Plasma-arc reactor for production possibility of powdered nano-size materials

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Abstract. Nano-size materials of various chemical compositions find increasing application in life nowadays due to some of their unique properties. Plasma technologies are widely used in the production of a range of powdered nano-size materials (metals, alloys, oxides, nitrides, carbides, borides, carbonitrides, etc.), that have relatively high melting temperatures. Until recently, the so-called RF-plasma generated in induction plasma torches was most frequently applied [1-3].

The subject of this paper is the developments of a new type of plasma-arc reactor, operated with transferred arc system for production of disperse nano-size materials. The new characteristics of the PLASMALAB reactor are the method of feeding the charge, plasma arc control and anode design.

The disperse charge is fed by a charge feeding system operating on gravity principle through a hollow cathode of an arc plasma torch situated along the axis of a water-cooled wall vertical tubular reactor. The powdered material is brought into the zone of a plasma space generated by the DC rotating transferred plasma arc.

The arc is subjected to Auto-Electro-Magnetic Rotation (AEMR) by an inductor serially connected to the anode circuit.

The anode is in the form of a water-cooled copper ring. It is mounted concentrically within the cylindrical reactor, with its lower part electrically insulated from it.

The electric parameters of the arc in the reactor and the quantity of processed charge are maintained at a level permitting generation of a volumetric plasma discharge. This mode enables one to attain high mean mass temperature while the processed disperse material flows along the reactor axis through the plasma zone where the main physico-chemical processes take place. The product obtained leaves the reactor through the annular anode, from where it enters a cooling chamber for fixing the produced nano-structure.

Experiments for AlN synthesis from aluminium power and nitrogen were carried out using the plasma reactor described here above.
1. Introduction

During last several years PLASMALAB (Plasma Metallurgy Research Laboratory) developed a new construction of plasma arc reactor for reduction processing of raw and waist materials [5-8], based on Bethlehem’s “Falling film” (FFP)-reactor [4, 5].

The development of the FFP-reactor idea, according to PLASMALAB’s concept, involves mainly the method of feeding the processed raw materials and the plasma arc organization [6-9].

In the PLASMALAB FFP-reactor (figure1) charge particles are sucked by the rotating arc at the exit of the hollow graphitic cathode, and are transferred to the wall of the concentrically mounted tubular reactor-anode [6-10]. It is assumed that the particles will be heated and melted in the volume of the arc column, while depositing on the reactor-anode wall. The melted metal-slag suspension flows down the reactor anode wall and reduction processes run. The obtained metal-slag melt drops into the reactor tundish, where final reduction and gravitational separation of the two liquids take place.

Preliminary experiments with the designed and manufactured laboratory FFP-plasma reactor PLASMALAB (figure 1) [6-10] showed that its productivity and efficiency of processing the powder charge depends mainly on the way of organizing the plasma arc rotation and on arc geometrical dimensions.

Figure 2 shows a scheme of the original “PLASMALAB” system for Auto-Electro-

![Scheme of a stand for the experimental study of the rotation speed of the anode and cathode spots of a plasma arc in FFP-plasma reactor “PLASMALAB”](image)

K5(1)-semi-cathode upper (section); K5(2)-semi-cathode lower (section); 6-refractory insulator; A7(1)-upper semi-section of a graphitic tubular anode; A7(2)-lower semi-section of a graphite tubular anode; 8-inductor; 9-contact terminals; 10, 11-connecting cables (anode); 12-transferred plasma arc; 13-transducer (anode); 14-transducer (cathode); E-DC power supply; 15, 16-connecting cables (cathode)
Magnetic-Rotation (AEMR) of plasma arc [6]. The DC transferred plasma arc is generated by the current that runs in the inductor. This figure illustrates also the idea of finding the rotating speed of the anode and cathode spot [8].

2. “Rotating arc column” and “arc volume”
An interesting phenomenon is observed during the experimental determination of the arc rotation speed [9]. Consider a reactor “PLASMALAB” operating in “idle regime” (i.e. without supply of material for processing) and under fixed arc current value. Then, an arc with a constant length performing an Auto-Electro-Magnetic Rotation in the reactor practically keeps constant revolutions during a specific period of time (in this specific case - about 1÷5 min after the start of arc burning). After the expire of that time, the transducer abruptly stops registering rotation of the cathode spot, and in a while it stops registering the anode spot rotation, too, i.e. it seems that “the arc stops rotating”.

As found, that effect is accompanied by change of the noise produced by the burning arc. When arc spots rotate, the sound released is louder than that released by a freely burning arc (without electro-magnetic control). Moreover, sound pulsations are registered for low current values. At the moment when “the arc stops rotating”, intensity of arc sound and pulsation notably decreases.

