The impact of high voltage electric discharge treatment in ethanol on the dispersity and phase composition of Cu – Al powder mixture

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Abstract *Studies of the impact of high voltage electric discharge (HVED) treatment on the dispersion and phase composition of 87,5 % Al + 12,5 % Cu powder system were performed. It was shown that HVED treatment in ethanol with specific treatment energy of 5 MJ/kg leads to the decrease of mean diameter of treated powder from 15 to 11 μm, and the increase of specific treatment energy to 20 MJ/kg leads to the decrease of mean diameter of treated powder from 15 to 6 μm. X-ray diffraction analysis shows that CuAl2 and Al4C3 are synthesized in all considered treatment regimes, and the quantity of these phases depend on the specific treatment energy.*
The use of "three point – plane" electrode system instead of "point – plane" during HVED treatment of 87,5 % Al + 12,5 % Cu powder system in ethanol leads to the increase of quantity of synthesized Al_4C_3 and $CuAl_2$ phases with the slight decrease in the dispersion efficiency. Up to 35% of particles in powder mixture, treated by HVED in ethanol with the use of "three point – plane" electrode system, have diameter close to the diameter of the initial powder mixture.
It is shown that the preparation of powders with an initial composition of 87.5% Al + 12.5% Cu using HVED treatment in kerosene or ethanol with subsequent consolidation by SPS method allows obtaining metal-matrix composites of the Al – Cu – C system with increased indicators of hardness, electrical conductivity and wear resistance.
Keywords*: HIGH VOLTAGE ELECTRIC DISCHARGE, POWDER, POWDER METALLURGY, SYNTHESIS, GRINDING, ALUMINUM, CUPRUM, SPARK PLASMA SINTERING*

1. Introduction

In Ukraine, there is an urgent need to replace outdated urban electric transport and ensure its reliability. Leading European manufacturers of electrical materials for this sector are Morgan Carbon, PanTrac, and Hoffman. Their electrical contact inserts predominantly use carbon and copper components [1]. These current collection elements, housed in aluminum frames, are secured with adhesives or bolted joints and often use copper clamps. Sliding electrical contacts are the most prevalent in urban electric transport. Technical analysis indicates that materials for these sliding elements must offer high electrical conductivity, arc resistance, hardness, strength, and wear resistance [1]. Additionally, the longevity of both the current collector and the contact wire, influenced by a low coefficient of friction, is crucial.

New materials based on carbon-containing powders clad with copper for manufacturing current collector inserts were developed in Ukraine. However, these materials exhibit high electric resistivity, low wear resistance, and unsatisfactory strength. Given these shortcomings, new materials with enhanced performance properties, particularly high wear resistance, are required for the manufacture of current collection elements. The existing materials fall short of meeting the technical and operational requirements, especially regarding system durability. They have specific resistances ranging from 3×10^{-5} Ω·m to 8×10^{-5} Ω·m, wear resistance characterized by a diameter loss of 0.1 to 0.4 mm over 20 hours, and durability ranging from 40 to 82 MPa. Consequently, developing materials that can withstand extreme operational conditions remains a pressing scientific challenge.

Materials consolidated through spark plasma sintering (SPS), self-propagating high-temperature synthesis (SHS), or selective laser sintering exhibit physical and mechanical properties that are 1.5–2 times higher than those produced by conventional metallurgical methods. This enhancement is due to reduced high-temperature exposure, which preserves the grain heterophase structure of the powder mixture within the consolidated material [2]. However, the properties of these materials depend not only on the consolidation method but also on the powder preparation technique. The high-voltage electric discharge (HVED) method for preparing initial powders for metal-matrix composite materials in a hydrocarbon liquid, developed at the IIPT of NAS of Ukraine, achieves significant particle dispersion and synthesizes dispersion-strengthening phases, such as carbides [2–9].

