Rev. Adv. Mater. Sci. 31 (2012) 62-67

EFFECT OF HEAT TREATMENT ON NANOSTRUCTURING IN HIGH-STRENGTH ALUMINUM ALLOY BY SEVERE PLASTIC DEFORMATION

O. Sitdikov[1], S. Krymskiy[1], M. Markushev[1], E. Avtokratova[1] and T. Sakai[2]

[1]Institute for Metals Superplasticity Problems RAS, 39 Khalturin St., Ufa, Russia
[2]UEC Tokyo (The University of Electro-Communications), Chofu, Tokyo 182-8585, Japan

Received: April 20, 2012

Abstract. Effect of prior heat treatment on nanostructuring of high-strength 7475 Al alloy, subjected to high-pressure torsion (HPT) at ambient temperature up to strain of ~7, was investigated. The alloy was (i) solution treated and water quenched, and then (ii) artificially aged before HPT. In distinction with the state (i), the state (ii) was characterized by the presence of nanoprecipitates of strengthening η' - phase in Al matrix. It was shown that the initial alloy state significantly affected the morphology and parameters of nanocrystalline structures evolved. Mixed (sub)grain/cellular structure composed by both equiaxed and elongated crystallites, predominantly separated by dislocation boundaries, was developed in the aged state, whereas more equiaxed and coarse crystalline structure with more equilibrium boundaries formed in the as-quenched material. It was concluded that high density precipitates in the alloy can delay the formation of nanograins in the deformation structures due to inhibition of dynamic recovery and homogenization of the dislocation slip.

1. INTRODUCTION

Recent investigations have shown that there is a great potential for extraordinary improvement in the chemical, physical and mechanical properties of many metals and alloys by grain refinement, and especially by nanostructuring with processing (sub)grain size smaller than 100 nm [1]. Among other methods of nanostructuring severe plastic deformation (SPD) has received much attention due to the simplicity and applicability for all classes of crystalline materials. During last decade few effective SPD techniques have been developed and used [1-9], such as equal-channel angular pressing, high-pressure torsion (HPT) and multidirectional forging. However, gaps in understanding the mechanisms of grain refinement as well as the factors affecting nanostructuring upon SPD processing still exist. It may be related to the fact that many important data on nanostructure formation are still lacking. In particular, the effects of microstructural variables associated with the initial material, e.g. second phase particles of different origin, are not well documented in the scientific literature. Most research on influence of these particles has focused on their strengthening effects at conventional deformation [10,11] and much less information has been given on their influence on the features of grain refinement and development of nanostructures during severe straining [1,12-16]. Moreover, there are some opposite data in the literature on the role of second phase particles in nanostructuring under SPD. For instance, it has been shown in [12] for some aluminum-base alloys that the high densities of alumina particles result in strong increase in dislocation density and accelerate new finer grain formation in a strained material. In other works [13-15], in contrast, it has

Corresponding author: O. Sitdikov, e-mail: sitdikov@nm.ru

© 2012 Advanced Study Center Co. Ltd.

