Features of electron-beam processing of metal-ceramic powders in the forevacuum

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Abstract. We present the results of sintering of a metal-ceramic sample consisting of alumina ceramics and titanium. Sintering has been conducted via the surface scanning by the electron beam generated by a plasma electron source. It has been shown that the bake-out temperature affects the formation of the layer with a uniform distribution of elements. We have determined the sintering regimes that enable fabrication of samples with uniformly distributed elements over volume.

1. Introduction

Modern industrial trends pose problems for researchers to develop materials with improved tribological properties. Because of the heavy duty conditions these materials have to operate, they must meet a number of often contradicting requirements, for example be resistant to heat impacts, complex mechanical stress, chemical corrosion and erosion. It is often required to combine incompatible properties in the same material, such as high hardness at high temperature and mechanical strength at low temperature. One of the ways to satisfy those requirements is fabrication of functionally graded materials (FGM) [1] with properties gradually varying from layer to layer. As a rule, the FGMs are fabricated using a combination of various metals, or metals and ceramics.

Most FGM production technologies are based on the combination of traditional well-reputed treatment techniques, such as powder metallurgy [2–4], laminated object manufacturing [5] and stereolithography [6], chemical vapour deposition and various coating techniques: centrifugal casting [7–8], slip casting, tape casting [9], and thermal spraying [10–11]. The choice of the production technology is mostly dictated by the required combination of materials, properties, and geometry of the finished product. Powder metallurgy (PM) is one of the most widely used processing methods because of its control over the wide range of the component composition and microstructure, and its ability to form a close-to-net shape structure. This method is characterized by low time and energy expenditures, as well as by variety and availability of the source powder materials. In the powder technology, the required gradient is usually achieved by mixing various powders in different proportions and arranging the obtained mixes in separate layers. Sintering is performed in an electrical furnace by high-frequency inductive heating or using the spark plasma sintering (SPS) technique [12]. Despite the variety of the available methods of sintering, the search for new ones for FGM applications is still of topical interest. The electron-beam treatment of metal-ceramic materials in the forevacuum range of pressure may turn out to be a promising approach. The so-called forevacuum plasma electron sources can be used for the electron-beam sintering of ceramic [13–14] and metal-
ceramic materials [15]. As shown in [13, 16], the electron-beam sintering of ceramic by a forevacuum plasma electron source enables one to fabricate a dense material in a shorter time than the adopted techniques.

The purpose of the present work is to study the effect of the electron-beam irradiation regimes on sintering of a multi-layer aluminum and titanium powder mixture.

2. Experimental setup
The experimental setup includes the vacuum chamber equipped with a gas evacuation system, necessary diagnostic equipment, and a forevacuum plasma electron source (figure 1). Electron source 1 was mounted on the upper flange of the vacuum chamber 2. The electron source generated a continuous cylindrical electron beam 3 with a diameter less than 1 mm, the current varied from 15 to 50 mA by changing the hollow cathode discharge current. The electron beam energy varied from 1 to 20 keV. The source design and principal parameters are discussed in more details in [17].

![Figure 1. Experimental setup. 1 – plasma source of a focused electron beam, 2 – vacuum chamber, 3 – electron beam, 4 – magnetic focusing and deflecting coils, 5 – infrared pyrometer, 6 – sintered sample, 7 – crucible, 8 – thermal shield.](image)

Magnetic coils 4, located immediately behind the source extractor, were used for the beam focusing and deflection. The sintering temperature was monitored by infrared pyrometer 5 (RAYTEK 1MH). The pyrometer can measure temperature in real time in the range 550–3000°C with a step 0.1°C. During the irradiation process, the electron beam raster scanned over the square area greater that the dimensions of the sintered sample. The scanning frequency set the speed of passing through the irradiated area and the sweeping coefficient set the area size. Sintered sample 6 was placed in the crucible located on the electron beam axis in the vacuum chamber. Since the ceramic sintering temperature is over 1200°C, the crucible must be made of heat-resistant material. So we selected graphite because of its high evaporation temperature and ease of working. To decrease the heat losses, crucible 7 was strung up from two tungsten wire holders (not shown in figure). The entire assembly part was covered by thermally shield 8 to diminish the heat radiative losses.

