Experiment on Injection of SIC and BN Nanoparticles into Liquid Aluminum Using MHD Stirring with Subsequent Crystallization of the Melt

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Abstract. The paper describes the technique and results of experiments on the introduction of SIC and BN nanoparticles into an aluminum melt contained in a cylindrical crucible with a water-cooled bottom and heated walls. The crucible was placed in an MHD stirrer, which generated a travelling and rotating magnetic fields ensuring in such a way bidirectional stirring of the liquid metal in the crucible. After introducing the reinforcing particles, aluminum was stirred and directionally crystallized. Reinforcing particles were introduced into molten aluminum as a component of pressed pellets. Pellets with diameter of 20 mm and thickness of 15 mm were made from a mixture of aluminum powder (100-156 µm) and reinforcing nanoparticles (100-200 nm) by pressing in a special pellet die. The concentration of reinforcing particles in the pellets was selected using preliminary experiments and was 5% of their total weight. In some experiments, the aluminum was reinforced with SIC nanoparticles introduced into the melt in the amount of 0.59%, 1.04%, 1.77%, while in other experiments the concentration of hexagonal BN nanoparticles introduced into aluminum was 7%, 1%, 1.8% of the ingot weight. As the experiment showed, the reinforcing particles formulated into tablets can be introduced into aluminum by means of MHD stirring. The dependence of the ultimate strength and electrical resistance of aluminum on the concentration of the reinforcing nanoparticles introduced into it has been obtained experimentally. It has been found that with an increase in the concentration of nanoparticles in aluminum, its ultimate strength and specific electrical resistance increase. The dependences of the ultimate strength and electrical resistance of aluminum on the concentration of SIC and BN nanoparticles are very similar and close to linear (in the investigated range of concentrations of introduced particles. With an increase in the concentration of introduced nanoparticles the mechanical strength of the resulting material grows faster than the electrical resistivity. In the experiments, the ultimate strength of the samples increased by 22%, while the electrical resistance increased by 5-6%. This opens up new possibilities for industrial applications of such materials, for example, in manufacture of self-supporting wires in power lines.

1. Introduction

One of the trends in the development of modern technologies in metallurgy is designing new structural materials, which could meet the increased requirements for strength, reduction of weight and metal content, high endurance and service life under extreme conditions of temperature and force impact. These properties are inherent in metal composites obtained by introducing the reinforcing micro or nanoparticles in the matrix metal, such as aluminum. In this case, the combination of the source elements gives the effect of enhancing the strength properties of the obtained metal composite in comparison with the properties of the elements comprising it. This is the reason why nowadays all leading industrialized countries show the growing interest in composite materials, in particular, in cast aluminum matrix composite materials filled with dispersed powder particles. These composites are heterogeneous systems consisting of two or more phases. One of the components, showing continuity throughout the
Composite materials, which consist of an aluminum matrix reinforced with finely dispersed particles non-soluble in the matrix metal (dispersion reinforced materials) increase the rigidity of the structure while reducing its metal content [1-4]. Today in fabrication of aluminum matrix composite materials different methods are used to introduce reinforcing particles into the melt:

- Mechanical stirring of particles into the melt, including the injection of powder through an electromagnetic funnel [5].
- Stirring of powders followed by pressure sintering.
- Impregnation of porous frames made of reinforcing powders with aluminum.
- Plasma spray deposition of metal and ceramic particles.
- Synthesis of endogenous reinforcing materials in a liquid matrix. (Such technology provides continuous and tight contact of phases and sufficient strength of bonds between them in the cast composite material). The idea behind this technique is that disperse particles of reinforcing phase are not introduced into the melt from some external source, but are synthesized in it due to controlled chemical reactions between the introduced components.
- Injection of particles by means of a special gas into liquid aluminum. [6,7].
- Plasma-jet injection of reinforcing particles into the melt through a plasmatron [8] and subsequent MHD stirring of the melt.

In recent years, engineering investigations have focused on the development of new techniques for introducing reinforcing particles into aluminum melt using for this purpose only MHD action on liquid metal [9,10]. However, all these methods are either inefficient, producing a large amount of waste, or require expensive equipment.

