Rev.Adv.Mater.Sci. 25 (2010) 74-81

THERMAL STABILITY OF THE ULTRAFINE-GRAINED Al-Cu-Mg-Si ALUMINUM ALLOY

M.A.Nikitina, R.K.Islamgaliev and A.F.Kamalov

Institute of Physics of Advanced Materials, Ufa State Aviation Technical University, 12 K.Marx Str., Ufa 450000, Russia

Received: December 15, 2009

Abstract. Microstructure and mechanical properties of ultrafine-grained (UFG) samples of the Al-Cu-Mg-Si aluminum alloy processed by high pressure torsion have been investigated. Transmission electron microscopy and electron back scattering diffraction were used for structural investigations. The ultimate tensile strength and ductility were evaluated using tensile tests of small samples. Dynamic dissolution and precipitation in UFG samples has been observed. Special attention was paid to influence of precipitates on thermal stability of UFG samples.

1. INTRODUCTION

It is known that application of high pressure torsion (HPT) [1] contributes to formation of ultrafine-grained structure and achievement of a high strength in various metals and alloys [1-4].

At the same time a coarsening of the grains is observed in many HPT metals even at low homological temperatures [5].

The reasons for grain growth in UFG materials are the enhanced grain boundary energy and high grain boundary mobility [6].

It is known that stabilization of UFG structure can be achieved by two basic ways.

The first is the reduction of the grain boundaries mobility by second phase drag and solute drag. For example in the work [7] it has been shown that the presence of scandium-bearing particles in model aluminum alloys of the Al-Mg-Sc system allow for retaining the enhanced microhardness up to the temperatures of 300-350 °C. Alongside with that the investigation of mechanical properties on tension has not been carried out in this work, which would be important, as it is known that the linear relation between the yield strength and microhardness is violated after annealing of nanomaterials at elevated temperatures [8].

The second is the lowering of the specific grain boundary energy by solute segregation to the grain boundaries. It can be achieved by segregation of the alloying elements on the grain boundaries [2], thereby reducing the elastic strain energy. To induce the lowering of the grain boundary energy and stabilize the grain size against coarsening the atomic radius of the alloying elements should be much larger

Corresponding author: R.K.Islamgaliev, e-mail: saturn@mail.rb.ru

Table 1. Chemical composition of the AK4-1 aluminum alloy, %.

	Cu	Mg	Fe	Ni	Si	Ti	Mn	Zn
EDS	2.46	1.48	0.89	0.92	0.22	0.04	0.04	-
Literature data [11]	1.9-2.7	1.2-1.8	0.8-1.4	0.8-1.4	0.35	0.02-0.1	0.2	0.3

or smaller than the atomic radius of aluminum, and the enhanced content of alloying elements will contribute to the decrease of the grain boundary energy to the minimum. For example, the use of the second approach allowed for increasing the stability of nanostructures in the Pd-Zr, Fe-Zr, Cu-Nb alloys up to 0.8 T_m [6], where T_m – is the melting temperature.

Alongside with that it has not been reported about a stabilization of UFG structure in HPT samples of commercial aluminum alloys at temperatures over 150 °C.

The aim of this work was to investigate the stability of UFG structure in HPT samples by the example of the commercial aluminum alloy of the AL-Cu-Mg-Si-Fe-Ni system, which contains the alloying Mg and Cu elements, the atomic radius of which differs from the atomic radius of aluminum by 0.017 nm, and 0.015 nm, correspondingly. In addition this alloy contains fine Al_9FeNi particles, which are stable up to the temperature of 450 °C [9].

2. EXPERIMENTAL

The commercial aluminum AK4-1 alloy has been chosen as a material for investigations, its chemical composition in accordance with the energy-dispersive analysis (EDS) and literature data is represented in Table 1.

To produce the UFG samples the high pressure torsion technique on the original die-set, allowing for manufacturing samples under the force of 200 tons, was used [10]. Before the deformation the initial billets were subjected to solid solution treatment at the temperature of 530 °C for 1 hour with a following water quenching.

The HPT treatment of the samples with a diameter of 20 mm and a thickness of 1 mm was carried out under the pressure of 6 GPa at a temperature of 20 °C. The annealing of HPT samples was carried out at the temperatures of 50, 100, 150, 175, 200, 225, 250, 275, and 300 °C for 30 min.

