

Installation for Preparing of Nanopowders by Target Evaporation with Pulsed Electron Beam

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Abstract Production of weakly agglomerated nanopowders with the characteristic size of about 10 nm and a narrow particle size distribution is still a topical problem especially if the matter is an acceptable output (>50 g/hour), a high purity of the final product, and a low (energy consumption. The available experience and literature data show that the most promising approach to production of such powders is the evaporation-condensation method, which has a set of means for heating of the target. From this viewpoint the use of pulsed electron accelerators for production of nanopowders is preferable since they allow a relatively simple adjustment of the energy, the pulse length, and the pulse repetition rate. The use of a pulsed electron accelerator provides the following opportunities: • a high-purity product; only the target and the working gas will interact and their purity can be controlled; • evaporation products will be removed from the irradiation zone between pulses; as a result, the electron energy will be used more efficiently; • adjustment of the particle size distribution and the characteristic size of particles by changing the pulse energy and the irradiated area. Considering the obtained results, we developed a design and made an installation for production of nanopowders, which is based on a hollow-cathode pulsed gas-filled diode. The use of a hollow-cathode gas-filled diode allows producing and utilizing an electron beam in a single chamber. The emission modulation in the hollow cathode will allow forming an electron beam 5 to 100 ms long. This will ensure an exact selection of the beam energy. By now we have completed the design work, manufactured units, equipped the installation, and began putting the installation into operation. A small amount of nanopowders has been produced.

Keywords : Nanopowders, Pulsed electron beam, Target evaporation

1. Introduction

Production of weakly agglomerated nanopowders with the characteristic size of about 10 nm and a narrow particle size distribution (the geometric mean deviation $\sigma < 1.5$) is still a topical problem especially if the matter is an acceptable output (> 50 g/hour), a high purity of the final product, and a low (not more than two sublimation energies) energy consumption.

A weak agglomeration makes it relatively easy to obtain compacts having a high density when bulky nanosized materials are produced and provides uniform mixtures of nanopowders. A narrow particle

size distribution and small dimensions of particles allow the utmost realization of unique properties of these particles (high activity, low sintering temperature, high mutual solubility of substances, large sorption ability, etc.).

Therefore, the main objective of this project consists in development of an efficient method for production of pure weakly agglomerated nanopowders with particles about 10 nm in size and the particle size distribution with $\sigma < 1.5$.

It is desirable that the method allows the use of relatively cheap components comprising a wide spectrum of starting materials (conducting and nonconducting substances, alloys and metals, mechanical

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mixtures, etc.).

The available experience and literature data show that the most promising approach to production of such powders is the evaporation-condensation method, which has a set of means for heating of the target (resistive heating, low-temperature plasma heating, heating of conducting materials with a high current pulse, laser beam heating, heating with beams of accelerated electrons and ions, eddy currents, etc.).

From the viewpoint of a minimum contamination of the final product and universality of starting materials used as the target (coarse powders, powder mixtures, metals, alloys, mixtures of metals and nonmetals, etc.), heating with a laser beam or an electron beam is most universal. However, lasers provide a very low efficiency of conversion of the electric power to the beam and their cost is large as compared to an electron accelerator (by a factor of 3 to 5 larger) when the average output power of the installation is over 1 kW. Moreover, laser radiation has larger cross-sections of the interaction with vapors and the plasma than accelerated electrons. This incurs additional energy losses.

The available literature data on the use of lasers^{1,2)} and continuous radiation electron accelerators³⁻⁵⁾ for production of nanopowders show that in both cases the efficiency of conversion of the radiation energy to the heat for heating of the target does not exceed 10%. The energy loss is due mostly to absorption and dissipation of radiation by the plasma and vapors of the evaporated material over the target surface.

The energy deposition time (the pulse length) should be shorter than the time of considerable expansion of heated vapors of the material to the gas medium surrounding the target. Only expansion to a low-pressure gas provides high expansion rates ensuring the decrease in the vapor concentration and production of small-size particles.

From this viewpoint, the use of pulsed electron accelerators for production of nanopowders is preferable since they allow a relatively simple adjust-

ment of the energy, the pulse length, and the pulse repetition rate.

