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AT REDUCED AIR PRESSURE

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GENERATION OF AEROSOLS BY THE ELECTRICAL EXPLOSION OF WIRES AT
REDUCED AIR PRESSURE

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ABSTRACT

The exploding wire method of particle production allows us to model the high speed formation of aerosols because of the fast heating and evaporation rates inherent to this technique. The method is also of interest from the viewpoint of controlling the production of aerosols of a particular material with a specific particle size distribution at a specific efficiency.

The electrical explosion of iron, aluminum, titanium, and copper wires has been investigated in various gases at pressures from 0.01 to 1 atm. In these experiments the energy density introduced into the material, w , is normalized to the sublimation energy of the material, w_s . The energy density also controls the heating rate. Particle and agglomerate sizes were determined using transmission electron microscopy and laser scattering methods. The specific surface area of the powder was measured by low-temperature adsorption. The phase composition was determined by X-ray diffraction.

The specific energy introduced into a material, w/w_s , the pressure (density) of the surrounding medium, and the exothermic effect due to oxidation reactions are controlling parameters in particle formation.

Increasing the energy density increases the internal energy of the material, the expansion velocity and the number of condensation centers, while the final particle size decreases. With an exothermic oxidation reaction, the optimum energy density can be less than the sublimation energy of the material. As a result, metal oxides are formed. As the density of the surrounding medium is increased, the particle size decreases because of an increase in the frequency of collisions with the ambient gas and more rapid cooling of the particles.

Electrical explosion of wires, at reduced air pressures, allows for the production of ultra-fine powders of oxides of various metals with particle sizes of less than 50 nm. The method is environmentally safe and does not require excess energy expenditures. The electrical explosion of wire at reduced pressure allows for new possibilities in the production of ultra-fine powders (UFP).

1. INTRODUCTION

The current interest in UFPs is predicated on the promise of producing materials with novel or improved properties. Various methods for the production of UFPs with a particle size of 1-100 nm have been developed. For example, methods utilizing laser [1], plasma [2], and electric arc [3] are reported in the literature.

The greatest advantage of the EEW (electrical explosion of wire) method over other evaporation techniques is that the electric energy is transferred directly into heat because of the extremely rapid energy release involved in this process. Depending on the experimental parameters,

the EEW products can be liquid, vapor-droplet, vapor, or vapor-plasma. The velocity of expansion of the exploded wire and its temperature may exceed 1000 m/s and 10,000 K, respectively. Because of this, the EEW method has been used to investigate the formation of particles at high heating and cooling rates [4-7].

The size, composition, and properties of EEW powder particles depends on many factors. The fineness depends, primarily, on the energy density introduced into the wire [6, 8]. The chemical activity of the environmental medium [8, 9] and its density [10] are also of importance. At high material expansion velocities, the number of collisions leading to growth of the nuclei is restricted. Lowering the environmental density decreases the size of particles produced and a narrowing of the particle size distribution is also expected. Research along this line is both of scientific and engineering interest.

2. INITIAL DATA

The surrounding medium causes a counterpressure which hinders expansion of the exploded wire. R.B. Baksht, et al. [11] who simulated the explosion of copper wires in water and vacuum with specific input energies up to 9 kJ/g noted a substantial difference in the expansion velocities: 10^5 cm/s for explosion in water as compared with 2×10^6 cm/s for explosion in vacuum. Because the surrounding medium hinders scattering of the material, the probability of attachment of a new atom to a condensation nucleus increases and an increase in the density of the surrounding medium should increase the average particle size. From this viewpoint, the work of G.P. Glazunov et al. [10] is illustrative. These workers investigated the formation of powders in an inert gas at pressures of 1-600 atm.

Figure 1. The effect of the surrounding gas (argon) pressure on the particle size distribution for the Ti powder (data of Ref. [10]) for the pressure $p = 5$ (1), 20 (2), 100 (3), and 600 atm (4).

As shown in Fig. 1, the mean particle size increases with increasing argon gas pressure while the particle size distribution broadens. In Fig. 2, the increase in the particle diameter with the gas pressure is shown using data from Fig. 1. The diameters plotted in Fig. 2 correspond to the distribution maxima in Fig. 2. Electron-diffraction and X-ray analyses performed by the authors of Ref. [10] showed that the particles are spherical monocrystals.

Figure 2. Ti and Al_2O_3 particle diameter versus pressure.

Also in Fig. 2 a similar relationship for Al_2O_3 is shown using data provided by Lerner [12]. Lerner exploded 0.31 mm diameter aluminum wires in argon-12% oxygen gas mixtures to produce aerosols of aluminum oxide. For Al_2O_3 the mean-square diameter d_s determined from the measured specific surface area, s , for the gathered powder is presented. In Lerner's experiments the ratio between the energy density introduced into the wire, w , and the sublimation energy, w_s , was $w/w_s = 1.3$. A decrease in particle size with decreasing gas pressure was also observed in these experiments.