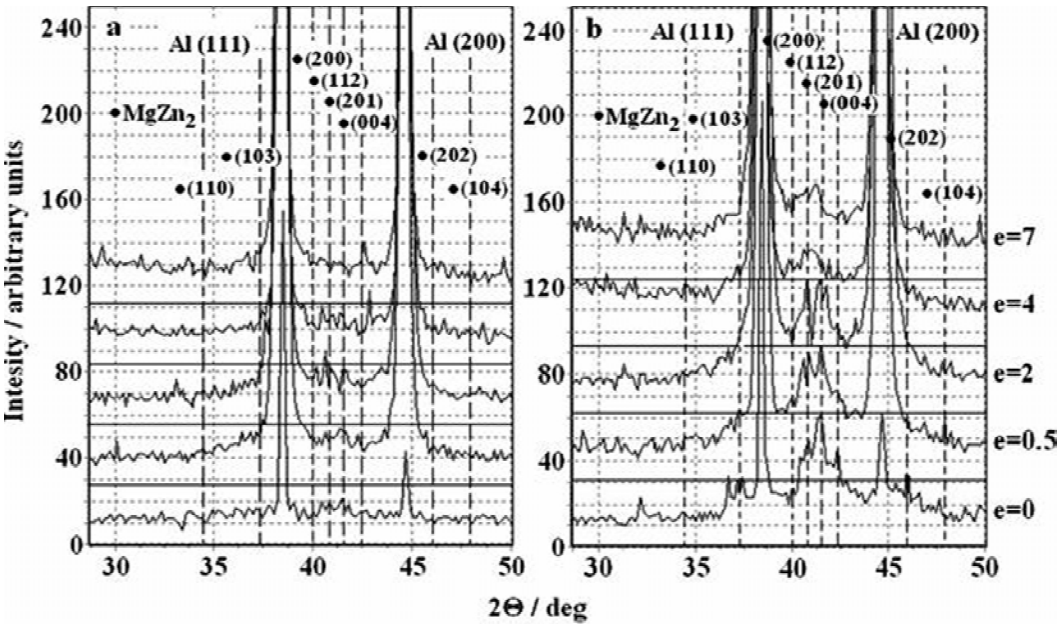

Fig. 1. XRD patterns of (a) as-quenched and (b) subsequently aged samples before and after HPT to different strains, *e*.

been pointed out that the presence of high densities of dispersoids of aluminides of transition metals and/or precipitates of the main strengthening phases inhibits the development of new high-angle boundaries and the formation of a nanocrystalline grain structure.

The aim of this work was to contribute to understanding the processing-structure relationship in the high-strength 7475 aluminum alloy subjected to HPT. For this purpose the deformation structure developed in the alloy containing the supersaturated solid solution has been studied and compared to that in the same alloy artificially aged before SPD. The material in both of the states was deformed under identical conditions by HPT to an effective strain of ~7 at ambient temperature. Effects of initial phase structure on the microstructure transformations taking place during SPD are discussed in detail.

2. EXPERIMENTAL PROCEDURE

The material used was a chill cast and homogenized at 490 °C for 20 hrs ingot of Zr modified 7475 aluminum alloy with the following chemical composition (in mass %): Al - 6.0Zn - 2.5Mg - 1.8Cu - 0.23Cr - 0.16Zr - 0.03Si - 0.04Fe - 0.03Mn. The ingot structure was presented by coarse lamellar grains and coarse particles of excess phases (see more details in [17,18]). The specimens 10 mm in diameter and 0.3 mm in thickness were machined from the ingot and heat-treated by two different routes, i.e. (i) water quenching from 475 °C to provide supersaturated solid solution [10,11] (hereafter "as-quenched samples") and (ii) quenching and aging at 120 °C for 8 hrs to provide η' - phase ($MgZn_2$) precipitates [10,11] (hereafter "aged samples"). Then the samples were subjected to HPT at ambient temperature up to strains (e) of 0.5, 1, 2, 4 and 7 under an applied pressure of 5 GPa. Strains imposed were calculated using equations represented in [2]. The hardness and structure of deformed samples were examined at a distance of about 3 mm from their center. The microhardness was evaluated using MHT-10 Vickers hardness tester with a load of 50-100 g on the diamond pyramid indenter. TEM structure was examined using a JEM-2000FX transmission electron microscope operating at 200 kV. The coherent domain size was determined by X-ray diffraction (XRD) analysis using the Williamson-Hall method.

