CAVITATIONAL IRON MICROPARTICLES GENERATION BY PLASMA PROCEDURES

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Abstract
The paper presents the experimental installation for the production in argon plasma, of cavitational iron microparticles (pore microspheres, microtubes and octopus-shaped microparticles). Experimental results are presented and discussed and it is shown that absorbant particles with a minimum iron content are obtained by the plasma procedures.

Keywords: iron microparticles, microspheres, pores, octopus-shaped microparticles, plasma, microtubes.

1. Introduction

Diagnosing and therapy of a number of diseases can be also achieved by the use of ferri-ferromagnetic microparticles. Refs.[1-5] show the advances as well as the limits to magnetic microparticles applicability in medicine. Relatively recent research has revealed that iron microparticles having ad/absorbant properties are of interest for the magnetically controlled transport of anticancer drugs in tumor [6], magnetic hemosorption [7] etc.

Absorbant iron microparticles are of the Ferro-Carbon-4 type [6-8]. They are obtained by the oxidoreduction reaction of iron oxides in the presence of carbon, followed by the formation of an iron nucleus on which carbon is deposited as nanotubes [6]. The quantity of iron in the nucleus of the adsorbant microparticle is an essential parameter in the control of the toxicity of the anticancer terapy with absorbant iron microparticles [7, 8].

Transport of substances in general and that of drugs in particular can also be achieved by means of cavitational microparticles, namely: pore microspheres [9,10], microtubes [11] and octopus-shaped microparticles [12]. We consider that the cavitational microparticles can be used as well to produce magnetorheological suspensions (MRSs). In Refs.[13-21] are presented the adequate methods for high stability MRSs production and properties.
In what follows, we set ourselves to describe the procedure for obtaining cavitational microparticles in plasma and to show that by the plasma procedure the quantity of iron can be controlled.

2. Experimental installation

The overall configuration of the installation for the production of cavitational microparticles by plasma procedures is the one shown in Figure 1a. The installation includes the plasma generator A, the current source B, the material advance system C, the material fixing-positioning system P, and the plasmagen gas storage area (G).

The plasma generator shows laminary stabilization. The power source has a descending characteristic (Figure 1b). The idle operation voltage is $260 \text{V}_{\text{dc}} \pm 5\%$. It yields a current through the electric arc, which is continually adjustable between $70 \text{A}_{\text{dc}} \pm 10\%$ and $350 \text{A}_{\text{dc}} \pm 5\%$.

![Figure 1.](image)

(a) Overall configuration of the installation utilized for iron micro-tube production in plasma: A-plasma generator; B-current source; C-material advance system; P-positioning-fixing system for material; 1-cathode; 2-nozzle; 3-material (carbon steel rod); 4-contact nozzle cooled with water; $d$-nozzle –material distance; $\alpha$-attack (incidence) angle; $v$-material advance speed; $I$-current intensity; $U$-tension on arc; K-switch; Ar-argon admission; $\leftrightarrow$-rod advance direction; $\uparrow$-rod up-down positioning.

(b) Static characteristics of the power source for various values of the command current $I_c$ of the magnetic amplifier.

The material advance system together with the material fixing-positioning system allows the uniform insertion of material in the plasma and the positioning of the electrode rod.
The advance velocity of the electrode rod can be continually adjusted between $0.20 \times 10^{-3}$ m/s ±1% and $1.5 \times 10^{-3}$ m/s ±1%. By means of device P, the angle of incidence of the electrode rod and the plasma jet can be adjusted between $\pi/6$ rad and $\pi/2$ rad and the distance $\delta$ between $1.5 \times 10^{-3}$ m ±1% and $25 \times 10^{-3}$ m ±2%.

The plasma generator can function as plasma jet (closed K contact) or as transferred arc (open K contact).

The plasmagen gas storage area consists of argon tubes (150 bar). A pressure reductor (150 bar/1.5 bar) with flowmeter has been mounted on each plasmagen gas tube. The argon flow can be continually adjusted between $0.10 \times 10^{-3}$ m$^3$/s ± 5% and $60 \times 10^{-3}$ m$^3$/s ±2%.

3. Experimental Results and Discussion

The installation in Figure1a is used for the production of cavitational microparticles in plasma. The material from which cavitational microparticles are obtained in plasma is a carbon steel rod. The diameter $d_e$ of the rod is $3 \times 10^{-3}$ m±10%. The chemical composition of the steel (% mass) is C: 0.19; Mn: 10.85; P: 0.045; S: 0.045; Si: 0.40; Fe: 88.47.

The plasma generator has a nozzle of diameter $d=2.0 \times 10^{-3}$ m. The distance between the nozzle of the plasma generator and the rod is fixed at $\delta=6.0 \times 10^{-3}$ m and $\alpha=\pi/2$ rad (Figure1a). The intensity of the electric current through the plasma arc transferred onto the electrode rod is $I=175 \ A_{dc}$. The velocity $v$ of the rod and the argon flow $D$ are values which change during the experiment.

