

Figure 7 shows a plot of hardness versus equivalent strain and, as in Fig. 5, all of the datum points from many tests now superimpose to give an essentially continuous curve [28]. In practice, this curve is essentially a mirror-image of the standard result for strengthening as shown in Fig. 5 because the points now decrease to a saturated minimum hardness at Hv ≈ 23 which is achieved at equivalent strains greater than ∼ 40 and thereafter the hardness values remain reasonably con-
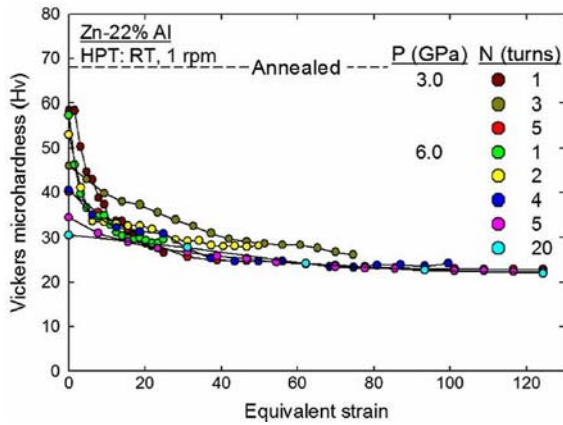
Fig. 7. Values of the Vickers hardness plotted against equivalent strain for a Zn-22%Al alloy processed by HPT for up to 20 turns [28].

stant. In Fig. 7, all of the points superimpose although data are shown for two different HPT pressures of 3.0 and 6.0 GPa.

4. Discussion

The procedures of ECAP and HPT are regularly used to achieve grain refinement in bulk metals. In both procedures, a material is deformed under an applied pressure and generally this produces a significant reduction in the average size of the grains. It follows from the Hall-Petch relationship in Eq. (1) that this grain refinement will usually produce a strengthening of the material and there are now many examples showing this effect. But for some materials the imposition of SPD processing may instead produce a weakening effect. Examples of weakening are shown in Fig. 3 for an Al-7034 alloy processed by ECAP and in Fig. 6 for a Zn-22%Al alloy processed by HPT.

The recording of hardness measurements provides a simple and expedient procedure for determining the occurrence of strengthening or weakening in HPT. A recent comprehensive review summarizes the various effects that may occur during HPT processing and describes the occurrence of weakening when processing by HPT [38]. Accordingly, it is important to note that, although most materials are strengthened by HPT, there remain some materials where the processing produces grain refinement but at the same time the material is weakened. In practice, therefore, care must be taken in all investigations to determine whether SPD processing strengthens or weakens a material.

Fig. 8. The author with Zuzana and Palko on a bridge in Karlovy Vary in 1988.

Fig. 9. In a hotel in Moscow in May 1989 showing standing (from left) Mikhail Myshlyaev of Moscow, Ruslan Valiev of Ufa, Zuzana, Palko, Mady and the author.

5. Postscript

It is a pleasure for me to have this opportunity to contribute to this special issue of *Kovove Materialy* celebrating the 80th birthday of Professor Pavel ("Palko") Lukáč. I have known Palko for almost forty years, dating back to the 1970s when my research was concentrated on high temperature creep and I made several trips to the Institute of Physical Metallurgy in Brno for discussions with the late Josef Čadek and Václav Sklenička. Always I traveled with my wife, Mady, and we would arrive in Prague before taking the bus or train to Brno. On almost every visit, we were met in Prague by Palko, usually with Zuzana Trojanová, and we spent one or two days at Charles University or exploring the sights of Bohemia.

The photo in Fig. 8 was taken in 1988 on a trip to Karlovy Vary and it shows Zuzana and Palko with me on one of the bridges. The second photo in Fig. 9 was taken in a hotel in Moscow in May 1989 on my first visit to the Soviet Union and it shows (from the left) Mikhail Myshlyaev of Moscow, Ruslan Valiev of Ufa, Zuzana, Palko, Mady and me. A few days later we traveled to Ufa by plane with Zuzana and Palko accompanied by Ruslan Valiev.

I want to take this opportunity to wish Palko a very happy 80th birthday with good health and much happiness in the years ahead. I am very much looking forward to seeing him again on my next visit to Prague.

Acknowledgements

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