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FORMATION OF GRADIENT METALLOCERAMIC MATERIALS USING ELECTRON-BEAM IRRADIATION IN THE FOREVACUUM

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Results of using an electron beam formed by a forevacuum plasma electron source to sinter metalloceramic materials in powder form are reported. As the materials to be sintered, we used mixtures of titanium powder and an aluminum-oxide or zirconium-oxide based ceramic powder. Sintering was performed using a narrowly focused beam directed onto the surface of the metalloceramic powder. It has been shown that using a mixture of finely dispersed zirconium dioxide or aluminum oxide powder with titanium allows one, by using the electron-beam method in the forevacuum pressure region, to obtain a metalloceramic sample with a titanium concentration gradient over the volume of the sample.

Keywords: electron beam sintering, metalloceramic, gradient ceramic materials.

INTRODUCTION

The current pace of development of technology and increased demands on parts and mechanisms requires a full-scale optimization of existing materials, alternatively the creation of fundamentally new materials, possessing enhanced parameters and combining within themselves the required properties of the indicated materials. The need for such materials is most urgent in the creation of durable, corrosion-resistant coatings for aviation and space technology, nuclear reactors, etc. [1–4]. Often, a metal substrate with a ceramic coating enters as the durable material with enhanced properties. It is well known that abrupt changes in the composition of the material and, consequently, in its physical properties leads to substantial surface and internal stresses [5]. However, if the transition from one material to another is smooth, then the stresses in the bulk of the material due to redistribution between layers of different materials are attenuated significantly. The given concept lies at the basis of functional gradient materials [6], these being comprised of different components, such as ceramic, metal, glass, and so on, with a metallic bond, the contents of which vary continuously over the bulk of the material [7]. The functional properties of such materials vary smoothly or with a jump over the bulk of the material [8–11]. In such cases, functional gradient metalloceramic materials can have both the advantages of a ceramic – high hardness, and of a metal, i.e, high impact strength [12].

At the present time, there exist several methods for fabricating parts from functional gradient materials. Such methods include the high-energy action of energy fluxes – selective laser sintering and electron-beam sintering, and also methods of acting on the bulk of the material, such as microwave heating, spark plasma sintering (SPS), etc. [13–17]. The SPS method and the method of layered laser sintering have gained the most widespread use. There are not many technologies of creating metalloceramic gradient materials using an electron beam. This is possibly due to the necessity of ensuring charge bleed-off from the irradiated ceramic surface. The low electrical conductivity of ceramic powder leads to the accumulation of charge on its surface, which can lead to a lowering of the efficiency of electron-beam action and, as a final result, deflection of the electron beam from the work spot. The solution of this problem can be

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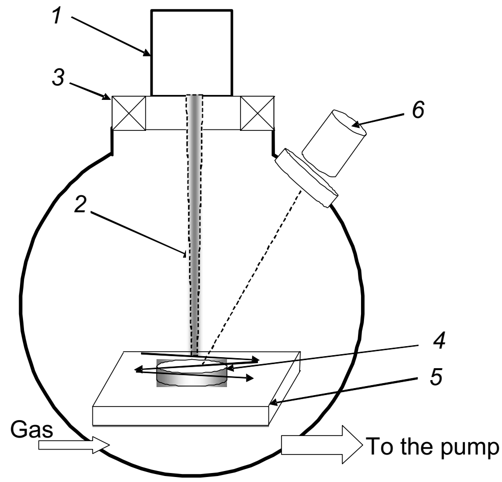

Fig. 1. Schematic depiction of the experimental setup: *1*) plasma source of a focused electron beam, *2*) electron beam, *3*) magnetic focusing and deflection system, *4*) powder mixture that is being sintered, *5*) graphite crucible, and *6*) infrared pyrometer.

facilitated to some degree by use of a forevacuum plasma electron source. The working pressure range of such a source lies in the range 1–100 Pa. As was shown in [18, 19], the secondary plasma formed upon propagation of the electron beam in such a pressure range makes it possible to efficiently remove the charge from the irradiated dielectric surface. The aim of this work was to investigate the possibility of using an electron beam generated by a forevacuum plasma source to obtain a gradient material based on an oxide ceramic and titanium.

