ECAP PROCESSED SILUMIN AK9 MODIFIED BY LIGATURE Al ALLOY:
MICROSTRUCTURE AND MECHANICAL PROPERTIES

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ABSTRACT

The studies of metallic materials of an ultrafine-grained or nanosized structure formed by intensive plastic deformation have occupied in recent decades a leading position in the ratings of the citations of scientific papers in the field of materials science. Irrespective of their large number there are still many questions associated with obtaining optimum mechanical properties of materials after equal-channel angular pressing (ECAP). The latter effect on the microstructure and the mechanical properties of modified by ligature silumin AK9 containing nanostructured carbon in the form of fullerene black and glassy carbon particles formed during the ligature preparation is investigated. It is shown that ECAP plastic deformation proceeds at 500°C within 3 passes despite the fact that silumins are non-deformable casting alloys. An essential structure refinement occurs relative to the initial silumin AK9 modified by ligature. It is shown that ECAP application provides crushing of the silicon inclusions, which have technological heredity and do not undergo plastic deformation in the course of conventional metal forming and heat treatment.

Keywords: microstructure, ECA-pressing, silumin AK9, microhardness.

INTRODUCTION

It has been recently recognized that the technologies based on intensive plastic deformation methods are perspective for production of bulk nanostructured construction materials, in particular alloys based on Al [1 - 2], Ni [3], Ti [4 - 5], Cu [6 - 8], as well as low carbonaceous [9] and stainless [10] steels. These technologies provide obtaining workpieces, sheets, rods, and wires of record high values of structural strength, yield strength, electrical conductivity, corrosion resistance and other attractive properties, while maintaining their technological plasticity. The methods of intensive plastic deformation (IPD), consisting in crimping with large degrees of deformation at relatively low temperatures and high pressures, result in obtaining bulk non-porous nanocrystalline metals and alloys, which cannot be achieved by compacting nanopowders [11 - 14]. When evaluating the important advantages of intensive plastic deformation methods, it should be noted that metals and alloys initial chemical composition does not change in the process of formation of their ultrafine-grained structure. The methods of intensive plastic deformation, in fact being a new application of metal forming methods, can be integrated into the existing workflows of the stages of metallurgical “ingot - blank” or “blank - finished product” conversion [15, 16].

A regulated structure and enhanced properties can be obtained by alloying with small additions of rare earth and/or transition elements. This method is used for different alloys production. The addition of small amounts of modifiers in the form of nanopowders or ligatures containing nanostructured areas provides the stimulation of non-dendritic crystallization and formation of nanostructures [17]. The necessity to create new composite aluminium materials and technologies for their production is dictated by the need of competitive products, as well as by the gradual depletion of the natural elite raw materials and their increased cost. The cheapest and most reliable are the materials based
on aluminium alloys subjected to modification and reinforcement by refractory dispersed particles. The formation of submicrorcrystalline and nanostructures in such materials would lead to significant improvement of their properties. IPD is a simple and effective way of refining the structure. It is the most widely used ECAP technology among the existing methods, providing three-dimensional nanostructuring, i.e. obtaining workpieces of a specified structure in the entire bulk [18 - 20].

The aim of this research is to increase the durability and strength of products from silumins by refining the material structure through modification and to stabilize them in the course of subsequent plastic deformation and heat treatment.

EXPERIMENTAL

A casting and deformation technology (in-suit technology) was used. It included mixing of the powder components of the charge, mechanical activation of the resulting mixture, charge extrusion to obtain a ligature and casting. Industrial hypoeutectic silumin AK9 (GOST 1583-93) was used. The choice of this material was dictated by its availability, the presence of highly developed industrial capacities and their use in our country facilitating the implementation. Furthermore, the large volume of the waste (swarf) generated during the products processing stimulated the application of powder technologies. The chemical composition of the silumin used in the work is shown in Table 1.

The modification and the simultaneous hardening of AK9 alloy was carried out using nanostructured carbon. Fullerene soot, fullerene black, as well as fullerene C60 and microcrystalline graphite were used as modifiers. The samples were prepared from aluminium powders having a particle size of the main fraction of 5 μm - 100 μm or AK9 alloy shredded shavings and some nanocarbon materials of a content of up to 10 mass % in the initial mixture. AK9 alloy was used as a base to obtain the developed composite material. The melt was produced in a VIS 0.004 induction furnace.

