# Formation of ultrafine grains and twins in the β-phase

# during superplastic deformation of two-phase brasses

Anastasia V. Mikhaylovskaya1\*, Olga A.Yakovtseva1, Natalia Yu. Tabachkova1,2 and
Terence G. Langdon3

1National University of Science and Technology “MISIS”, National University of Sciences and Technology “MISIS”, Leninskiy Ave. 4, 119049  Moscow, Russia

2 Prokhorov General Physics Institute of the Russian Academy of Sciences, Vavilova 38, 119991 Moscow, Russia

3Materials Research Group, Department of Mechanical Engineering, University of Southampton, Southampton SO17 1BJ, U.K.

\*corresponding author e-mail: mihaylovskaya@misis.ru

**Abstract**

During superplastic deformation of microduplex-structured brasses, the strain is primarily localized in the β-phase where deformation occurs by grain boundary sliding (GBS) and dislocation slip/creep mechanisms. Dynamic recrystallization and twinning were observed in the b.c.c. β-phase leading to a transformation of the initial coarse β-phase grains with average sizes of ~4-5 µm to ultrafine grains with sizes below 1 µm. Alloying with Al refined the β-grains, improved the superplastic behavior and reduced the occurrence of residual cavitation.

**Keywords:** brass; deformation mechanisms; grain boundary sliding; superplasticity; twinning.

Two-phase brasses are classic microduplex-structured materials which exhibit superplastic behavior [1][2] [3][4]. The superplastic deformation of alloys with duplex-type structures is fundamentally complicated because insufficient data are at present available documenting the microstructural changes that occur in each phase during superplastic flow. Brasses consist of f.c.c. α and b.c.c. β phases having similar values of staking fault energies but different crystalline structures leading to differences in the ductilities and diffusivities of the two phases [5][6]. For this reason, the hot deformation behavior of the α and β phases are different and the overall deformation process may be controlled by the deformation of the ductile and highly diffusive β phase.

Knowledge about the superplastic deformation mechanisms of brasses are limited by a clear understanding that grain boundary sliding (GBS) is dominant in superplastic flow [7][8]. Experiments showed that the sliding rate for α/β interfaces is about 1.5 times slower than for α/α boundaries and about 2.5 times slower than β/β boundaries [9]. From this research it was concluded that increasing the β-phase fraction leads to an improvement in the superplasticity due to the well-developed GBS and the occurrence of only limited residual cavitation. The importance of the β-phase fraction as a cavitation controlling parameter was confirmed in several reports [1][10] [11]. Since it is more ductile and diffusive than the f.c.c. α-phase, the b.c.c. β-phase should stimulate the accommodation of GBS by dislocation slip/creep and/or diffusional creep mechanisms. This is consistent with the fact that the β-phase b.c.c. lattice has multiple dislocation slip systems and high diffusivity and thereby improves the occurrence of dislocation creep mechanisms. Additionally, in α/β brass the deformation occurs at a higher homologous temperature for the β phase and this suggests a higher diffusivity. However, only limited experimental evidence is at present available to provide a comprehensive understanding of the precise roles of the different phases.

Several studies emphasize the importance that alloying by Al improves superplasticity and decreases the residual cavitation of brasses [12][13][14]. Thus, small additions of Al (~0.5-1%) insignificantly influences the grain and phase structure but Al alloying decreases the residual cavitation and leads to higher elongations-to-failure by about two times compared with the binary alloy [12]. In practice, the volume fraction of residual cavities is of the order of ~5% and ~0.5% for binary and Al-bearing alloys, respectively [12]. Although the precise nature of this Al effect is not yet understood, it is reasonable to anticipate that the presence of Al influences the occurrence of GBS and the phase diffusivities [14][15].

