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# Effect of Particles of the Eutectic Phases on the Structure of Superplastic Aluminium Alloys

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The fine grain structure and high parameters of superplasticity can be achieved by structure heterogeneity controlling. The use of eutectic particles for the grain structure control is very actual. Aluminium base materials of the Al – Ni, Al – Mg – Si, Al – Ni – Ce and Al – Cu – Ce systems were investigated. The alloys were found in the hypoeutectic area on the phase diagrams. The investigated materials included 0 – 27 volume per cent particles of eutectic phase with size of 0.3 – 4 µm. The particles influence on the grain growth control and stimulated grain nucleation was researched. Theoretical Zener-Smith`s law dependence of grain size on the particle parameters was confirmed and experimental coefficients were found. But experimental coefficients of Zener-Smith`s equation, which obtained in this work, depend on the particle size and differ from theoretical coefficients of this law.

Some alloys having a grain size of 3 µm demonstrated vary good superplasticity indicators. The strain rate sensitivity index m = 0.5-0.6 and elongation over 400 % were obtained at optimum constant strain rate $5 \times 10^{-3} s^{-1}$.

***Keywords:*** *Aluminium alloys, recrystallization, grain refinement, superplastic deformation*

## 1. Introduction

The phenomenon of superplasticity is used in the manufacture of products using superplastic forming, in which the whole product is obtained a given matrix of complex shape under the influence of gas per one molding operation. This technology decreases the number of seams in the product, reduces the weight of products and equipment costs. Superplasticity is the ability of polycrystalline materials to exhibit in an isotropic manner extensive tensile deformations prior to failure.Superplasticity effect occurs in alloys with grain sizes less than 10-12 microns, and at particular temperature and stain rate [1, 2]. The grain size less than 10 µm after recrystallization can be received in aluminum by optimizing of structure heterogeneity to use the hard particle phases of various origins.

A lot of research has been directed at the development of thermomechanical processes for the grain refinement of aluminium alloys by static or dynamic recrystallization. The deformation ratio, the temperature of deformation and recrystallization and the heating rate are parameters influencing on the recrystallization process and the grain size. Particle size, particle volume fraction and interparticle space significantly influence on the recrystallization [3-5]. Many investigations have been carried out on the effect of a dispersion of non-deformable particles oxides, carbide and other non-crystallization character of particles on the recrystallization [6–8]. The role of eutectic phase particles on the recrystallization and the grain size in aluminium alloys was investigated poor. Very perspective to use the eutectic alloys for the grain size control and superplasticity. For example, the eutectic alloys of Al – Ca, Al – Ca – Zn, Al – Ca – Si [9], systems have the grain size of 1-3 µm and very perspective for superplasticity The most of authors to investigate of recrystallization heterogeneous alloys assume the large undeformable (rigid) particles (> 1µm) as a nucleation sites and increased the rate of recrystallization (PSN); and the small particle (< 0.1 µm) are retarding recovery and primary recrystallization, inhibiting low angle boundaries migration (Zener pinning) [ 3-5,10,11]. The both of them influence on the grain structure. The main target of this work is to research influence quantitative of the eutectic particles on the nucleations of recrystallization and pinning boundaries for micro size grain structure formation and superplastisity.

## 2. Experimental details

The aluminium base materials of Al – Ni, Al – Mg – Si [12], Al – Cu – Ce [13] and Al – Ni – Ce system in aluminium rich corner were the objects of this study. The alloys of this system were consisted of the $Al_3Ni$, $Mg_2Si$, $Al_8CeCu_4$ and $Al_4Ce$ eutectic phase (Table 1) and aluminum solid solution. The alloys are selected in hypoeutectic concentrations regions for receiving various volume fraction of the different particles from $f_{min}$ to $f_{max}$. (Table 1).

