J Mater Sci (2008) 43:7360–7365
DOI 10.1007/s10853-008-2771-2

The development of internal cavitation in a superplastic zinc–aluminum alloy processed by ECAP

Megumi Kawasaki · Terence G. Langdon

Received: 11 February 2008 / Accepted: 2 June 2008 / Published online: 13 July 2008
© Springer Science+Business Media, LLC 2008

Abstract A Zn-22% Al eutectoid alloy was processed by Equal-Channel Angular Pressing (ECAP) to produce an ultrafine grain size and then pulled in tension at elevated temperatures to evaluate the role of internal cavitation under superplastic conditions. Tensile testing yielded a highest elongation of 2,230% at a strain rate of $1.0 \times 10^{-2}\ s^{-1}$ at 473 K representing high strain rate superplasticity. Quantitative cavity measurements were taken to investigate the significance of the internal cavities formed during superplastic deformation. The results demonstrate that cavity nucleation occurs continuously throughout superplastic flow, and there is a transition in the cavity growth mechanism from superplastic diffusion growth at the smaller cavity sizes to plasticity-controlled growth at the larger sizes.

Introduction

It is well-documented that superplastic ductilities are achieved in metallic materials when the grain sizes are very small, typically <10 μm, and when specimens are pulled at high temperatures at or above $\sim 0.5T_m$ where T_m is the absolute melting temperature of the material [1]. Although commercial superplastic alloys generally have grain sizes within the range of ~2–5 μm, procedures were developed recently for the fabrication of bulk materials having exceptionally small grain sizes, usually within the submicrometer or even the nanometer range, through the application of Severe Plastic Deformation (SPD) [2].

M. Kawasaki (✉) · T. G. Langdon
Departments of Aerospace & Mechanical Engineering and Materials Science, University of Southern California, Los Angeles, CA 90089-1453, USA
e-mail: mkawasak@usc.edu

T. G. Langdon
Materials Research Group, School of Engineering Sciences, University of Southampton, Southampton SO17 1BJ, UK

Several different SPD processing techniques are now available but the most promising, and the method receiving the most attention, is Equal-Channel Angular Pressing (ECAP), where a sample is pressed repetitively through a die without incurring any change in the cross-sectional dimensions [3]. This technique is now widely used for the production of ultrafine-grained (UFG) materials with grain sizes generally in the range of ~100 to 1,000 nm. Accordingly, and as first suggested several years ago [4], it should be possible to make use of the grain refinement introduced by ECAP to produce materials having excellent superplastic properties and, because of the exceptionally small grain sizes, with the superplastic regimes occurring at very rapid strain rates. The occurrence of high strain rate superplasticity in materials processed by ECAP was demonstrated in several reports [5–7] and there are now direct demonstrations of the capability of developing superplastic flow in a range of metallic alloys processed by ECAP [8]. A recent comprehensive review provided a tabulation of the numerous reports describing the occurrence of superplasticity after processing by ECAP [9].

Very early research on the Zn-22% Al eutectoid alloy provided the first evidence for the occurrence of cavitation during superplastic flow [10], and subsequently a detailed evaluation of the cavitation behavior was conducted for this alloy [11]. More recently, there have been numerous reports of cavity development in conventional superplastic metallic alloys. For example, a recent investigation described the development of cavities leading to premature fracture in a magnesium–lithium two-phase alloy [12]. There are also numerous reports of cavity nucleation and growth during superplastic flow in several different alloys [13, 14].

To date, there are only a small number of reports describing the development of internal cavitation during superplastic flow in materials processed by ECAP [15–20], and the characteristics of these cavities are not well documented. Accordingly, it is important to investigate the role of cavitation in representative materials processed by ECAP where the grain sizes are exceptionally small. The objective of the present research was to examine the characteristics of cavity development in the Zn-22% Al eutectoid alloy after processing by ECAP. This material was selected for two reasons. First, the Zn-22% Al alloy was used extensively in very early studies of cavitation behavior in superplastic flow [10, 11, 21, 22]. Second, a recent investigation showed this alloy exhibits excellent superplastic properties after processing by ECAP [23].