The cheapest and simplest (for fabricating alumina matrix composites) technologies are based on the methods of mechanical stirring of fillers into the matrix melt. A distribution of reinforcing filler in this case depends on wetting of the filler by the matrix melt, stirring conditions and subsequent processing. Wetting ensures a continuous physical contact between the phases, which is necessary to achieve strong adhesive bonds. During the stirring process, most of the injected powder is not absorbed and is rejected, which greatly reduces the efficiency of the methods. In addition, there is the problem of making an impeller, which will be resistant to liquid aluminum and prevent increased formation of oxide. As noted above, the main difficulty in creating an alumina matrix composite by the simplest methods of mechanical stirring of reinforcing particles into liquid metal is the problem of wetting the particles with liquid aluminum. Liquid aluminum, due to its high surface tension, is unable to provide the required wetting of most of the materials intended for injection as reinforcing elements. However, such particles as SiC, Al₂O₃ and BN particles are wettable, but only at high temperatures. For example, wetting of SiC and Al₂O₃ occurs at temperatures above 900°C. For this reason the main difficulty with introducing particles into liquid aluminum is the problem of passing through the air - liquid metal interface. Therefore, it is necessary to introduce the particles right under the surface of liquid metal followed by intense stirring of the latter to ensure a uniform distribution of particles throughout the entire melt. Note that in this case the simplest way of introducing particles by placing them in bags of aluminum foil does not lead to a positive result. When the aluminum foil melts, almost all of the powder is ejected to the surface.

A search for simple ways of introducing the reinforcing particles into aluminum is an urgent problem. We already tested a method of introducing boron nitride microparticles in fusing aluminum tubes followed by intense MHD stirring of the melt [11]. In this method, an aluminum tube filled with a powder of reinforcing microparticles was gradually immersed into liquid aluminum melting in the crucible. The end of the tube melted, and the particles immediately got into the stirred liquid metal bypassing its surface. In such a manner, the problem of particle penetration through the liquid metal - air interface was eliminated. Nevertheless, a part of the injected powder was carried to the surface and did not penetrate into the stirred metal. During these experiments it was found that if reinforcing particles were mixed with aluminum powder before filling the tubes, the mass of rejected reinforcing particles decreased. In addition, there are also difficulties with introducing reinforcing particles when they are reduced to nanosize. Their injection into the stirred aluminum leads to undesirable effect (the mass of rejected particles increases, and in the liquid metal the particles are collected into clusters). Our
experiments showed that the introduction of nanoparticles into liquid aluminum by means of melting aluminum tubes leads to significant ejection of the introduced powder on the melt surface. One of the ways to solve this problem is to attempt preliminary mechanical briquetting of injected particles and aluminum particles [12], as well to expose the metal containing the injected particles to ultrasound. The aim of this work is to test a simple method of introducing alloying nanoparticles (Fig. 1) as a component of pressed pellets into liquid aluminum. The procedure is accompanied by MHD stirring of the melt leading to a directional crystallization of aluminum. We conducted a series of experiments, in which the alloying particles were introduced into liquid aluminum in the form of pellets prepared from the mixture of reinforcing powder of SiC and BN nanoparticles and aluminum micropowder. We supposed that during MHD stirring, the pellets would penetrate into the metal volume, and the reinforcing particles would be detached in small amounts from the pellets, gradually eroded by the metal and distributed throughout the melt.

![Fig. 1. Powder particles (hexagonal BN - a) have an oblong shape. Powder particles (SiC - b) are segments of nanofilaments](image)

2. The experimental setup
The setup consists of the following units:
1. resistance furnace, in which aluminum A0 was melted in a graphite crucible (furnace temperature is controlled by a platinum-platinum-rhodium thermocouple);
2. MHD-stirrer capable of generating separately controlled travelling (wave number 20.9 m⁻¹ frequency 50Hz) and rotating magnetic field (two-pole inductor 50Hz) Fig.2a [8,11] in the working volume of the crucible;
3. crucible with water-cooled bottom installed in the working volume of the stirrer (Fig.2a)
4. ring heater enclosing the crucible side walls (Fig.2a)
5. controlled power sources (for resistance furnace, MHD-stirring, and ring heater)
6. pressing machine generating force up to 200000N with a pellet die (for fabrication of pellets with diameter of 20mm and thickness of 10-15mm)

