Production of tungsten powders by electrical explosion of wires

A.P. ILYIN, O.B. NAZARENKO[*], D.V. TIKHONOV
Tomsk Polytechnic University, Lenin str. 30, 634050, Tomsk, Russia

The results of the study of phase and dispersed composition of the powders produced by electrical explosion of tungsten wires in the different gases are presented. The influence of electrical parameters (the value of energy input in wire and the arc stage of the explosion) is studied. The factors that make for increasing dispersiveness of nanopowders are the lower pressure of surrounding gas ambient and the use of addition of chemically active gas - nitrogen.

1. Introduction

Tungsten powder is widely applied as additive for producing special steels, main component high-strength and high-temperature alloys, that are used for production of the cutting instrument, stamps and parts of machines and mechanisms operating in conditions of high temperature. The considerable improvement of characteristics of functional material can be reached with the use of nanopowder. One of the methods to obtain the metal nanopowder is an electric explosion of wires (EEW).

The electrical explosion of wires is the process of explosive destruction of a metal wire under the action of great density current ($>10^{10}$ A/m^2) [1]. This process is accompanied by the scattering products, shock waves and electromagnetic radiation. Material of the wire transmutes into particles of nanosized range (10…100 nm) in accordance with certain conditions. The formation of particles in EEW process occurs in the presence of strongly nonequilibrium conditions – the large gradients of temperature and speeds of cooling ($> 10^7$ K/s), intensive heat transfer and mass transfer, ionization, mixture, influence of electric and magnetic fields.

Applicability of EEW for obtaining tungsten powders was shown in [2]. It was stated that phase composition of electroexplosive tungsten powders includes polymorphic modifications α-W и β-W, and the surface layer of tungsten particles forming at passivation in air, consists of oxides W$_3$O. The peculiarity of tungsten powder, which was produced with method EEW, is formation β-W. This phase has a lower X-ray density (19.1 g/cm^3) in comparison with α-W (19.3 g/cm^3). The oxide layer of the finest fraction of powder is well-crystallized oxide W$_3$O [3]. The largest particles are covered with amorphous oxide layer, element composition of which is closed to WO$_2$.

Content of β-W in electroexplosive tungsten powder amounts to 30-40 mass. %. The increase of the specific energy input in the wire causes rise of contents β-W. This fact is explained with the increase of the velocity of the EEW products expansion, and, consequently, with the increase of velocity of cooling the scattering products of explosion. It brings to stabilization of high temperature modification β-W [2]. At the same time, in this work there is not clear information about the influence of the composition of surrounding gas and the value of arc stage energy on characteristics of tungsten powder.

This paper reports the study of the influence of the EEW energy parameters such as energy input in the wire and energy of the arc stage, the composition of gas and the gas pressure on dispersiveness of powder, produced by electric explosion of tungsten wires.

2. Experimental details

The principal scheme of the industrial installation UDP-4G for producing electroexplosive powders is shown in the Figure 1. The installation works as follows. The capacitor battery 2 is charged from the high-voltage power source 1. The wire driving mechanism 3 is used for automatic feed of the exploding length of wire 4 in the electrodes gap. When the wire reaches the high-voltage electrode 5, the commutator 6 operates, and the electric discharge of the capacitor battery occurs on this part of wire: the wire explodes. Obtained powder collects in the powder collector 7. Gas refined from powder is given back in the explosive chamber 9 by means of the ventilator 8. The explosive chamber 9 is vacuumed before working, and then the chamber is filled with working gas atmosphere by means of system 10. As working gas for production of tungsten powder the gases were used: 1) the gases at the pressure P of 1.5·10^5 Pa: argon (Ar), Ar with addition of 10 vol. % nitrogen (N$_2$), pure N$_2$, N$_2$ with addition of 5 vol. % hydrogen (H$_2$), 2) N$_2$ at the pressure of 0.3·10^5 Pa and 0.1·10^5 Pa. The metal powder produced in inert gases self-ignites immediately after the contact with 100%-air. Thus, the passivation procedure is required

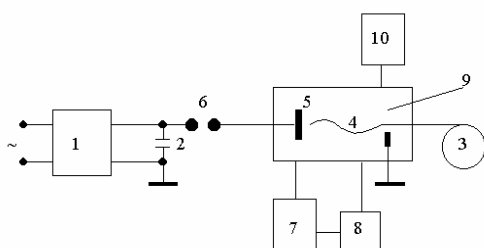with low oxidation by following medium: working gas + 0.1 vol. % air [3].

