INVESTIGATION OF THE PROPERTIES OF MONODISPERSED MICROGRANULES OF REFRACTORY METALS PREPARED IN A PULSED ARC

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The formation of monodispersed metallic particles in a pulsed arc is described. The characteristics of the high-temperature oxidation of copper and molybdenum drops and their effect on the properties of microgranules are analyzed.

The development of modern technologies confronts investigators with new problems concerning the production of monodispersed drops of heat-resistant metals and alloys as well as purposeful control of the properties of monodispersed systems.

In this work we study the physical principles of the production of monodispersed drops of refractory metals by the electric-arc method [1] and we study the possibility of obtaining prescribed surface and phase properties of metallic monodispersed systems. The method for producing monodispersed spherical metallic particles is based on fusion of a thin cylindrical blank electrode by a pulsed electric arc. The properties of the particles are determined primarily by their phase composition and size, which are prescribed beforehand. Drops that are formed at the tip of the electrode are detached by forces arising in the arc column. The initial velocity of the drop formed is directed along the arc column at the moment of detachment. The heat effect of and the force exerted by a pulsed arc was studied by the method of photographing different stages of the process of melting of the electrode. The method is based on comparing the configurations of the melted electrode after it is exposed to arc pulses of definite width with the initial configuration. Thus we studied the action of pulses, whose width varies from 0.1τ up to τ with a step of 0.05τ, where τ is the width of a pulse after which drop detachment occurs, on an electrode. The successive stages of melting of the copper electrode are shown in Fig. 1. Analysis of such photographs showed that the rate of melting is established virtually immediately and is equal to, for example, 0.40 g/sec for copper and 0.24 g/sec for tungsten with arc power of 400 W per pulse. Such rates of melting, as the calculations show, indicate that the losses of energy to heat removal into the cold part of the melting electrode during the arc pulse are negligibly small and they make it possible to regard the heating of the end of the melting electrode as adiabatic. As a result of this, a large temperature gradient is observed on the boundary between the melted and solid part of the melting electrode. This gradient rapidly equalizes the temperature when the arc is switched off. Thus the drop formed up to some moment of action of the arc solidifies, without having enough time to change shape, when the heat source is switched off. At the finishing stages of melting of the drop and before the drop becomes detached, it is possible to see a displacement of the center of mass of the drop. Starting at this moment, before the pulse ends a drop of mass M is accelerated by the forces arising in the arc up to the initial velocity v_0 with which the drop departs. This makes it possible to estimate the order of magnitude of the force acting on the drop from the relation

$$F\Delta t = Mv_0,$$

where Δt is the time from the moment at which the center of mass of the drop starts to move up to the moment at which the drop becomes detached. The initial velocity of the drop v_0 was recorded on the basis of the particle tracks, recorded through an obturator. For copper particles with a radius of 140 μm the order of magnitude of the force was found to be $F = 2 \cdot 10^{-4}$ N. Obviously, this force must agree with the surface tension forces which hold back the drop. Detachment of the drop is opposed by the tangential component of the surface tension, which for copper particles with a radius of 140 μm is equal to $F_\sigma = 1.8 \cdot 10^{-4}$ N, comparable to the experimentally measured force; this indicates that the force can be determined by the method described.

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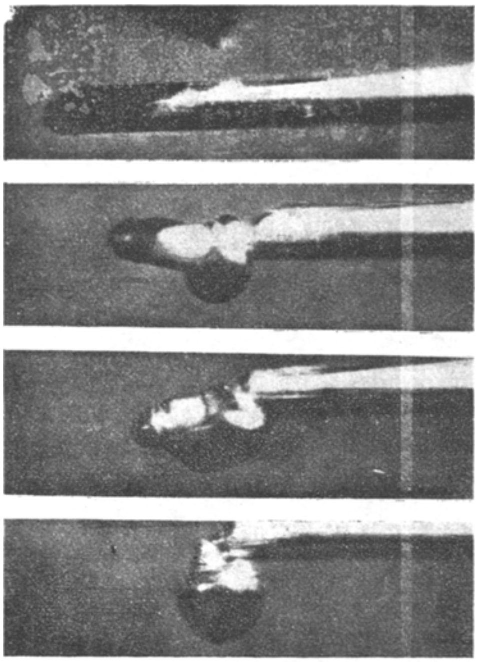