The ligatures containing 10 mass % of carbon were introduced to AK9 alloy at temperatures of 750°C - 780°C. The melting time was 3 min - 5 min. The amount of the injected ligature was calculated on the ground of the condition referring to 1 mass % carbon in the composite. The ligature production included mechanical activation of the initial materials in a planetary mill, compaction in rigid molds and hot extrusion. The mechanically activated powders were compacted into tablets at P = 450 MPa. They were then extruded at a temperature of 450°C - 500°C at a stretching ratio ≥ 10. The ligature was obtained in the form of rods.

The mechanical activation treatment was carried out for 30 min - 40 min at a rotation frequency of the central shaft of 400 min⁻¹ - 600 min⁻¹. The mass ratio of the grinding bodies to the weight of the loaded components was equal to 20:1.

The extrusion was carried out at the speed of 0.005 m/s. The pressing force required was increased by temperature increase above 500°C. The melting of the structural components took place in an aluminium matrix. The material was transferred into a borderline state.

The main structural components of AK9+C alloy cast state were dendrites of a solid solution of aluminium (α-phase) and aluminium-silicon eutectic. The workpieces obtained were subjected to annealing aiming stabilization of the structure. It was decided to investigate the microstructure of the alloy after quenching recognizing that it was recommended as the alloy initial thermal treatment prior to deformation. Thus, two basic thermal operations, annealing and quenching, were applied to AK9+C alloy. The corresponding temperature values are given in Table 2.

Following the preliminary thermal treatment, the samples of a section of 15mm x15mm x70mm were subjected to ECAP in an equal channel step die with a junction

<table>
<thead>
<tr>
<th>Alloy</th>
<th>Si</th>
<th>Mg</th>
<th>Cu</th>
<th>Mn</th>
<th>Zn</th>
<th>Ni</th>
<th>Fe</th>
<th>Al</th>
</tr>
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<tbody>
<tr>
<td>AK9</td>
<td>10.0</td>
<td>0.4</td>
<td>1.0</td>
<td>0.4</td>
<td>0.5</td>
<td>0.3</td>
<td>1.2</td>
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angle of the channels of 125° [21]. Be route with canting of the workpiece at 90° around the longitudinal axis [22] was used. The experiment was conducted on a hot forging single column crank press model ПБ 6330-02 with the force of 1000 kN at temperatures of 25°C and 500°C.

All the deformed workpieces were labelled. Then their transverse and longitudinal sections were subjected to metallographic analysis by using light and scanning microscopy. The aluminium mechanical properties were evaluated in the course of microhardness and tensile tests. All the samples were investigated in their middle plane to avoid the influence of the peripheral areas. The preparation of the microsections for the metallographic analysis was performed by standard methods. The observations were carried out on Leica light microscope equipped with a micro durometer providing the measurement of the Vickers microhardness of aluminium.

JEOL JSM 5910 scanning electron microscope was used to estimate the smaller structural elements.

The chemical composition of the aluminium alloy was determined on a portable X-ray fluorescence analyser, Olympus Delta XRF Innov-X Systems.

The tensile machine MI40KU was used to evaluate torsionally the mechanical properties of the aluminium alloys. 32 standard samples of a cylindrical shape were tested (diameter of the working part of 3 mm, length of 15 mm). The tensile speed of the samples was 0.5 mm/min corresponding to a strain rate of 0.56·10⁻³ s⁻¹. The workpieces of significant cracks obtained in the course of deformation and not healed during the pressing process were not used for further deformation cycles.

**RESULTS AND DISCUSSION**

Research results referring to the initial components of the charge

*Aluminium powder.* Samples were prepared from aluminium powders having a particle size of the main fraction of 5 μm - 100 μm. The topograms of the initial aluminium powder are shown in Fig. 1a.
 Fullerene soot was obtained by arc evaporation of graphite. This was an amorphous carbon with a bulk density of 250 kg/m³ and fullerene content of 11%. It was a product of Ioffe Institute. The results obtained by scanning electron microscopy are shown in Fig. 1(b). The powder consisted of dispersed soot particles and large fullerene particles [23].

 Fullerene black (FB). It was a product of the Ioffe Institute. It was a black powder with a particle size of 40 nm - 50 nm [24], a bulk density of 500 kg/m³ and fullerenes content not greater than 0.1%. It was in fact extracted fullerene soot after removal of non-polar organic solvents and subsequent treatment with steam to remove the organic solvent. The fullerene black consisted of 100% black carbon with no impurities. The scanning electron microscopy results are shown in Fig. 1©.

 Microcrystalline graphite. It was a product of Asbury Graphite Mills, Inc. United States [25]. The results of the observations of microcrystalline carbon powders of different dispersion revealed that the carbon particles had the form of plates and flakes, typical for the hexagonal crystalline structure. Fig. 1(d) shows a topogram of microcrystalline carbon.