Fig. 1. Principle scheme of installation UDP-4: 1 - high-voltage power source, 2 - capacitor battery, 3 - wire driving mechanism, 4 - exploding wire, 5 - high-voltage electrode, 6 – commutator, 7 - powder collector, 8 – ventilator, 9 – chamber, 10 – system of filling with working gas.

The tungsten wires with diameter $d = 0.2…0.3$ mm and length $l = 50…80$ mm were used in the experiments. Electrical explosion of wires was carried out under conditions of "fast" explosion with an arc stage. Energy parameters of EEW were regulated with change of charging voltage and geometric characteristics of exploding wires. The specific electrical energy input in the wire (e) was changed from 0.4 to $1.5e_s$ (e_s is the sublimation energy of the wire material); the energy of the arc stage (e_a) – $(0.7…1.7)e_s$. Parameters of electric circuit: capacitance $C = 2.25$ µF; charging voltage $U = 15…30$ kV; inductivity $L = 0.58$ µH.

The phase analysis of the obtained powder was performed using CuK$_\alpha$-radiation of a DRON-3.0 X-ray diffractometer. Particle shapes and the dispersiveness were determined by means of a JSM-840 scanning electron microscope. The determination of specific surface area (S_{sp}) was carried out by using a method of low temperature nitrogen adsorption (BET). The differential-thermal analysis (DTA) and thermogravimetric analysis (TGA) was performed with apparatus Q-1000.

3. Results and discussion

It is known [4, 5], that the increase of the energy input in the wire leads to the rise of dispersiveness of electroexplosive powder. To investigate the influence of energy characteristics of EEW on the dispersiveness of the tungsten powders gas argon at the pressure $1.5·10^5$ Pa was used as working atmosphere. The conditions of the powder production and the values of specific surface area are shown in the Table 1. The diameter of the wires in these experiments was 0.2 mm.

Figure 2 shows the dependences of the specific surface area of the electroexplosive tungsten powders on the specific electrical energy input in the wire (*1*) and the specific electrical energy of the arc stage (2). The increase both of the energy input in the wire and of the energy of the arc stage causes the rise of dispersiveness.

Table 1. Specific surface area of electroexplosive tungsten powders depending on energy parameters.

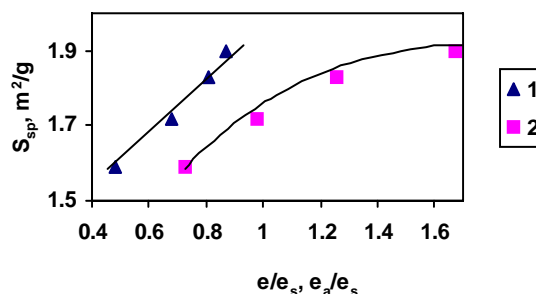| №	U, kV	l, mm	e/e_s	e_a/e_s	S_{sp}, m^2/g
1	28	80	0.87	0.76	1.90
2	28	60	0.81	1.26	1.83
3	28	50	0.64	1.67	1.90
4	23	50	0.68	0.98	1.72
5	20	50	0.48	0.73	1.59

Fig. 2. Dependence of specific surface area of electroexplosive tungsten powders on the specific electric energy input in the wire (1); the specific electric energy of the arc stage (2) .

It should be noted that for $U = 28$ kV (Table 1, the samples 1–3) the decrease of the wire length from 80 to 50 mm causes to the comparatively slow reduction of electric energy input in the wire, and to intensive rise of energy of the arc discharge. At the same time, the specific surface area of powder is practically constant $(1.83…1.9$ m^2/g). For constant length of wires (50 mm) the decrease of voltage entered in the wires from 28 to 20 kV causes to the reduction both of electric energy input in the wire and the energy of the arc stage. The specific surface area of the electroexplosive tungsten powders in this case decreases from 1.9 to 1.59 m^2/g.