3. RESULTS

3.1. Phase constituents

Typical XRD patterns of the as-quenched and aged samples before and after HPT to various strains are depictured in Fig. 1 with indication of aluminum and hexagonal η-phase peaks and indexes. It is seen that the as-quenched alloy before and after SPD appears mostly as a continuous solid solution with a minor quantity of second-phases, which are usually retained after solution treatment [6]. Among them, η-phase excess particles are reflected in Fig. 1a by

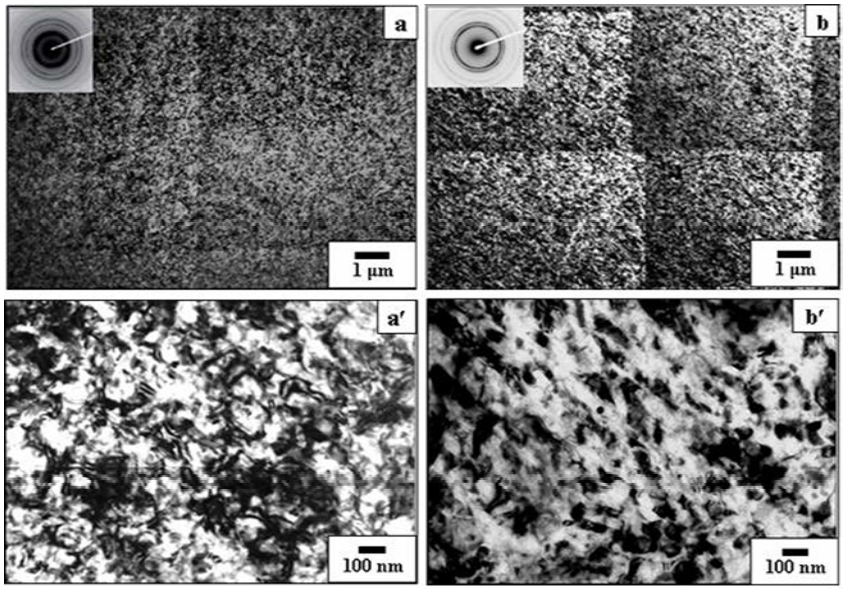

Fig. 2. Typical TEM micrographs of 7475 Al alloy in (a-a‘) as-quenched and (b-b‘) aged state processed by HPT to e ~ 7.

very weak peaks between Al (111) and (200) ones. X-ray patterns for aged samples exhibit a well-defined broad peak that is resolved into three other peaks, whose angular positions are a bit lower than those of η-phase (Fig. 1b). This may be a reflection from the metastable hexagonal η‘-phase, partially coherent with the Al matrix having larger than η-phase lattice parameter [10,19]. It is also evident in Fig. 1b that intensities of the peaks are scarcely changed after HPT to *e* ~2 and then somewhat decrease with subsequent straining to *e* ~4-7. This suggests that the strengthening phase particles may be partially dissolved in Al matrix at large strains. However, even after severe deformation, the difference in the phase structure of the as-quenched and aged samples still remains remarkable to affect the microstructural development.

3.2. Microstructure

Typical TEM structures for the as-quenched and aged samples subjected to HPT to *e* ~7 are represented in Fig. 2. For both states, the relatively uniform nanocrystalline structures can be seen with ring-like diffraction patterns (taken from the selected area of about 5 μm in diam.) suggesting predominant high-angle misorientations of boundaries. However, it can be also seen that there is the significant effect of the initial alloy state on the morphology and character of the microstructure evolved. That is, the essentially equiaxed crystallites with size of about 80-100 nm outlined by relatively sharp intercrystalline boundaries with distinct extinction fringes are frequently developed in the as-quenched state. In the aged state a finer mixed structure, composed of both equiaxed and elongated crystallites separated by dislocation subboundaries is formed. The size of equiaxed crystallites is about 50-80 nm and the elongated ones are up to about 300 nm in length. Thus, the present data suggest that HPT of the as-quenched material leads to more equiaxed crystallite structure with more equilibrium intergranular boundaries. It should also be noted that the microstructure formed at high strains in the aged alloy may be similar to that observed earlier in some cubic materials subjected to SPD at low-to-intermediate temperatures [7,8,20] and can be associated with development of a spatial net of microshear bands that are located in different directions and intersect the elongated cellular substructures with high density of lattice dislocations. Shearing by microshear bands leads to an apparent bending of the cell and microshear bands evolved by SPD, resulting in a characteristic wavy substructure (Fig. 2b‘).