The use of a pulsed electron accelerator provides the following opportunities:

- a high-purity product; only the target and the working gas will interact and their purity can be controlled;
- a high coefficient of conversion of the electric power to the electron energy, up to 50%;
- a low cost, availability and diversity of targets (metals, alloys, powder mixtures, conducting and nonconducting materials);
- the installation will cost 2 to 3 times less than a laser installation, while the output power will be the same;
- evaporation products will be removed from the irradiation zone between pulses (the blow rate of only 10 cm/s will be required at the operating frequency of 30 Hz and the diameter of the beam on the target equals 3 mm); as a result, the electron energy will be used more efficiently, while the particle size will be preserved at a level of 10 nm;
- adjustment of the particle size distribution and the characteristic size of particles by changing the pulse energy and the irradiated area;
- since parameters of the pulse beam can be easily changed, the energy input can be matched to the moment the vapor cloud appears when a different target material is used; therefore, the energy is used very efficiently.

Considering the obtained results, we developed a design⁶⁾ and began making an installation for production of nanopowders, which is based on a hollow-cathode pulsed gas-filled diode (HCPGD)⁸⁾.

The use of a hollow-cathode gas-filled diode allows producing and utilizing an electron beam in a single chamber. Therefore, it is unnecessary to solve such a complicated technical problem as the transport of a low-energy electron beam to the atmosphere. The emission modulation in the hollow cathode will allow forming an electron beam 5 to 50 ms long.

This will ensure an exact selection of the beam energy.

2. Design and brief Description of Operation of the Installation for Production of Nanopowders

The HCPGD installation for production of nanopowders (hereinafter referred to as “the installation”) is intended for experimental studies concerned with refinement of the technology, improvement of production regimes, fabrication of pilot batches of nanopowders, and final development of the installation units. Technical characteristics of the installation are given in Table 1.

By design, the installation consists of 3 basic components.

1) A vacuum chamber on a pedestal (Fig. 1). The vacuum chamber houses an electron gun, an evaporation chamber, beam focusing and deflection coils, a powder collection system, and a target unit.

2) A technical rack, which mounts a vacuum pump, vacuum system commutation valves, a gas cylinder, a reducer and a leak valve of the gas inflow system.

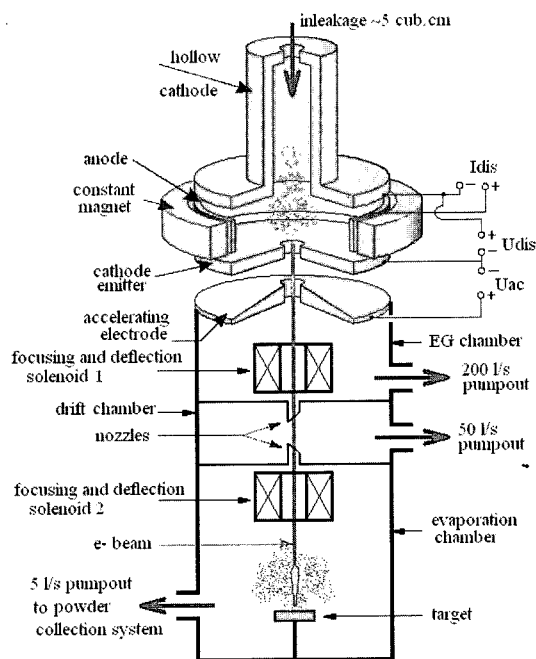


Fig. 1. Installation diagram.

3) A control rack, which mounts a system controlling the installation and all its units and systems, and an accelerating voltage source.

The electron gun (EG) of the installation is based on a hollow cathode with the discharge plasma initiated by different methods. By design, EG is fitted on top of the vacuum chamber. The hollow-cathode electron gun is shown schematically in Fig. 1. Time diagrams of EG are given in Fig. 2.

Until the moment $t = 0$ the discharge voltage source (Fig. 1) provides a negative potential U_{dis} of the emitter cathode relative to the hollow cathode. The emission current at this moment is $I_{beam}(0) = 0$.

At the moment $t = 0$ a voltage is applied from the discharge current source (Fig. 2) to the electrodes in the discharge chamber and a discharge with the current I_{dis} is initiated. The emission current at this moment is also zero thanks to the potential between the cathodes.