Other methods for synthesizing fine powders show a similar trend of increasing particle size with increasing gas pressure. One such study by V.N. Antziferov, *et al.* [1] involved laser vaporization of aluminum in air to produce aluminum oxide. In this work, the specific surface area of the powder produced at an air pressure of 0.1 atm is $120 \text{ m}^2/\text{g}$, and at atmospheric pressure it decreases to $40 \text{ m}^2/\text{g}$. The object of this work is to investigate possibilities of ultra-fine powders production by the EEW method at lower pressures.

3. EXPERIMENTAL TECHNIQUES

The electrical explosion of Fe, Al, Cu, and Ti wires in air, nitrogen or a mixture $\text{N}_2 + \text{O}_2$ at pressures between 0.01 and 1 atm was investigated. The experimental equipment used to produce and analyze the powders are described below.

3.1. Powder Production

The electrical circuit used for the EEW is shown in Fig. 3. To perform an explosion the capacitor, C , is discharged through the wire to be exploded, EW. To begin the process the

Figure 3. (a) The electric circuit of the set. L is the inductance and R is the resistance of the circuit. (b) The current oscillograms for shorting (1) and the electrical explosion (2).

capacitor, C , is charged and the wire is automatically fed between the electrical contacts. Next, the switch, S , closes and the capacitor, C , is discharged. The current flows along the electric circuit, producing Joule heating and explosive evaporation of the wire, EW. After the first explosion, the process is recycled.

In Fig. 3b a typical oscillogram for the discharge current is shown. The Joule energy introduced into the wire was determined using the current oscillogram, $I(t)$:

$$W = QV_0 - \frac{Q^2}{2C} - \frac{LI^2}{2} - R \int I^2 dt,$$

where $Q = \int I dt$ and V_0 is the charging voltage on the capacitor. In this study the heating rate was also controlled, and conditions for fast electrical explosion [13] were fulfilled. To achieve fast electrical explosion the energy deposited into the wire must be of the order of the sublimation energy and the heating time shorter than the time required for development of instabilities (capillary and magnetohydrodynamic) but longer than the time required for the current propagation across the wire radius. Under fast electrical explosion conditions, heating occurs simultaneously over the entire wire length and radius.

Explosions were performed in a hermetic chamber filled with either air, nitrogen or a mixture $N_2 + O_2$. The pressure was controlled and maintained at a designated level. The powder produced by this method was collected and analyzed.

3.2. Powder Analysis

Ordinary methods of analysis were used to determine the powder size, phase composition,

Figure 4. The histograms of the particle size distributions for the samples Nos. 7 (a), 8 (b), 9 (c), 11 (d), and 12 (e) in Table I, respectively.

and chemical composition. Particle shape and size distribution were determined by transmission electron microscopy (TEM) using EM-125 and Tesla-BS 500 microscopes and carbon replicas. Carbon replicas were prepared by first forming a powder suspension by intensive mechanical stirring of oxide powders in water. For pure metals the powder suspension was formed by stirring in methanol. Next, a glass substrate was coated with the powder from a suspension and a carbon replica was sprayed. The replica was subsequently removed from the glass substrate and examined by TEM. In Fig. 4 histograms of the powder size distribution are plotted from measurements of no less than 300–500 particles. In addition to TEM studies, the specific surface area of the samples was measured by the Klyachko-Gurvich method [14]. This method is based on adsorption of dry air at the temperature of liquid nitrogen.

Ultra-fine powder agglomerates very readily. Agglomerate size was determined using laser scattering. Phase composition was determined by electron and X-ray diffraction methods. In some instances chemical analysis was performed to determine the total nitrogen content.

4. EXPERIMENTAL RESULTS

The experimental results are listed in the Tables I and II as well as in Figs. 4 and 5. In Table I the particle sizes are characterized by the specific surface area, s , the surface median diameter, d_s (SMD), and the count median diameter, d_m (CMD). SMD was determined from the known relation $d_s = 6/(\gamma s)$, where γ is the density and the particle is assumed to be spherical. CMD was determined from the TEM measurements plotted in Fig. 4.

Figure 5. Particle size versus the ambient gas pressure. The data for Al wire explosion from this work is designated by crosses. The reminder of the data is from Fig. 2.

4.1. Particle Size

The effect of introduced energy density on particle size can be observed from the data obtained for the explosion of aluminum in nitrogen at pressures of 150 and 350 mm of Hg. Under these conditions the amount of AlN formed is small; the total nitrogen content is no more than 2% of the total mass. The specific surface area of the powder increases with increasing introduced energy density, w/w_s . The specific surface area, s , also increases with decreasing pressure. When

Al was exploded in nitrogen, the specific surface area of the powder, s , varied within a range of 50 to 130 m²/g and the SMD varied within a range of 20 to 40 nm.

