

Available online at www.sciencedirect.com

jmr&t
Journal of Materials Research and Technology
www.jmrt.com.br



Original Article

Comparisons of self-annealing behaviour of HPT-processed high purity Cu and a Pb–Sn alloy[☆]



Yi Huang^{a,*}, Shima Sabbaghianrad^b, Abdulla I. Almazrouee^c, Khaled J. Al-Fadhlah^d, Saleh N. Alhajeri^c, Nian Xian Zhang^a, Terence G. Langdon^a

^a Materials Research Group, Faculty of Engineering and the Environment, University of Southampton, Southampton, United Kingdom

^b Departments of Aerospace & Mechanical Engineering and Materials Science, University of Southern California, Los Angeles, United States of America

^c Department of Manufacturing Engineering, College of Technological Studies, P.A.A.E.T., Shuwaikh, Kuwait

^d Department of Mechanical Engineering, College of Engineering & Petroleum, Kuwait University, Safat, Kuwait

ARTICLE INFO

Article history:

Received 13 June 2017

Accepted 13 July 2017

Available online 10 August 2017

Keywords:

High-pressure torsion

High purity Cu

Pb–62%Sn alloy

Self-annealing

ABSTRACT

Early published results have demonstrated that high purity Cu and a Pb–62% Sn alloy exhibit very different behaviour during high-pressure torsion (HPT) processing at room temperature and subsequent room temperature storage. High purity Cu showed strain hardening behaviour with a refined grain structure during HPT processing whereas a Pb–62% Sn alloy displayed a strain weakening behaviour because the hardness values after HPT processing were significantly lower than in the initial as-cast condition even though the grain size was reduced. During room temperature storage after HPT processing, high purity Cu with lower numbers of rotations softened with the time of storage due to local recrystallization and abnormal grain growth whereas the Pb–62% Sn alloy hardened with the time of storage accompanied by grain growth. Through comparisons and analysis, it is shown that the low absolute melting point and the high homologous temperature at room temperature in the Pb–62% Sn alloy contribute to the increase in hardness with coarsening grain size during room temperature storage.

© 2017 Brazilian Metallurgical, Materials and Mining Association. Published by Elsevier Editora Ltda. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

1. Introduction

High-pressure torsion (HPT) is one of the most popular severe plastic deformation (SPD) techniques due to the significant grain refinement and mechanical strength enhancement in

processed metallic materials [1–3]. Many different metals and alloys were successfully processed by HPT at room temperature [4] and different materials display different strength (hardness) evolutions during HPT processing [5]. There are three typical hardness evolution tendencies: microhardness evolution with no recovery in most metals and alloys,

[☆] Paper was a contribution part of the 3rd Pan American Materials Congress, February 26th to March 2nd, 2017.

* Corresponding author.

E-mail: y.huang@soton.ac.uk (Y. Huang).

<http://dx.doi.org/10.1016/j.jmrt.2017.07.002>

2238-7854/© 2017 Brazilian Metallurgical, Materials and Mining Association. Published by Elsevier Editora Ltda. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

microhardness evolution with softening in some materials with high stacking fault energy (such as high purity Al), microhardness evolution with weakening in two phase alloys (such as Zn–Al and Pb–Sn alloys) [5].

In order to benefit from the grain refinement and strength enhancement developed from HPT processing, it is expected that HPT-processed materials should have good thermal stability to maintain the ultrafine-grained microstructures and high strength at the application environment. There are very limited studies on the microstructural stability during room temperature (RT) storage in HPT-processed metals and alloys although this is an important issue and has great influence on the material mechanical properties and therefore on the potential applications.

There are several reports on the thermal stability of HPT-processed Cu at various annealing conditions [6–9]. Long-term (~1.5 years) room temperature (RT) annealing of cryogenically-rolled Cu was examined [10] and it was found that the RT annealing processes include grain boundary migration (accompanied by annealing twinning) and recovery. There are also reports on the RT self-annealing of HPT-processed Cu for 100h [11] and up to 6 weeks [12] and both reported a continuous hardness decrease during the RT storage.

For the two-phase Pb–62% Sn alloy, several reports demonstrate significant grain refinement by ECAP and HPT [13–17]. Since the Pb–62% Sn alloy has a low absolute melting point of 456 K, the thermal stability was investigated through self-annealing behaviour at RT [18,19] and it was reported that the hardness increased during RT storage after HPT processing.