 Research results of the charge after its mechanical activation

 The study shows that plastic deformation processes of the initial powder components occur in absence of any changes of the elemental and phase compositions in course of the mechanical activation of Al-C system charge. Thus, the obtained topograms of the charge powders containing various carbon additives are similar. Fig. 2a presents as an example the topogram of the charge powder Al + 10% fullerene soot.

 Research results of Al-C compositions after charge extrusion

 The structural state obtained by ligatures extrusion is studied. In this case, superhard grey-coloured particles of various modifications are revealed in the samples. They are unusual for Al-C alloy. An indentation recovery effect indicating very high elastic properties is detected in the course of this phase microhardness measurement. The superhard phase determined by micro X-ray spectral analysis EDX is identified as carbon (Fig. 3). Grey phase particles of a wavy surface (a globular relief) of no traces of grinding or polishing (Fig. 3(a)) are observed in the microstructure of a number of samples (particularly those of the series with fullerene black). They have a very high microhardness: the indenter imprints in the image are hardly visible, while the imprints slip off the particles leaving crosses with cleavage. The behaviour of this phase indicates that its hardness is close to that of diamond.

 This phase is observed in all samples containing nanocarbon additives, i.e. those with fullerene soot, fullerene black, and C_{60} fullerenes (Figs. 3(a-d)). The analysis shows that the dimensions, the shape and the number of the particularly solid pure carbon phases of high elasticity differ in ligatures compositions.

 The analysis of the structural state of he Al-microcrystalline carbon samples after charge extrusion shows uniform distribution of the carbon component (black and grey inclusions) in the aluminium matrix (Fig. 3(d)). The small size of the carbon inclusions does not provide microhardness measurements, which in turn makes it impossible to identify them as superhard carbon phases as those obtained in presence of nanocarbon additives.

 All composites of Al-C system have a modified structure of the metallic base with a dispersed distribution of inclusions of intermetallic compounds. As already mentioned above, the intermetallic compounds considered are characterized by a significantly higher microhardness in comparison with that of the base. Thus, on the basis of the foregoing, it can be concluded that the dispersed distribution of the aforementioned intermetallic compounds has a strengthening effect on...
Al-C composites structure.

The Raman spectroscopy study shows that these phases are amorphous, similar to vitreous carbon. Aluminium (Al$_4$C$_3$) and/or silicon (SiC) carbides are also identified in the ligatures. The results obtained do not reveal fundamental differences in the structure of the aluminium composites containing costly fullerenes in comparison with those obtained with cheap nanocarbon materials (fullerene soot, fullerene black). Furthermore, the difference observed in the mechanical properties is insignificant. The advantage of the hypoeutectic silumin AK9 modified with ligature refers to the content of nanostructured carbon in the form of fullerene black and synthesized particles of glassy carbon formed during the production of the ligature. Therefore, the effect of the pressing process in the equal channel step die on the mechanical properties and structure will be followed only in case of this composite.

**Results referring to the alloy obtained**

The resulting alloy is investigated by JEOL JSM - 5400 scanning electron microscope with EDS X-ray microanalyzer. The results obtained are presented in Fig. 4.

**Results referring to AK9+C alloy after ECAP**

AK9 silumin modified by ligature is subjected to pressing at a room temperature which results in its destruction. An experiment aiming at deformation by compression with automatic control of the temperature and the degree of deformation as well as heating and cooling rates recording is carried out with AK9+C composite alloy. Baehr Thermoanalyse (Germany) DIL805A/D dilatometer is used. The results obtained suggest carrying out the ECAP process at the temperature of 500°C and a deformation rate of 1 mm/s with heating of the workpieces to 500°C after each deformation cycle. A possibility of the sample plastic deformation up to 3 passages followed by its destruction is shown. No visible damage is obtained in the course of the three ECAP cycles. The workpieces of significant cracks appearing during the deformation process and not sealed in the extrusion process are excluded from the further deformation cycles.

The macroscopic observation shows that the number of the siliceous inclusions decreases, while the structure becomes more thorough and evenly distributed after the
first pass of ECAP. The microstructures of the samples obtained after the first pass and after three cycles of ECAP are shown in Fig. 5.

Significant refinement of the aluminium structure occurs following the three deformation passes after ECAP. It refers not only to the surface but also to the centre of the workpiece. Hence, the initial grain size of 22 μm is decreased to 3 μm after the deformation brought about by the technology used.