EXPERIMENTAL SETUP

To sinter the metalloceramic powder, we used an ÉLU-1 electron-beam setup, equipped with a forevacuum plasma electron source, an electric current source, and a vacuum pump. A schematic depiction of the setup is shown in Fig. 1. The plasma electron source (*1*) consists of a cylindrical hollow stainless-steel cathode, a flat anode, and an extractor [20]. A glow discharge was ignited between the cathode and the anode. Electrons are extracted from the discharge plasma with the help of an accelerating system consisting of an anode with a perforated electrode and an extractor. The perforated electrode in the anode is realized in the form of a removable grid, fabricated from a tantalum plate 1 mm in thickness with an emission opening 0.8 mm in diameter.

The electron source formed narrowly focused beam *2* with energy in the range 2–18 keV and current in the range 15–45 mA. Focusing and deflection of the electron beam were realized by magnetic coils located directly behind the electron source extractor. In the process of irradiation, we used a raster scan of the electron beam over a square area with dimensions exceeding those of the sample being sintered (*4*). The assigned scan frequency determined the rate of passage of the beam over the entire irradiated area, and the sweep scale determined the size of the given area. It is worth mentioning that the use of a scanning beam is significantly more effective than using a wide beam with cross section commensurate with the area of the sample. The point here is that the beam power density has a Gaussian distribution, which does not allow uniform heating of the entire surface of the sample.

In the course of this work, it was necessary to take into account the thermal properties of the material that the samples are fabricated from and the fact that high temperatures do not allow the use of a crucible of just any material. In the experiments, we used graphite crucible *5*. To reduce the loss of heat, the crucible was positioned on thin holders made from tantalum wire 1 mm in diameter. This structure was covered by a screen acting as a thermal shield in order to reduce radiative heat loss.

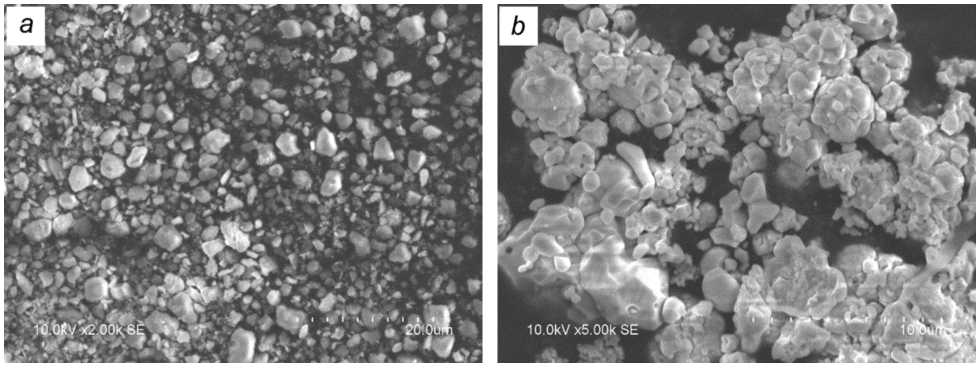

Fig. 2. Images of the aluminum oxide powder (*a*) and zirconium oxide powder (*b*) used in the sintering.

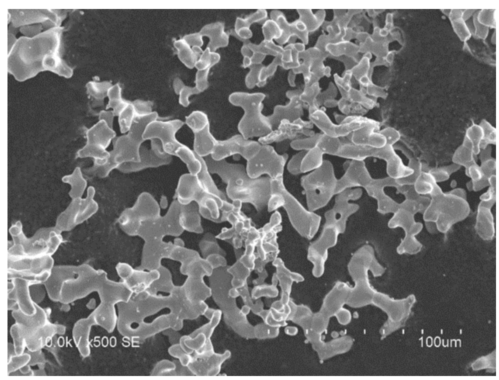

Fig. 3. Image of the titanium powder.