Structural investigations were performed on the JEM-100B and JEM-2100 transmission electron microscopes. The average grain size *D* was calculated by the results of diameter measurements of over 100 grains. Selected area diffraction patterns were taken from the area of 2 μm^2.

The misorientation spectra and the chemical composition of the samples were examined in the JSM-6390 scanning election microscope, which is equipped by electron back scattering diffraction (EBSD) and energy-dispersive analysis.

The X-ray analysis has been performed on the Rigaku diffractometer with the use of Cu$K\alpha$-radiation at 50 kW and 40 mA. The dimensions of the coherent-scattering domains (*D*) and the values of root-mean square strains ($\langle\varepsilon^2\rangle^{1/2}$) were calculated with the application of the Williamson-Hall method. In order to determine the crystal lattice parameter *a* of the alloy before and after HPT the extrapolation procedure by Nelson-Reilly was used [9].

The temperatures of precipitation and dissolution of precipitates were determined by the differential scanning calorimeter Netzsch STA 409 PC in the air atmosphere at a heating rate of 10 K/min.

The microhardness measurements were performed with the load of 100 g for 10 sec, on the Micromet-5101 device, equipped with a digital camera and computer software for pattern analysis.

The tensile tests at room temperature were performed on the die-set, equipped with a horizontal measuring-power device [10], with a strain rate of $10^{-3}\,s^{-1}$. The mechanical properties were evaluated on the samples with the dimension of the gage section of 2x1x0.4 mm^3 which had been cut by the electrospark machine.

All investigations of microstructure and mechanical properties were performed on the areas, positioned on the distance of a half a radius of HPT samples.

3. RESULTS AND DISCUSSION

According to the results of the investigations in the optical microscope the average grain size in the initial coarse-grained samples after water quenching was about 40 μm.

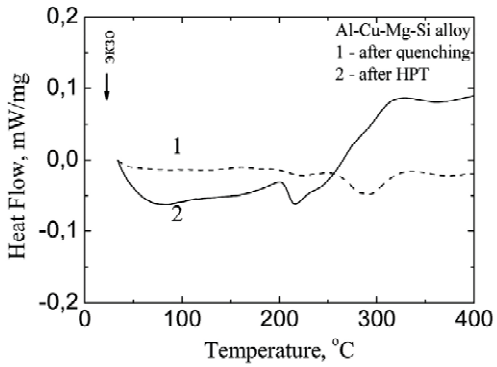


Fig. 1. Dependence of heat release on the temperature in the Al-Cu-Mg-Si alloy, observed in the differential scanning calorimeter at heating rate of 10 K/min: dashed line – after quenching; solid line – after HPT.

An explicit exothermic peak within the temperature range of 270-290 °C (Fig. 1) has been observed on the dependence of heat release from the temperature, which is connected, obviously, with precipitation of particles contributing to strengthening of the material.

In the HPT sample of the Al-Cu-Mg-Si alloy a wide exothermic peak in the temperature range of 200-270 °C (Fig. 1) consisting of two peaks, has been recorded. In order to analyze the origin of these peaks the structural investigations of HPT samples using transmission electron microscopy (TEM) were carried out.

It has been established that HPT resulted in the grain refinement to the average grain size of 200 nm (Fig. 2). A complex diffraction contrast on the bright-field and dark-field images testified to the presence of high internal stresses in the structure of the samples.

After annealing at a temperature of 175 °C no visible changes in the structure of HPT samples have been observed.

Annealing at a higher temperature of 200 °C was accompanied by relaxation of internal stresses and a beginning of grain growth to 300 nm with precipitation of globular particles with an average size of 50 nm (Fig. 3).

The following annealing of HPT samples at a temperature of 225 °C led to an additional grain growth to 570 nm and the increase of the size of particles to 100 nm (Fig. 4a). The analysis of the electron-diffraction pattern from these particles showed that they are the Al_9FeNi phase particles (Fig. 4b). Along-