![Fig. 2. a - MHD-stirrer and crucible with water-cooled bottom. MHD-stirrer-1, stainless steel crucible-2, water-cooled bottom-3, ring heater with external thermal insulation-4, b - turbine wheel to determine rotational speed of liquid aluminum in the crucible, c - ingot cross-section showing the parts of the ingot used to cut out the slab - 1 (2 - the surface of the slab was polished and etched) and the samples for determining electrical resistance and testing mechanical characteristics – 3](image)
The crucible with a water-cooled bottom (Fig. 2a) was made of a sheet of stainless nonmagnetic steel 1 mm thick. The inner surface of the crucible was covered with a thin layer of special coating to protect the walls against liquid aluminum. The side walls of the crucible were heated with an embracing ring heater and thermally insulated by a 10 mm layer of mullite cardboard. The crucible bottom was cooled by flowing water with adjustable flow rate and inlet and outlet temperatures. The power supply source allowed the MHD stirrer to produce separately adjustable travelling and rotating magnetic fields. The ring heater could create heat flux of 1020–1330w.

3. Preparative works
Preparative works involved the fabrication of pellets, which contained reinforcing particles intended for injection into liquid aluminum. Reinforcing nanoparticles together with aluminum micro powder were placed in a container with steel balls, where they were mixed by shaking. Then the mixture was pressed into pellets with a diameter of 20 mm and height of 15 mm by applying the force of 50000N (Fig.3). Few mixtures of BN, SiC nanoparticles (100-200nm) with PA 4 aluminum powders (100-140μm) were prepared to determine the required quantitative composition of the pellets. The content of reinforcing powder in the mixtures was 0%, 5%, 10%, 15%, and 20%. The pellets were placed in a muffle furnace and heated to 800°C. Pellets containing 0 and 5% particles were easily smashed with a spatula, 10% pellets crumbled by applying some force and 15% pellets crumbled when pressed with a spatula. Pellets containing 20% of particles did not crumble and remained monolithic. Pellets with the same content of reinforcing powder were injected into liquid aluminum at 810°C during MHD stirring. Pellets with 0% and 5% concentration of particles dissolved, while pellets with 10% particle concentration dissolved partially. Pellets with 20% concentration of particles and higher did not dissolve even after 35 minutes of stirring and remained monolithic.

![Fig. 3. Distribution of nanoparticles BN-a and SiC-b (100-200nm) among microparticles (100-140μm) of aluminum in pellets prepared for injection into the melt](image)

4. Results of experiment
In the experiments, the pellets including SiC (100-200nm) and BN (100-200nm) nanopowders were introduced into Ak7 liquid aluminum alloy heated to a temperature of 810°C, and stirred by the downward travelling magnetic field (12.96mT) and rotating (8.08mT) magnetic fields (50Hz), which corresponded to electric currents of 12A and 8A in the inductors of travelling and rotating fields, respectively. A control experiment was carried out to determine the rotational speed of the liquid metal in the selected operating mode of the MHD stirrer. Measurements of speed were made using a special turbine (Fig.2.b). The speed was determined by the time it took for the turbine to make 40 revolutions. Speed of the rotating liquid metal for the selected stirring mode was in the range of 0.8 -1 m/s. Similar results were obtained in the numerical experiments in [13]. According to calculations, the estimated maximum velocity of the poloidal flow in our experiments was approximately 0.4 m/s.

Aluminum was melted in a crucible (Fig. 2a), the side walls of which were heated by a 1225W heater. The pellets were pressed from a mixture of reinforcing nanopowder (100-200nm) and aluminum micro powder with particle sizes up to 100-140μm. The content of reinforcing nanoparticles in the pellets was 5% of their weight. The pellets were introduced into the stirred metal for 2 minutes by sequential injections. After introducing the pellets, the crucible was closed with an insulating lid and then the metal was stirred for 45 minutes. At the end of this time the water cooling of the crucible bottom was started. Directional crystallization of the ingot occurred during MHD stirring of the melt. The ingot after completion of crystallization (15-20 minutes) and cooling was taken out of the crucible and cut into
pieces (Fig.2c). One of the parts of the ingot was used to make a thin slab, which was etched to display its structure.

Fig.4,5 shows the structure of the longitudinal section of the ingot with different contents of injected SiC nanoparticles and BN nanoparticles.