During HVED treatment, metal powder particles are subjected to mechanical (shock wave, hydro flows, cavitation) and thermoelectric (discharge channel plasma, microplasma formations between particles, discharge current) factors, resulting in their dispersion. The use of hydrocarbon liquid prevents oxidation of the metal particles. Additionally, as demonstrated in [3–9], HVED in a hydrocarbon liquid induces pyrolysis (breaking of chemical bond chains) due to the temperature of the plasma discharge channel and pulse pressure, forming active carbon nanoparticles up to C70 [2, 6]. This environment facilitates the synthesis of dispersion-strengthening compounds, such as carbides [3–9]. However, the application of HVED treatment in hydrocarbon liquid for preparing starting powders of metal-matrix composites in the Al–Cu system, specifically for manufacturing current collector inserts, remains insufficiently studied. There is a lack of information regarding the patterns of changes in the dispersion and phase composition of Al–Cu system powders under HVED treatment.

Therefore, the **goal of the work** is to investigate the influence of high-voltage discharge treatment in ethanol on the dispersion and phase composition of 87.5% Al + 12.5% Cu powder system, as well as to study the properties of composites of Al – Cu – C system, obtained by consolidation of treated powder mixtures.

2. Methodology

Elemental Al powders PA1 GOST 6058-73 (ПА1 ГОСТ 6058-73) with d_0 = 25 μm and Cu PMS-1 GOST 4960-75 (ПМС-1 ГОСТ 4960-75) with d_0 = 15 μm were selected as initial materials for the study, on the basis of which a mixture of 87.5 % Al + 12.5% Cu mass composition with an initial diameter of 22 μm was prepared. Ethanol (DSTU (ДСТУ) 4221:2003) was used as a working liquid. The ratio of solid to liquid phases in the studies was 1:15 (the weight of the processed powder was 100 g, the volume of the working liquid was 1.5 l).

The scheme and detailed description of the experimental stand for high-voltage electric discharge processing and, in particular, the working chamber, are given in [3–5]. Investigations of HVED in "liquid dielectric – metal powder" disperse system were performed at a voltage U = 50 kV, the inductance of the discharge circuit L from 0.5 to 1 μH (limit technological values) and the discharge interval l_p ~ 17 mm. In the process of research, the specific energy of processing was varied by changing the number of discharges n. The capacity C was 0.8 μF, so the value of the stored energy of a single discharge W_1 was 1 kJ.

Two structural types of electrode systems (ES) were used in the research, namely "point – plane" (P – P) and "multipoint anode – plane" (MP – P) with 3 tips. These ESs were used due to the fact that according to the results of [3–6, 8–10] the change of the electric field configuration due to the use of different types of ESs allows to create conditions for changing the distribution of plasma formations in the volume of processed powder.

In turn, if the plasma formations are distributed over the entire volume of the processed powder, it will intensify the processes of erosion and hydrodynamic dispersion and synthesis of carbide and intermetallic compounds during HVED treatment [3–6, 8, 9]. When using the point–plane system, HVED develops according to the classical mechanism of spark discharge (SD) in a dispersed system, while the use of "multipoint anode – plane" type ES allows the implementation of the mode of volume-distributed multi-spark discharge (VMD).

The use of different electrode systems allowed controlling the distribution of the intensity of the influence of the main factors of HVED impact [3–6, 8–10]. Thus, if in the case of the implementation of SD most of the accumulated energy was transformed into shock waves, the implementation of VMD using multi-point anodes allows to increase the intensity of thermal and current impact factors.

In order to evaluate the degree of influence of HVED in ethanol on the morphology and particle size of powders, the following hardware and software was used –BIOLAM-I (БИОЛАМ-I) optical microscope with a maximum magnification of × 1350, REMMA-102 (РЕММА-102) raster electron microscope with a magnification range from 10 to × 250000 (IPM NASU), Canon digital camera. Powder samples for optical microscopy were taken in accordance with GOST 23402-78 (ГОСТ 23402-78). After obtaining a sharp image, magnification was recorded and photographs were taken, which were processed for further analysis.

Since the average diameter of particles and their shape factor are the most informative in materials science in terms of describing the morphology of the powder, the diagnosis of morphological characteristics was performed before and after HVED treatment by average diameter (d_m) and shape factor (F) in accordance with GOST 25849-83 (ГОСТ 25849-83). The number of particles that came into the field of view of the microscope eyepiece was not less than 2000 pieces, and at least five samples were considered for each studied mode. This allowed to present data in the form of graphs and histograms with a confidence interval of 0.96.