In preparation for sintering, the vacuum chamber was pumped down to a pressure of 2–4 Pa and then perfused with helium for 5 minutes. Sintering was performed in an inert-gas medium (helium) at a pressure of 30 Pa. The process of electron-beam irradiation of the crucible and powder lasted 40–60 min, depending on the temperature, before which it was necessary to heat the material. The heating rate was 30 deg/min. The sample was kept at a constant temperature for 10 min, after which it was cooled by gradually lowering the beam power. The cooling rate was 50 deg/min. The sintering time was chosen on the basis of our experience with sintering pressed tablets of aluminum oxide and zirconium oxide [21–23].

MATERIALS AND METHODS OF ANALYSIS

As the basis for the fabrication of a gradient metalloceramic, we used aluminum oxide (Al_2O_3) and titanium (Ti) powders, these being the most widely used materials for additive production of ceramic parts. We used two types of ceramic powders – one based on aluminum oxide, and the other, based on zirconium oxide. The ceramic powders contained particles with dimensions of 1–10 μm (Fig. 2). The particles of the titanium powder had an irregular shape and a developed surface (Fig. 3), which made it possible to form blanks using comparatively low pressing pressures.

Two variants of the powder mixtures containing a different mass fraction of ceramic and metal were prepared for sintering: 1) aluminum oxide 80 mass% and titanium 20 mass%; 2) zirconium dioxide 80 mass% and titanium 20 mass%. The powder mixtures were placed in a press form and pressed at a pressure of 5 MPa. The pressing pressure was chosen to be the minimum necessary so that the pressed samples would not fall apart under manual manipulation.

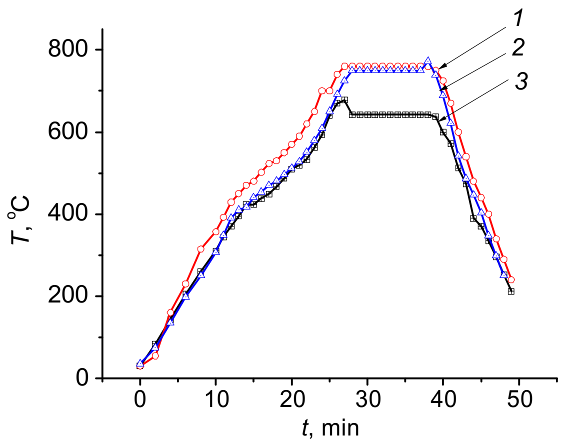

Fig. 4. Dependence of temperature on irradiation time for three different powder mixtures: *1*) aluminum oxide 80 mass% and titanium 20 mass%, *2*) zirconium dioxide 80 mass% and titanium 20 mass%, and *3*) titanium powder.

The microstructure of the surface of a transverse thin section and the elemental composition of the sintered samples were examined on a Hitachi S3400N scanning electron microscope, equipped with a Bruker X'Flash 5010 attachment. The temperature of the irradiated surface was recorded using infrared pyrometer *6* (Fig. 1) (RAYTEK 1MH) with temperature measurement range 550–3000°C.

Figure 4 shows the dependence of the surface temperature of samples prepared from powders of various composition on the time of electron-beam irradiation during sintering. The heating curves of the ceramic/metal mixtures differ insignificantly. The difference consists only in the temperature up to which it is possible to heat the powdered material for the same power level of the electron beam (the saturation segment in Fig. 4 occurs at the irradiation time 30–40 min). Thus, for the titanium powder without admixture of ceramic the temperature that can be reached is lower, which has to do with the difference in the thermophysical properties of the materials. Due to its high thermal conductivity, titanium transfers heat obtained from the electron beam to the holder and heats up less.

EXPERIMENTAL RESULTS AND ANALYSIS

As a result of sintering of mixtures of titanium with aluminum-oxide and zirconium-oxide ceramic, we obtained samples in the form of disks. The parameters of the sintered samples are given in Table 1. It is remarkable that in the course of irradiation, we did not detect any deflection of the electron beam from the surface of the irradiated target, which testifies to the possibility of using electron-beam irradiation of dielectric powders in the forevacuum pressure region.

