

## ARC PLASMA PROCESS FOR PRODUCING HIGH-QUALITY CERAMIC MATERIALS FROM SOLUTIONS AND MELTS

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### ABSTRACT.

Scientific and technical experience in obtaining disperse oxides by plasma decomposition of liquid raw materials is generalized. Capacity for work of powerful plasma apparatuses and problems of producing pure oxide ceramics are discussed.

### INTRODUCTION

Plasma processing nitric solutions was used for obtaining oxides of U, Cr, Mg and oxide compositions. The process has been developed for U-technology but later used for obtaining other ceramic materials. Technical level of this technology should be analyzed, especially: completeness, power of the reactor, capacity of the apparatus, feasibilities to separate powder and gas, to produce ceramic materials required.

### GENERAL TECHNOLOGICAL SCHEME OF THE PROCESS

The flow diagram of the plasma process is shown in Fig. 1.

Nitric solutions or melts of nitric salts are dispersed by a sprayer into the air plasma. The solution drops mix with plasma and decompose to oxides of dissolved metals, steam,  $\text{NO}_x$ ,  $\text{N}_2$ ,  $\text{O}_2$ . Powders and gases are separated under conditions when the recombination processes are thermodynamically forbidden or kinetically slowed down. Then gas flow is directed into a condenser-absorber where steam and nitric oxides recombine to  $\text{HNO}_3$ .

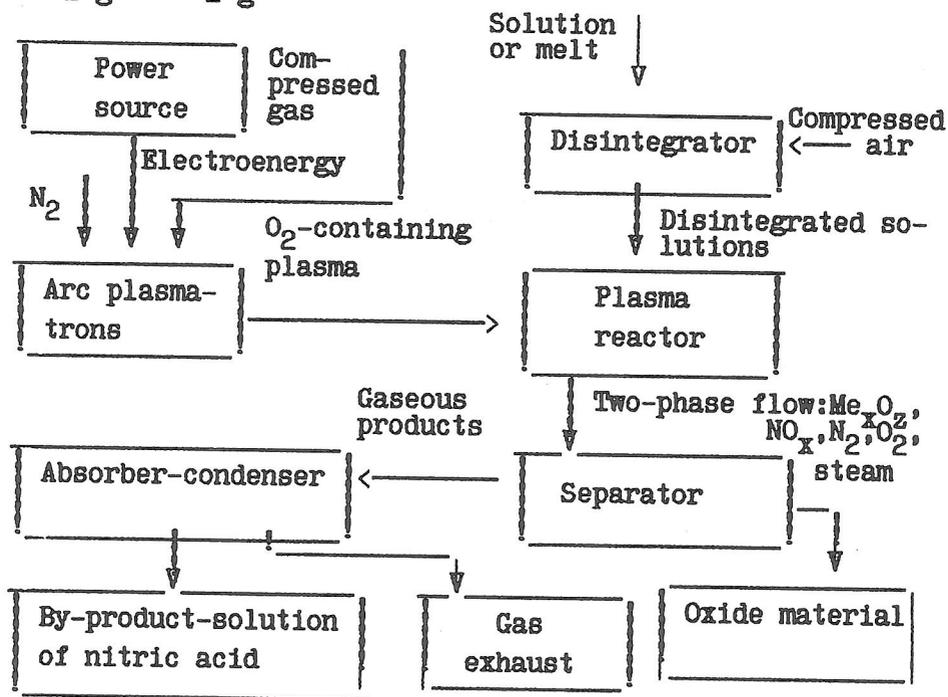
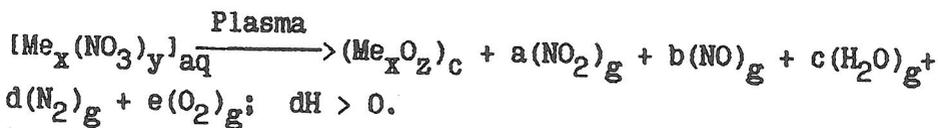


Fig.1. Process flow diagram.

