

Electron beam sintering of gradient $\text{Al}_2\text{O}_3\text{-ZrO}_2$ ceramics with the forevacuum plasma electron source

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Abstract. The paper presents electron beam sintering of functionally gradient (FGM) $\text{Al}_2\text{O}_3/\text{ZrO}_2$ materials. An electron beam generated by a pre-vacuum plasma electron source for sintering was used. FGM were manufactured with different number of layers of aluminum oxide and zirconium dioxide powders. The layers differed in the percentage of components by volume. It is shown that the sintering of the composite sample occurs during electron-beam heating. After sintering for 10 min at 1600°C , the sample density increases up to 4 g/cm^3 . The presented photos of cross-sections of the samples indicate low pore content after sintering.

1. Introduction

Functional gradient materials (FGM) are characterized by a nonlinear distribution of composition and properties (hardness, density, thermal conductivity, modulus of elasticity) by volume, which distinguishes them from traditional materials [1, 2]. The need for FGM is related to their ability to operate under the thermal stress created by the temperature difference between the sample sides [3]. Gradient ceramics, and in particular the system based on $\text{Al}_2\text{O}_3\text{-ZrO}_2$, currently displaces a number of metals and alloys [4]. Zirconium and aluminum ceramics play an important role in medicine, namely in joint prosthetics, implantology and other very important branches of medicine [5, 6]. Alumina ceramics has a high hardness and wear resistance among structural materials, the disadvantage of this material is its high fragility. Zirconium dioxide ceramic combines high strength and crack resistance. FGM based on aluminum and zirconium oxides are used in medicine for implants that combine in one material the high strength of inert ceramics with biocompatibility of bioactive ceramics and its ability to participate in metabolic processes of the body. An important factor is the lack of immune response of the body to ZrO_2 , as well as Al_2O_3 [7]. Despite the success in this direction when using FM at high temperatures (coating, individual units and assemblies) there are characteristic problems of interfacial reaction, which largely determine the reliability and durability of these materials.

Currently various methods of obtaining FGM based on ZrO_2 and Al_2O_3 have been developed: dry pressing followed by heat treatment [8], diffusion welding [9], electrophoretic deposition [10], tape-cast alumina-zirconia laminates [11], slip casting [12], printing with “ceramic inks” [13]. The choice of FGM creation method depends on the required level of properties for a particular practical application.

There is no universal way to obtain FGM based on ZrO_2 and Al_2O_3 with any given composition and gradient profile properties and, therefore, it is necessary to use a set of different methods. In work [14] successes in use of a forevacuum plasma electron source for creation of FGM of metal-ceramic materials on the basis of aluminum oxide and titanium are presented. A narrowly focused electron beam propagating in vacuum at pressures from units to tens of Pascal [15] is capable of rapidly heating the



workpiece surface to the sintering temperature (about 1600°C). The plasma formed during the beam propagation in the gas neutralizes the negative charge of the surface irradiated by the electron beam [16]. For vacuum plasma electron sources are successfully used for sintering of ceramics, electron beam welding of metal-ceramic units, processing of quartz glass and other technologies of processing of dielectrics [17, 18].

The purpose of this study is to carry out electron-beam sintering of a gradient material based on aluminum oxide and zirconium using a forevacuum plasma electron source.

2. Materials and methods

The experiments were carried out using a plasma electron source, a schematic image of which is shown in Figure 1. The design and basic parameters of the electron source are presented in [19]. In the forevacuum pressure range, the electron source generated a beam with a diameter of 0.5 mm with a current of 50 mA and electron energy of 20 kV. Focusing of the electron beam was carried out by means of a magnetic focusing system, and deflection of the beam to the processed sample – by means of a magnetic deflecting system. The plasma source was placed on the flange of the vacuum chamber. All the experiments were in the medium of helium. The choice of helium is due to its chemical inertness.

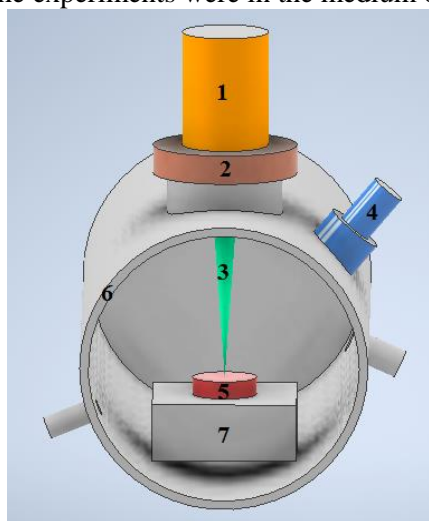


Figure 1. Scheme of the experimental setup: 1 – plasma source of focused electron beam, 2 – magnetic focusing and deflecting coils, 3 – electron beam, 4 – infrared pyrometer, 5 – graphite crucible and sintered sample, 6 – vacuum chamber, 7 – collector.