Fig. 1

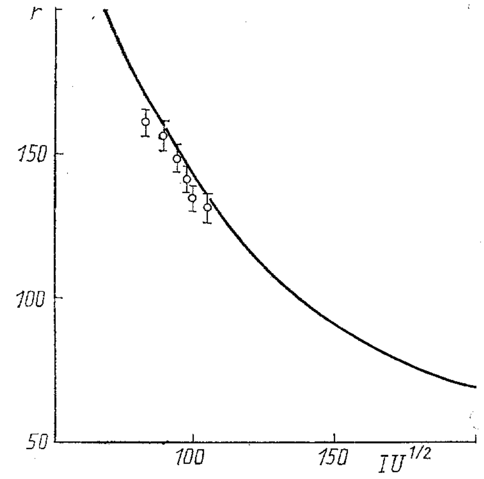

Fig. 2

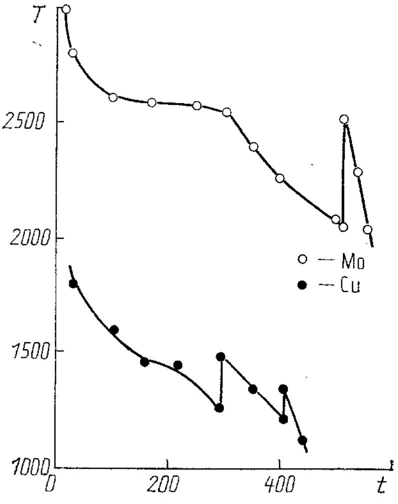

Fig. 3

Fig. 1. Successive stages of melting of a copper electrode. ×75.

Fig. 2. Theoretical and experimental dependences of the radius of particles obtained at the maximum possible temperature on the parameters of the electric discharge. r, μm; $IU^{1/2}$, A·$V^{1/2}$.

Fig. 3. Plots of the change in the temperature of copper (r = 140 μm) and molybdenum (r = 100 μm) particles as a function of time. T, °C; t, msec.

To clarify the nature of the dominant force which is capable of detaching the drop we analyzed the magnitude of the pinch-effect forces arising in an arc discharge, the pressure of the arc plasma, the electrostatic forces, and the electronic-pressure forces. Comparing these forces showed that only the force of the electron pressure determined [2] by

$$F_e = I\sqrt{\frac{2m}{e}U}$$

can compete with the surface tension in the cases studied. Taking into account the competion of the forces and making the assumption that the surface tension varies linearly as a function of the temperature [3] makes it possible to write the following expression for the radius of the drop r:

$$r = \frac{2\pi r_e^2}{\left(\frac{2m}{e}U\right)^{1/2} I}\left[\sigma_m + (T_0 - T_m)\left(\frac{d\sigma}{dT}\right)_m\right].$$

Figure 2 shows the dependence of the drop radius r on the parameter $IU^{1/2}$ calculated from this formula for the case of dispersing of copper particles with the radius of the melting electrode r_e = 0.07 mm. The initial drop was equal to 2500°C, which corresponds to the maximum possible temperatures for copper particles, since the boiling of copper is equal to 2560°C. In the case when particles are obtained in air with the maximum achievable temperature intense vapor-phase combustion of the particle is recorded experimentally; this indicates that its temperature is close to the boiling point. Figure 2 shows the measured values of the radius of particles which are obtained with the maximum temperature and the corresponding values of the parameter $IU^{1/2}$.

The agreement between the computed and experimental dependences in the interval 80 A × $V^{1/2} \leqslant IU^{1/2} \leqslant 110$ A·$V^{1/2}$ shows that the assumption that the electron pressure dominates in the process of drop detachment is correct.

The subsequent formation of the properties of the particles is determined by the character of the processes in which the particles are involved in the gas medium directly after

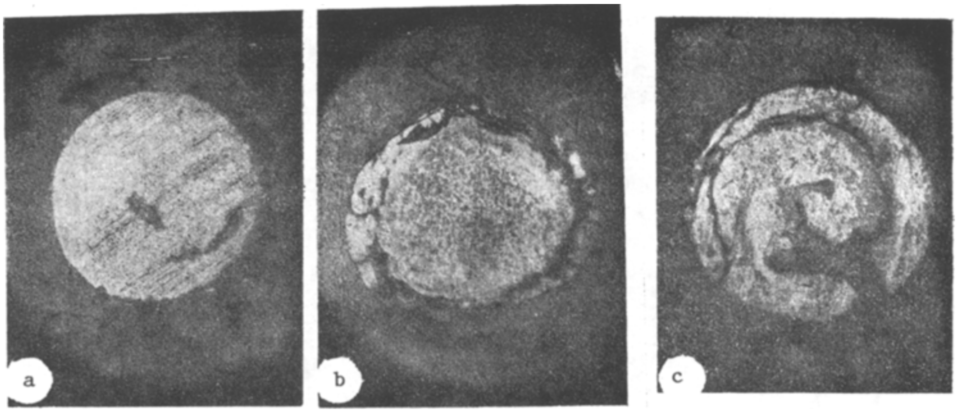

Fig. 4. Microsections of copper particles extinguished at different stages of burning: 10 msec (a), 200 msec (b), and 300 msec (c). ×150.

the particles are produced. Oxidation processes, occurring in an oxygen-containing atmosphere and, in particular, in air, are especially important. In the case of exothermal oxidation (combustion) of metallic particles, the phase composition and the structure of the particles change. For Cu and Mo particles studied, the combustion process divides into two or three (in the case of copper) stages with a sharp change in brightness at the transition between stages.