The present research was undertaken to clarify both the role of the α and β phases in the superplastic deformation behavior and the influence of Al on the superplasticity of these brasses. The focused ion beam (FIB) technique has been used in experiments for the production of regular surface grid markers that may contribute to the overall understanding of the superplastic deformation mechanisms in Al [16][17][18][19], Ti [20] and Fe [21] based alloys. A similar approach was used in this research to study the strain-induced evolutions of FIB-grids in cooperation with corresponding strain-induced changes of the dislocation structures in a Cu–40Zn α+β brass and in a Cu–38Zn-1Al (wt%) α+β brass.

Microstructural analysis was performed using a TESCAN Vega 3 LMN scanning electron microscope (SEM) and a JEOL-JEM 2100 transmission electron microscope (TEM). Specimens for microstructural SEM examination were prepared by mechanical grinding on SiC papers and polishing using a Struers LaboPol 5 machine. Samples for TEM study were prepared by ion-milling using a STRATA-FIB-205 FEI200 focused ion beam (FIB) microscope. This microscope was also used to produce the grids. Samples were pre-strained to a logarithmic strain of 0.69 at a temperature of 550°C under a constant strain rate of
1.0 × 10-3 s-1 and subsequently ground and polished. FIB grids with a size of 40 × 40 μm, a step of 2.5 μm and a depth of 0.2 μm and microgrids with a step of 0.5 μm and a depth of 0.1 μm were milled on the pre-polished gauges of the samples. Mechanical tensile tests were performed using a Walter Bai LFM 100 tensile machine in an argon atmosphere at a temperature of 550°C under a strain rate of 1.0 × 10-3 s-1. These conditions were considered optimal [12] and provided an α/β ratio close to 50/50, a maximum elongation-to-failure and a strain rate sensitivity, *m*, of ~0.5-0.6 for both alloys. Before testing, the mean grain sizes of the binary alloy were ~7.6 ± 0.7 µm for the α-phase and ~5.1±0.8 µm for the β-phase. For the Al-bearing alloy, the initial mean grain size was ~5.9 ± 0.7 µm for the α-phase and ~3.8±0.6 µm for the β-phase [12]. The volume fractions of the β-phase at 550°C were ~0.46±0.07 and ~0.50±0.06 for the Cu-Zn and Cu-Zn-Al brass, respectively. It is important to note that the α/β phase ratio remained unchanged during the deformation.

The strain-induced microstructural changes occurring on the sample surfaces during superplastic deformation are shown in detail in Fig. 1. Thus, a grain neighbor switching led to shifts of the grid lines (blue-colored dashes lines in Fig. 1) and α-grain rotations (green-colored arrows in Fig.1) were observed in both alloys. The maximum grain rotation angle was 35° with an increase in strain from 0.69 to 0.97-1.07. An intergranular strain was developed at α/β but rarely at the α/α grain boundaries (see blue-colored arrows in Fig.1). The intragranular strain for the α-phase grains was insignificant but several local areas demonstrated evidence of strain localization (see evolution of grains A in Figs.1 a-c, B and C in Fig.1d-f).

Many large cavities were developed on the surface of the binary brass (in areas marked with white arrows in Figs. 1b,c) in which cavitation clearly accompanied GBS. Generally, this cavitation was insignificant for the Al-bearing brass (Figs. 1e,f). Thus, an Al effect on cavitation weakening during superplastic deformation of brasses is in agreement with the studies of sample volumes reported earlier [12][15]. The difference in cavitation was mainly related to differences in the deformation behavior of the β-phase for these alloys. Thermal etching and oxidation effects were significant in the β-phase of the Al-free binary brass and its surface with fine grid lines became partially evaporated. The remaining β-phase demonstrated less significant strain localization than that of the Al-bearing brass (Fig.1). Owing to strain localization, the β-phase surface in the Al-bearing brass was strongly folded and the grid lines were partially distorted at a strain of 0.18 and primarily disappeared at a strain of 0.39. In both alloys, curvatures of the grid lines in the β-phase areas and grain rotations (red arrows and lines in Fig.1) indicated that intergranular deformation by GBS also occurred at β/β grain boundaries. To understand the strain-induced microstructural evolution for the β-phase, FIB-milled trenches (positions indicated with lines in Figs. 2a,c) were processed for the deformed samples.