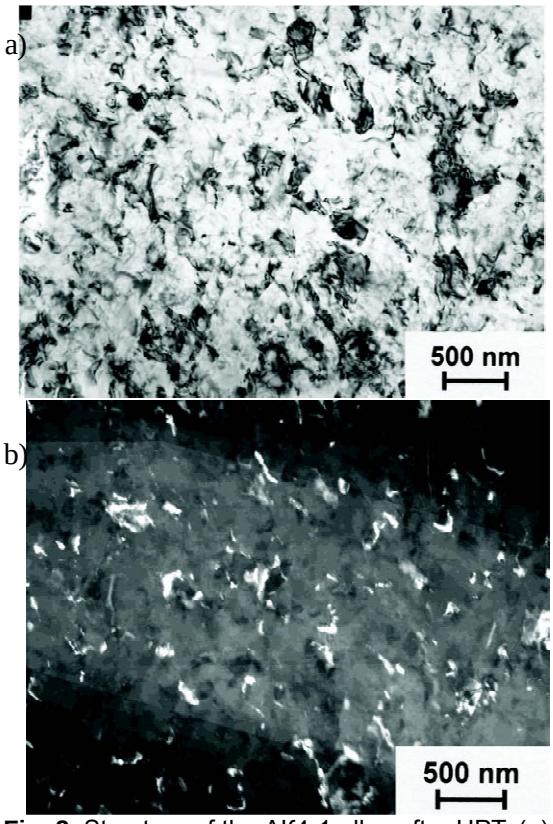


Fig. 2. Structure of the AK4-1 alloy after HPT: (a) bright-field image; (b) dark-field image.

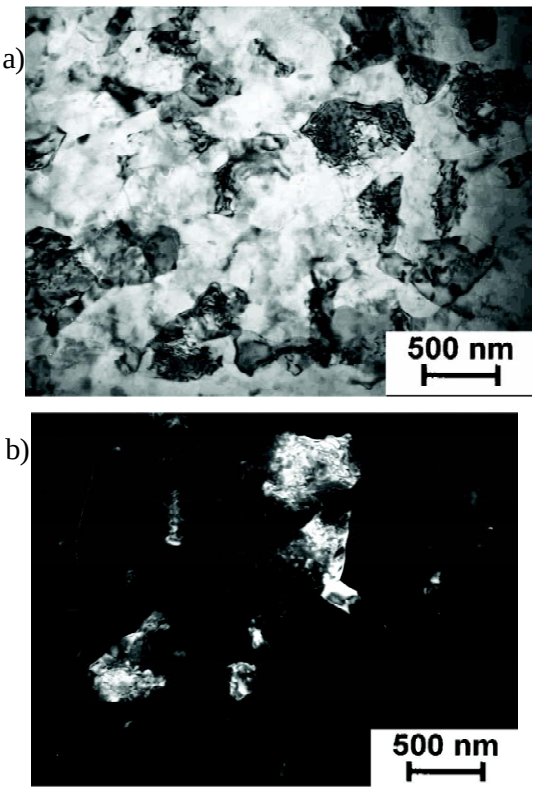


Fig. 3. Structure of the AK4-1 alloy after HPT and annealing at the temperature of 200 °C for 30 min: (a) bright-field image; (b) dark-field image.