After sintering, the overall dimensions of all the samples decreased with a corresponding increase in the density of the material. Shrinkage of the sintered sample is an indirect confirmation of occurrence of the process. A study of images of transverse sections of sintered samples obtained using an electron microscope with a micro-analyzer is shown in Fig. 5. The magnification for the samples of Ti (20 mass%) + Al_2O_3 (80 mass%) was 45. The magnification for the samples of Ti (20 mass%) + ZrO_2 (80 mass%) was 60. As the photomicrographs reveal, the sample containing zirconium oxide is more uniform in comparison with the sample containing aluminum oxide. The central region of the aluminum-oxide sample was subjected to more shrinkage, which appears to be the result of the lower sintering temperature for this type of ceramic and also the difference in the degree of shrinkage upon heating of these types of ceramic powders.

Both sintered samples have a porous structure. The elemental distribution with depth of the sintered samples shows that the oxygen, zirconium, and aluminum content for samples with the corresponding composition is quite

TABLE 1. Parameters of Metalloceramic Samples Before and After Electron-Beam Sintering

Sample	Sintering	Mass m, g	Diameter d, mm	Height h, mm	Density, g/cm^3
Ti(20%) + ZrO_2(80%)	Before	0.500	10.23	2.02	3.24
	After	0.439	8.22	1.47	5.33
Ti(20%) + Al_2O_3(80%) (fine)	Before	0.500	10.25	3.52	1.63
	After	0.427	8.6	2.76	2.52

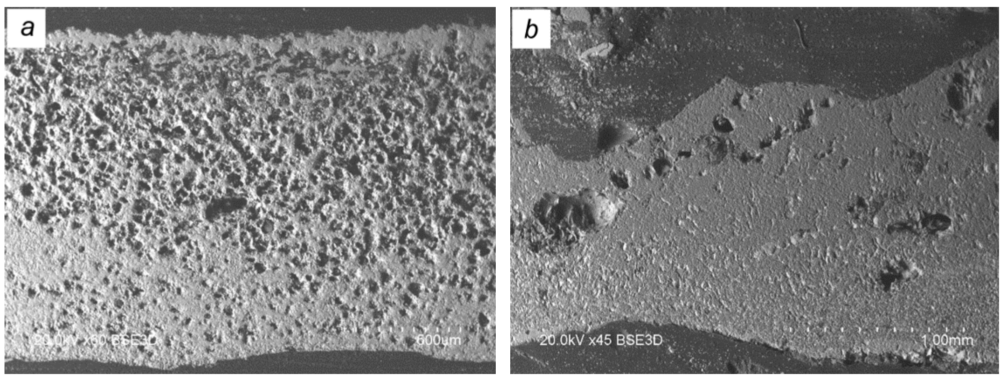

Fig. 5. Photomicrographs of sections of sintered samples: *a*) Ti (20%) + ZrO_2 (80%), *b*) Ti (20%) + Al_2O_3 (80%).

uniform (Fig. 6). The titanium content, at the same time, varies with depth (Figs. 6*e* and *f*). Its highest content is observed in the lower part of the sintered samples.

The variation of titanium concentration with depth of the samples allows one to speak of the possibility of using an electron beam generated by a forevacuum plasma electron source to create gradient metalloceramic materials. The technology of obtaining multilayer metalloceramic parts requires further improvement. However, already at this stage it is possible to conclude that the electron-beam method can be used to fabricate gradient materials in the forevacuum pressure region.

CONCLUSIONS

In this paper we have presented results of using electron-beam irradiation of mixtures of metalloceramic powders in the forevacuum pressure region in a helium medium. We have shown that as a result of heating of a metalloceramic powder consisting of finely dispersed aluminum oxide with titanium or of zirconium dioxide with titanium, a gradient material is formed having varying titanium content with depth. The results presented here testify to the fundamental possibility of applying a forevacuum plasma electron source to create materials with composition varying over the volume of the sample.