It is apparent that pure Cu and the Pb–Sn alloy display different hardness evolution tendencies during RT storage after HPT processing [12,13,18,19]. Considering the self-annealing behaviour in pure Cu and the Pb–Sn alloy, the hardness and microstructures recorded in the HPT-processed materials may depend upon the time that elapses between the HPT processing and the microstructure/mechanical strength investigations. This type of delay may be of the order of several weeks, months or even years, and would have great impact on any understanding of microstructure/mechanical strength development during HPT processing. Accordingly, it is important to compare the self-annealing behaviour between HPT-processed high purity Cu and the Pb–62% Sn alloy during RT storage. Through these comparisons, it is possible to achieve two objectives. The first is to construct whole scenarios of microstructure and mechanical strength development in HPT-processed high purity Cu and the Pb–62% Sn alloy during RT storage where this will be useful for RT practical applications. The second is to clarify whether the observed microstructures and measured mechanical properties originate from HPT processing or from RT storage after HPT processing. It is necessary to pay special attention to the time when the microstructure/mechanical strength investigations are conducted in order to successfully correlate the microstructure/mechanical strength investigated to the HPT processing or to the self-annealing process during post-HPT RT storage.

2. Self-annealing behaviour in HPT-processed high purity Cu and Pb–62% Sn alloy

Early publications reported the self-annealing behaviour in HPT-processed high purity Cu [11,12] and Pb–62% Sn alloy [18,19] respectively. We will review and compare the self-annealing behaviour in HPT-processed high purity Cu and Pb–62% Sn alloy in this section.

2.1. Self-annealing behaviour in HPT-processed high purity Cu

The microhardness evolution and microstructures development within 48 h after HPT processing to 1/2 turn and subsequent RT storage for periods of 1, 3 and 4 weeks reported earlier [12] were utilised to demonstrate the self-annealing behaviour in HPT-processed high purity Cu.

Before HPT processing, the annealed high purity Cu had an initial hardness of ~41 Hv and average grain size of ~85 μm . The hardness measured within 48 h of HPT processing was taken as the representative hardness of HPT processing. Inspection of Fig. 1 shows that after 1/2 turn HPT processing the hardness values increase significantly around the disc edge area but there are lower hardness values near the disc centre area, and the hardness values increase from the centre to a position ~1.5 mm away from the centre in order to attain a plateau level. This is consistent with the strain hardening behaviour in most metals during HPT processing [5,20]. Hardness measured in a planned time schedule during RT storage for 1, 3 and 4 weeks in Fig. 1 shows the self-annealing behaviour during RT storage. From the disc centre to a position ~1.5 mm from the centre there was almost no hardness change from week 1 to week 4. But from a position ~1.5 mm from the disc centre to the edge the hardness values showed significant changes from week 1 to week 4. Thus, after 1 week there was a slight hardness drop, after 3 weeks the hardness drop became more significant, and after 4 weeks the hardness values attained a stable state without any further drop.

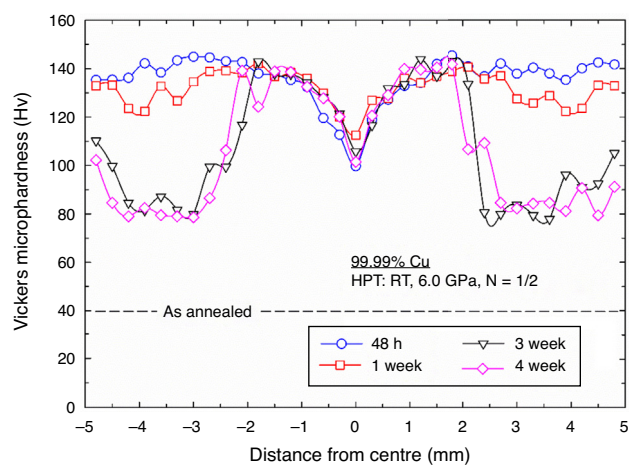


Fig. 1 – Hardness recorded along disc diameters after HPT processing for 1/2 turn in high purity Cu [12].