The study of the phase composition of the powders was performed by X-ray phase analysis. The diffraction patterns were recorded using a DRON-4-07 (ДРОН-4-07) diffractometer under CuKα radiation. A Ni β-filter was used to reduce the background on the diffractograms as well as to reduce the probability of β-lines. Phase identification on diffraction patterns was performed using POW_COD databases [11]. A standard made of quartz was used to determine the instrumental function.

The consolidation of powder materials was performed using a spark plasma sintering (SPS) method in a vacuum on the universal experimental complex "GEFEST" [12], which is based on passing a superposition of direct and pulsating current with a frequency of 10 kHz through the powder at an operating voltage of U = 2 V. The consolidation temperature was selected according to [13] and was 500 - 600ºC, the holding time was 20 min at a pressure of 60 MPa. Vickers hardness, specific electric resistance and wear resistance were studied for consolidated specimens.

3. Results and discussion

The study of the effect of HVED treatment on the change in the properties of 87.5% Al + 12.5% Cu powder system was performed in the modes shown in table 1. The results of the high-voltage electric discharge treatment in the modes with different types of used ES and different values of specific treatment energy were compared. The initial mixture of 87.5% Al + 12.5% Cu powders had an average diameter of 22 μm and was characterized by a monomodal size distribution, the main peak of which was at the point of 20 μm (~ 55%) (see Fig. 1, curve 1).

Table 1: *Modes of HVED treatment of 87.5% Al + 12.5% Cu powder system in ethanol.*

No. of mode	Number of points of the electrode system	W_s, MJ/kg
1	1	20
2	1	10
3	1	5
4	3	20
5	3	10
6	3	5

HVED treatment of this powder mixture in ethanol with single-point ES (SD mode) at W_s = 5 MJ / kg leads to a change in dispersion (see Fig. 1, curve 2) – the particle size distribution retains a monomodal appearance and shifts to the area of smaller diameters, and the average diameter decreases to ~ 11 μm. Increasing the specific processing energy to 10 MJ / kg leads to an increase in the dispersion efficiency (see Fig. 1, curve 3) – the particle size distribution undergoes a significant shift to the area of smaller diameters, and the average particle diameter of the mixture decreases to 8 μm. Increasing specific processing energy to 20 MJ / kg leads to the further decrease of the average particle diameter of the mixture to 7 μm.

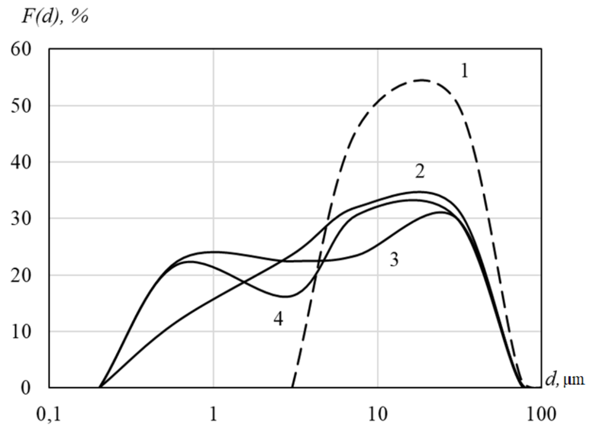


Fig. 1 *Particles size distribution of initial 87.5% Al + 12.5% Cu powder (1) and after HVED treatment in ethanol in SD mode with different values W_s = 5 MJ/kg (2), W_s = 10 MJ/kg (3) and W_s = 20 MJ/kg (4)*

Changing the type of used ES from one-point to three-point (and thus changing HVED mode from SD to VMD) insignificantly decreases the dispersion efficiency (see Fig. 2). Treatment with W_s = 5 MJ / kg allowed reducing the average particle diameter to 14 μm (see Fig. 3). Treatment with W_s = 10 MJ / kg reduced the average diameter of the powder mixture particles after treatment to ~ 10 μm, and with W_s = 20 MJ / kg to 8 μm.

Comparison of these results with the results of studies of the impact of high voltage electric discharge treatment in kerosene on the dispersity and phase composition of Cu – Al powder mixture, presented in [1], show, that HVED treatment of a powder mixture of 87.5% Al + 12.5% Cu in kerosene with W_s=5 MJ/kg leads to a decrease in the average diameter of the particles of the treated mixture from 15 μm to 13 μm, while treatment with a similar mode in ethanol leads to a decrease in the average diameter of the particles to 9 µm. The divergence between treatment results is even smaller when processing a mixture of powders 87.5% Al + 12.5% Cu with W_s = 20 MJ/kg – the average diameter of the particles after processing decreases to 6 µm when using kerosene and to 8 µm when using ethanol.