Experimental material and procedures

A commercial Zn-22% Al eutectoid alloy was used in the present experiments. The material was provided in the form of a plate with a thickness of 25 mm and having the following composition in ppm: Cr <10, Cu 20, Fe 70, Mg <10, Mn <10, and Si 70. The plate was machined into rods having diameters of 10 mm and then cut into billets with lengths of ~60 mm. Each billet was annealed in air for 1 hour at 473 K to give a mean linear intercept grain size of ~1.8 μm.

Processing by ECAP was conducted at 473 K using a solid die having two channels, each with a diameter of 10 mm, which intersected at an internal angle of $\Phi = 90°$ and with an additional outer arc of curvature at the point of intersection of $\Psi = 20°$. It is known from first principles that these values of Φ and Ψ lead to an imposed strain of ~1 on each separate pass through the die [24]. Each billet was pressed repetitively through 8 passes using processing route B_C where the billet is rotated in the same direction by 90° about its longitudinal axis between each pass [25]. Thus, the total imposed strain for these specimens was equal to ~8. As shown in an earlier report, the longitudinal mean linear intercept grain size, $\bar{L}$, immediately after pressing is ~0.8 μm [26].

Tensile specimens were machined from the pressed rods having gauge lengths of 4 mm and cross-sectional areas of 3×2 mm^2. The gauge lengths of these samples were cut parallel to the pressing direction. Each of these specimens was heated to a temperature of 473 K, held at temperature for 1 hour, and then pulled in tension using a testing machine operating at a constant rate of cross-head displacement and with initial strain rates in the range from 1.0×10^{-4} to 1.0 s^{-1}. Most of these tests were continued to failure but an additional test was also conducted at a strain rate of 1.0×10^{-2} s^{-1} and this test was terminated at a preselected elongation of 500%.

Quantitative cavity measurements were undertaken on the ECAP samples after tensile testing to failure or to the predetermined elongation of 500%. One side of the specimen surface was polished to a mirror-like finish without etching, and then the cavities were examined and measured using an optical microscope connected through an image monitor and a personal computer. All the quantitative measurements were performed using Image Measure IM5100 software which automatically evaluates all cavities by scanning a selected area displayed on the image monitor. Thus, each cavity was scanned and the size and other parameters determined automatically by the software program. In order to obtain consistent data for all measurements of every specimen, the scanning was performed at selected points close to the central axis of the elongated gauge lengths. For the samples pulled to failure, the scanning was performed in areas close to the fracture tips but excluding the edges of the gauge sections.

For each set of measurements, the intensity threshold was adjusted manually so that all visible cavities appeared on the monitor with consistent and equal images. Measurements were made for a total of 20 different measurement windows where each window was rectangular with an area of 0.05 mm^2. Thus, measurements were taken on each sample in an area corresponding to 1 mm^2. The minimum detectable cavity size was set at 1 μm^2 to avoid any difficulties with dirt or small artifacts on the polished surfaces.

From the quantitative measurements, the following numerical information was recorded on the personal computer: (a) the cumulative total number of all cavities in each measurement window, (b) the area of each individual cavity, (c) the perimeter of each individual cavity by measuring the length around the periphery, (d) the maximum length of any diameter within the cavity which was determined by taking any two points on the cavity perimeter, (e) the orientation of each cavity determined by the angle between the maximum diameter and the tensile axis, and (f) the roundness coefficient for each cavity defined as $\{4\pi \times \text{area}\}/(\text{perimeter})^2$, where a spherical cavity has a coefficient of 1.0 and lower values represent increasing deviations from a circular configuration.

The cavity morphologies within the polished gauge sections were also examined visually, and photomicrographs were taken, using an Olympus Vanox AHMT3 photomicrographic facility.