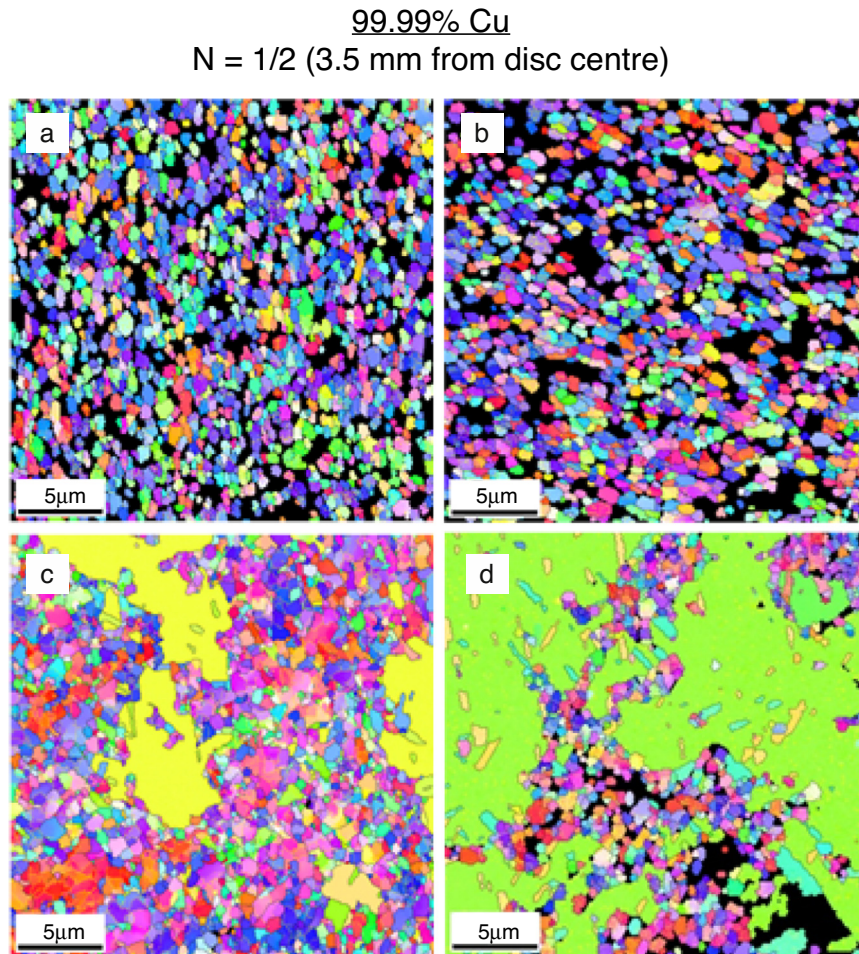


Fig. 2 – Microstructure development in HPT processed high purity Cu during RT storage at (a) 48 h, (b) 1 week, (c) 3 weeks and (d) 4 weeks [12].

In summary, for the 1/2 turn high purity Cu sample there exists significant local hardness drops from a position ~ 1.5 mm from the disc centre during 4 weeks of RT storage, such that the hardness drops from ~ 140 Hv within 48 h after HPT to ~ 80 Hv in 4 weeks after HPT.

The corresponding microstructures development after HPT processing (within 48 h) and subsequent RT storage for the periods of 1, 3 and 4 weeks in the 1/2 turn high purity Cu sample are shown in Fig. 2. Significant grain refinement occurred in the disc edge area (3.5 mm from disc centre) after only 1/2 turn of HPT processing because the grain size was refined from ~ 85 μm before HPT processing to ~ 0.5 μm after HPT processing. During the subsequent RT storage, in week 1 the microstructure showed similar characteristics to the microstructure within 48 h after HPT processing maintaining a significantly refined grain structure. With RT storage times up to 3 and 4 weeks, the disc edge area encountered significant microstructure changes such that a few grains grew to very large sizes while most grains remained as fine grain structures. These large grains continued to grow from week 3 to week 4 and expanded to occupy almost two-thirds of the observation area after week 4 where abnormal grain growth occurred. The abnormal grain growth at the disc edge

area during RT storage (Fig. 2) correlates well with the local hardness drop shown in Fig. 1.

2.2. Self-annealing behaviour in HPT-processed Pb–62% Sn alloy

The microhardness evolution and microstructure development within 24 h after HPT processing to 1 turn and subsequent RT storage for certain periods of time published earlier [18] were chosen to depict the self-annealing behaviour in HPT-processed Pb–62%Sn alloy.