The temperature of a moving particle was measured with a pyrometric gauge, which was placed along the trajectory of the particles. For each section of the trajectory a series of measurements of the brightness temperature of a definite sample of particles was performed. The results of the measurements were averaged and corrections taking into account the emissivity of the matter in the working wavelength range were introduced. Plots of the temperature as a function of time for copper and molybdenum particles are presented in Fig. 3. The sharp peaks in the graphs are not the result of averaging, but rather they were observed in the output pulses of the pyrometric gauge and they correspond to the transition between stages of combustion. As one can see from the plots, the combustion regime changes in the case of molybdenum particles at a temperature of 2500°C, corresponding to the temperature of crystallization of molybdenum $T_m = 2615°C$, while for copper particles both jumps start at the same temperature of 1250°C, equal to the crystallization temperature of copper oxide Cu_2O.

For copper particles the change in the structure and composition of the particles at different temperatures was investigated as a function of the oxidation time. To this end particles moving in air for a known time were selected, and microsections of the particles were prepared and examined under a metallographic microscope. Photographs of the characteristic microsections are presented in Fig. 4. The oxidation time of the particles (Fig. 4a) is equal to 10 msec and the temperature at which the particles were extinguished was equal to 2200°C. For the particles (Fig. 4b and c) the times are equal to 200 and 300 msec and the quenching temperature are equal to 1400 and 1350°C, respectively. Examination of the microsections showed that in the case of particles moving in air at high temperatures an oxide film is not formed on the surface of the particles (Fig. 4a). After the particles cool to temperatures $T < 1800°C$ a layer of Cu_2O starts to grow on the surface (Fig. 4b); the cracks in the oxide film, which can be seen in the photographs, were formed when the sections were prepared.

At the second stage of combustion (the particles remained in motion for more than 300 msec) intense oxidation of the interior of the particles, which occurs nonuniformly and spreads, as a rule, from one zone, is observed. Taking into account the fact that the second stage starts at the moment of crystallization of the oxide and that this is observed for particles of different sizes makes it possible to propose the following interpretation of the phenomenon. For sufficiently low temperatures 1500-1200°C the rate of oxidation of a copper particle is determined by diffusion of Cu through the Cu_2O layer. When the oxide layer crystallizes a crack forms as a result of thermal settling and the oxidizer flows directly into the central unoxidized region of the particle. The reaction is sharply activated and the temperature of the particle rises. Next, the straight channel for the oxidizer is closed by a new layer of Cu_2O and the particle starts to cool. With the next drop in temperature to 1250°C crystallization of the oxide once again results in analogous processes, which are significantly weaker owing to the fact that by this time almost the entire volume of the copper particle is oxidized.

The questions associated with high-temperature oxidation of copper particles moving in air are especially important in connection with the development of controllable systems for depositing coatings with prescribed electrophysical properties. In the study of the combustion of copper and other particles there opens up the possibility of controlling the phase composition and size of the particles and forming an oxide coating with different thickness, which substantially changes the surface and volume properties of the particles, by varying the conditions of combustion and the moment of extinguishment. A regulatable, high initial temperature of the drops makes it possible to achieve good adhesion to the substrate and to form, in air under normal conditions, unoxidized coatings, since an oxide layer starts to grow on copper particles at temperatures $T < 1800°C$, and in the process there is enough time for the particles to traverse a significant distance, which makes it possible to employ electrostatic control devices.

NOTATION

τ, width of the pulse making it possible to detach the drop; M, mass of the particle; v_0, velocity of emergence; F, force; Δt, time from the start of displacement at the center of mass of the drop up to detachment of the drop; F_σ, surface tension force; F_e, force due to the electronic pressure; I, current; U, voltage; m and e, electron mass and charge; r, radius of the drop; R_e, radius of the electrode; T, temperature; T_0, initial temperature of the drop; T_m, melting point of the metal; σ_m, surface tension of the metal at the melting point; and $(d\sigma/dT)_m$, derivative of the surface tension of the metal at the melting point.

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APPLICATION OF FLUORESCING SOLUTIONS FOR TEMPERATURE DIAGNOSTICS OF MONODISPERSED MACROPARTICLES

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A method for measuring the temperature of monodispersed particles is studied. The method is based on the fact that the fluorescence of water solutions of fluorochromes is temperature dependent. The employed method makes it possible to reduce significantly the instability of the excitation light flux and of the optoelectronic recording apparatus.

In order to study heat transfer of photodispersed macroparticles which are moving in a gas flow the temperature must be determined by a contact-free method, since existing methods of measurement with the help of thermocouples do not permit investigating drops smaller than 400 μm in size [1]. In [2] a method based on the temperature dependence of the fluorescence of water solutions of some substances was proposed. The character of this dependence for different substances and concentrations of the same substance is different, since it is determined by different physicochemical processes occurring in the solutions. We employed as temperature indicators water solutions of rhodamine 6G and riboflavin.

For low-concentration solutions of fluorochromes the fluorescence intensity is proportional to the intensity of the excitation light

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