Experimental results

Mechanical properties after ECAP

In earlier experiments [27], and in order to check on the grain size in the Zn–Al alloy immediately prior to the tensile testing, a tensile sample was pressed through 8 passes, heated to a temperature of 473 K, and then held for one hour and cooled in air. The mean linear intercept grain size in this condition was measured along a longitudinal traverse as $\bar{L} \approx 0.9$ μm where this corresponds to the condition before tensile testing. These measurements demonstrate there is a very small increase in the grain size, from 0.8 to 0.9 μm, during the time required to establish equilibrium prior to testing. In practice, the spatial grain size, d, is given by $d = 1.74 \times \bar{L}$ so that the spatial grain size at the beginning of the tensile testing was $d \approx 1.6$ μm.

Several samples of the Zn-22% Al alloy were processed by ECAP and pulled to failure at various strain rates to determine both the flow stress, σ, and the elongation to failure for each specimen. The results are recorded in Fig. 1 which shows the elongation to failure (upper) and the flow stress (lower) versus the initial strain rate. The highest elongation recorded in these experiments was 2,230% at an initial strain rate of 1.0×10^{-2} s^{-1}. This result provides a clear demonstration of the occurrence of high strain rate superplasticity, since this is defined as superplastic flow at strain rates at or above 10^{-2} s^{-1} [28]. From the upper plot in Fig. 1 there is evidence for the three conventional regimes of superplasticity corresponding to region I at the lower strain rates below $\sim 2 \times 10^{-3}$ s^{-1}, region II at immediate strain rates up to $\sim 10^{-1}$ s^{-1}, and region III at even higher strain rates. It is also apparent that the optimal superplastic strain rate occurs in the vicinity of $\sim 1.0 \times 10^{-2}$ s^{-1}.

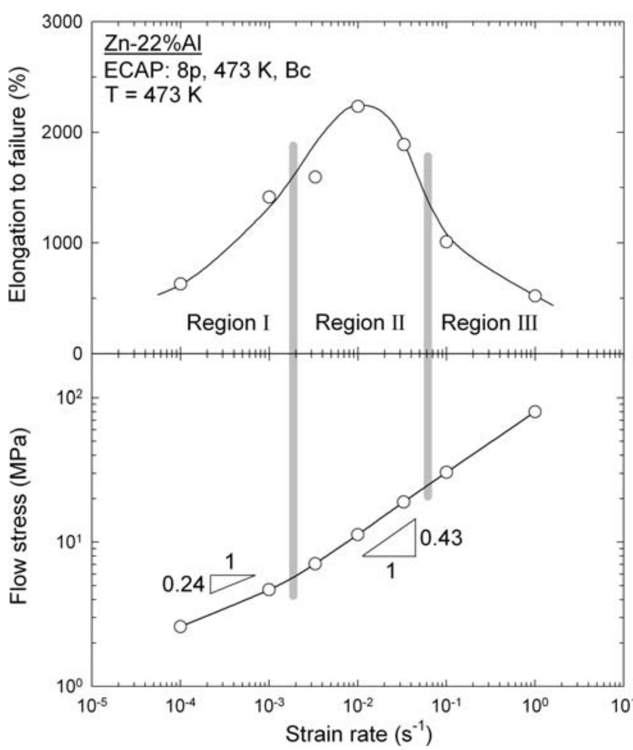

Fig. 1 Variation in the elongation to failure (upper) and the flow stress (lower) with strain rate for a Zn-22% Al eutectoid alloy tested at 473 K

The strain rate sensitivity, m, is denoted by the slope of the logarithmic plot of flow stress against strain rate shown in the lower section of Fig. 1, and this indicates values of $m \approx 0.43$ at intermediate strain rates in region II and $m \approx 0.24$ at lower strain rates in region I.