The Pb–62% Sn alloy had a hardness of ~ 10 Hv in the as-cast condition. The hardness measured on the same day as the HPT processing is labelled as 0 day and is regarded as the representative hardness of the HPT processing. Fig. 3 shows the hardness development after 1 turn of HPT processing and subsequent RT storage for 4 and 9 days in the Pb–62% Sn alloy. After 1 turn of HPT processing, the disc centre has higher hardness values than the edge area but both the disc centre and the edge area have lower hardness values than the as-cast condition. This is typical of strain weakening behaviour as reported also in the Zn–22% Al alloy [15,21]. During subsequent RT storage, the hardness in the disc edge area increased after 4

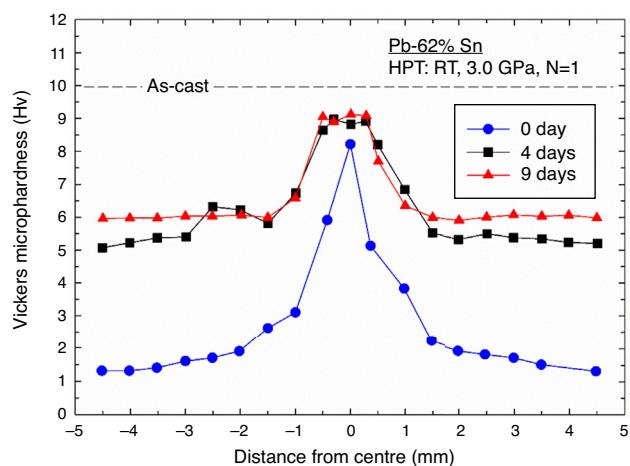


Fig. 3 – Hardness recorded along disc diameters after HPT processing for 1 turn in Pb–62% Sn alloy [18].

days of RT storage and slightly increased further after 9 days, whereas in the disc centre area the hardness also increased after 4 days of RT storage but essentially saturated after 9 days.

The microstructure development after HPT processing (0 day) and subsequent RT storage for 2 and 11 days in the 1 turn Pb–62% Sn alloy sample are shown in Fig. 4 where the top and bottom row correspond to the Sn grains and Pb grains, respectively. After HPT processing, the disc edge area has a reasonably refined structure of fine Sn grains and Pb grains compared to the coarse structure in the as-cast condition. During RT storage, the grain size of both the Sn grains and the Pb grains increased after 2 days of RT storage and then exhibited a further increase after 11 days. The grain growth at the disc edge area during RT storage (Fig. 4) and the local hardness increase (Fig. 3) are not consistent with the Hall–Petch relationship.

In summary, during HPT processing, high purity Cu showed strain hardening behaviour whereas the Pb–62% Sn alloy displayed strain weakening behaviour. With subsequent RT storage, a microstructure coarsening occurred in both high purity Cu and the Pb–62% Sn alloy but the hardness evolution with storage time displayed different tendencies. Thus, in high purity Cu there was abnormal grain growth and a corresponding hardness drop with storage time whereas in the Pb–62% Sn alloy there was normal grain growth but an increase in hardness with storage time.

3. Self-annealing mechanism in HPT-processed high purity Cu and Pb–62% Sn alloy

Based on comparisons of the different self-annealing behaviour in HPT-processed high purity Cu and Pb–62% Sn alloy, the mechanisms behind the different self-annealing behaviours are now discussed in this section.

3.1. Softening behaviour during self-annealing in HPT-processed high purity Cu

The microstructure development after HPT processing and subsequent RT storage in high purity Cu demonstrates that abnormal grain growth occurs after 3 and 4 weeks of RT storage. Based on the EBSD data, the number fraction of $\Sigma 3$ boundaries was $\sim 2.4\%$ within 48 h after HPT processing, $\sim 2.4\%$ after 1 week, $\sim 5.4\%$ after 3 weeks and $\sim 14.3\%$ after 4 weeks [12]. In FCC metals, the occurrence of $\Sigma 3$ boundaries is dominated by the formation of annealing twins [22]. Thus, based on the increase of $\Sigma 3$ boundaries with longer RT storage and significant hardness drop from 140 Hv to 80 Hv in the 1/2 turn sample shown in Fig. 1, it is reasonable to conclude that recrystallization occurred during RT storage. Considering that many short twins exist within the abnormal growth of grains in Fig. 2c and d, it is assumed that either grain boundary migration or crystal lattice rotation may make contributions to the disappearance of grain boundaries and the coalescence of adjacent grains to form the abnormal growth of grains. Abnormal grain growth leads to a softening behaviour during self-annealing in HPT-processed high purity Cu.