The subsurface structure in the trenches showed a channeling contrast between grains of the α- and β-phases (Figs.2b,d,e). Specifically, coarse grains were found in the β-phase areas for the binary brass whereas many fine grains were found in the β-phase area for the Al-bearing brass (Figs.2b,d,e).

A TEM study provided a detailed understanding of the microstructural evolution during the superplastic deformation. The Cu-Zn and Cu-Zn-Al alloys exhibited similar microstructures before the beginning of the superplastic deformation (Figs.3a,b) with both phases almost free of dislocations and with typical coarse-grained α and β phase selected area electron diffraction (SAEAD} patterns. Coarse twins with typical lengths of ~1 to 5 µm and thicknesses of ~100-400 nm were observed only in the α-phase.

The dislocation density was low and only several dislocation walls were visible in the α-phase after straining to 1.0 in both alloys (Figs.4a,c). The weak strain-induced changes for the dislocation structures are consistent with insignificant intragranular strains on the surfaces of the α-phase. It appears, therefore, that a dislocation slip/creep mechanism accommodates GBS in the α-phase. The α-grains also became coarse and exhibited the same diffraction pattern as in the initial stage before the onset of deformation.

By contrast, the strain-induced changes in the β-phase were significant. Many grain boundaries and nanoscale twins were formed in the β-phase after deformation (Fig. 4) and there was a ring-type diffraction from the b.c.c. phase due to the formation of many grains in the studied area of 1 µm after superplastic deformation. Thus, the grain size of the β-phase decreased after straining from an initial value of ~4-5 µm to an ultrafine-grained state <1 µm in both alloys. An important difference was found also between the β-phase grain sizes in the Cu-Zn and Cu-Zn-Al alloys. Thus, the β-phase grain size ranged from ~300-800 nm in the binary alloy while grain sizes of ~100-400 nm were observed in the Al-bearing alloy (Fig. 4b). The widths of the nanocsale twins (denoted by arrows in Fig. 4) varied in the range from several nanometers to tens of nanometers. There are also other reports of similar fine twins formed during severe plastic deformation of brasses [22][23].

The β-phase structure remained unchanged and consisted of dislocation-free coarse grains within the grip parts of the tensile samples. Twins in the β-phase and the grain refinement effect were thus induced by superplastic deformation and this was attributed to the advent of dynamic/post-dynamic recrystallization [24][25][26][27]. Therefore, this study firstly and clearly confirms evidence for dynamic recrystallization (DRX) and twinning in the b.c.c. β-phase during the superplastic deformation of brasses.

Dynamic recrystallization in the β-phase is a principal process that significantly influences the superplastic deformation behavior of brasses. The difference between the β-phase grain size of the two alloys clearly explains the difference in their behavior during superplastic deformation. Thus, it is clear that there is improved superplasticity of in Al-bearing brass and the inherent low cavitation is a direct consequence of grain refinement in the β-phase during straining. The Al is predominantly a solute in the β-phase and the increased solute content provides uniform deformation with finer DRX grains for the β-phase during the superplastic deformation. Furthermore, it is established that high solute alloys usually demonstrate a weaker dynamic grain growth effect [16]. Thus, the finer grains of the β-phase simplify the GBS on the β/β grain boundaries this decreases the residual cavitation and increases the elongation for the Al-bearing brass.

In summary, grain boundary sliding on α/β and α/α boundaries contribute about 70 and 40%, respectively, for binary brass and Al-bearing brass, respectively. The 70% and 40% need to be included in the text. Significant cavitation was developed in the binary brass but this was not observed for the Al-bearing brass. The α-phase grains demonstrated low intragranular strain and limited dislocation activity for both alloys. Strain was localized in the β-phase this demonstrated both GBS and intragranular deformation accompanied by DRX. Localized deformation in the β-phase provided about 20% of the total strain for binary brass and about 40% of the total strain for Al-bearing brass. You need to include this information in the text. Due to DRX, ultrafine β-phase grains with nanoscale twins were formed. The β-phase grain size was in the range of ~300-800 nm for binary brass and ~100-400 nm for the Al-bearing brass. Thus, the finer β-phase grain structure leads to a lower residual cavitation and improved superplasticity for the Al-bearing brass.