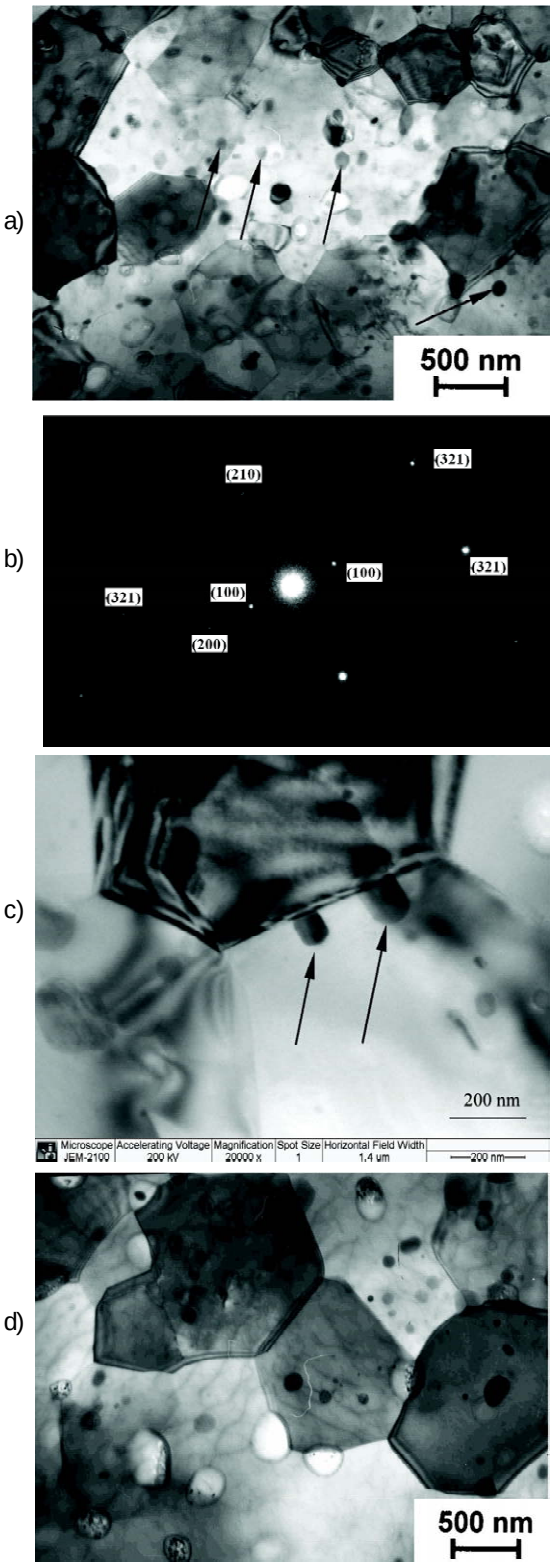


Fig. 4. Structure of the AK4-1 alloy after HPT and annealing at a temperature of: (a, b, c) 225 °C; (d) 250 °C for 30 min; (b) electron-diffraction pattern from the globular Al_9FeNi particles indicated by arrows in the pattern (a); (c) arrows illustrated the elongated Al_2CuMg particles.

side with that in the structure there were also particles of an elongated shape, belonging to the Al_2CuMg phase (Fig. 4c).

After annealing at 250 °C coarsening of both the grains up to 900 nm and precipitates up to 200 nm has been observed (Fig. 4d).

According to EBSD analysis there is a number low-angle grain boundaries in coarse-grained billets of the AK4-1 alloy, subjected to a solid solution treatment at the temperature of 530 °C, for 1 hour with the following water quenching and ageing at the temperature of 180 °C for 12 hours (Fig. 5).

After high pressure torsion and additional annealing at 250 °C for 30 min the structure of samples was characterized not only by the ultrafine grains

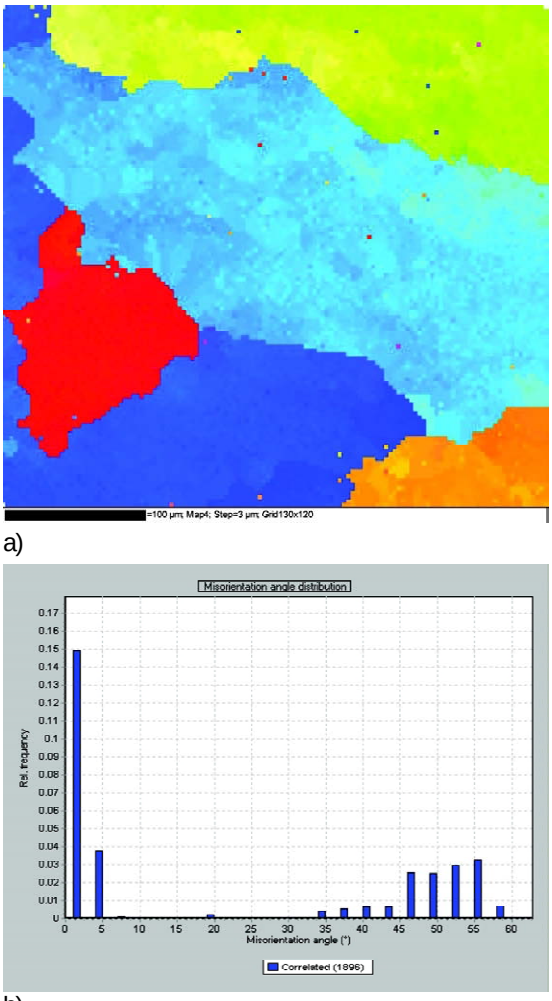
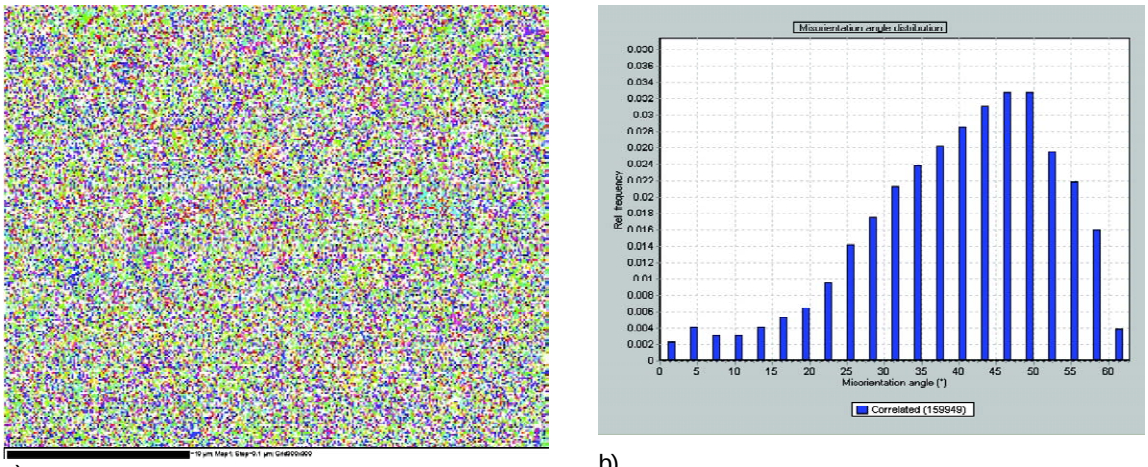