3.2. Hardening behaviour during self-annealing in HPT-processed Pb–62% Sn alloy

The Pb–62% Sn alloy displayed grain growth (Fig. 4) and the hardness increased (Fig. 3) during RT storage. Thus, HPT-processed Pb–62% Sn alloy showed a hardening behaviour during the self-annealing process after HPT processing but the grain size and mechanical strength were not consistent with the Hall–Petch relationship in which fine grains have higher strength and coarse grains denote lower strength.

The grain size is the most important and dominant structural parameter in polycrystalline materials. In the low-temperature regime, the yield stress σ_y varies with the grain size, d , through the Hall–Petch relationship [23,24]

$$\sigma_y = \sigma_0 + k_y d^{-1/2} \tag{1}$$

where σ_0 is the lattice friction stress and k_y is a constant of yielding. Conversely, in the elevated temperature regime, generally above $\sim 0.5T_m$ where T_m is the absolute melting temperature, diffusion is important and the steady-state strain rate during plastic flow is described by the phenomenological creep equation [25]:

$$\dot{\epsilon} = \frac{ADGb}{kT} \left(\frac{b}{d}\right)^p \left(\frac{\sigma}{G}\right)^n \tag{2}$$

where A is a dimensionless constant, D is the appropriate diffusion constant ($D = D_0 \exp(-Q/RT)$), where D_0 is a frequency factor, Q is the activation energy, R is the gas constant and T is the absolute temperature), G is the shear modulus, b is the Burgers vector, k is Boltzmann’s constant, σ is the flow stress and n and p are the stress and inverse grain size exponents, respectively.