Appearance of the cavities

First, it should be noted that a recent report discussed the internal cavities present in an as-received Al-7034 alloy prior and subsequent to ECAP [20]. Careful inspection showed a very small number of extremely small cavities in the as-received sample which were below the cut-off limit generally used for the quantitative measurements in cavity investigations. Furthermore, there was no evidence for any increase in the level of cavitation during ECAP. These observations are also supported by earlier reports demonstrating a decrease in the level of porosity in cast aluminum alloys when using ECAP [29, 30]. From these various reports, it is reasonable to conclude that the occurrence of cavitation in the present samples is a direct consequence of superplastic flow at the elevated testing temperature of 473 K.

In order to investigate the appearance of cavities both during superplastic flow and at the point of failure, an additional sample of the Zn-22% Al alloy was processed by ECAP and then pulled in tension at 473 K at the optimal superplastic strain rate of 1.0×10^{-2} s^{-1} to a predetermined elongation of 500%. This specimen and the specimen taken to failure at this same strain rate were polished and examined optically and the results are shown in Fig. 2 for (a) the sample taken to 500% and (b) the sample pulled to failure at 2,230%: in both photomicrographs the tensile axis, and thus the pressing direction, is horizontal. These photomicrographs were taken close to the center of the deformed gauge length for the specimen pulled to 500% and close to the fracture tip but not close to the edge of the gauge section for the failed specimen.

Several conclusions are reached from the inspection of these photomicrographs. First, cavity nucleation occurs at random locations which are distributed reasonably uniformly throughout the gauge lengths of the samples. The random nature of cavity nucleation is consistent with earlier reports after tensile testing of a Zn-22% Al alloy without processing by ECAP [31] and an Al-7034 alloy after processing by ECAP through 6 passes [20]. Second, larger cavities are visible at the higher elongation in Fig. 2b thereby demonstrating the occurrence of cavity growth and cavity coalescence. Third, there is clear

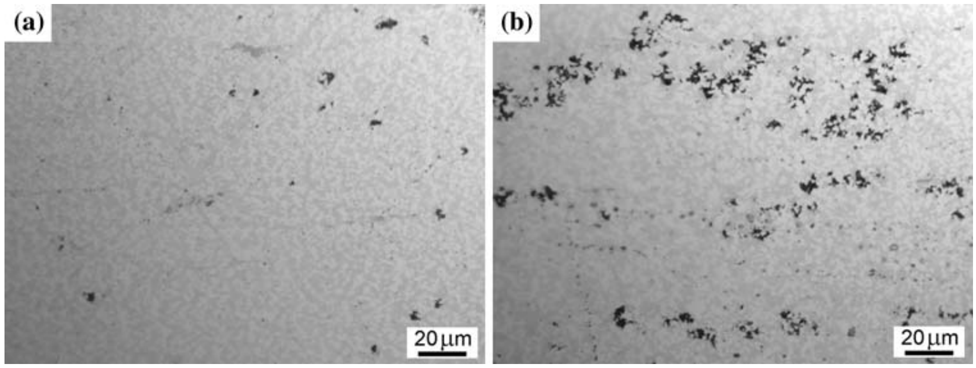

Fig. 2 Appearance of cavities in the Zn-22% Al alloy after ECAP and pulling at a strain rate of 1.0×10^{-2} s^{-1} (**a**) for a specimen pulled to an elongation of 500% and (**b**) for a specimen pulled to failure at 2,230%; in both photomicrographs the tensile axis is horizontal

evidence for the formation of cavity stringers in Fig. 2b where stringers are defined as lines of cavities aligned parallel to the tensile direction. Cavity stringers are a common feature of conventional superplastic materials and they have been documented both in Zn-22% Al without processing by ECAP [10, 11, 32–36] and in several superplastic Al alloys processed by ECAP [17, 37, 38]. The presence of these stringers is directly influenced by the locations of impurities, oxidized particles, and precipitates which may become broken and aligned parallel to the stress direction during thermo-mechanical processing such as in any rolling or extrusion operations [39, 40].