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**References**

[1] S. Sagat, D.M.R. Taplin, Fracture of a superplastic ternary brass, Acta Metall. 24 (1976) 307–315. doi:10.1016/0001-6160(76)90005-5.

[2] M.. Willis, J.. Jones, Creep mechanisms in a dual-phase brass, Scr. Mater. 44 (2001) 31–36. doi:10.1016/S1359-6462(00)00554-6.

[3] E. Farabi, A. Zarei-Hanzaki, M.H. Pishbin, M. Moallemi, Rationalization of duplex brass hot deformation behavior: The role of microstructural components, Mater. Sci. Eng. A. 641 (2015) 360–368. doi:10.1016/j.msea.2015.06.042.

[4] K. Neishi, Z. Horita, T.G. Langdon, Achieving superplasticity in a Cu-40%Zn alloy through severe plastic deformation, Scr. Mater. 45 (2001) 965–970. doi:10.1016/S1359-6462(01)01119-8.

[5] H. Miyamoto, T. Tanaka, T. Mimaki, R. Matsubara, N. Ashie, S. Miura, Interphase boundary sliding of two-phase (α/β), (β/γ), and (γ/α) CuZnSn alloy couples produced by solid-to-solid diffusion bonding, Mater. Sci. Eng. A. 380 (2004) 34–40. doi:10.1016/j.msea.2004.03.048.

[6] T. G. Nieh, J. Wadsworth, O.D. Sherby, Superplasticity in metals and ceramics, 1st ed., Cambridge university press, NewYork, 1997. doi:10.1017/CBO9780511525230. This is not a good reference here. You need to refer to a fundamental experimental paper.

[7] T.G. Langdon, A unified approach to grain boundary sliding in creep and suerplasticity, Acta Metall. Mater. 42 (1994) 2437-2443.

[8] T.G. Langdon, An evaluation of the strain contributed by grain boundary sliding in superplasticity, Mater. Sci. Eng. A. 174 (1994) 225–230. doi:10.1016/0921-5093(94)91092-8.

[9] N. Chandra, Constitutive behavior of superplastic materials, Int. J. Non. Linear. Mech. 37 (2002) 461–484. doi:10.1016/S0020-7462(01)00021-X.

[10] C.W. Humphries, N. Ridley, Cavitation during the superplastic deformation of an α/β brass, J. Mater. Sci. 13 (1978) 2477–2482. doi:10.1007/BF00808064.

[11] A.R. Ragab, Modeling of the effect of cavitation on tensile failure of superplastic alloys, Mater. Sci. Eng. A. 454–455 (2007) 614–622. doi:10.1016/j.msea.2006.11.093.

[12] O.A. Yakovtseva, A.V. Mikhaylovskaya, A.V. Pozdniakov, A.D. Kotov, V.K. Portnoy, Superplastic deformation behaviour of aluminium containing brasses, Mater. Sci. Eng. A. 674 (2016) 135–143. doi:10.1016/j.msea.2016.07.053.

[13] O.A. Yakovtseva, A. V. Mikhailovskaya, A.D. Kotov, V.K. Portnoi, Effect of alloying on superplasticity of two-phase brasses, Phys. Met. Metallogr. 117 (2016) 742–748. doi:10.1134/S0031918X16070188.

[14] S.S. Xuanxiang S. , Shiyou G., Superplasticity in Aluminium Brass(HAl 66-6-3-2), J Mater Sci Technol. 8 (1992) 440–442.