To throw light on that phenomenon, we perform control of arc spot rotation in time considering different values of arc current, while the rest of the arc parameters are kept constant. As seen in figure

![Figure 3](image)

**Figure 3** Change of arc rotation speed for the time of constant arc current values 100, 150, 200 and 250 A

![Figure 4](image)

**Figure 4** Plasma Arc in FFP-Reactor at different mode of burning
A - Spiral motion of the arc column; B - Plasma volume
7, the effect “termination of arc rotation” occurs faster when higher current is applied. Consider current of 100 A. Then, arc rotation is observed for 4.2 min at the average. Consider now current of 250 A. Then, arc rotation is not observed after 1.1 min at the average. Visually, arc rotation ceases long before counters (13 and 14 in figure 2) stop registering impulses, which are generated by rotation of the anode and cathode spot, respectively. Another observed phenomenon is that cathode spot rotation ceases earlier than anode spot rotation. The following hypothesis is formulated as a result of measurements performed and phenomena observed.

The arc, having significant power, burns in an arbitrary small space bounded by the wall of the tubular reactor-anode. The temperature of the reactor working surface, even when applying water cooling, rises comparatively quickly up to values higher than 1500-1600 °C (if no material is fed for processing, this takes several minutes). At such temperature and under high speed of rotation of the arc column, gas volume in the reactor is ionized to a degree which provokes the generation of a number of additional gas electric discharges outside the basic arc column. At a specific moment, thanks to the high temperature, i.e. to the increased degree of ionization, the arc column looses its characteristic contours and transforms into a plasma volume, which fills the reactor working space.

Considering further technological development of the design of the plasma reactor “PLASMALAB”, it is extremely important to find out how the transformation of the rotating arc column into plasma volume will affect the effective transport of powder charge from the aperture of the hollow graphitic cathode to the wall of the tube reactor-anode. One should also find the efficiency of the heat-exchange running between the powder charge and the plasma volume.

Experiments of processing the powder charge (a mixture of converter slag, fine coke and fluxes) are performed for that purpose. The charge particle size is lower than 0.1 mm, and charge is uniformly fed into the improved plasma reactor “PLASMALAB” [9] by means of the powder-feeding system “PLASMALAB” [7].

Some modifications of the initial idea of the design and operation of the system “plasma torch with hollow graphite cathode - water-cooled copper tubular reactor anode” were made on the basis of numerous preliminary experiments [6, 8, 11-13], results found and analysis of a number of undesirable effects. These structural modifications of the improved “PLASMALAB” reactor are shown in figure 5.

A graphite crucible of special design 10 is placed along the axis of reactor 3 (figure 5), at its lower end, under the water cooled graphite anode 6 and in the tundishe. Products of the pyro-metallurgical process are collected in it. These are metal-slag melt and eventually unprocessed charge, unmelted and not transported by the arc to the reactor wall.

Figure 5 Scheme of the improved plasma reactor "PLASMALAB"

1 – hollow graphite cathode; 2 - nozzle; 3 – concrete reactor; 4 – water-cooled inductor serially connected in the anode chain; 5 – skull layer; 6 – graphite bush-anode; 7 – contact clamp; 8 – refractory insulator; 9 – tundishe roof; 10 – special design graphite crucible; 11 – thermocouple; 12 – slag; 13 – metal; 14 – unprocessed charge.
Copper ring-wise water-cooled inductor (figure 5) plays the role of a classical copper water-cooled reactor. Reactor space is walled by refractory concrete, and the inductor serves as reinforcement. Thus, thermal losses in the reactor are drastically decreased. Note that the reactor wall is already electrically non-conducting, and the anode power supply is connected to the graphite bush-anode through the inductor. Hence, maximal length of the burning arc is naturally attained.

Experiments were performed in the following sequence:
- the arc was ignited the operating current to 250 A was set up. The reactor operated in “idle regime” for 3 min, which was enough time to heat the internal surface of the reactor wall to a temperature higher than 800°C (figure 6) (heating was controlled by thermocouple 11 (figure 5));
- the arc passed from regime “rotating arc column” into regime “plasma volume”. The moment of that transition was found as corresponding to the change of the arc sound and to a slight voltage drop;
- charge feeding through the hollow graphitic cathode and into the reactor volume started via the powder-feeding system “PLASMALAB”;
- fed particles were taken by the plasma volume where they melted and were transported to the wall of the tube reactor. Skull was formed (figure 5) together with metal-slag melt which started flowing towards the reactor exit and entered the graphitic special design crucible placed in the tundishe. Since the water cooled power supplying graphitic bush (figure 5) was mounted in the lower section of the reactor, increase of the thickness of the skull layer 5 was observed. This cooling effect yielded a characteristic profile of the reactor longitudinal cross section (figure 7);
- the experiment proceeded for 15 more minutes after the reactor wall attained a constant temperature. This time was enough to reliably estimate how much charge was melted and landed on the reactor wall and how much of it fell non-processed into the crucible below the reactor.
- after the experiment conclusion, a profile of the tube reactor-anode was taken. The quantity of charge having melted and flown from the reactor into the crucible, as well as that of non-processed charge, were collected and weighted.

3. Results
For further technological development of the FFP reactor design, as well as for the extension of its application area, it is very important to find out what will be the effect of the described “plasma arc” metamorphosis in the “plasma volume” on the efficient transfer of powdered charge from the outlet of the hollow graphite cathode to the wall of the tubular reactor anode.

The experiments for reduction processing of the powder charge were performed under current constant value 250 A, with three different rates of charge feeding into the reactor working space – 10, 30 and 50 g/min.