### PLASMA GENERATORS

The process was developed for producing oxides of regenera-

ted U. Power of one apparatus was defined in 4000 kW. We designed the multi-torch reactor to attain necessary power having arc plasma torches of 800-1000 kW.

Power supply consists of two thyristorized rectifiers; each of them operates on two plasma torches. Parameters of the rectifier are:  $V_{\text{rect.}}=1050$  V,  $I_{\text{rect.}}=1050$  A. The rectifier has abruptly falling VI-characteristics providing stable arc burning. Efficiency of the rectifier is 95 %.

The cathode of the 1000 kW plasma torch is made of W; the interelectrode inserts, the anode are made of Cu. The cathode is protected by Ar (0.78 g/s) or pure  $N_2$ . The length of the arc is more than 0.5 m, anode spot is rotated by magnetic coil. Bulk temperature of the plasma is of 3500-4000 K at  $I \sim 950-1000$  A at the air feed of 128 g/s, operating power is of 880-950 kW. Efficiency of the plasma torch at  $I \sim 700$  is equal to 85 %.

## PLASMA REACTOR

The reactor made of stainless steel consists of mixing chamber and reaction tube. The sprayer and plasma torches are installed at the top of the mixing chamber. The reactor has double cooling jackets: internal one is cooled with air, external - with water. Geometry of the reactor for producing U-oxides is correlated with the nuclear safety requirements.

## FEED OF RAW MATERIAL.

We have used three types of sprayers. Centrifugal sprayers has rough dispersion (over 100  $\mu$ ) and uncertain cone of spraying; pneumatic sprayers atomize solutions up to micron size, have controlled cone of spraying but introduce into reactor ballast gas worsening reaction kinetics. But pneumatic-centrifugal sprayer has all the advantages of the previous sprayers and minimizes all their disadvantages.

## SYSTEM OF SEPARATION OF PHASES, PURIFICATION OF OFF-GASES.

System of separation of powder and gas comprises centrifugal separator, metal cloth and metal-ceramic filters, receiver for shock blowing away, screw conveyers etc.

System of purification of gas consists of condensers, absorbers, pumps hydrogate, sanitary filter etc.

## TECHNICAL PARAMETERS OF POWERFUL PLASMA APPARATUS FOR PRODUCTION OF OXIDES OF REGENERATED URANIUM.

1. Output on nitric solutions of U- up to 1500 l/h.
2. Output on U-oxides - up to 700 kg/h.
3. Gas volume leaving the reactor at 873 K up to 12000 m<sup>3</sup>/h.
4. Composition of gases leaving the reactor(vol %): air - 49; steam - 49.3; NO<sub>x</sub> - 1.7;
5. Recombination degree of nitrogen to NO<sub>x</sub> ~50 %.
6. Purification degree of gas at the entry into filter,% -98.
7. Power of the plasma reactor ~4 MW.
8. Efficiency of the plasma torch - up to 0.9.
9. Air supply for the process - 500-1500 nm<sup>3</sup>/h.
10. Cooling water supply - up to 40 m<sup>3</sup>/h.
11. Extraction degree of U into product - not less 99.9 %.
12. Ar supply for protection of the W-cathodes 3-4 nm<sup>3</sup>/h.

## GRANULOMETRY AND MORPHOLOGY OF DISPERSED MATERIALS.

When drops sprayed into a reactor do not crush, there is a simple ratio of diameters of an oxide particle and an initial drop:  $d_{\text{oxide}}/d_{\text{drop}} = (\beta \cdot \rho_{\text{oxide}}/\rho_{\text{drop}})^{1/3}$  where  $\beta$  is a stoichiometric content of oxide in a raw material,  $\rho$ - density. As a matter of fact there is secondary crushing of initial drops. There are two mechanisms of crushing drops: "external" and "internal".