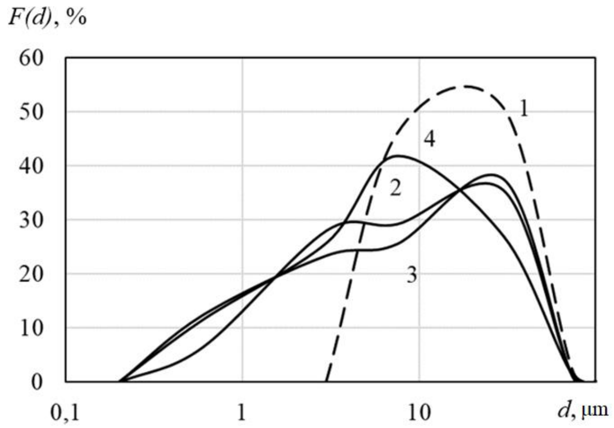


Fig. 2 *Particles size distribution of initial 87.5% Al + 12.5% Cu powder (1) and after HVED treatment in ethanol in VMD mode with different values* W_s *= 5 MJ/kg (2),* W_s *= 10 MJ/kg (3) and* W_s *= 20 MJ/kg (4)*

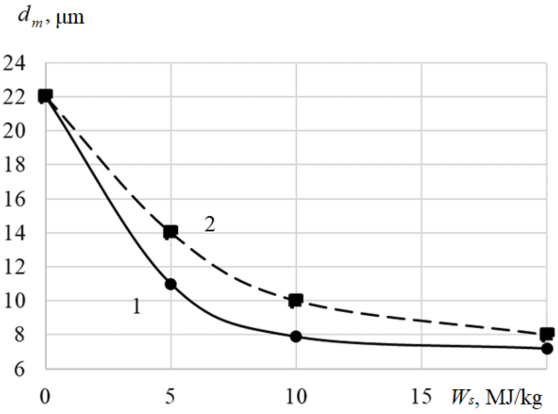


Fig. 3 *Dependence of the average particle diameter of 87.5% Al + 12.5% Cu powder on the specific processing energy when using SD mode (1) and VMD mode (2) in ethanol*

X-ray phase analysis of the initial mixture of 87.5% Al + 12.5% Cu powders confirmed that only aluminum and copper are present in the composition of the initial mixture (see Fig. 4), no significant impurity content was detected.

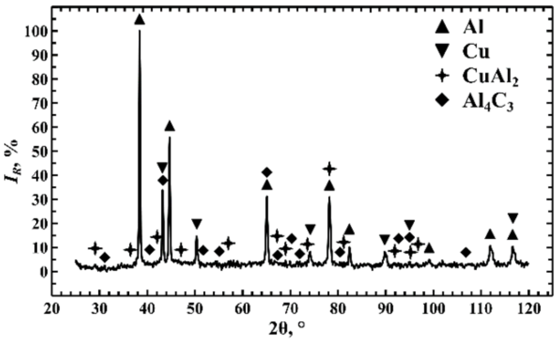
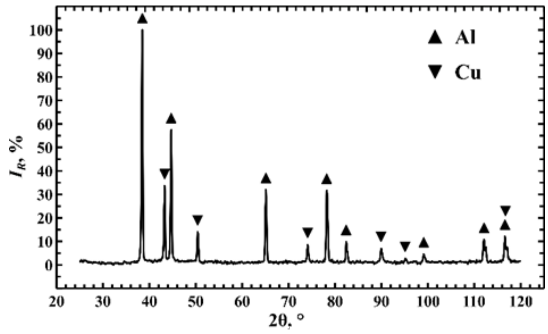
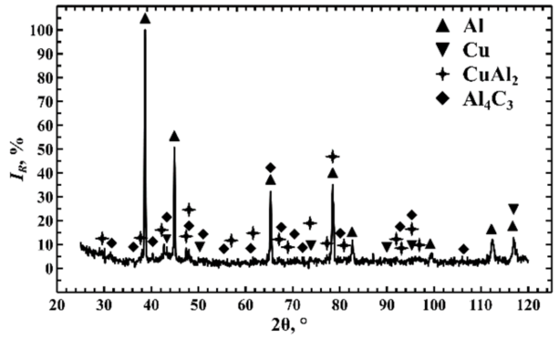