Operating voltage varied from 50-55 V at arc ignition to 60-70 V when different charge quantities were fed. Reactor electric power varied within ranges 12.00 – 17.25 kW.

The experimental results are shown in figure 6 and figure 7. Significant difference in the degree of charge processing was observed for consumption of 10 and 30 g/min (figure 6). While for minimal
consumption of 10 g/min the quantity of processes charge was hardly 54%, for much larger consumption - 30 and 50 g/min, the quantity of processed charge grew up to 86 and 88%, respectively. That fact being illogical at first sight, can be explained with the arc burning mechanism, i.e. different mode of arc burning provided different charge transportation.

Charge quantity of 10 g/min was insufficient to cool the arc volume and to reverse arc burning regime from “plasma volume” to “rotating arc column”. Considering the latter, it is probable that the density of negatively charged particles moving from the cathode to the wall of the tube reactor is significantly larger. This is the probable reason for the significantly better transport capabilities of the “rotating arc column” as compared to those of the “plasma volume”. That supposition is backed up by the fact that for charge consumption of 10 g/min, the skull layer is quite thin (figure 7), melt accumulated in the crucible is homogeneous and the degree of reduction of the metal oxides is low – 61-66%, i.e. metal-slag melt has been strongly overheated, it has low viscosity and it has flown down into the crucible for a short time. The insufficient time is also the reason for low reduction degree. Besides, charge non-processed particles melted when moving along the reactor axis through the plasma volume and then entered the central section of the special crucible. Large part of those non-processed oxide particles were spheroidized. This fact confirms the extremely high temperature of the “plasma volume”.

* Consider material fed into the reactor. We adopt the term “processed charge”, meaning material which the arc has transported in melted state to the reactor wall and which has flown down the wall as a thin film. Then, the partially reduced metal-slag melt accumulates in the external ring-shaped section of the graphite crucible 10 (figure 5).
As seen in figure 6, charge losses for 10 g/min consumption are the greatest ones. This is so, since evaporation of the processed material is most intensive in this case.

The increase of charge quantity yields decrease of the temperature of the “plasma volume” and it is transformed into a “rotating arc column”. This is seen in the schemes of the cross sections cut along the reactor height (figure 7). Significant difference between the reactor profile for charge consumption of 10 g/min and that for charge consumption of 30 g/min is seen.

The lower reactor temperature for charge consumption of 30 and 50 g/min has led to the accumulation of a skull with significant thickness, especially in the reactor lower section where cooling was more intensive.

Consider significant increase of the quantity of charge fed for processing (over 45-50 g/min for the specific experiments discussed). Then, a tendency of extreme skull increase and a risk for reactor block up occur. Besides, increase of skull thickness leads to decrease of the reaction area of the melted charge layer where the reduction processes run most intensively. This effect is confirmed by the fact that for charge consumption of 50 g/min, the degree of reduction is 78-81%, while for charge consumption of 30 g/min it is 88-91%.

4. Nano-materials production

The results obtained gave us confidence that the PLASMALAB FFP-reactor, after proper redesign, could be successfully used to produce powdered nano-sized materials. For the purpose, the anode design was changed and the diameter of the water-cooled tubular FFP-reactor was increased (figure 9). The anode (figure 9) was shaped as a water-cooled ring and made of copper tube. It was mounted concentrically to the cylindrical reactor, in its lower part and was electrically insulated from it.

The principle of device operation is as follows. The arc is subjected to Auto-Electro-Magnetic Rotation by the inductor serially connected to the anode circuit (figure 9). The disperse material subject to processing and the technological gas (if required) are fed through the hollow graphite cathode by the powder feeding system (figure 1).

The energy parameters of the arc in the reactor and the quantity of processed charge are kept in proportions so as to maintain a “plasma volume” combustion mode. That mode permits achievement of high average mass temperature (over 2000°C), and the dispersed processed material passes along the reactor axis, through the plasma space where the main physico-chemical processes were running. The product leaves the reactor through the annular anode, after which it enters a cooling chamber where the obtained nano structure is fixed.

Figure 9 Schema of PLASMALAB FFP-reactor with hollow graphitic cathode and electromagnetic control of arc for nano-materials synthesize
The reactor described was used in experiments for the synthesis of AlN from aluminum powder and nitrogen (figure 10). During each experiment 100 g Al powder was processed. An average yield of 76% was achieved. The non-reacted aluminum powder was 22%. 78% of the produced AlN with average particle size 35 nanometers (figure 11) and average specific surface 39 m²/g separated in the cooling chamber. About 6% deposited on the reactor walls and the anode, and the rest of that quantity left the reactor with the off gases. The total charge loss with the off gases was about 16%.

5. Conclusions
- The first experiments demonstrated that the new design of PLASMALAB FFP-reactor is suitable for the production of nano-size powdered materials.
- The geometrical dimensions of the working space of reactor and the anode will be optimized in order to improve the technical and economic parameters of the process.

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Figure 10 SEM photograph of synthesized AlN powders collected in the collection vessel.

Figure 11 Size distribution of the AlN powder measured with a particle size analyzer.
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