The sintering process was carried out as follows: the sample 5 was placed in a working vacuum chamber on a graphite crucible. The crucible was located at a distance of 45 cm from the electronic source. The working chamber was pumped to a pressure of 3 Pa, after which the chamber was filled with helium to a pressure of 30 Pa. The sample was heated for 20 minutes. During heating, the power of the electron beam was increased from 20 to 400 W with a heating rate of 80 deg/min. When the temperature of the treated surface reached 1600°C the heating stopped and the sample was kept at a constant temperature for 10 minutes. The surface temperature of the sintered sample was measured using a pyrometer 4. After exposure, the power of the electron beam gradually decreased to 20 watts for 10 minutes. The cooling rate of the sample was 100 deg/min. After switching off the plasma electron source, the sample was cooled in vacuum for 10-15 minutes.

Samples of gradient ceramics in the form of discs were made for sintering. Samples were made from fine-grained powders of aluminum oxide and zirconium dioxide. Cold static pressing in a closed mold was used [20]. This method of pressing is relatively simple in terms of technological implementation and universal for pressing products of different shapes from powders of any composition, allowing to implement a multi-place scheme of pressing in production conditions. The composition of the samples varied discretely in volume. Sample 1 consisted of three layers – the first layer is aluminum oxide, the second layer is a mixture of powders of aluminum oxide 50% and zirconium dioxide 50% (wt), the third layer is zirconium dioxide. Sample 2 consisted of six layers. The first layer – aluminum oxide, layers 2-

5 consisted of a mixture of powders of aluminum oxide and zirconium dioxide, 6th layer consisted of zirconium dioxide (table. 1). The aluminum oxide content in the second sample varied by 20% from layer to layer. The third sample contained 11 layers and the content of aluminum oxide in each layer varied by 10% (table 1). After sintering, the elemental composition and microstructure of the samples were studied by scanning electron microscopy using a Hitachi s-3400N microscope.

Table 1. Composition of sintered samples.

No	Sample 1		Sample 2		Sample 3	
	Al ₂ O ₃ , mass. %	ZrO ₂ , mass. %	Al ₂ O ₃ , mass. %	ZrO ₂ , mass. %	Al ₂ O ₃ , mass. %	ZrO ₂ , mass. %
1	100	0	100	0	100	0
2	50	50	80	20	90	10
3	0	100	60	40	80	20
4			40	60	70	30
5			20	80	60	40
6			0	100	50	50
7					40	60
8					30	70
9					20	80
10					10	90
11					0	100

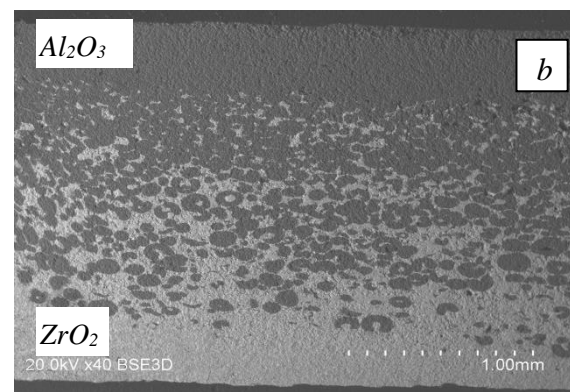
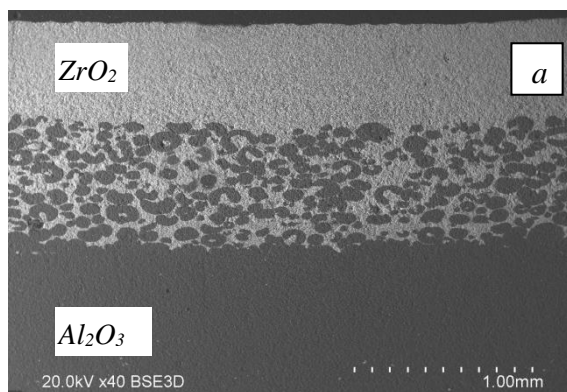
3. Experimental results and discussion

As a result of electron beam sintering, samples were obtained, the parameters of which before and after sintering are given in table 2. As can be seen from table 1, the volume of all samples after sintering decreased and the density increased. The highest density of 4 g/cm³ was found in the Sample 2 consisting of six layers. The low density value after sintering in Sample 3 may be due to insufficient heating.