Fig. 5. The AK4-1 alloy after the after the solid solution treatment: (a) orientation map; (b) distribution of misorientation angles.



a) b)

Fig. 6. The AK4-1 alloy after HPT and annealing at the temperature of 250 °C for 30 min: (a) orientation map; (b) distribution of misorientation angles.

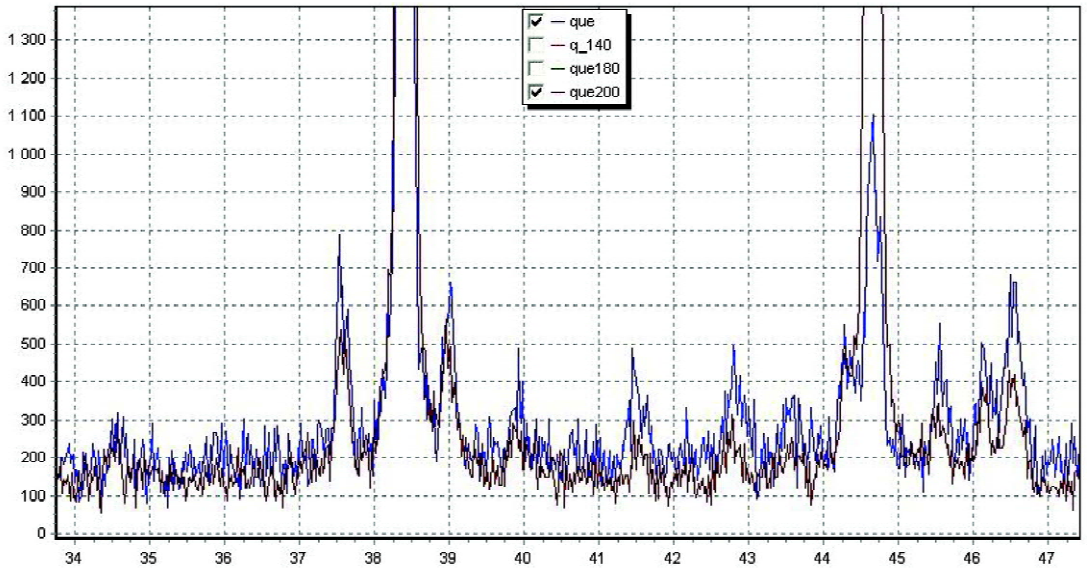


Fig. 7. X-ray diffraction patterns of the aluminum AK4-1 alloy after quenching (blue color) and additional annealing at 220 °C for 5 hours (red color).

with the average size of about 900 nm, but also by predominantly high-angle grain boundaries, the share of which exceeded 95 % (Fig. 6).

X-ray analysis showed that after quenching there are additional peaks (Fig. 7), the location of which coincided with the location of peaks for intermetallics containing aluminum, magnesium and copper (Al_2CuMg, $Mg_2Cu_6Al_{15}$, $AlCu_3$, $AlCu_4$, $Mg_{25}Cu_{27}Al_{23}$).

The annealing of the quenched samples at 220 °C did not lead to changes of quantity and location of X-ray peaks, but only to a slight redistribution of their intensity (Fig. 7).