Table 1 Characteristics of the investigated alloys

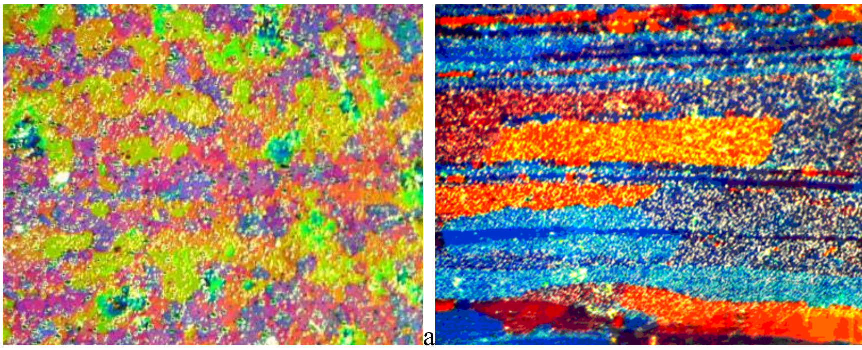
| | Melting point*, °C | Eutectic concentration (maximum for use) | Eutectic phase | $f_{min}$, vol. % | $f_{max}$, vol. % | $\overline{d}_{min}$, μm | $\overline{d}_{max}$, μm |
|---|---|---|---|---|---|---|---|
| Al-Ni [12] | 645 | 6 wt.% Ni | $Al_3Ni$ | 3 | 10 | 0.30±0.03 | 2.2±0.1 |
| Al-Mg-Si (quasi-binary section) [12] | 585 | 8.2 wt.% Mg and 4.75 wt.% Si | $Mg_2Si$ | 8 | 18 | 0.70±0.05 | 4.0±0.2 |
| Al-Cu-Ce (quasi-binary section) [13] | 610 | 14 wt.% Cu and 7 wt.% Ce | $Al_8CeCu_4$ | 6 | 21 | 0.7±0.1 | 2.1±0.1 |
| Al-Ni-Ce | 627 | | $Al_3Ni$ and $Al_4Ce$ | 3 | 27 | 0.5±0.1 | 2.0±0.2 |
| Al-Zn-Mg-Cu-Ni | 545 | 4 wt.% Ni | $Al_3Ni$ | f ~ 9 | | $\overline{d}$ = 1.8±0.2 | |

* - determined by thermal analysis differential scanning calorimeter Seteram Labsys DSC-1600 on laboratory patterns

These particles have different properties and may have different effects on the recrystallization. For example, the large particles $Al_3Ni$ of greatest hardness would be intense generate local lattice rotations. Moreover, the alloys of Al – Ni and Al – Ni – Ce has a low solute aluminum matrix, and other Al – Mg – Si, Al – Cu – Ce has more high solute aluminum matrix.

Alloys were cast in water-cooled copper moulds. ingots had a size of 100 × 40 × 20 mm. The cooling rate during casting was about 15 $K \times s^{-1}$. Casting temperatures were in the range of 750–800 C. All ingots were exposed homogenization at 450 - 620 °C. After this heat treatment particles have different size (from $\overline{d}$ min up to $\overline{d}$ max) (Table 1). Subsequent rolling was carried out at temperature 450 °C with a reduction of 83%. Then cold rolling with a reduction of 67% followed. The final sheet thickness was 1 mm.

Microstructural characterization was carried out using optical (OM) (Neophot-30) and scanning electron (SEM) microscopes (JSM35-CF). Specimens for optical and scanning electron metallography were mounted in polystyrene and mechanically polished using the Struers Labopol polishing system. Samples etched in Keller's reagent for research the particle characterization or subjected to oxidation for the analysis of grain structure. The average grain size $\boldsymbol{D}$, particle size $\boldsymbol{d}$, volume fraction of particles $\boldsymbol{f}$ and interparticle space were determined by the linear intercept method. Interparticle space was counted using Eq. 1 [14] and volume fraction of particles was calculated by TermoCalc software (version TALL5). The calculated and measured value both of volume fraction and interpaticle spacing are equal.



a b

Fig. 1 The structure after 20 min annealing at the $0.95T_{melt}$ of the alloy of system Al-Ni consisted $\boldsymbol{f}$ = 10% particles of phase $Al_3Ni$ (a) - the size of particle $\boldsymbol{d}$=1.2 μm, (b) - the size of particle $\boldsymbol{d}$=0.3 μm (OM, ×500)

$$\boldsymbol{l = 2/3\, d\, (1/f - 1),} \qquad (1)$$

Specimens for transmission electron microscopy (TEM) were sliced from parallel to the plane of rolling. Thin foils were jet polished to perforation with a Struers Tenupol polishing system using 20 per cent perchloric acid in methanol at the temperature 5 - 10 °C below zero.