Table 2. Sample parameters before and after sintering.

	Sample 1		Sample 2		Sample 3	
	before	after	before	after	before	after
<i>m</i> , g	0.541	0.526	0.569	0.549	0.805	0.779
<i>d</i> , mm	10.19	9.8	10.14	8.630	10.22	9.89
<i>h</i> , mm	2.43	2.25	2.500	2.300	3.56	3.44
ρ , g/cm ³	2.731	3.442	2.819	4.083	2.758	3.265

In studies it was found that ceramic tablets under the influence of unilateral irradiation are deformed (become concave) due to more intense shrinkage on the irradiated side.



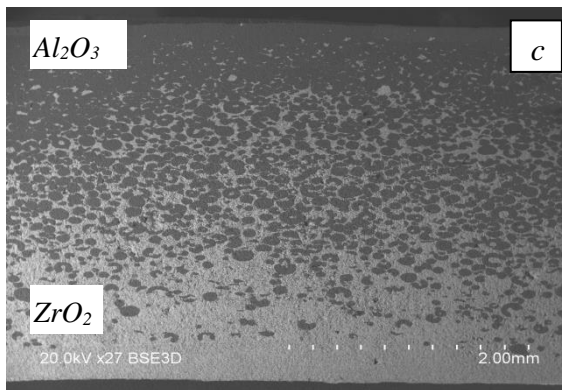


Figure 2. A photo of a section of sintered samples with different number of layers: a – 3, b – 6, c – 11.

As can be seen from figure 2, Sample 1 has clearly defined layer boundaries. Irradiation of the sample was carried out from the side of zirconium dioxide. As the number of layers increases, the distribution of elements becomes smoother, allowing the creation of multi-layered materials with a gradient structure. The presence of pores and cracks on sintered samples is obviously associated with inhomogeneous heating of the sample and requires the selection of sintering modes. The heterogeneity of the material distribution near the side surfaces is obviously related to the method of pressing the sample. As is known, the friction of the pressed material on the walls of the mold leads to anisotropy and uneven properties of the product in different directions, which can be a source of macrodefects in sintered products [21].

Figure 3 shows the depth distribution of the elements for a six layer sample.

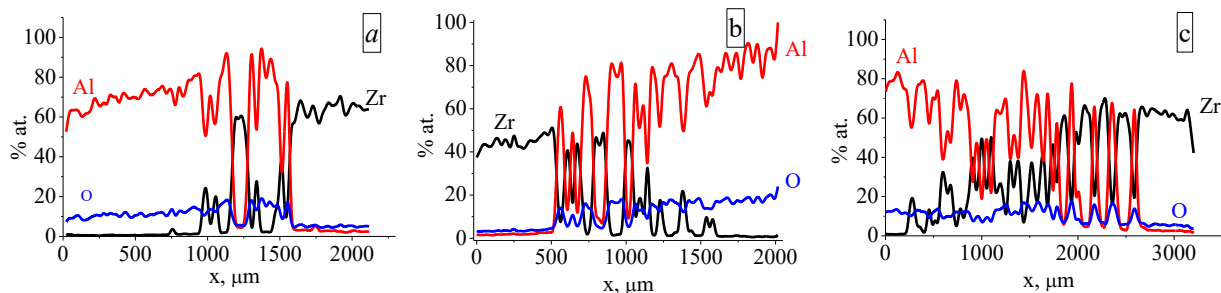


Figure 3. Distribution of elemental composition in depth of samples (a – Sample 1, b – Sample 2, c – Sample 3).

As expected, the Sample 1 with three layers is characterized by a sharp change in concentration with clearly defined boundaries on the cut. The Sample 2 is characterized by a smooth change in concentration, which may tell us about a stronger bond. In the future, it is planned to conduct a process of thermal cycling of samples and test the strength of the bond between the layers.

4. Conclusion

As a result of the research, samples of gradient ceramics were obtained. The density of the obtained samples increased by an average of 30%, which is associated with shrinkage by reducing the pore size during sintering. The distribution of elements in the sample volume showed that with the growth of the number of layers, the distribution of elements becomes smoother. The samples are durable and do not break down during manual manipulation. Electron beam sintering of multilayer gradient samples from Al_2O_3 - ZrO_2 ceramics allows obtaining dense sintered materials with a non-uniform distribution of the

concentration of elements that can be used in the manufacture of prostheses and other branches of medicine.

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