According to the electronic microscopy investigation, the particles of the tungsten powder have spherical shape and smooth surface (Figure 3). There is the fraction of particles in powder, diameters of which are less than 100 nm. It is noticeable that all particles present spheres that point to passing them through liquid state and on the action of the surface tension forces.

The conditions of experiments and the values of specific surface area of the tungsten powders, which were obtained under the explosion in different gases, are presented in the Table 2. The length of the exploding wires in all experiments was 60 mm.

Fig. 4 represents the specific surface area values of obtained powders. According to these results, the composition of surrounding gas ambient in the explosive chamber influences greatly on the dispersiveness of the tungsten powder. At the gas pressure $1.5·10^5$ Pa the powder with large specific surface area was produced under the use of N_2 as surrounding ambient for the wires

with diameter 0.2 mm (the sample 3, Table 2) and under the addition of N_2 in Ar for the wires with diameter 0.3 mm (the sample 6, Table 2). The tungsten powder with the largest dispersiveness was produced under the use of N_2 at the pressure $0.3 \cdot 10^5$ Pa. The specific surface area of electroexplosive tungsten powder in this case was 2.59 m^2/g.
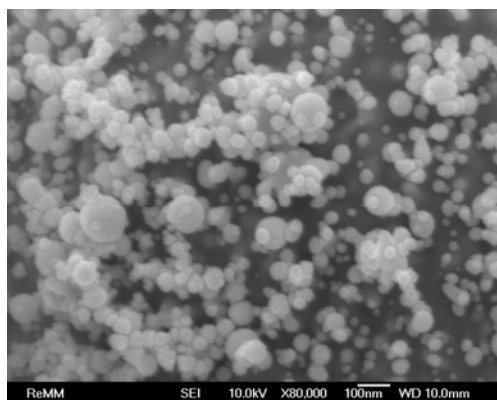
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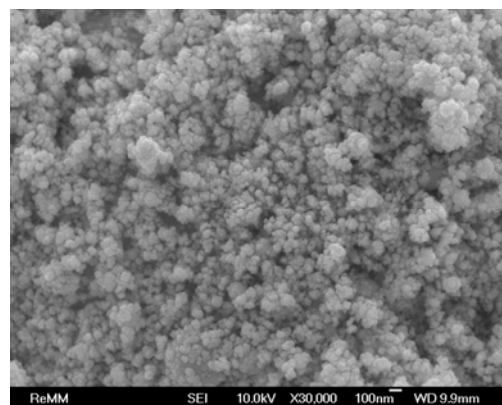
b

Fig. 3. Microphotos of electroexplosive tungsten powder, obtained in mixture N_2 and Ar at $P = 1.5 \cdot 10^5$ Pa (a), in N_2 at $P = 0.3 \cdot 10^5$ Pa (b)

Table 2. Specific surface area of electroexplosive tungsten powders depending on composition of surrounding gas ambient in the explosive chamber.

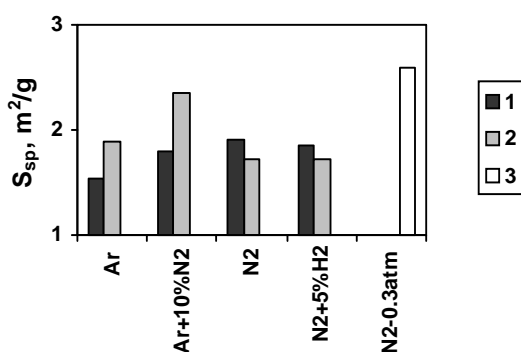| №	d, mm	U, kV	e/e_s	e_a/e_s	Gas	P, $\cdot 10^5$ Pa	S_{sp}, m^2/g
1	0.2	13.7	0.45	0.78	Ar	1.5	1.54
2	0,2	13,7	0,50	0,74	Ar+10 vol. % N_2	1.5	1,80
3	0,2	13,7	0,52	0,72	N_2	1.5	1,91
4	0,2	13,7	0,53	0,71	N_2+5 vol. % H_2	1.5	1,86
5	0,3	22,9	0,35	1,12	Ar	1.5	1,89
6	0,3	22,9	0,35	1,13	Ar+10 vol. % N_2	1.5	2,35
7	0,3	22,9	0,69	0,82	N_2	1.5	1,72
8	0,3	22,9	0,67	0,84	N_2+5 vol. % H_2	1.5	1,68
9	0,3	22,9	0,43	0,93	N_2	0.3	2,59