Fig. 4 *X-ray diffraction patterns of 87.5% Al + 12.5% Cu initial powder mixture*

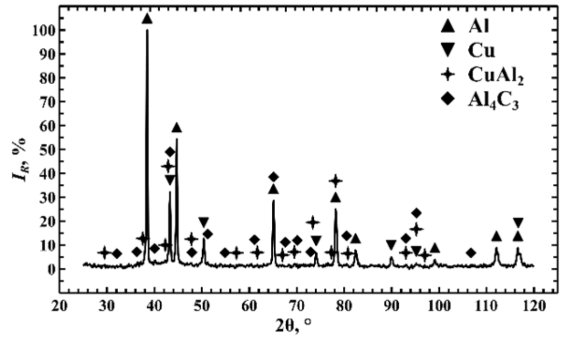
HVED treatment of 87.5% Al + 12.5% Cu powder mixture in ethanol with SD leads to a change in its phase composition (see Fig. 5, a, c, e). In the composition of the powder, in addition to the initial Al and Cu, as a result of HVED treatment with a specific energy of 5 MJ/kg, reflexes appear, corresponding primarily to $CuAl_2$ intermetallic phase and aluminum carbide Al_4C_3 (see Fig. 5, a). Increasing the specific energy to values of 10 and 20 MJ/kg increases the quantity of synthesized phases, as evidenced by some increase in the corresponding peaks on X-ray diffraction patterns (see Fig. 5, c, e).



a



b



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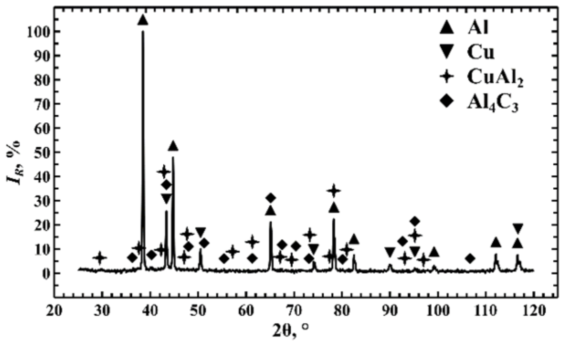
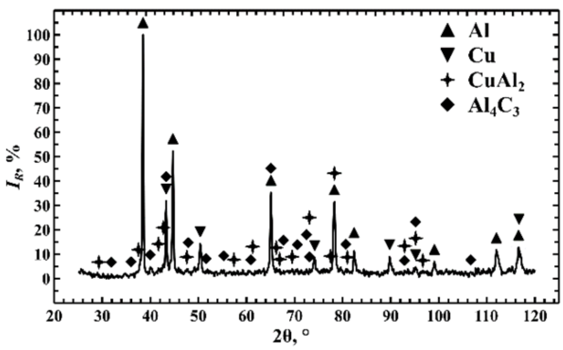
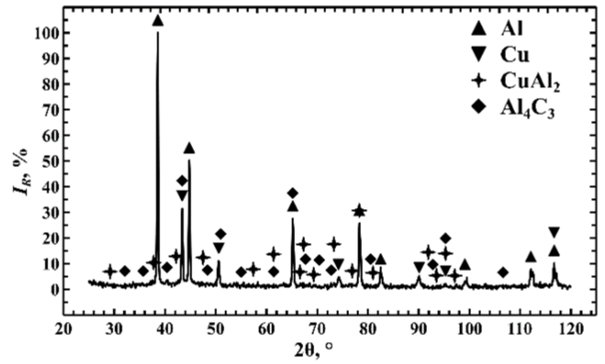