At the moment $t_1 = t_1$ (t_1 is equal to ~ 30 ms, which is necessary for formation of a stationary discharge) the $U_{dis}(t_1) = 0$. The discharge chamber cathodes

Table 1. Technical characteristics of the installation

No.	Parameter	Value
1	Power consumption, kWA, maximum	5
2	Power mains	380 V (3 phases), 50 Hz
3	Pulse length, μ s	5-50
4	Repetition rate, Hz	up to 500
5	Accelerating voltage, kV	up to 50
6	Beam current, A	0.3
7	Beam diameter, mm	1.5
8	Chamber pressure, Pa	10^{-1} - 10^5
9	Gas inflow rate, l/s	up to 63
10	Weight of the installation, kg, maximum	500
11	Floor space, m*m	2*2

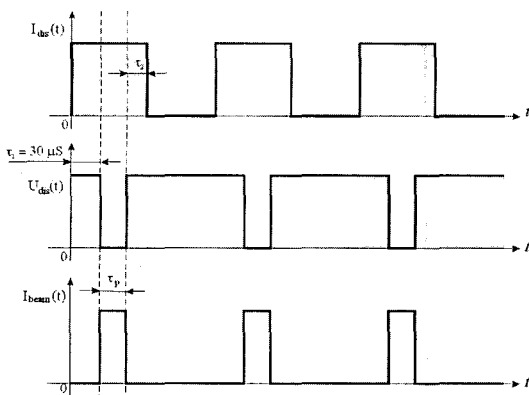


Fig. 2. Time diagrams of EG.

acquire a similar potential and, as a result, the I_{beam} arises and is accelerated by voltage U_{ac} .

At the moment $t_2 = t_1 + t_p$ (t_p is the pulse length) the U_{dis} again acquires the maximum value and the emission current stops.

At the moment $t_3 = t_2 + t_2$ the voltage is removed from the I_{dis} source and the discharge is quenched.

Processes in EG proceed at a certain pressure of an assigned gas. Therefore, a vacuum is produced first in the vacuum chamber and then a gas is leaked into the vacuum. The vacuum system produces a vacuum. It includes a vacuum pump (2NVR-5DM, the capacity of 5 l/s), a vacuum measuring system, pipelines and valves commutating gas flows.

A vacuum is produced by a vacuum system including (Figs. 1 and 3):

- diffusion vacuum pumps for EG pump-out (2 units type AVP 100/100 with 2NVR-5DM pumps rated at 100 and 5 l/sec respectively);
- an AVR50 unit (50 l/sec) for the drift chamber pump-out;
- a 2NVR-5DM pump for the evaporation chamber pump-out;
- a vacuum measuring system.

All pumps in places of connection have cryogenic traps of vapour of vacuum oil.

The gas is bled in by a gas inflow system, which consists of a gas cylinder, pipelines, a reducer, a leak valve, and a control system.

The control rack mounts the following units and devices:

- a discharge supply unit;
- a beam focusing and deflection unit;
- an accelerating voltage source (voltage source 1 in Fig. 1);

a control panel.

The control rack mounts the following units and devices:

- a discharge supply unit (Fig. 1), which consists of a pulsed current source (for powering of the discharge) and a pulsed voltage source (for the bias between the hollow and emitter cathodes);

The discharge supply unit is shown schematically in Fig. 1. It comprises a pulsed current source 1 (for discharge power supply) and a pulsed voltage source 2 (for a potential between the hollow and emitter cathodes).

The pulsed voltage source (hereinafter referred to as “the potential voltage unit, BVU”) consists of two sources and serves to apply a potential between the hollow and emitter cathodes:

- a beam focusing and deflection unit, which serves for focusing and aiming of the electron beam on the upper gas-dynamic window and, also, focusing and deflection of the beam after it passes the lower window;
- an accelerating voltage source, which supplies the accelerating voltage;
- a control panel.

The powder collection system consists of a cylindrical cooler, which is cooled by liquid nitrogen, a pusher-scraper, and a powder collection vessel.

The electron beam current was measured using three transducers – two current transformers and a shunt. One transformer measured the current passing through the accelerating voltage source, i.e. the full beam current. The other transformer was installed in the current lead, which connected the target and the vacuum chamber frame, and measured the current of the beam hitting the target. The shunt was fitted between the front flange (electric-

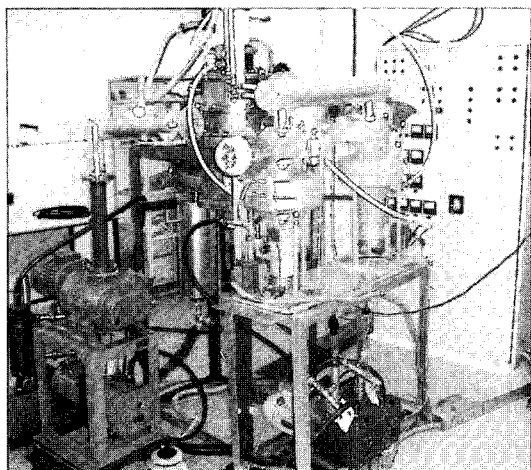


Fig. 3. External appearance of the installation.

cally insulated) of the drift chamber and measured the current lost during the beam transport.