3.3. Coherent diffraction domain size

Strain dependence of the mean crystallite size *D*, calculated from the XRD peak broadening, is shown in Fig. 3. It is seen that *D* in the as-quenched and aged states gradually decreases with strain and finally approaches roughly saturated values of about 20-50 nm. Besides, *D* remains always smaller in the aged state than in the as-quenched one at all strains investigated. It should be noted that its ab-

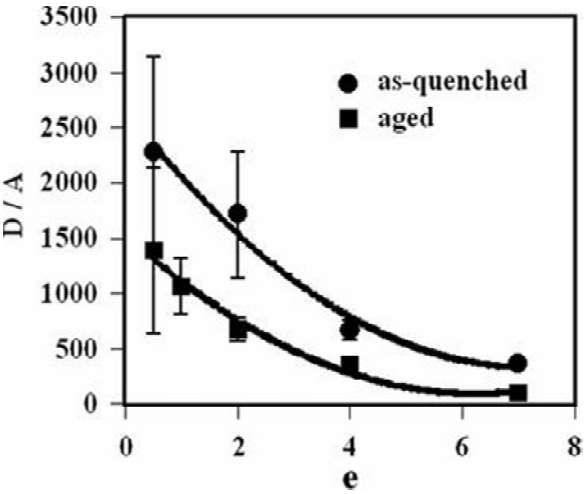

Fig. 3. Changes in coherent diffraction domain size in 7475 Al alloy on strain, as determined by X-ray analysis.

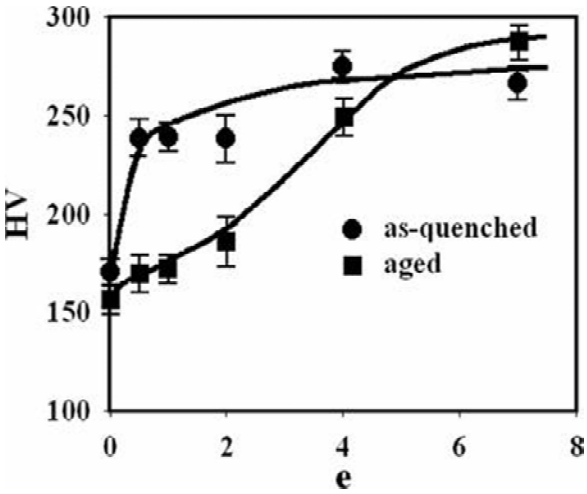

Fig. 4. Effect of strain on room-temperature Vickers hardness of HPT processed 7475 Al alloy.

solute value is significantly smaller than the crystal size derived from TEM observations, because XRD measurements give the coherent diffraction domain size, e.g. (sub)grain/dislocation cell size [19].

3.4. Microhardness

The variation of room-temperature alloy microhardness with strain is shown in Fig. 4. It is seen that in both alloy conditions, hardness increases with strain and reaches near the same values of Hv ~270-280 after *e* ~7. Besides, the as-quenched samples reveal more intense hardening, practically completed to *e* ~ 0.5-1, with no significant hardness changes with further straining. It is noted that such attenuating strain hardening behavior of the microstructures produced by HPT can be comparable with that of some cubic materials processed by SPD and typical of severe straining at low to intermediate temperatures [7,21]. The drastic increase in hardness at the earlier stages of deformation may be due to a strong work hardening caused by the rapid increase in the density of dislocations occurring with intense shear straining in the original grain interiors [21]. The reason why the hardness is slightly changed with further strain increase may be due to the rearrangement of dislocations in subboundaries assisted by dynamic recovery occurring during HPT. The hardness of the aged samples, in contrast, increases continuously with straining with roughly a constant hardening rate at *e* > 2. This suggests that more homogeneous deformation can be introduced in the aged material and then result in more uniform dislocation distribution in the deformation microstructure [22].

4. DISCUSSION

The experimental results described above testify that HPT of cast coarse-grained 7475 Al alloy, carried out at ambient temperature, results in formation the relatively uniform nanocrystalline structures and that the initial alloy state significantly affects the morphology and parameters of the SPD-ed microstructure. Let us consider the reasons of the alloy behavior observed.