No visible X-ray peaks of intermetallic phases has been revealed in the HPT samples which had been observed in the quenched sample, that testified to a complete dynamic dissolution of particles as a result of HPT processing (Fig. 8). Their dissolution was also confirmed by a significant increase of the lattice parameter from 4.0540 Å after quenching to the value of 4.0561 Å after HPT (Table 2).

Alongside with that the appearance of new X-ray peaks of a low intensity has been revealed in HPT samples, the location of which coincided with the location of peaks for the Al_9FeNi phase, which play

Table 2. Lattice parameter, size of coherent-scattering domains and mean-root square strains in the AK4-1 alloy after various treatments.

Sample	*Treatment*	*Lattice parameter,* Å	*Size of coherent-scattering domains,* nm	*Mean-root square strains,* %
Coarse-grained	quenching	4.0540		
	quenching + annealing 200 °C	4.0536		
Ultrafine-grained	HPT	4.0561	62 ± 4	0.183 ± 0.012
	HPT + annealing 140 °C	4.0536	68 ± 5	0.169 ± 0.013
	HPT + annealing 180 °C	4.0519	102 ± 3	0.068 ± 0.005
	HPT + annealing 200 °C	4.0515	178 ± 8	0.038 ± 0.003

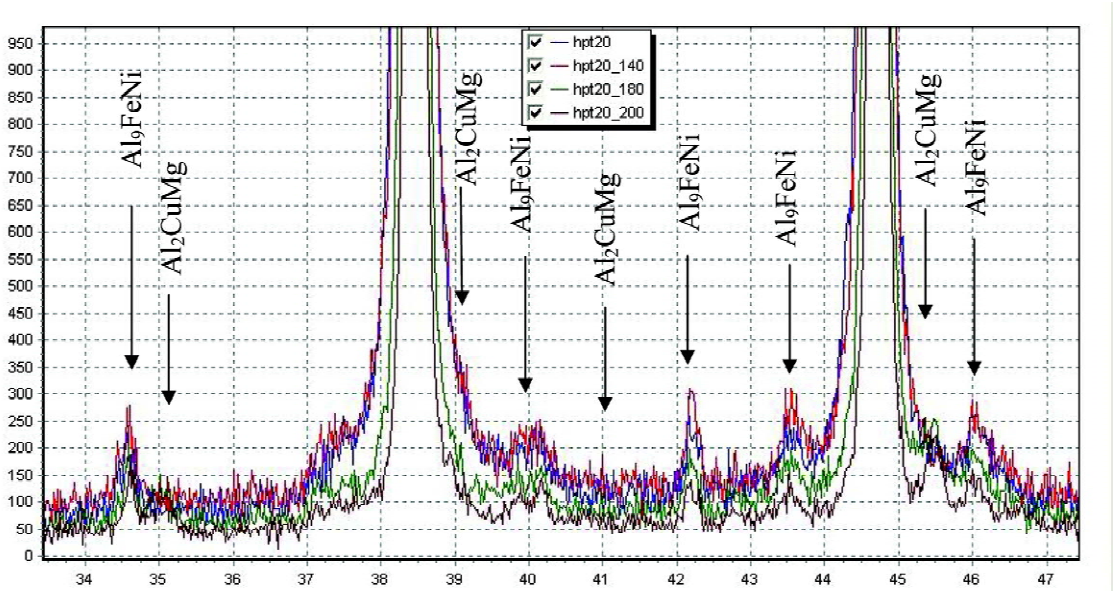


Fig. 8. X-ray diffraction patterns of the aluminum AK4-1 alloy after HPT (blue color) and additional annealing at a temperatures of 140 °C (red color), 180 °C (green color), 200 °C (brown color) for 5 hours.

an important role in formation of high strength in this alloy in the process of ageing at elevated temperatures [11]. This phase is represented in the form of globular particles with the size of about 50 nm in the corresponding TEM micrographs (Figs. 4a and 4b). Appearance of these peaks testified to the dynamic precipitation of these particles during HPT at room temperature, because these precipitates were absent in coarse-grained samples annealed at 200 °C (Fig. 8).

Annealing of HPT samples at the temperatures of 140, 180, and 200 °C for 5 hours revealed that location and intensity of peaks of Al_9FeNi phase particles remain stable up to a temperature of 200 °C, though there was a considerable decrease of the lattice parameter from the value of 4.0561 Å in HPT sample to the value of 4.0515 Å after additional annealing at 200 °C. This decrease of the lattice parameter could be connected with appearance of additional peaks of the Al_2CuMg phase particles (Fig. 8), which are observed in the form of elongated particles in TEM micrographs (Fig. 4c).