Quantitative cavity measurements

Quantitative cavity measurements were undertaken on the Zn-22% Al alloy after ECAP and pulling to elongations of 500% and failure at 2,230%. The results from these measurements are displayed in Figs. 3–5.

The distributions of the cumulative numbers of cavities are plotted against the individual cavity areas in the histograms shown in Fig. 3. Almost all the cavities have a cavity area of less than 10 μm^2 in the sample pulled to an elongation of 500%, whereas in the specimen pulled to failure many of the cavities have areas of $\sim$10–40 μm^2 and there are some cavities with even larger areas of $>$70 μm^2.

The significance of the shapes of the individual cavities is shown in Fig. 4 where the normalized cavity number is plotted against the measured roundness coefficient, and a normalized cavity number is used because it permits a direct comparison of samples having large differences in the total numbers of cavities. These results show a large fraction of cavities having roundness coefficients of >0.8 but with the occurrence of more rounded cavities at the lower elongation. A corresponding plot is shown in Fig. 5 for the orientations of the cavities with respect to the tensile axis. These histograms demonstrate there is a clear transition from a peak in the angular range of 75–90° for the sample terminated at 500% and a peak at the lowest angular range of 0–15° for the specimen pulled to failure.

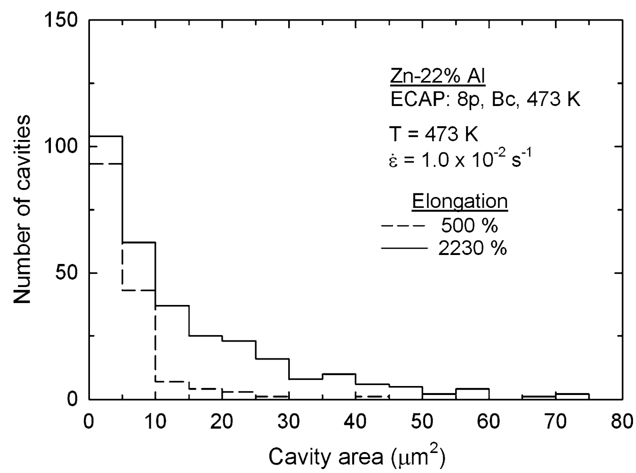

Fig. 3 Distribution of cavity areas for the Zn-22% Al alloy after ECAP and pulling to 500% or pulling to failure at 2,230% at a strain rate of 1.0×10^{-2} s^{-1} at 473 K

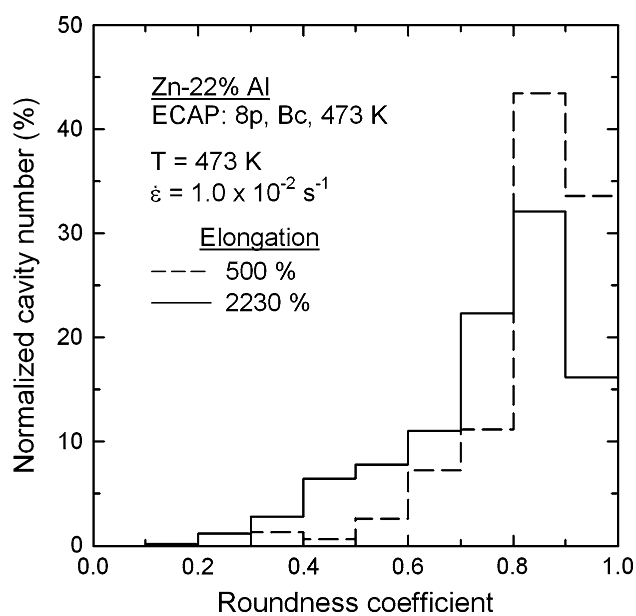

Fig. 4 Distribution of roundness coefficients for the Zn-22% Al alloy after ECAP and pulling to 500% or pulling to failure at 2,230% at a strain rate of 1.0×10^{-2} s^{-1} at 473 K