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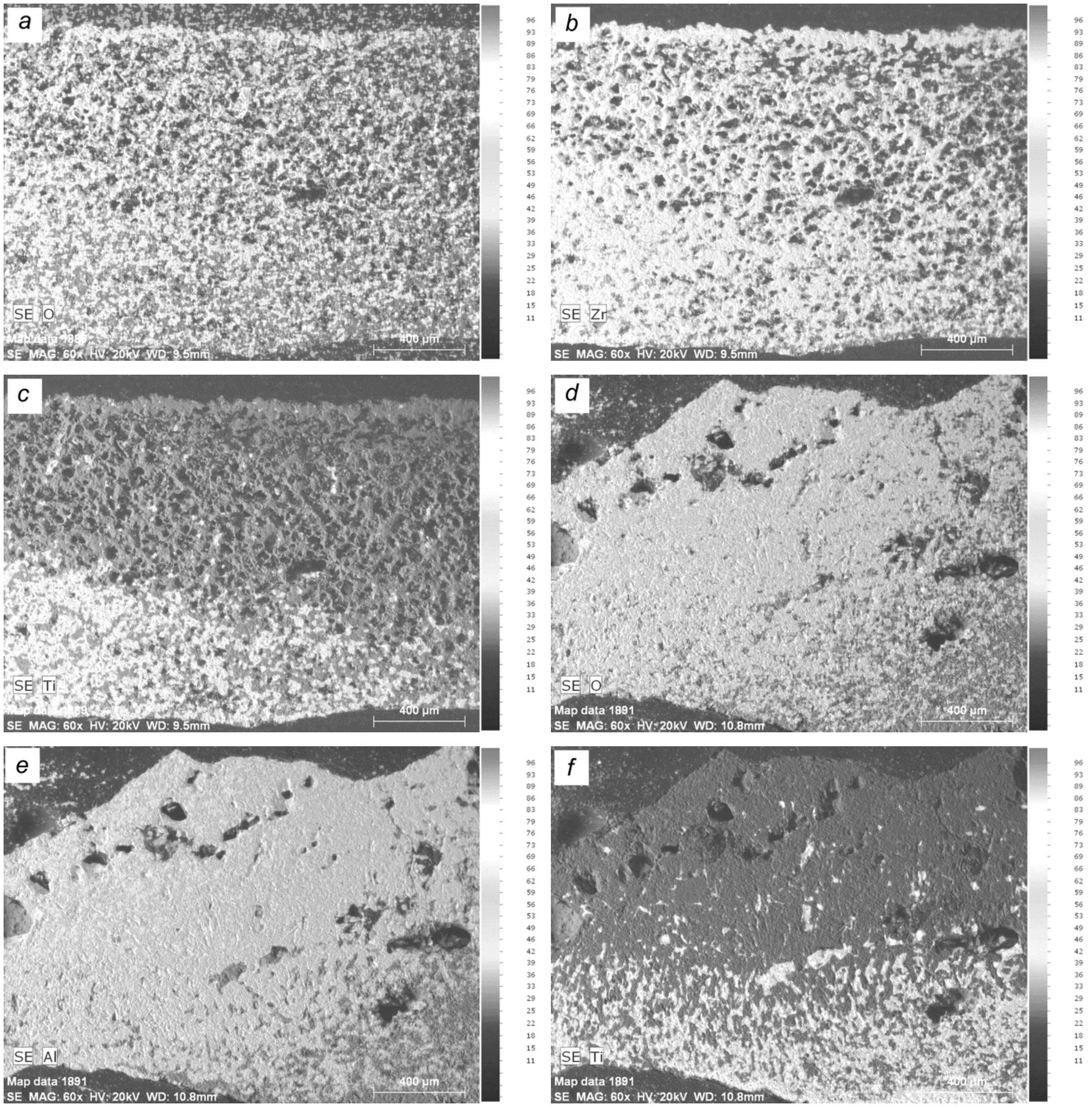

Fig. 6. Elemental distribution with depth of the sintered samples: *a*–*c* are for Ti (20%) + ZrO_2 (80%), and *d*–*f* are for Ti (20%) + Al_2O_3 (80%). Elemental content: *a* and *d* – oxygen, *b* – zirconium, *c* and *f* – titanium, and *e* – aluminum.

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