The analysis by the Williams-Hall method showed that there is a relaxation of elastic strains from the value of 0.183% to the value of 0.038% after additional annealing at 200 °C. High values of elastic strains have been observed in HPT samples of other aluminum alloys [2-4]. Their relaxation with the increase of annealing temperature can be caused by decrease of concentration of atoms of

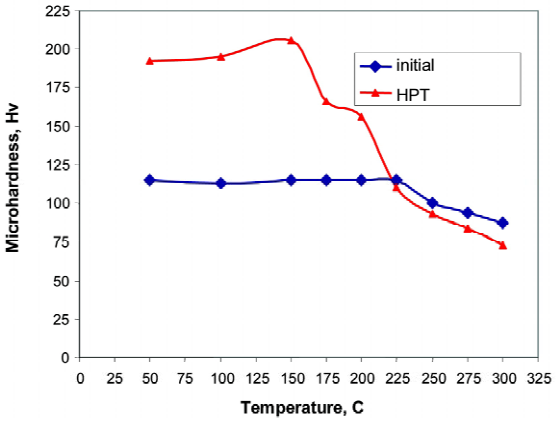


Fig. 9. Dependence of the AK4-1 alloy microhardness on the annealing temperature for 30 min.

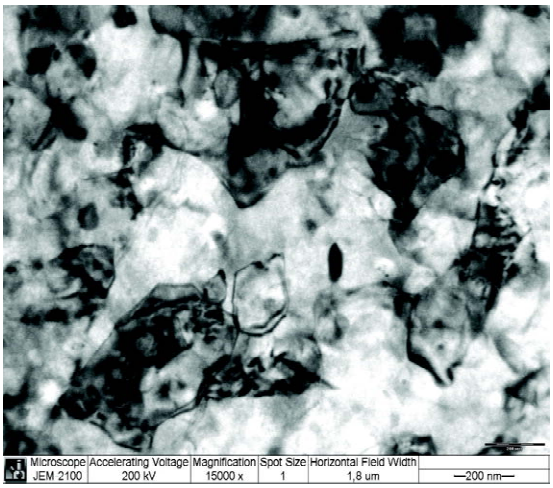


Fig. 10. Structure of the AK4-1 alloy after HPT and annealing at 180 °C for 5 hours.

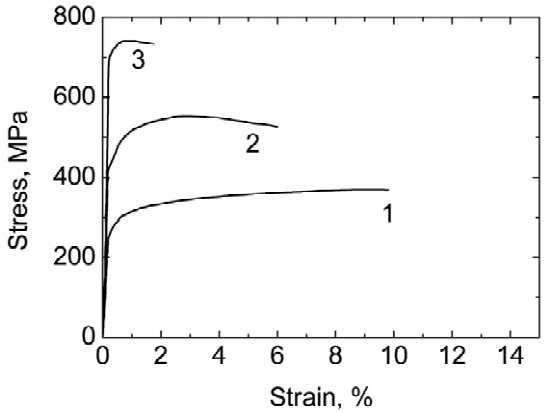


Fig. 11. Engineering stress–strain curves of the AK4-1 alloy after various treatments: (1) quenching and ageing at a temperature of 180 °C for 12 hours; (2) HPT and annealing at a temperature of 180 °C for 5 hours; (3) HPT and annealing at a temperature of 180 °C for 1 hour.

alloying elements in a solid solution as a consequence of their precipitation in the form of segregations and particles in the quantity below the sensitivity of X-ray analysis.

In the UFG samples, obtained by HPT technique, the value of microhardness was equal to 1950 MPa, which is considerably higher than the microhardness of 1110 MPa in the initial coarse-grain material (Fig. 9). After annealing at the temperature of 150 °C there was a considerable increase of microhardness to 2100 MPa, which is probably connected with the beginning of segregation of alloying elements at grain boundaries [2].

After annealing at 175 °C the decrease of microhardness caused by relaxation of elastic strains (Table 2), which usually proceeds to beginning of grain growth, was observed. The considerable grain growth at higher temperatures (Figs. 3 and 4) led to the following lowering of microhardness (Fig. 9).