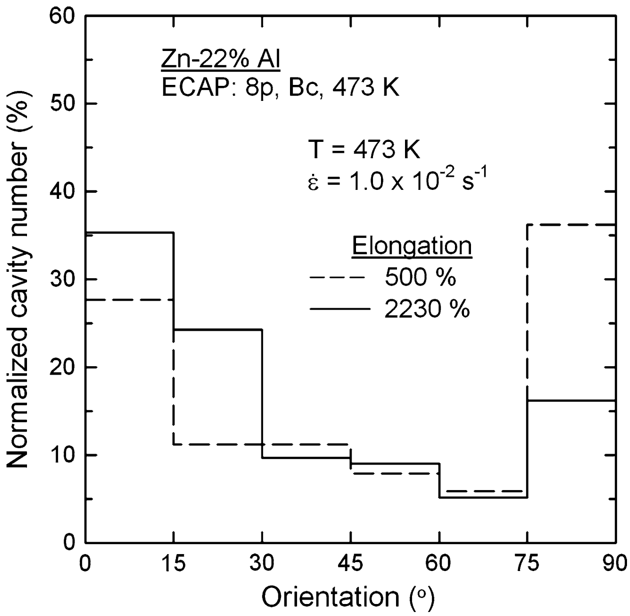

Fig. 5 Distribution of cavity orientations for the Zn-22% Al alloy after ECAP and pulling to 500% or pulling to failure at 2,230% at a strain rate of 1.0×10^{-2} s^{-1} at 473 K

Discussion

Several important conclusions may be reached from this investigation.

First, processing of the Zn-22% Al eutectoid alloy by ECAP provides an opportunity to introduce submicrometer grain sizes into the bulk alloy where these very small grain sizes cannot be reached using conventional thermo-mechanical processing. Furthermore, because of the two-phase nature of the microstructure, these ultrafine grains exhibit exceptionally good stability at elevated temperatures.

Second, this ultrafine-grained alloy exhibits excellent superplastic properties with elongations up to >2,000% under optimum conditions at a testing temperature of 473 K. The present results in Fig. 1 are very similar to those reported earlier at the same testing temperature for an alloy unprocessed by ECAP and having a spatial grain size of 2.5 μm [31]. Specifically, there is the same division into three distinct regions of flow with the highest elongation occurring in the vicinity of $\sim 10^{-2}$ s^{-1}. The value of $m \approx 0.43$ in the superplastic region II is similar to, but a little lower than, the value of $m = 0.5$ anticipated for the superplastic region if flow occurs by the conventional mechanism for superplasticity [41].

Third, it is readily apparent from Fig. 3 that the total numbers of cavities increase between 500% and the point of failure at 2,230%. Nevertheless, and despite this well-documented increase, the total number of small cavities recorded within the lowest range up to a cavity area of 5 μm^2 remains almost identical between the elongations of 500% and 2,230%. It follows, therefore, that these measurements provide very clear evidence that cavity nucleation is a continuous and dynamic process taking place throughout superplastic flow up to the ultimate failure of the material. It is also apparent that the largest cavity sizes are due to the coalescence of individual adjacent cavities.