The effect of volume fraction and particle size on the recrystallized grain size and tensile tests for superplastic testing have been studied at elevated temperature $0.95T_{melt}$ after 20 minutes. The samples with test portion gage by 14×6×1 were used for superplastic test. Elongation and flow stress were determined in tests with constant strain rate. Factor of strain rate sensitivity m was determined during tests with jump increasing strain rate.

## 3. Results and discussion

The structure of the alloys after cold rolling consists of the grains elongated along the deformation axis. The result of analyzes fine structure was reveal the some differences in alloy consist another size of particles. All the materials containing 12 - 24 vol. % of the particles size of 0.7 - 4 μm had a subgrains structure with low density of free dislocations. In alloys consisted 3 - 8 % of the particles formed cellular dislocation structure with a much larger number of dislocations.

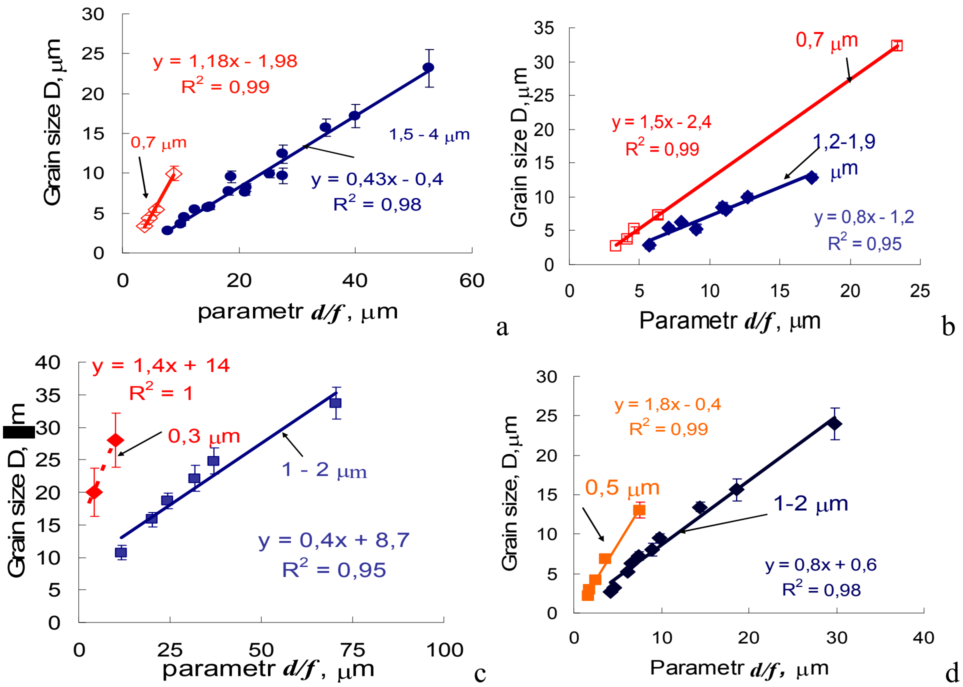



Fig. 2 The dependences of grain size on equitation $\boldsymbol{d/f}$ for alloys of Al-Mg-Si (a), Al-Cu-Ce (b), Al-Ni (c), Al-Ni-Ce (d) systems

After annealing during 20 minute at the temperature of 0.95 $T_{melt}$ the structure of almost alloys were clear recrystallized (Fig. 1 a). Except for the alloy with a particle size of 0.3 µm and the volume fraction of 10 % (Al - Ni) in the structure of which remain nonrecrystallized zone (Fig.1 b).