"EXTERNAL MECHANISM" is determined by interaction of drops with turbulent streams of plasma. A stable diameter of a drop is defined by several parameters. One of them is Weber's criterion -  $We = f_1(\rho, w, L, \sigma)$ , defining ratio of inertia forces of gas medium to surface tension where  $w$ -re-

lative velocity,  $L$  -characteristic dimension,  $\sigma$ -surface tension. The next parameter is Laplas's criterion  $L_p = f_2(\mu, \sigma, \rho, L)$  defining ratio of viscosity to surface tension.

At meanings of  $We < (Re)^{0.5}$  occurs deformation mechanism of destruction of a drop. Crushing starts at the critical value of velocity estimated from the relationship  $We_{cr.} = f_3(L_p)$ , while the initial diameter of a drop is accepted as inherent size in both criterions.

At the meanings of  $We > (Re)^{0.5}$  the breaking off mechanism of destruction occurs which consists of blowing off from peripheral regions of a drop ripple wave crests and of inertional throwing down a portion of intradrop bordering layer into the jet. A stable diameter of drop estimated on a critical  $We$ -number is equal to  $\sim 10 \mu$ ; a diameter of a particle is of 3-4  $\mu$ .

"INTERNAL MECHANISM" OF CRUSHING DROPS. Oxide particles produced by plasma processing liquid drops have various morphology: solid or hollow spheres, shapeless fragments etc. Feasibility to obtain solid spheres is determined by properties of raw material and operating modes of the process. According to experience, the next properties of a raw materials are essential:

TEMPERATURE COEFFICIENT OF SALT SOLUBILITY. At processing solutions of salts having positive temperature coefficient of solubility there is high probability to obtain solid particles.

MOLECULAR STRUCTURE OF A PRECURSOR. A probability of solid particle increased when an internal coordination sphere of metal-ion consists of OH-groups or (H-OH)- molecules.

FUSED AND PLASTIC PRECURSOR. One should avoid salts forming fused and plastic intermediate states.

INFLUENCE OF EXPERIMENTAL CONDITIONS ON PARTICLE STRUCTURE. More profitable to rely on plasma conditions which can provide active control over morphology of the particles: all

the hollow spheres and their fragments can be remelted depending on temperature and stay-time in plasma.

#### SOLVING OF THE OXIDES PURITY PROBLEM.

The plasma process aforesaid ensures high purity of product. Cathodes of the plasmotrons used had specific erosion of  $1.5 \cdot 10^{-13}$  kg/Kl and operated without replacement 550-800 hours. The specific erosion of anodes was of  $1.5 \cdot 10^{-10}$  kg/Kl. When production rate of a reactor is of 100 kg/h, the W-content in the product can not exceed  $3,3 \cdot 10^{-6}$  % and Cu not over  $3,3 \cdot 10^{-3}$  %. In fact the yield of W and Cu is 10-20 times less due to the condensation of W and Cu close to electrodes.

#### SOME APPLICATIONS ON INDUSTRIAL AND PILOT SCALE.

URANIUM OXIDES. Industrial lots of U-oxides have been produced and used for manufacturing fuel for nuclear reactors.

(U-Cr-O)-COMPOSITION AS AN EXPERIMENTAL NUCLEAR FUEL. The industrial lots composition  $UCrO_4$  have been produced and used for manufacturing new kind of nuclear fuel.

MAGNESIA FOR COATINGS OF TRANSFORMER STEEL. Magnesia used for this aim had to have bulk density of 0,2-0,4 g/cm<sup>3</sup> and activity determined by a special parameter—a citric number of 70-90 sec. Such material has been produced and used for industrial manufacturing transformer steel.

#### HIGH TEMPERATURE SUPERCONDUCTING OXIDE COMPOSITIONS.

Compositions  $Y_1Ba_2Cu_3O_{7-x}$  and  $Bi_2Sr_2Ca_2Cu_3O_{10}$  were obtained by plasma decomposition of mixed nitric solutions and used for manufacturing various kinds of HTSC-ceramics.