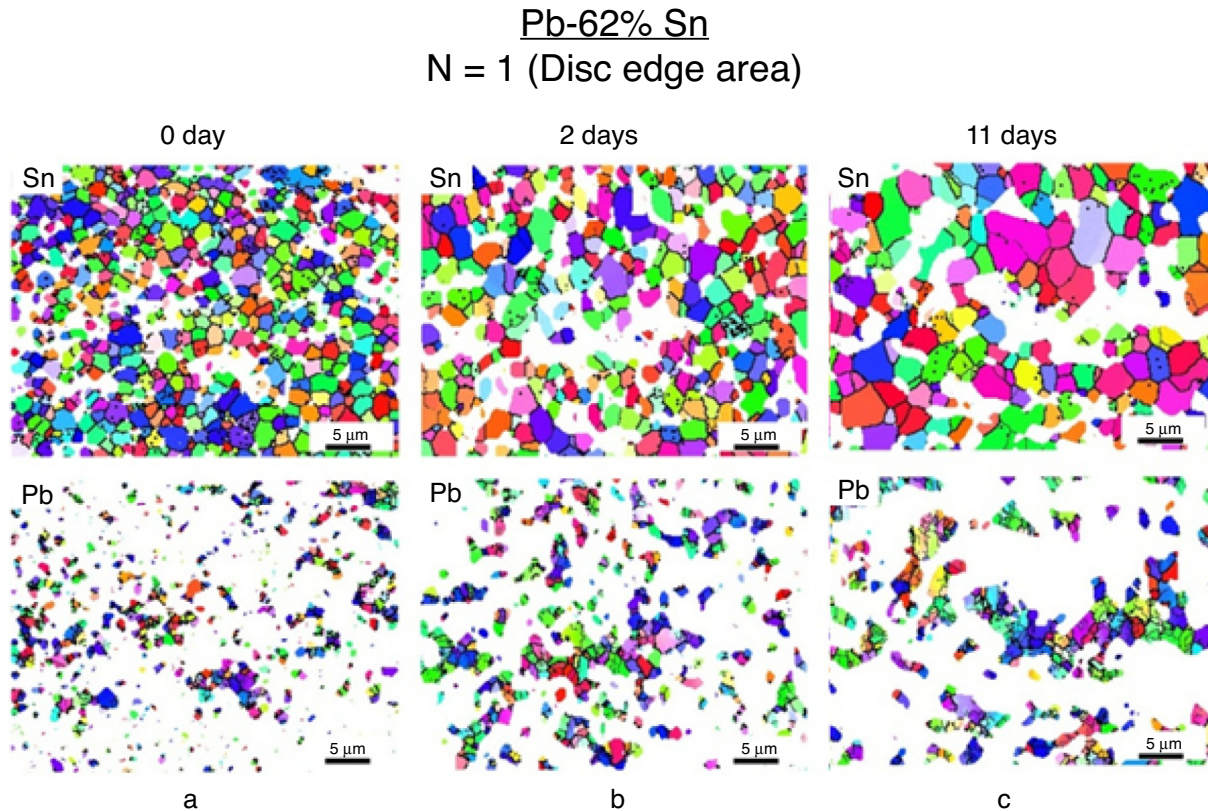


Fig. 4 – Microstructure development in HPT processed Pb–62% Sn alloy during RT storage at (a) 0 day, (b) 2 days and (c) 11 days [18].

The Pb–62% Sn alloy has a low absolute melting temperature, T_m , of ~ 456 K, so that room temperature (298 K) is equivalent to $\sim 0.65T_m$ and the hardness measured at RT represents the mechanical behaviour at $\sim 0.65T_m$. Therefore, the high homologous temperature at RT indicates that the Pb–62% Sn alloy follows the high temperature deformation behaviour described in Eq. (2) even at RT.

The load applying speed of the FM-300 microhardness tester is $\sim 60 \mu\text{m/s}$ and this exerts a very fast deformation strain rate of $>10^{-2} \text{ s}^{-1}$ to the Pb–62% Sn alloy during the hardness measurement. The strain rate sensitivity measurements at RT showed that only at strain rates below $\sim 10^{-2} \text{ s}^{-1}$ the HPT-processed Pb–62% Sn alloy had strain rate sensitivities of ~ 0.5 [17,19]. Therefore, during the hardness measurement, due to the fast load applied by the microhardness tester, the Pb–62% Sn alloy has a low strain rate sensitivity. This means that the strain rate, imposed during the hardness measurements, is not the critical factor for the flow stress in Eq. (2) but rather the grain size has a large effect on the material strength. In the case of the Pb–62% Sn alloy, a coarse grain structure leads to a high strength at temperatures above $\sim 0.5T_m$. When considering the high homologous temperature of the Pb–62% Sn alloy at room temperature, the measured hardness values are a reflection of the high temperature deformation behaviour, therefore leading to the phenomenon that a hardness increase is accompanied by grain coarsening during RT storage.

4. Summary

1. High purity Cu shows strain hardening behaviour during HPT processing. Self-annealing is important in the 1/2 turn sample and there is a significant drop in the hardness values near the edges of the discs after RT storage for 3–4 weeks where this is associated with abnormal grain growth.
2. The Pb–62% Sn alloy displays a strain weakening behaviour after 1 turn of HPT processing. Grain growth occurs at the disc edge area during subsequent RT storage but the hardness increases with increasing grain size.
3. The high homologous temperature at RT, $\sim 0.65T_m$, plays an important role in the hardness increase with increasing grain size during RT storage in the HPT-processed Pb–62% Sn alloy.

Conflicts of interest

The authors declare no conflicts of interest.

Acknowledgements

This research was presented at the 3rd Pan American Materials Congress held as part of the TMS Annual Meeting in

San Diego, California, on February 27–March 2, 2017. The work was supported in part by the Kuwait University General Facility under Grant No. GE 01/07, in part by the National Science Foundation of the United States under Grant No. DMR-1160966 and in part by the European Research Council under ERC Grant Agreement No. 267464-SPDMETALS.