Fourth, Fig. 4 shows that over 70% of the total cavities have a roundness coefficient in the range of 0.8–1.0 for the sample pulled to an elongation of 500%, whereas over 50% of the total cavities have roundness coefficients of <0.8 in the sample pulled to failure. This plot shows that the cavities are initially reasonably spherical but they become elongated when the sample pulls out to achieve the maximum superplastic elongation. Furthermore, it is apparent from Fig. 5 that the preferred orientation of the cavities is close to 90° at the lower elongation of 500% so that the longer axes of the cavities lie perpendicular to the tensile direction. However, this trend changes with increasing strain. For the sample pulled to failure at an elongation of 2,230% a total of ~60% of the cavities have orientations in the range of 0–30°, and there is a peak in the orientation range at the lowest angular increment of 0–15°. These results demonstrate a clear transition in cavity morphology so that the cavities ultimately pull out preferentially along the tensile axis. The explanation lies in a transition in the cavity growth process from the superplastic diffusion growth process when the cavities are very small [42] to a plasticity-controlled growth mechanism when the cavities are large [43]. Similar transitions were also noted in earlier reports of cavity measurements in an aluminum metal matrix composite [19] and a spray-cast Al 7,034 alloy [20] both processed by ECAP to produce ultrafine grain sizes.

Summary and conclusions

1. Processing of a Zn-22% Al eutectoid alloy by equal-channel angular pressing produced a spatial grain size of ~1.6 μm and superplastic elongations when pulling at a temperature of 473 K, including an optimum elongation of 2,230% when using an initial strain rate of 1.0×10^{-2} s^{-1}.
2. Inspection of polished sections of samples after tensile testing showed the presence of many small cavities that developed internally during superplastic flow. Cavities were present both after pulling to the optimum superplastic elongation of 2,230% and after pulling to a preselected elongation of 500% at the same initial strain rate.
3. Using quantitative measurements of the cavity numbers and their shapes and sizes, it is shown that cavity nucleation must occur continuously throughout

superplastic flow. In addition, the shapes of the cavities provide evidence for a transition in the growth process from superplastic diffusion growth to growth by a plasticity-controlled mechanism.

Acknowledgement This work was supported by the U.S. Army Research Office under Grant No. W911NF-05-1-0046