d



e

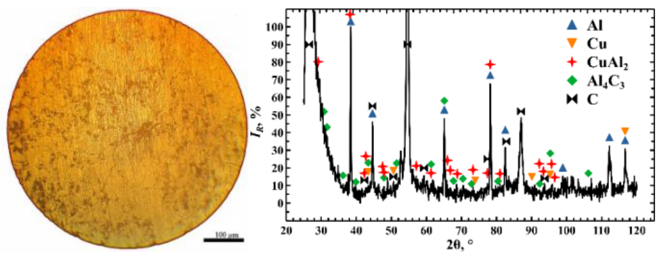


f

***Fig. 5** X-ray diffraction patterns of 87.5% Al + 12.5% Cu powder mixture after HVED treatment in ethanol in the modes of SD (a, c, e) and VMD (b, d, f) with a specific energy of 5 MJ/kg (a, b), 10 MJ/kg (c, d) and 20 MJ/kg (e, f)*

The change of HVED type from SD to VMD led to a slightly larger quantities of synthesized phases particles. At the same time, the decrease in the intensity of the reflexes corresponding to copper is quite noticeable (see Fig. 5, e and Fig. 5, f). Comparison of obtained results with the results of studies of the impact of high voltage electric discharge treatment in kerosene on the dispersity and phase composition of Cu – Al powder mixture, presented in [1], show, that the most intensive synthesis of new compounds is observed after processing a mixture of 87.5% Al + 12.5% Cu powders in ethanol in the VMD mode with W_s= 20 MJ / kg.

Consolidation of treated powder mixtures by SPS method allows obtaining composites with low porosity, dispersion-strengthened by $CuAl_2$ and Al_4C_3 compounds (see Fig. 6).



a b

***Fig. 6** Structure (a) and X-ray diffraction pattern (b) of material consolidated by the SPS method from the powder charge obtained by HVED treatment of a mixture of 87.5% Al + 12.5% Cu initial composition in the SD mode in ethanol with a specific energy of 5 MJ/kg*

Preparation of powders with the initial composition of 87.5% Al + 12.5% Cu by HVED in the SD mode in ethanol with a specific energy of 5 MJ/kg and subsequent consolidation by the SPS method for 10 min made it possible to increase the hardness of the obtained material of the Al – Cu – C system from 650 MPa to 900 MPa relative to the material obtained by SPS of the original mixture of 87.5% Al + 12.5% Cu and the material obtained by SPS from the powder mixture, treated with 10 MJ/kg, however, the resistivity of the obtained material increased to 0.1 $\Omega \cdot mm^2/m$.

The preparation of a powder mixture with an initial composition of 87.5% Al + 12.5% Cu by means of HVED in the VMD mode in kerosene with a specific energy of 5 MJ/kg makes it possible to obtain a material with a hardness of 600 MPa and a specific electrical resistance of 0.03 $\Omega \cdot mm^2/m$ after consolidation by the SPS method for 10 minutes, which corresponds to the level of specific resistance of cast aluminum. Increasing the consolidation time to 20 min increases the hardness of the samples to 1000 MPa, but their specific electrical resistance increases to 0.06 $\Omega \cdot mm^2/m$, which corresponds to consolidated copper of PMS-1 brand. The mass loss during abrasive wear for such material is 0.12 g per 5 km of travel, while for the material obtained from the charge of the original composition of 87.5% Al + 12.5% Cu by SPS for 10 min, this indicator is 0.99 g for a 5 km of wear.

4. Conclusions

1. The regularities of high-voltage electric discharge treatment of 87.5% Al + 12.5% Cu powder mixture in ethanol were studied and it was shown that HVED treatment with W_s = 5 MJ/kg leads to a decrease in the average particle diameter of the treated mixture from 15 µm to 11 µm (in SD mode), and increasing the specific processing energy to W_s = 20 MJ / kg allows to reduce the average particle diameter of the treated mixture to 7 µm.

2. It is found out that regardless of HVED mode, treatment of 87.5% Al + 12.5% Cu powder mixture in ethanol leads to the synthesis of $CuAl_2$ intermetallic compound and Al_4C_3 carbide, the amount of which increases with increasing specific processing energy.

3. It is shown that the preparation of powders with an initial composition of 87.5% Al + 12.5% Cu using HVED treatment in kerosene or ethanol with subsequent consolidation by SPS method allows obtaining metal-matrix composites of the Al – Cu – C system with increased indicators of hardness, electrical conductivity and wear resistance.

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