REFERENCES

- [1] Langdon TG. Twenty-five years of ultrafine-grained materials: achieving exceptional properties through grain refinement. *Acta Mater* 2013;61:7035–59.
- [2] Zhilyaev AP, Langdon TG. Using high-pressure torsion for metal processing: fundamentals and applications. *Prog Mater Sci* 2008;53:893–979.
- [3] Huang Y, Langdon TG. Advances in ultrafine-grained materials. *Mater Today* 2013;16(3):85–93.
- [4] Edalati K, Horita Z. A review on high-pressure torsion (HPT) from 1935 to 1988. *Mater Sci Eng A* 2016;652: 325–52.
- [5] Kawasaki M. Different models of hardness evolution in ultrafine-grained materials processed by high-pressure torsion. *J Mater Sci* 2014;49:18–34.
- [6] Jiang HG, Zhu YT, Butt DP, Alexandrov IV, Lowe TC. Microstructural evolution, microhardness and thermal stability of HPT-processed Cu. *Mater Sci Eng A* 2000;290:128–38.
- [7] Schafner E, Kerber MB. Microstructural investigation of the annealing behaviour of high-pressure torsion (HPT) deformed copper. *Mater Sci Eng A* 2007;462:139–43.
- [8] Čížek J, Janeček M, Srba O, Kužel R, Barnovská Z, Procházka I, et al. Evolution of defects in copper deformed by high-pressure torsion. *Acta Mater* 2011;59:2322–9.
- [9] Zhilyaev AP, Sergeev SN, Langdon TG. Electron backscatter diffraction (EBSD) microstructure evolution in HPT copper annealed at a low temperature. *J Mater Res Technol* 2014;3:338–43.
- [10] Konkova T, Mironov S, Korznikov A, Semiatin SL. On the room-temperature annealing of cryogenically rolled copper. *Mater Sci Eng A* 2011;528:7432–43.
- [11] Zhilyaev AP, Langdon TG. Long-term self-annealing of copper and aluminium processed by high-pressure torsion. *J Mater Sci* 2014;49:6529–35.
- [12] Huang Y, Sabbaghianrad S, Almazroue AI, Al-Fadhalah KJ, Alhajeri SN, Langdon TG. The significance of self-annealing at room temperature in high purity copper processed by high-pressure torsion. *Mater Sci Eng A* 2016;656:55–66.
- [13] Kawasaki M, Mendes AdeA, Sordi VL, Ferrante M, Langdon TG. Achieving superplastic properties in a Pb–Sn eutectic alloy processed by equal-channel angular pressing. *J Mater Sci* 2011;46:155–60.
- [14] El-Danaf EA, Khalil KA, Soliman MS. Effect of equal-channel angular pressing on superplastic behavior of eutectic Pb–Sn alloy. *Mater Des* 2012;34:235–41.
- [15] Kawasaki M, Ahn B, Langdon TG. Microstructural evolution in a two-phase alloy processed by high-pressure torsion. *Acta Mater* 2010;58(3):919–30.
- [16] Zhang NX, Kawasaki M, Huang Y, Langdon TG. Microstructural evolution in two-phase alloys processed by high-pressure torsion. *J Mater Sci* 2013;48:4582–91.
- [17] Zhang NX, Kawasaki M, Huang Y, Langdon TG. Influence of grain size on superplastic properties of a two-phase Pb–Sn alloy processed by severe plastic deformation. *J Mater Metall* 2015;14(4):255–62.
- [18] Zhang NX, Kawasaki M, Huang Y, Langdon TG. The significance of self-annealing in two-phase alloys processed by high-pressure torsion. *IOP Conf Series Mater Sci Eng* 2016;63, 012126(1-9).
- [19] Zhang NX, Kawasaki M, Huang Y, Langdon TG. Self-annealing in a two phase Pb–Sn alloy after processing by high-pressure torsion. *Mater Sci Eng A* 2016;666:350–9.
- [20] Kawasaki M, Figueiredo RB, Huang Y, Langdon TG. Interpretation of hardness evolution in metals processed by high-pressure torsion. *J Mater Sci* 2014;49:6586–96.
- [21] Kawasaki M, Ahn B, Langdon TG. Significance of strain reversals in a two-phase alloy processed by high-pressure torsion. *Mater Sci Eng A* 2010;527:7008–16.
- [22] Field DP, Bradford LT, Nowell MM, Lillo TM. The role of annealing twins during recrystallization of Cu. *Acta Mater* 2007;55:4233–41.
- [23] Hall EO. The deformation and ageing of mild steel: III. Discussion of results. *Proc Phys Soc B* 1951;64:747–53.
- [24] Petch NJ. The cleavage strength of polycrystals. *J Iron Steel Inst* 1953;174:25–8.
- [25] Langdon TG. Creep at low stresses: an evaluation of diffusion creep and Harper–Dorn creep as viable creep mechanisms. *Metall Mater Trans A* 2003;33:249–59.