References

1. Langdon TG (1982) Metall Trans 13A:689
2. Valiev RZ, Islamgaliev RK, Alexandrov IV (2000) Prog Mater Sci 45:103. doi:10.1016/S0079-6425(99)00007-9
3. Valiev RZ, Langdon TG (2006) Prog Mater Sci 51:881. doi:10.1016/j.pmatsci.2006.02.003
4. Ma Y, Furukawa M, Horita Z, Nemoto M, Valiev RZ, Langdon TG (1996) Mater Trans JIM 37:336
5. Valiev RZ, Salimonenko DA, Tsenev NK, Berbon PB, Langdon TG (1997) Scr Mater 37:1945. doi:10.1016/S1359-6462(97)00387-4
6. Komura S, Berbon PB, Furukawa M, Horita Z, Nemoto M, Langdon TG (1998) Scr Mater 38:1851. 10.1016/S1359-6462(98)00099-2
7. Furukawa M, Ma Y, Horita Z, Nemoto M, Valiev RZ, Langdon TG (1998) Mater Sci Eng A 241:122. doi:10.1016/S0921-5093(97)00481-4
8. Kawasaki M, Figueiredo RB, Xu C, Langdon TG (2007) Metall Mater Trans 38A:1891
9. Kawasaki M, Langdon TG (2007) J Mater Sci 42:1782. doi:10.1007/s10853-006-0954-2
10. Ishikawa H, Bhat DG, Mohamed FA, Langdon TG (1977) Metall Trans 8A:523
11. Ahmed MMI, Mohamed FA, Langdon TG (1979) J Mater Sci 14:2913. doi:10.1007/BF00611474
12. Kawasaki M, Kubota K, Higashi K, Langdon TG (2006) Mater Sci Eng A 429:334. doi:10.1016/j.msea.2006.05.043
13. Taplin DMR, Dunlop GI, Langdon TG (1979) Annu Rev Mater Sci 9:151. doi:10.1146/annurev.ms.09.080179.001055
14. Jiang XG, Earthman JC, Mohamed FA (1994) J Mater Sci 29:5499. doi:10.1007/BF00349941
15. Xu C, Lee S, Langdon TG (2001) Mater Sci Forum 357–359:521
16. Park K-T, Myung SH, Shin DH, Lee CS (2004) Mater Sci Eng A 371:178. doi:10.1016/j.msea.2003.11.042
17. Musin F, Kaibyshev R, Motohashi Y, Itoh G (2004) Metall Mater Trans 35A:2383
18. Musin F, Kaibyshev R, Motohashi Y, Itoh G (2004) Scr Mater 50:511. doi:10.1016/j.scriptamat.2003.10.021
19. Kawasaki M, Huang Y, Xu C, Furukawa M, Horita Z, Langdon TG (2005) Mater Sci Eng A 410–411:402. doi:10.1016/j.msea.2005.08.073
20. Kawasaki M, Xu C, Langdon TG (2005) Acta Mater 53:5353. doi:10.1016/j.actamat.2005.08.012
21. Miller DA, Langdon TG (1978) Metall Trans 9A:1688
22. Miller DA, Langdon TG (1979) Metall Trans 10A:1869
23. Kawasaki M, Langdon TG (2008) Mater Trans 49:84. doi:10.2320/matertrans.ME200720
24. Iwahashi Y, Wang J, Horita Z, Nemoto M, Langdon TG (1996) Scr Mater 35:143. doi:10.1016/1359-6462(96)00107-8
25. Furukawa M, Iwahashi Y, Horita Z, Nemoto M, Langdon TG (1998) Mater Sci Eng A 257:328. doi:10.1016/S0921-5093(98)00750-3
26. Kumar P, Xu C, Langdon TG (2006) Mater Sci Eng A 429:324. doi:10.1016/j.msea.2006.05.044
27. Kumar P, Xu C, Langdon TG (2005) Mater Sci Eng A 410–411:447. doi:10.1016/j.msea.2005.08.092
28. Higashi K, Mabuchi M, Langdon TG (1996) ISIJ Int 36:1423. doi:10.2355/isijinternational.36.1423
29. Lapovok R (2002) Int J Fract 115:159. doi:10.1023/A:1016399111787
30. McKenzie PWJ, Lapovok R, Wells P, Raviprasad K (2003) Mater Sci Forum 426–432:297
31. Ishikawa H, Mohamed FA, Langdon TG (1975) Philos Mag 32:1269. doi:10.1080/14786437508228105
32. Park K-T, Yang ST, Earthman JC, Mohamed FA (1994) Mater Sci Eng A 188:59. doi:10.1016/0921-5093(94)90356-5
33. Chokshi AH, Langdon TG (1989) Acta Mater 37:715. doi:10.1016/0001-6160(89)90255-1
34. Yousefiani A, Earthman JC, Mohamed FA (1998) Acta Mater 46:3557. doi:10.1016/S1359-6454(98)00030-5
35. Yousefiani A, Mohamed FA (1999) Philos Mag A 79:1247. doi:10.1080/01418619908210359
36. Tanaka T, Higashi K (2004) Mater Trans 45:2547. doi:10.2320/matertrans.45.2547
37. Park K-T, Hwang D-Y, Chang S-Y, Shin DH (2002) Metall Mater Trans 33A:2859
38. Park K-T, Hwang D-Y, Lee Y-K, Kim Y-K, Shin DH (2003) Mater Sci Eng A 341:273. doi:10.1016/S0921-5093(02)00216-2
39. Chokshi AH, Langdon TG (1990) Acta Metall Mater 38:867. doi:10.1016/0956-7151(90)90040-N
40. Park K-T, Mohamed FA (1990) Metall Trans 21A:2605
41. Langdon TG (1994) Acta Metall Mater 42:2437. doi:10.1016/0956-7151(94)90322-0
42. Chokshi AH, Langdon TG (1987) Acta Mater 35:1089. doi:10.1016/0001-6160(87)90056-3
43. Hancock JW (1976) Meat Sci 10:319. doi:10.1016/0036-9748(76)90084-3