Tensile tests showed that there was a very high ultimate tensile strength, reaching a value of 800 MPa in UFG samples of the AK4-1 alloy, processed by HPT, but with that the ductility decreased to 1%. For comparison in the coarse-grained samples, subjected to conventional treatment T6 (quenching and ageing at 180 °C, 12 hours) the ultimate tensile strength was equal to 370 MPa with ductility of 10%. The enhanced strength in HPT samples can be explained by formation of UFG structure, while the decrease of ductility is conditioned obviously by a high level of internal stresses near grain boundaries and precipitates, which raise difficulties in generation of dislocations and development of dislocation slip. In order to increase ductility the HPT samples of the AK4-1 alloy were subjected to an additional annealing at 180 °C for 1 and 5 hours to reduce the internal stresses without visible changes in the average grain size (Fig. 10). As a result the UFG samples have demonstrated the enhanced ultimate tensile strength of 750 MPa and 550 MPa with ductility of 2 % and 6%, respectively (Fig. 11). It should be noted that favorable effect from lowering of internal stresses on ductility of Al alloys processed by HPT has been observed also in [4].

4. CONCLUDING REMARKS

The application of high pressure torsion to the aluminum AK4-1 alloy leads to a various change in microstructure: a strong grain refinement from 40 μm to 200 nm, increase of the lattice parameter to 4.0561 Å, enhancement of mean-root square strains to 0.183%, formation of high-angle grain boundaries on majority of grains and dynamic precipitation of Al_9FeNi particles.

UFG structure has been stable up to 250 °C though relaxation of elastic strains with subsequent grain growth and precipitation of Al_2CuMg particles has been observed.

The UFG samples of the AK4-1 alloy have demonstrated an enhanced microhardness of 1950 MPa, which is higher by 70% in comparison with the microhardness of the initial coarse-grained state. The enhanced value of microhardness of the HPT samples retained after annealing at a temperature of 200 °C.

The application of HPT and subsequent annealing at 180 °C allowed for processing of ductile samples with high ultimate tensile strength of 750 MPa which is more than in two times higher in comparison with strength of 370 MPa in the coarse-grained samples subjected to solid solution treatment.

ACKNOWLEDGEMENT

The authors are appreciate to A.R. Kilmametov for execution of X-ray investigations. This work was supported by Federal Target-oriented Program “Research and **development of priority directions** of elaboration of the scientific and technological **complex** of **Russia** on **2007-2012** years” within the framework of contract No. 02.513.11.3471.

REFERENCES

[1] R.Z.Valiev, R.K.Islamgaliev and I.V.Alexandrov // *Progr. Mater. Sci.* **45** (2000) 103.

[2] G.Nurislamova, X.Sauvage, M.Murashkin, R.Islamgaliev and R.Valiev // *Phil. Mag. Lett.* **88** (2008) 459.

[3] M.Y.Murashkin, A.R.Kilmametov and R.Z.Valiev // *Phys. Met. Metallogr.* **106** (2008) 93.

[4] R.K.Islamgaliev, N.F.Yunusova, I.N.Sabirov, A.V.Sergeeva and R.Z.Valiev // *Mat. Sci. Eng.* **A319-321** (2001) 874.

[5] N.M.Amirkhanov, J.J.Bucki, R.K.Islamgaliev, K.J.Kurzydlowski and R.Z.Valiev // *Journal of metastable and nanostructured metals* **9** (2001) 21.

[6] C.C.Koch, R.O.Scattergood, K.A.Darling and J.E.Semones // *J. Mater. Sci.* **43** (2008) 7264.

[7] S.V.Dobatkin, V.V.Zakharov and L.L.Rokhlin // *Mater. Sci. Forum* **503-504** (2006) 399.

[8] R.K.Islamgaliev, I.Y.Pyshmintsev, V.A.Khotinov, A.V.Korznikov and R.Z.Valiev. *Phys. Met. Metallogr.* **86**, 4 (1998) 115.

[9] N.A.Belov and V.S.Zolotarevsky // *Russian chemical journal* **5-6** (2001) 15.

[10] R.K.Islamgaliev, V.V.Latysh, A.R.Kilmametov, V.U.Kazykhanov, Y.T.Zhu, T.C.Lowe and R.Z.Valiev, In: *Ultrafine Grained Materials IV*, ed. by Y.T.Zhu, T.G.Langdon, Z.Horita, M.J.Zehetbauer, S.L.Semiatin and T.C.Lowe (The Minerals, Metals and Materials Society, 2006), p. 183.

[11] *Industrial aluminum alloys*, ed. by F.I. Kvasov and I.N. Fridlander (Metallurgy, Moscow, 1984).