An unexpected result was observed when oxygen was added to nitrogen or when air was used as the environmental medium. All other factors being the same, a small oxygen addition (5 %) to the nitrogen produced considerably finer powder. This is evident, when the specific surface areas of samples No. 5 and No. 6 (see Table I) are compared with ones for No. 7 and No. 8, respectively. In going to oxygen-containing gas, the powder fineness is elevated and the particle size decreases. The specific surface area of alumina varies within the range of 90 to 150 m²/g, equivalent to a SMD for alumina spherical particles of 10–18 nm.

Table I. Experimental Results

No.	Powder sample	Wire material	Powder material	Gas, p , mm Hg	w/w_s	s , m ² /g	d_s , nm	d_m , nm	Fig 4.
1	T 1	Ti	TiO ₂	Air, 350	1.5	43	33		
2	T 2	Ti	TiO ₂	Air, 150	1.5	37	38		
3	N 5	Al	Al	N ₂ , 150	1.0	128	18		
4	N 6	Al	Al	N ₂ , 350	1.0	64	35		
5	N 7	Al	Al	N ₂ , 150	0.6	60	37		
6	N 8	Al	Al	N ₂ , 350	0.6	55	40		
7	N 9	Al	Al ₂ O ₃	O ₂ + N ₂ , 150	0.6	135	17	5.5	Fig. 4, <i>a</i>
8	N 10	Al	Al ₂ O ₃	O ₂ + N ₂ , 350	0.6	150	15	5.5	Fig. 4, <i>b</i>
9	F 1	Fe	Fe ₃ O ₄	Air, 150	1.1	20	58	15.5	Fig. 4, <i>c</i>
10	F 6	Fe	Fe ₃ O ₄	Air, 350	0.7	33	33		
11	A 2	Al	Al ₂ O ₃	Air, 250	1.1	96	17	15.5	Fig. 4, <i>d</i>
12	A 6	Al	Al ₂ O ₃	Air, 250	0.6	163	11	10.0	Fig. 4, <i>e</i>

From analysis of the data for the alumina powder, it is observed that particle size can be minimized according to the environmental pressure, to oxygen content in the environment, and to the energy introduced into the material. A comparison of the data for sample No. 11 with No. 12 (alumina) and for No. 9 with No. 10 (iron oxide) shows that the fineness is higher for a smaller introduced energy, which is lower than the material sublimation energy w_s . Exothermic effects of chemical reactions are also significant in the formation of oxide particles. The data of samples No. 1 and No. 2 for titanium oxides and No. 7 and No. 8 for alumina in Table I also confirm this fact. In these instances a reduction in the gas pressure from 350 to 150 mm of Hg resulted in a decrease in oxygen concentration and an increase in particle size. The exothermic effect and the fineness have been decreased. Chemical analysis also disclosed a decrease of the amount of oxide formed with decreasing pressure.

Table II. Size Distribution of Agglomerates, D μm

Sample		d_s , nm	d_m , nm	D μm , by percentile				
Name	No.			0.1	16.0	50.0	84.0	99.9
B157	11	17	15.5	0.29	0.69	2.57	7.66	60.52
B161	12	11	10	0.28	1.31	5.49	26.81	116.71

4.2. Particle Morphology

The analysis of ultra-fine particles is a difficult problem. Measurements of the specific surface area of the powder only provide accurate information about particle size if the morphology, smoothness and degree of equiaxialness of the particle are known. TEM analysis is capable of providing information on these properties. Particles of the order of 5 nm often form as thin plates or needles. There is a tendency to form spherical particles as dimensions increase. The synthesis of oxides also results in the formation of more equiaxed particles. For instance, examination of samples No. 11 and No. 12 (Table I) revealed that the vast majority of the particles formed had a near-ideal spherical shape.

The tendency of ultra-fine particles to form coarse agglomerates is the other problem which complicates analysis. The smaller the particle size, the larger the agglomerate size. This is illustrated with the data on the agglomerate size distribution measured by laser light scattering for two samples given in Table II. In the "sample" column the laser scattering file name and the corresponding sample number from Table I are given. It is seen that the ultra-fine particles having initial diameters of 17 and 11 nm form rather coarse agglomerates with mean diameters of 2.57 and 5.49 μm for the samples No. 11 and No. 12, respectively. Their maximum dimensions reach 60.52 and 116.71 μm , respectively.

5. CONCLUSIONS

The results presented in this paper show that particle formation is a complex process dependent on several parameter such as the energy introduced into the wire, the ambient gas pressure and the heat effect of chemical reactions. These factors are dependent rather than independent and must be considered together.

These experiments have shown that reduced pressure can decrease particle size considerably (see Fig. 5). The general character of relationships between the size of oxide particles and the gas pressure, the oxygen content of the gas, and the introduced energy density have been revealed. A detailed parametric study is necessary to optimize these variables. To obtain the most complete and accurate information about morphology and size it is necessary to use a variety of methods to analyze ultra-fine powders.

Electrical explosions of wires, at reduced air pressures, allows for the production of ultra-fine powders of oxides of various metals with particle sizes of less than 50 nm. This method is environmentally safe and does not require excess energy expenditures. The electrical explosion of wire at reduced pressure allows for new possibilities in the production of ultra-fine powders.

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