The dependences of the grain size on the interparticle spacing were linear. The Eq.2 are described these relationships with the magnitude squared $R^2$ = 0.95-0.99.

$$D = a \cdot l + c \tag{2}$$

The dependences of grain size on the ratio of ***d/f*** were linear too (Fig.2) ($R^2$ = 0.95-0.99) (Eq. 3)

$$D = k \cdot \left(\frac{d}{f}\right) + b \tag{3}$$

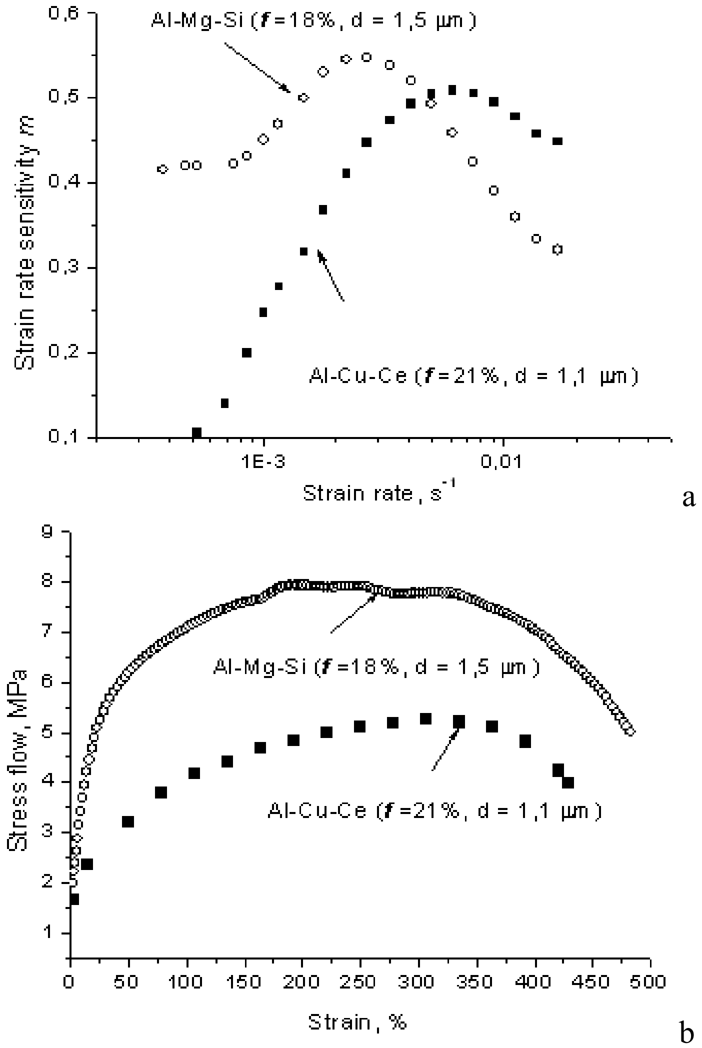



Fig.3 Superplastic indicators for Al-Mg-Si alloy (f = 18 vol. %, d = 1.5 µm) and Al-Cu-Ce alloy (f = 21 vol. %, d = 1.1 µm). (a) - The dependence of strain rate sensitivity from strain rate during superplastic deformation, (b) - the dependence of stress flow from elongation during superplastic deformation alloys with the constant rate of $8\times10^{-3}$ $s^{-1}$

Experimentally obtained different values of the coefficients *k* in Eq. 3 depended on the size of the particles. We have been received one relation of the grain size on ***d/f*** if the size of particles ***d*** less than 1 µm and if the size of particles more than 1 µm another relation from their angle coefficient. If the particle size in the range of 0.3 – 0.7 µm values the coefficients *k* in Eq. 3 more than one (k = 1.2 – 1.8). And with particle sizes in the range of 1.1 - 4 µm *k* = 0.4-0.8 less than one. As a result, at a fixed value of the parameter ***d/f***, the grain size in the case of particles of d = 1 - 4 µm is less than in the case of smaller particles with d = 0.3 - 0.7 µm. According to the Zener model then smaller of particle size we have, that the more they inhibit movement of borders, and the smaller the grain size should be able. This is the result that large particle size of 1 μm and more not only retard the growth of grains, but are centers of nucleation (PSN). The small particle size of d = 0.3 - 0.7 μm only inhibit recrystallization or retarded of growth of recrystallized grains. Eq. 3 was corresponds to the known theoretically obtained of Zener and Smith equation (Eq. 4) [15].