At $v=1.25 \times 10^{-3}$ m/s and $D=0.35 \times 10^{-3}$ m$^3$/s, parts of the rod melt. The melt, in continuous flow, is transported by the plasmagen gas. By solidification, a compact mass is obtained. By increasing the advance velocity of the rod to $v=1.10 \times 10^{-3}$ m/s, iron microparticles and solidified melt are obtained (Figure2a). We hold the advance velocity of the rod at the same value. But when we increase the argon flow to $1.00 \times 10^{-3}$ m$^3$/s, 95% iron microparticles result (Figure2b).

One can see through the optic microscope that some of microparticles have pores, as shown in Figure2c. Analysis of about 450 particles shows that 35% are pore microspheres. The mean microsphere has its diameter of 10 µm and its wall thickness of 0.75 µm.

Increase of argon flow by 286% determines instability of melt flow. So, the liquid metal moves through the plasma as drops, as shown in Ref.[22-24]. The shape of the drop is
spherical because $\sigma_{Fe} \gg \eta_j \cdot v_j$ (where $\sigma_{Fe}$ is the superficial tension of the melted metal, $\eta_j$ and $v_j$ are the dynamic viscosity and the plasma velocity.

![Figure 2](image1.png)

**Figure 2.** Shapes and sizes of the iron microparticles: (a) microparticles, microspheres and melted iron mass; (b) microparticles and microspheres; (c) pore microspheres.

Short after formation, the drop changes into vapors. A mixture of vapors and argon is formed. We have called this mixture “fluid sphere” [24]. The fluid sphere moves at the plasma velocity. At a certain moment, the fluid sphere-gas interface reaches the “dew drop” temperature for liquid iron. Then, the transformation of the fluid sphere surface into a membrane occurs [9]. A temperature gradient appears between the membrane and the fluid sphere center. The temperature gradient determines the movement of the vapour volume elements towards the membrane.

Each vapour element sets on the inner surface of the membrane. The contact time is short. Between the volume element and the membrane there occurs a transfer of substance
through diffusion in non-stationary condition [9, 24, 25]. After the transfer, another volume element takes its place. The process continues until no vapours are left in the fluid sphere.

The transferred vapours condense on the membrane. The result is a thickening of the membrane and the formation of the microsphere [9]. The velocity of the plasma jet is maximal in the discharge axis [10, 23, 24]. On decelerations of the microsphere [10], the inertial force of the gas in the microsphere becomes much greater by comparison with the force due to the superficial tension of the membrane.

Then, penetration of the membrane and formation of pores occur on solidification of the microsphere. In Refs. [9, 24] the condition for pore formation and their diameter are analytically determined.

At \( v=1.25 \times 10^{-3} \text{ m/s} \) and \( D=0.65 \times 10^{-3} \text{ m}^3/\text{s} \), there result microparticles, microspheres and, respectively, about 15% microparticles having a central nucleus (microsphere) out of which ligaments stick in the same plan. In Figure 3 one can see that the ligaments are empty. The particles displaying such shapes will be referred to by us as octopus-shaped microparticles. The mean sizes of the octopus-shaped microparticles are:

- for the nucleus (mean diameter: 12\( \mu \text{m} \) and wall thickness: 0.5\( \mu \text{m} \)) and,

- for the ligaments (length: 188\( \mu \text{m} \), equivalent diameter: 2\( \mu \text{m} \) and wall thickness: 0.35\( \mu \text{m} \)).

Bringing the advance velocity of the electrode rod to the initial value (\( v=1.25 \times 10^{-3} \text{ m/s} \)) and increase in the argon flow by 50% determine the formation of fluid spheres and vapours. At a certain moment and for a short time, because of the plasma viscosity, the fluid sphere becomes immobile.

At that moment, the hydrodynamic spectre of the fluid medium around the sphere consists of a movement around a circular obstacle, combined with the movement produced by a fixed-point whirl [12, 25]. The fixed-point whirl occurs around a circle resulting from the intersection of the sphere with the fluid medium plane of movement.

Stagnation points of the flow of the vapour and gas mixture form around the membrane. Ref. [10] shows that eight stagnation centers form around the fluid sphere, as shown in Figure 4. Current lines branch off each stagnation center consisting of vapours and gas, yielding closed a open-ended microtubes [10]. At \( v=1.15 \times 10^{-3} \text{ m/s} \) and \( D=0.75 \times 10^{-3} \text{ m}^3/\text{s} \), a movement of the melt is noticed in the plasma as continuous and parallel lines. Following solidification, fibres result as those in Figure 5a. Seen on the optic microscope, they appear like microtubes of shapes and sizes as shown in Figures 5b-5d. The mean microtube is characterized by: length of 100\( \mu \text{m} \); diameter of 3\( \mu \text{m} \) and 3wall thickness of 0.25\( \mu \text{m} \).
Figure 3. Octopus-shaped iron microparticles

Figure 4. [10]. Fluid sphere of r radius: 1-microsphere or/and liquid metal drop; 2-cylinder consisting of vapours and gas around the current line; a, b, ..., g-stagnation points.