[15] O.A. Yakovtseva, A. V. Mikhaylovskaya, A. V. Irzhak, A.D. Kotov, S. V. Medvedeva, Comparison of Contributions of the Mechanisms of the Superplastic Deformation of Binary and Multicomponent Brasses, Phys. Met. Metallogr. 121 (2020) 582–589. doi:10.1134/S0031918X20060186.

[16] K. Sotoudeh, P.S. Bate, Diffusion creep and superplasticity in aluminium alloys, Acta Mater. 58 (2010) 1909–1920. doi:10.1016/j.actamat.2009.11.034.

[17] M.A. Rust, R.I. Todd, Surface studies of Region II superplasticity of AA5083 in shear: Confirmation of diffusion creep, grain neighbour switching and absence of dislocation activity, Acta Mater. 59 (2011) 5159–5170. doi:10.1016/j.actamat.2011.04.051.

[18] H. Masuda, E. Sato, Diffusional and dislocation accommodation mechanisms in superplastic materials, Acta Mater. 197 (2020) 235–252. doi:10.1016/j.actamat.2020.07.042.

[19] A.V. Mikhaylovskaya, O.A. Yakovtseva, A.V. Irzhak, The role of grain boundary sliding and intragranular deformation mechanisms for a steady stage of superplastic flow for Al–Mg-based alloys, Mater. Sci. Eng. A. 833 (2022) 142524. doi:10.1016/j.msea.2021.142524.

[20] E. Alabort, P. Kontis, D. Barba, K. Dragnevski, R.C. Reed, On the mechanisms of superplasticity in Ti–6Al–4V, Acta Mater. 105 (2016) 449–463. doi:10.1016/j.actamat.2015.12.003.

[21] H. Masuda, H. Tobe, E. Sato, Y. Sugino, S. Ukai, Diffusional mass flux accommodating two-dimensional grain boundary sliding in ODS ferritic steel, Acta Mater. 176 (2019) 63–72. doi:10.1016/j.actamat.2019.06.049.

[22] Y. Li, Y.H. Zhao, W. Liu, C. Xu, Z. Horita, X.Z. Liao, Y.T. Zhu, T.G. Langdon, E.J. Lavernia, Influence of grain size on the density of deformation twins in Cu-30%Zn alloy, Mater. Sci. Eng. A. 527 (2010) 3942–3948. doi:10.1016/j.msea.2010.02.076.

[23] Y.B. Wang, X.Z. Liao, Y.H. Zhao, E.J. Lavernia, S.P. Ringer, Z. Horita, T.G. Langdon, Y.T. Zhu, The role of stacking faults and twin boundaries in grain refinement of a Cu-Zn alloy processed by high-pressure torsion, Mater. Sci. Eng. A. 527 (2010) 4959–4966. doi:10.1016/j.msea.2010.04.036.

[24] Y. Cao, Y.B. Wang, X.H. An, X.Z. Liao, M. Kawasaki, S.P. Ringer, T.G. Langdon, Y.T. Zhu, Grain boundary formation by remnant dislocations from the de-twinning of thin nano-twins, Scr. Mater. 100 (2015) 98–101. doi:10.1016/j.scriptamat.2015.01.001.

[25] L.K.L. Falk, P.R. Howell, G.L. Dunlop, T.G. Langdon, The role of matrix dislocations in the superplastic deformation of a copper alloy, Acta Metall. 34 (1986) 1203–1214. doi:10.1016/0001-6160(86)90007-6.

[26] T. Sakai, A. Belyakov, R. Kaibyshev, H. Miura, J.J. Jonas, Dynamic and post-dynamic recrystallization under hot, cold and severe plastic deformation conditions, Prog. Mater. Sci. 60 (2014) 130–207. doi:10.1016/j.pmatsci.2013.09.002.

[27] H.K. Zhang, H. Xiao, X.W. Fang, Q. Zhang, R.E. Logé, K. Huang, A critical assessment of experimental investigation of dynamic recrystallization of metallic materials, Mater. Des. 193 (2020) 108873. doi:10.1016/j.matdes.2020.108873.