$$D = k \cdot \left( \frac{d}{f} \right) \quad (4)$$

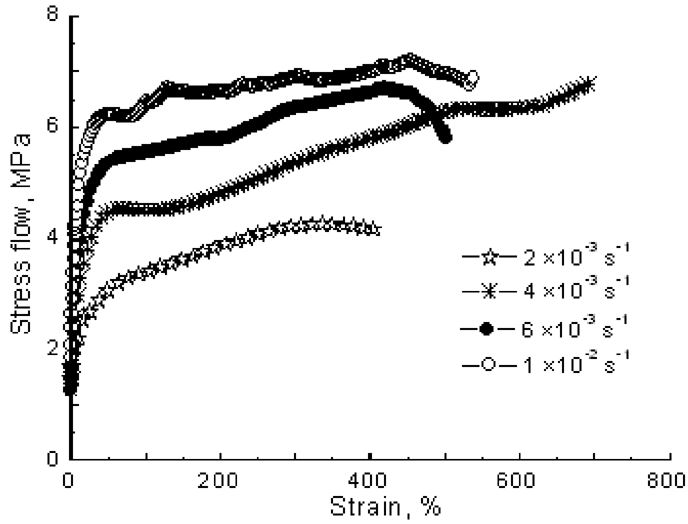



Fig. 4 The dependence of flow stress from elongation during superplastic deformation the alloy of Al-Zn-Mg-Cu-Ni system

However, the coefficient of slope k in Eq. 4 can vary according to calculated data from 0.1 to 1. In this work we obtained experimentally dependence of the coefficient on the ability of particles to the PSN. The described patterns are observed in the alloys of all of the systems, regardless of the hardness of the particles or the composition of the solid solution.

The number of particles per one grain at 0.95 $T_{melt}$ (in spherical approximation form of grains and particles) for the same of their volume fraction in the case of large particle size from 1 μm to 4 μm order of magnitude smaller than in the case of small particle size 0.3 - 0.7 μm. This fact also show of the predominant formation of recrystallization nuclei on the large particles.

The most promising in terms of superplasticity have the alloys with small recrystallized grains at 0.95 $T_{melt}$. The grain size about 3 μm, obtained in the systems Al-Mg-Si and Al-Cu-Ce and Al-Ni-Ce in the amount of eutectic particles 1-2 microns and the maximum volume fraction of particles. Superplasticity indicators of these alloys at 0.95 $T_{melt}$ were at high level. The strain rate sensitivity index m = 0.5 – 0.6 and elongation over 400 % were obtained at optimum constant strain rate $5 \times 10^{-3} s^{-1}$ (Fig.2). Stress flow in these did not exceed 8 MPa at the constant strain rate of $8 \times 10^{-3}$ $s^{-1}$.

The possibility of using eutectic particles for superplasticity in aluminium alloys was shown and the example of another model alloy system Al-Zn-Mg-Cu-Ni contains the large (2 μm) spherical particles phase $Al_3Ni$. The alloy shows high levels of SPD - 500-700% elongation at speeds up to 1 $10^{-2}$ $s^{-1}$ (Fig. 3). Whereas in the absence of large particles $Al_3Ni$ phase in this alloy was observed only 200 – 250 % elongation at the same strain rates.

## 4. Conclusions

The influence of formation of grain structure for the superplastic deformation in the alloys of systems Al - Ni, Al - Mg - Si, Al - Cu - Ce and Al - Ni – Ce, contained the particles of eutectic phases with sizes from 0.3 μm to 4 μm and the volume fraction of particles 3% up to 27% was studied.

It is founded that recrystallized grain size is depend on the ratio of particle size to volume fraction - *d*/*f* describes by a linear equation, like Zener – Smith low. It is shown that in this equation, if the angle factor k<1 the large particle size from 1 to 4 μm are stimulate the nucleation and inhibit the grain growth, and if the particle size from 0.3 to 0.7 μm k> 1, only retard of grain grown.

In the alloys of Al-Mg-Si and Al-Cu-Ce systems large particles of the eutectic phase the size of 1 - 2 μm when the volume fraction of 15-21% ensures the formation of small recrystallized grain size of about 3 μm, so these alloys have shown good performance during superplastic deformation at 0.95 $T_{melt}$: elongation about 400% at the optimal constant strain rate $5 \times 10^{-3}$ $s^{-1}$.

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