For $v=1.15 \times 10^{-3} \text{ m/s}$ and $D=0.25 \times 10^{-3} \text{ m}^3/\text{s}$ the mixture consisting of vapours and gas has a stable movement. Along each current line, vapour and gas cylinders are formed. They move at the speed of the plasma jet. At a certain moment, the cylinder-gas interface reaches
the dew-drop temperature and it changes into a liquid membrane. The liquid membrane set off the vapour-gas mixture from the rest of the gas.

![Image]

(a)  
(b)  
(c)  
(d)

**Figure.5.** Iron fibres (a) and iron microtubes (b), (c) and (d)

Movements of the mixture volume elements from the center of the tube towards the membrane occur. A substance transfer in a nonstationary condition occurs between the volume element and the membrane [11, 25]. As in the case of microspheres, on the completion of the process, the membrane changes into a wall surrounding an empty space. Refs [11, 25] show the formation condition and the dependence of the size of the iron microtube on the main technological parameters. The octopus shaped microparticles have the lowest volume iron concentration (Table 1).

<table>
<thead>
<tr>
<th>Type of microparticle</th>
<th>Microsphere</th>
<th>Microtube</th>
<th>Octopus-shaped microparticles</th>
</tr>
</thead>
<tbody>
<tr>
<td>Volume concentration (%)</td>
<td>∼12</td>
<td>∼35</td>
<td>∼10</td>
</tr>
</tbody>
</table>

Table 1. Volume concentration of iron in the cavitational microparticles

For the case of liquid transport, the inner volume of the microsphere and microtubes has been calculated. In the case of the octopus-shaped iron microparticles, the previous
volume of the empty central nucleus and the liquid volume between the branches, respectively, have been calculated. For all cases the mean value particles have been considered. The volume thus obtained was related to the volume iron concentration, for each type of cavitary micro-particles. The results obtained are the ones in Table 2.

Table 2. Mean volume of liquids by relation to the volume iron concentration, \( V(\mu m^3/\%) \)

<table>
<thead>
<tr>
<th>Type of micro-particle</th>
<th>Micro-sphere</th>
<th>Micro-tubes</th>
<th>Octopus-shaped micro-particles</th>
</tr>
</thead>
<tbody>
<tr>
<td>( V(\mu m^3/%) )</td>
<td>∼27</td>
<td>∼14</td>
<td>∼262</td>
</tr>
</tbody>
</table>

From Table 2 there results that the octopus-shaped microparticles could be of interest for biomedical purposes because the iron concentration is low as compared to the volume of liquids incorporated. Diminution of the iron quantity lowers the toxicity level in cancer therapies, as shown in Refs [7, 8]. The magnetization curve and the hysteresis curve of cavitary microparticles are shown in Figure 6.

![Magnetization curve and hysteresis curve](image)

Figure 6. Magnetization curve (a) and hysteresis curve (b) of iron cavitary microparticles in a quantity of \( 178 \times 10^{-6} \) kg, raised by means of the vibratory probe magnetometer, type VSM-880 (Physica). M-magnetization, H-magnetic field intensity.

From Figure 6 it results that cavitary microparticles belong to the category of soft magnetic materials. The value of the magnetization of the cavitary microparticles is comparable to that of the microparticles used for biomedical purposes [1-8]. The existence of iron oxides on the surface of cavitary microparticles (Figure 7) can be removed by means of an Ar plasma mixed with \( \text{H}_2 \) and collecting the particles in liquid matrices.
The cavitational microparticles (Figures 2,3 and 5) have their surface covered with Fe₃O₄ (Figure 7). The presence of Fe₃O₄ is due to the oxygen traces in the technical argon.

Taking into account 450 cavitational microparticles, the mean volume occupied by the iron layer is calculated. Further, the volume occupied by the liquid is determined and the iron concentration contained by a mean cavitational micro particle is calculated. The results obtained are those in Table 1.

4. Conclusions

• The installation in Figure 1a allows obtaining microparticles in general and cavitational microparticles in particular;
• By maintaining the discharge current at constant values, cavitational microparticles are obtained by modifying the argon flow and, respectively, the material advance velocity;
• The iron concentration of the microparticles can be controlled by using the plasma procedure (Table 1);
• The volume iron concentration by relation to the ad/absorbed liquids volume is much higher in iron microparticles by comparison with the other kinds of cavitary microparticles (Table 2);
• Cavitary microparticles are soft magnetic materials (Figure 6).

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References