3. Performance of Experiments on the Installation

The erection of the installation was followed by some commissioning work (CW) on the electron accelerator. CW included the study of the possibility to produce a pressure differential in the drift chamber, generate and focus the beam, and transport it to the target.

A pressure differential should be produced in the drift chamber, because the normal operation of the hollow-cathode pulsed gas-filled diode (HCP GD) requires a pressure of about 10^{-4} Torr. However, quick cooling of particles and their transport require a pressure of several units or tens of Torr.

Considering the literature data,⁷⁾ the design com-

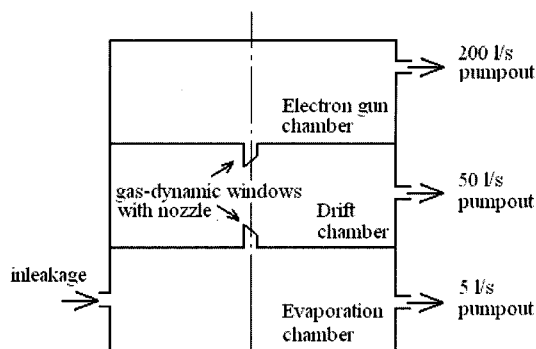


Fig. 4. Vacuum diagram.

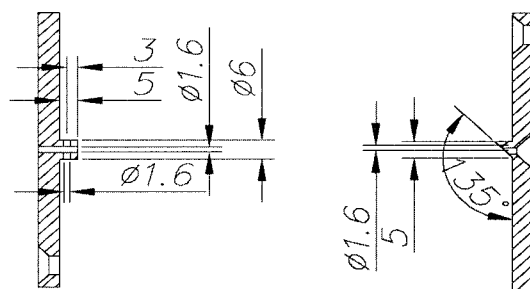


Fig. 5. The gas-dynamic windows with the nozzles.

prising two gas-dynamic windows installed at a distance of 100 mm opposite one to the other was chosen (Fig. 4). Each nozzle had a through hole 2 mm in diameter with the lower edge cut at an angle of 45 degrees (Fig. 5). The EG pressure was measured as a function of the pressure in the evaporation chamber at different rates of the gas leakage into the latter.

The measurement results are given in Table 2 and Fig. 6. It is seen that the chosen configuration of the channel for the beam transport provided a considerable pressure differential (over the required interval

Table 2. Results of the experiments on the pressure differential in the chamber

Chamber	Pressure, Torr			Remarks
	No inleakage	Inleakage of 5 dm ³ /hour	Inleakage of 60 dm ³ /hour	
Evaporation	0.72	1.5	4.1	5 l/s pumpout
Drift	$9 \cdot 10^{-3}$	$1.8 \cdot 10^{-2}$	$5 \cdot 10^{-2}$	50 l/s pumpout
Electron gun	$2.7 \cdot 10^{-4}$	$3.5 \cdot 10^{-4}$	$1.3 \cdot 10^{-4}$	200 l/s inleakage

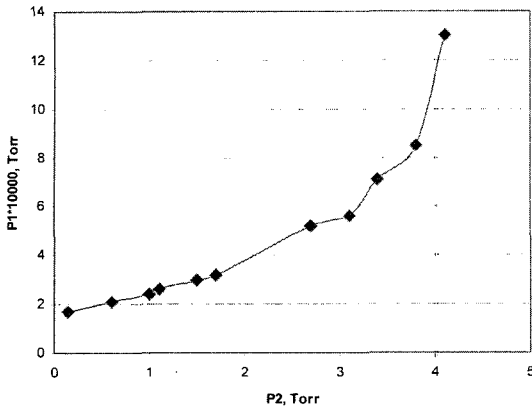


Fig. 6. EG pressure vs. the evaporation chamber pressure.

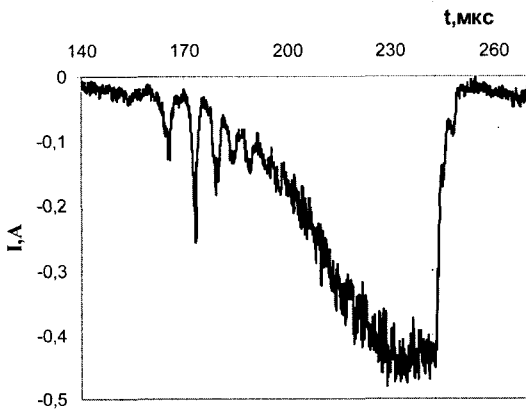


Fig. 7. Oscillograms of the full beam current.

of up to 10 Torr) at relatively small inleakage rates.