It should be noted, that the new fine grain formation and evolution processes, occurring in some cubic materials during cold-to-hot SPD, are well represented and discussed in the literature. For instance, in the previous works [7,8,13,17,18,20,23] it has been shown that high strain and misorientation gradients can be introduced into original coarse grains in the earlier stages of SPD and, concurrently, new low-to-moderate and moderate-to-high angle dislocation boundaries that are related mainly to the boundaries of microshear bands evolve often in the regions with high local lattice rotations. It has been also pointed out that evolution of microshear bands can play a key role in occurrence of grain refinement during SPD. Namely, several sets of these bands, which are developed in various directions, fragment original grains into small separate misoriented domains. A gradual increase in the number and misorientation of the boundaries of microshear bands and their conversion into high-angle boundaries can result in the development of the new ultra fine-grained structure with further straining.

A similar grain refinement process may also occur in the 7475 Al alloy, in which microshear bands can

be frequently developed by HPT (see Fig. 2 b'). However, the initial grain subdivision by microshear bands as well as rigid lattice rotations within these bands may be considered as some mechanically-induced events, and so these events may have athermal nature [7,8,23]. On the other hand, the strain-induced grain formation is usually resulted not only from the mechanical formation of high-angle boundaries at lower strains, but also from the frequent operation of dynamic recovery in these boundaries at larger strains [23], because the boundaries of microshear bands introduced by SPD are rather non-equilibrium and diffuse interfaces [7,13]. Dynamic recovery that occurs even at ambient temperature can assist the transformation of such strain-induced non-equilibrium boundaries to more equilibrium ones, leading to both an increase of grain boundary misorientation and a decrease of dislocation density in (sub)grain interiors. Microstructural changes during SPD are, therefore, considered to be effectively controlled by thermally activated rate processes at large strains [8,23]. The observed steady state-like hardness dependence on strain at $e \geq 1$ (Fig. 4) implies that dynamic recovery may play an important role in evolution of the deformation microstructure in the as-quenched state of the alloy. It can be generally considered that more rapid operation of dynamic recovery in this state may assist the formation of coarser and more equiaxed grains with more equilibrium boundaries. In the aged alloy, in contrast, dynamic recovery is more strongly repressed due to the presence of the dispersed secondary phases, which can prevent the rearrangement of lattice dislocation and/or migration of dislocation (sub)boundaries. The strong inhibition of recovery retards the formation of nanocrystalline grain structure at least until much higher strains, where some equiaxed crystallites are eventually formed.

Another factor that may hinder the nanostructure evolution in the aged alloy state is conditioned by the homogenization of dislocation slip due to a strong interaction of lattice dislocations with the (semi)coherent dispersed particles of η'-phase [13,15]. The latter may provide a higher resistance to localization of plastic flow, as compared with as-quenched alloy, and, hence, a lower density of more diffuse microshear bands can develop in the grain interiors during SPD [13]. It is safely to assume that the formation of more weakly misoriented and diffuse boundaries in the particle-containing alloy at the earlier stages of deformation can block the most important mechanism of high-angled boundary development mentioned above and thus delay the formation of a well-developed nanocrystalline grained structure at ultrahigh strains.

7. CONCLUSION

The 7475 aluminum alloy was subjected to HPT at ambient temperature up to strain of ~7. The deformation structure developed at high strains in the solution treated and water quenched alloy with preliminary supersaturated aluminum solid solution was compared to that in the same alloy artificially aged before HPT. In distinction with the as-quenched state, the microstructure of the aged alloy was characterized by the presence of high densities of nanoprecipitates of strengthening η'-phase in aluminum matrix. The main results can be summarized as follows.

(1) Relatively uniform nanocrystalline structures can be produced by HPT in both of the alloy states accompanied by significant increase in its hardness with a factor of about 1.8. In the as-quenched alloy, hardness increases rapidly in the earlier stages of deformation followed by saturation at higher strains. In contrast, in the aged state, hardness growths gradually with almost constant hardening rate, finally approaching the same saturation value at high strains.