**Fig. 4.** Half of the longitudinal section of an aluminum ingot with injected SiC powder (its height is 140 mm, width in the middle part is 60 mm)
- a- amount of the added SiC nanopowder in aluminum is 0.59% of ingot weight;
- b- amount of the added SiC nanopowder in aluminum is 1.04% of ingot weight;
- c- amount of the added SiC nanopowder in aluminum is 1.77% of ingot weight.

As is evident from Figures 4, 5, the ingots have fine crystalline and essentially homogeneous structure; pellets in cases a and b dissolved well in the aluminum ingot, whereas in case c a small amount of the pellets dissolved incompletely. The incompletely dissolved pellets can be seen in the vicinity of the ingot surface in Fig. 4c, 5c.

**Fig. 5.** Half of the longitudinal section of an aluminum ingot with injected nanopowder BN. (its height is 140 mm, width in the middle part is 60 mm)
- a- amount of injected BN nanopowder in aluminum is 0.7% of ingot weight
- b- amount of injected BN nanopowder in aluminum is 1% of ingot weight
- c- amount of injected BN nanopowder in aluminum is 1.8% of ingot weight
Fig. 6. Photographic images of the ingot structure (in the middle part of the longitudinal section) with introduced SiC nanoparticles; a – 0.59%; b – 1.04%; c – 1.77% of the ingot weight. The photos (a,b,c) show a scale section of 1µm length.

Fig. 7. Photographic images of the ingot structure (in the middle part of the longitudinal section) with injected SiC nanoparticles: a - 0.7%; b - 1.0%; c - 1.8% of the ingot weight. In the photos (a, b, c) the scale section is 1µm long.

The magnified images of ingot cross-sections (Fig.6,7) were obtained using the FEI Quanta 650FEG scanning electron microscope fitted with EDAX Octane Elite X-ray microanalysis add-on device. In the obtained photographs one can see small 100-200 nm particles distributed over the ingot cross-section. The ingots with different concentrations of SiC and BN reinforcing particles were used to fabricate samples, which were subjected to tensile tests to determine the ultimate strength of the obtained materials and their specific electrical resistance (Fig.8). The specific electrical resistance was determined using a four-point scheme.

Fig. 8. Dependence of the ultimate strength of aluminum on the concentration of reinforcing nanoparticles a - SiC nanoparticles; b - BN nanoparticles (Each point is the test result of three samples)

It should be noted that the concentration of nanoparticles in aluminum was determined by the total mass of particles introduced into liquid aluminum as a component of the pellets, but as can be seen from the experimental results (Fig. 4c, 5c), at the highest concentration of introduced particles, the dissolution of a small portion of the introduced pellets was incomplete. Therefore, the real concentration of particles in aluminum at the maximum concentration achieved in our experiments was slightly lower than the calculated one. The presence of particles in the ingot can be judged by their effect on the physical properties of the metal in the ingot. The mechanical strength of the ingot material and the specific electrical resistance increased linearly with increasing amount of nanopowder introduced into the metal of the ingot (Fig. 8, 9). The degree of homogeneity of reinforcing particle distribution over the ingot
volume can be indirectly determined from the scatter of values of the ultimate strength and electrical resistance (Fig. 8, 9) of test specimens cut from three different places of the ingot cross section, Fig. 2c.

Fig. 9. Dependence of the specific electrical resistance of aluminum on the concentration of nanoparticles a - SiC nanoparticles and; b - BN nanoparticles (Each point on the graphs is the result of processing nine measurements taken at three samples cut from each ingot)

5. Conclusion
The experiment showed that the reinforcing particles can be introduced into liquid aluminum as components of the pellets containing aluminum powder and dispersed in the melt by subsequent MHD stirring of the melt. As the concentration of nanoparticles in aluminum increases, its ultimate strength and specific electrical resistance increase. The dependences of the ultimate strength and electrical resistance of aluminum on the concentration of SiC and BN nanoparticles are very similar and close to linear (in the investigated concentration range of the injected particles). It should be noted that the mechanical strength of the material with increasing concentration of the introduced nanoparticles grows faster than the increase in the resistance. Thus, the introduction of 1.77-1.8% SiC or BN nanoparticles increases the ultimate strength of the metal by about 22%, while their electrical resistance is increased only by 5 - 6%, which opens new ways of using such materials for industrial applications, for example, for the manufacture of self-supporting wires in power transmission lines.

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