The measurement results demonstrated that a stable discharge with a current of up to 1.2 A was generated in EG. This discharge was used for formation of an electron beam, which was focused to a 1-mm diameter on the target and had the energy of up to 50 keV, the current of up to 0.46 A (Fig. 7), the length of up to 100 ms, and the repetition frequency of up to 500 Hz. Eighty percent of the beam was brought to the target.

Powders of zirconium oxide, and stainless steel were synthesized in initial experiments. First experiments for nanopowder preparation show next.

- The characteristic size of the 10YSZ ($ZrO_2 + 10\%Y_2O_3$) particles of the powders is about 2-5 nm (Fig. 8), they are amorphous (Fig. 9) and

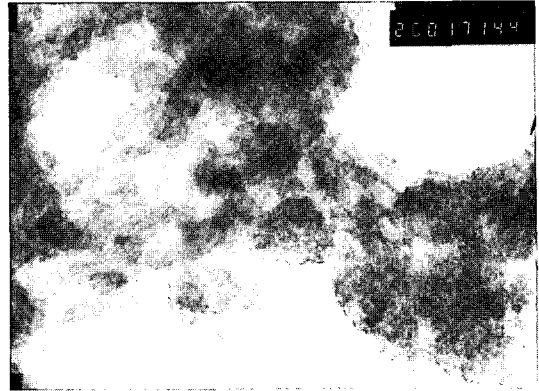


Fig. 8. The 10YSZ powders TEM photo from JEM-200 (Multiplying is 400 000).

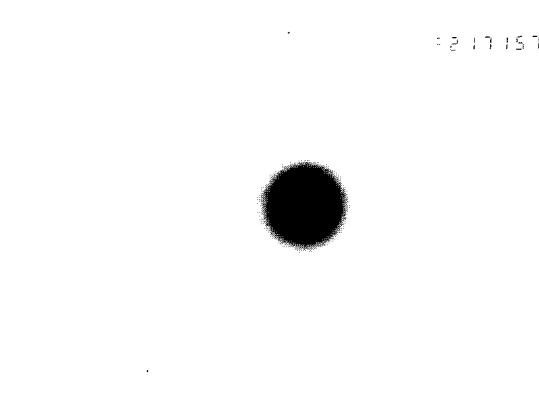


Fig. 9. Electron diffraction pattern of our amorphous particles.

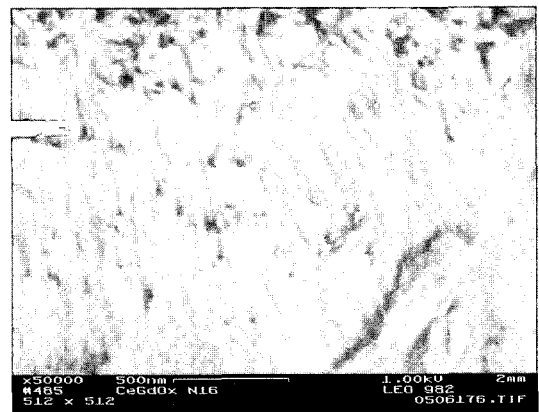


Fig. 10. the CeGdO_x powders SEM photo from JSM T220.

strong agglomerated.

- The characteristic size of the CeGdO_x particles of the powders is about 5 nm (Fig. 10). The par-

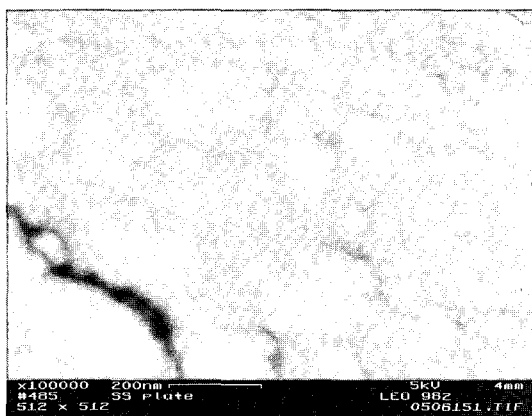


Fig. 11. the stainless steel powders SEM photo from JSM T220.

ticles are sintered in a stick.

- The characteristic size of the stainless steel powders is about 20 nm (Fig. 11).

Next steps of our work are:

- to change the powder collection system to exclude the sintering of particles;
- to use different starting materials.

Thereby, installation for prepare of nanopowders is created and first experiments are performed.

First experiments for nanopowder preparation show that nanopowders are not corresponded all requirements just now.

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