(2) The initial alloy state significantly affects the morphology and characteristics of the microstructure induced by HPT. Namely, a mixed (sub)grain/cellular structure, which is composed by both equiaxed and elongated crystallites separated by high-density dislocation subboundaries, is developed in the aged state, whereas SPD of the as-quenched material leads to formation of a rather uniform microcrystalline structure composed by more equiaxed and coarse crystallites with more equilibrium boundaries.

(3) It is concluded that the presence of the metastable η-phase precipitates in the deformation structure of the preliminary aged alloy may delay the formation of equiaxed nanoscale grains even at large strains due to inhibition of dynamic recovery and homogenization of dislocation slip.

REFERENCES

[1] R. Valiev, R. Islamgaliev and I. Alexandrov // *Prog. Mat. Sci.* **45** (2000) 103.

[2] V.A. Shabashov, A.V. Litvinov, A.G. Mukoseev, V.V. Sagaradze, D.V. Desyatkov, V.P. Pilyugin, I.V. Sagaradze and N.F. Vildanova // *Mat. Sci. Eng.* **A361** (2003) 136.

[3] A.P. Zhilyaev and T.G. Langdon // *Prog. Mat. Sci.* **53** (2008) 893.

[4] R.R. Mulyukov, R.M. Imayev and A.A. Nazarov // *J. Mater. Sci.* **43** (2008) 7257.
[5] C. Xu, S.V. Dobatkin, Z. Horita and T.G. Langdon // *Mat. Sci. Eng.* **A500** (2009) 170.
[6] P.V. Liddicoat, X.-Z. Liao, Y. Zhao, Y. Zhu, M.Y. Murashkin, E.J. Lavernia, R.Z. Valiev and S.P. Ringer // *Nature Communications* **1** (2010) DOI:10.1038 /ncomms1062
[7] C. Kobayashi, T. Sakai, A. Belyakov and H. Miura // *Phil. Mag. Lett*. **87** (2007) 751.
[8] T. Sakai, A. Belyakov ans H. Miura // *Met. Mat. Trans.* **A39** (2008) 2206.
[9] M.V. Markushev // *Letters on Materials* **1**(2011) 36 (in Russian).
[10] L.F. Mondolfo, *Aluminum Alloys: Structure and Properties* (Butter Worths, London – Boston, 1979)
[11] Information on http://aluminium.matter.org.uk
[12] C.Y. Barlow, N. Hansen, Y.L. Liu // Acta Mat. 50 (2002) 171.
[13] P.J. Apps, M. Berta and P.B. Prangnell // *Acta Mat.* **53** (2005) 449.
[14] M. Berta, P.J. Apps and P.B. Prangnell // *Mat. Sci. Eng*. **A410-411** (2005) 381.
[15] M.V. Markushev // *Phys.Met.Metallography* **108** (2009) 161.
[16] I. Nikulin, A. Kipelova, S. Malopheyev and R. Kaibyshev // *Acta Mat.* **60** (2012) 487.
[17] O. Sitdikov, T. Sakai, A. Goloborodko, H. Miura and R. Kaibyshev // *Philos. Mag*. **85** (2005) 1159.
[18] O. Sitdikov, T Sakai, H. Miura and C. Hama // *Mat. Sci. Eng*. **A516** (2009) 180.
[19] Y.H. Zhao, X.Z. Liao, Z. Jin, R.Z. Valiev and Y.T. Zhu // *Acta Mater*. **52** (2004) 4589.
[20] P. J. Hurley and F. J. Humphreys // *Acta Mat.* **51** (2003) 1087.
[21] S.-Y. Chang, J.G. Lee, K.-T. Park and D.H. Shin // *Mat. Trans*. **42** (2001) 1074.
[22] J. Gil Sevillano, P. van Houtte and E. Aernoudt // *Prog. Mat. Sci*. **25** (1980) 69.
[23] I. Mazurina, T. Sakai, H. Miura, O. Sitdikov and R. Kaibyshev // *Mat. Sci. Eng*. **A486** (2008) 662.