The scanning electron microscopy (SEM) study of the aluminium samples shows that the grains obtained are equiaxed with a size is in the range from 2 μm to 4 μm (Fig. 5(b)). It is evident from the picture that fragmentation of the inclusions due to their crushing takes place after the cycles of deformation applied. At the same time, significant crushing of the large lamellar particles occurs while that of the needle-shaped one is not as intense. The fine particles initially oriented along the grain boundaries distribute chaotically over the cross section of the sample with cycles number increase.

A quite subtly differentiated eutectic is registered in the samples after the first ECAP pass. Besides, the individual needle-like crystals of silicon are fragmented, while the boundaries between them become blurred and
hard to discern. Their colonial structure becomes visible as well. In general, the microstructure is not uniform and a coarser «needle» silicon phase forms alongside the dispersed areas. The proportion of the silicon phase in the general structure of the alloy decreases, while the aluminium phase it increases correspondingly with increase of the number of cycles. The subtly differentiated component of the eutectic is completely absent. There is displacement of α- solid solution of the silicon phase so that grain boundaries begin to form in the aluminium phase and grains start to identify. After three passes, the classical acicular silicon crystals are practically not observed. They are highly fragmented and spheroidized, their share in the overall structure of the alloy is markedly decreased, while the aluminium phase starts to dominate. The change of AK9+C alloy microhardness with increase of the degree of deformation during ECAP correlates with the structure evolution. After ECAP the microhardness increases almost two-fold compared with that of the initial state, i.e. from 451 MPa in the initial state to 882 MPa after the third pass. This can be explained with the higher degree of deformation. The microhardness at the head part of the samples is somewhat higher (up to 10 %) when compared with the average value. The deviation of the microhardness from the mean value in the central part of the sample along the axis and in the transverse direction does not exceed 10 %. These data confirms that ECAP provides an increase of AK9+C alloy strength characteristics. The analysis of the alloy hardening is conducted in the course of a tensile testing at a room temperature. The material used is an annealed condition.

The stress-strain graphs register surging changes of the stress during the process of stretching. They are caused by cyclic hardening-softening under the effect of movement and attachment dislocations on the barriers. The tensile tests of the samples show that the hardness of the cast composite material obtained with nanostructured carbon and glassy carbon particles, increases by 20 - 25 % after ECAP and reaches a maximum during the third passage. The increase of the number of passes results in hardness decrease due to the crushing of acicular silicon crystals. The temporary fracture resistance ($\sigma_{B}$) is increased from 299 MPa in the initial state to 400 MPa after the first pass and to 596 MPa after the third pass. This is in fact almost two-fold increase. The yield strength ($\sigma_{0.2}$) increases from 158 MPa in the initial state up to 220 MPa after the first pass, and up to 525 MPa after the third pass. Almost 2.5 times increase is obtained.

The experimental study of the plastic characteristics change (the relative elongation observed in the tensile test) shows that it is insignificant.

Thus, it is verified once again that the increased strength characteristics of the cast composites allow their use for the production of important parts in the field of mechanical engineering, while the achieved level of plasticity makes the composites suitable for subsequent plastic deformation, and hence for production of workpieces of a textured structure. The obtained properties are attained by dispersing all structural components and uniform distributing of the hardener in the form of nanostructured carbon.

**CONCLUSIONS**

The studies carried out show that a substantial structure refinement occurs referring not only to the surface but also the bulk of the workpiece after three deformation passes of the modified by ligature AK9 silumin containing nanostructured carbon in the form of fullerene black and glassy carbon particles formed during the production of the ligature. The initial grain size of 22 μm is decreased to 3 μm. Even the macroscopic observation shows that the number of siliceous inclusions decreases and the structure becomes more thorough and evenly distributed after the first ECAP pass. The classical acicular silicon crystals are practically not observed after the third pass. They are highly fragmented and spheroidized, their share in the alloy overall structure decreases significantly, while the aluminium phase starts to predominate.

The results of the mechanical testing of the samples deformed by the proposed method show that the strength of the cast composite materials obtained with the addition of nanostructured carbon and glassy carbon particles, increases by 20 % - 25 % after ECAP and reaches a maximum during the second pass. A further increase of the number of passes results in strength decrease due to the crushing of the acicular silicon crystals. The tensile test carried out shows that the plastic properties of the aluminium composites increase with about 6% after the first ECAP pass. A further increase of the passes number decreases the alloy ductility.

Thus, the cast composites increased strength char-
characteristics and level of plasticity resulting from their modification with nanostructured carbon and glass-like carbon particles, as well as by intensive plastic deformation provide obtaining workpieces of a textured structure of importance for the field of mechanical engineering. The obtained properties result to a great extent to the dispersion of the structural components and the even distribution of the hardener in the form of nanostructured carbon.

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