Fig. 4. Dependence of the specific surface area of electroexplosive tungsten powders on composition of the surrounding gas ambient in the explosive chamber: 1) d = 0.2 mm, P=1.5 $\cdot 10^5$ Pa, 2) d = 0.3 mm, P=1.5 $\cdot 10^5$ Pa, 3) d = 0.3 mm, P=0.3 $\cdot 10^5$ Pa.

In case of the lower pressure in the explosive chamber the density of EEW-products in process of expansion reduces quicker than under the explosion in conditions of higher pressure, and the probability of coagulation and sintering particles decreases.

Thus, the specific surface area of electroexplosive tungsten powders slightly depends on electrical parameters of explosion such as the energy input in the wire and the energy of the arc stage. The value of the specific surface area of the powders obtained in Ar at the pressure $1.5 \cdot 10^5$ Pa changes from 1.6 to 1.9 m^2/g. The use of N_2 as working gas permitted to increase the value of the specific surface area of the powder from 1.7 m^2/g at the pressure $1.5 \cdot 10^5$ Pa in the explosive chamber to 2.6 m^2/g at the pressure $0.3 \cdot 10^5$ Pa.

The activity of the prepared tungsten powders was estimated by the method of DTA-DTG under linear heating (15 ºC/min) in air with the following parameters: the temperature at which the first stage of oxidation started (T_1, °C) and the second stage of oxidation started (T_2, °C),

the degree of transformation (the oxidation level) of powders at temperatures $\leq 1000°C$ (α, %), and the maximum oxidation rate (v_{ox}, %/s). These parameters are presented in the Table 3.

Table 3. Thermal stability of powders prepared by electroexplosions of tungsten wires under the heating in air.

№	e/e_s	e_a/e_s	Gas	P, $\cdot 10^5$ Pa	T_1, °C	T_2, °C	α, % $\leq 1000°$ C	v_{ox}, %/s
1	0.45	0.78	Ar	1.5	310	465	23.7	0.0061
2	0,52	0,72	N_2	1.5	310	470	23.8	0.0070
3	0,53	0,71	N_2+5 vol. % H_2	1.5	320	475	23.9	0.0065
4	0,43	0,93	N_2	0.3	310	515	23.7	0.0097

The process of oxidation has three stages. The second stage is the stage of intensive oxidation. In the examined samples the temperature at which the second stage of oxidation started (T_2) depends on the surroundings and the gas pressure considerably: T_2 increases if the addition of chemically active gas were used and at the lower pressure. The powder obtained by electrical explosion in N_2 at the pressure $0.3 \cdot 10^5$ Pa has the maximum oxidation rate (v_{ox}, %/s) and the degree of transformation (α, %). This fact is explained with the greatest dispersiveness of this powder.

Thus, the parameters of thermal stability of the powders prepared by tungsten wire electroexplosions are closely correlated with their dispersiveness.

4. Conclusion

Dispersiveness of the tungsten powders produced by the electrical explosion of wires is defined by the ratio of the length to diameter of wires, the specific electrical energy input in the wire and energy of the arc stage. In studied range of energy the specific surface area of obtained powders slightly depends on both of the energy input in the wire and of the energy of the arc stage. The value of the specific surface area changes from 1.6 to 1.9 m^2/g. The most significant parameter of the dispersiveness regulation is the use of low pressure of working gas atmosphere in the explosive chamber (0.3×10^5 Pa and lower) and the use of addition of chemically active gases, such as nitrogen. In this case the specific surface area increases to 2.6 m^2/g, and at that the average diameter of the particles decreases to 0.196 μm.

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[*]Corresponding author: obnaz@mail.ru