The mixture of ceramic powders was irradiated by the electron beam in an inert gas atmosphere (helium) at a pressure 30 Pa. The heating rate was 30 deg-min⁻¹. The sample was exposed to a constant temperature for 10 minutes, following which it was cooling down by gradually diminishing the beam power. The cooldown rate was 50 deg-min⁻¹. The isothermal exposition time was selected based on our experience of sintering alumina and zirconium compacts [13, 16, 18].
To fabricate metal-ceramics, we selected the powders of alumina with an average particle size 10 µm and titanium with a particle size less than 60 µm. The choice of this particular combination of materials was dictated by their wide use in fabrication of functional graded materials. The alumina and titanium powders were being mixed in a mixer for 30 minutes. The resulting mixture contained 20 mass % of titanium and 80 mass % of alumina. The powder mixtures were placed in a mold and subjected to a pressure of 5 MPa. The selected pressure was minimally required so that the squeezed samples did not fall apart when manually handled. The sample height and diameter were respectively 10 mm and 3 mm.

A SEM Hitachi S3400N coupled with a Bruker XFlash 5010 SDD detector was used to analyze the polished section microstructure and the elemental composition of the sintered samples.

3. Experimental results and discussion

The experiments have demonstrated that the heating of powder mixtures under a gradual increase of the beam power occurs in a non-uniform fashion. Figure 2a shows the dependence of the sample surface temperature during sintering by the electron-beam irradiation. Since the pyrometer measurement range begins at 550 °C, the initial stage of baking is not reflected in figure 2a. During the heating, the electron beam power was gradually changing: increasing each minute by 15 W, thereby the sample temperature also was gradually increasing.

![Figure 2](image_url)

**Figure 2.** Dependence of the sample surface temperature on the sintering time as well as the time dependence of its thermal radiation power at \( T = 1400 \, ^\circ\text{C} \).

In figure 2a, one can distinguish four intervals of the temperature vs. time dependence, which have different slopes. The first and the second intervals correspond to the sample heating. The difference of the tangent angles is due to an increased radiative losses from the sample surface. For example, at 1200 °C, the percentage of the power radiated from the sample surface is over 10% and increases with increasing temperature. Figure 2b shows a typical dependence of the power versus time when the sample is heated to 1400 °C (curve 1). In the same figure, curve 3 gives the dependence of the power of thermal radiation and curve 2 plots the difference between the two powers. The thermal radiation power was determined from the Stephan-Boltzmann formula. For the surface emitting radiation, we took the total area of the sample and crucible. The emissivity was assumed to be 0.8 and the temperature was the experimental value.

As seen from the presented dependences, as early as 10 minutes after the start of the irradiation, the heat loss due to the thermal radiation transfer begins to affect the process of the sample heating.

The required sintering temperatures were reached by increasing the accelerating voltage. The electron beam current was kept constant in all experiments. The average density of samples before sintering was 1.75 g·cm⁻³. The sample density after sintering depended on the temperature of isothermal exposition and grew as this temperature increased (table 1).
Table 1. Sample parameters before and after sintering

|   | T (°C) | sintering | m (g) | d (mm) | h (mm) | ρ (g·cm⁻³) |
|---|--------|-----------|-------|--------|--------|------------|
| 1 | 1400   | Before    | 0.478 | 10.31  | 3.24   | 1.768      |
|   |        | After     | 0.463 | 9.95   | 3.24   | 1.861      |
| 2 | 1500   | Before    | 0.47  | 0.32   | 3.18   | 1.768      |
|   |        | After     | 0.451 | 9.45   | 3.05   | 2.109      |
| 3 | 1600   | Before    | 0.474 | 10.3   | 3.27   | 1.741      |
|   |        | After     | 0.454 | 9.08   | 3.05   | 2.3        |
| 4 | 1700   | Before    | 0.478 | 10.3   | 3.15   | 1.8        |
|   |        | After     | 0.451 | 9.05   | 2.92   | 2.345      |
| 5 | 1900   | Before    | 0.5   | 10.35  | 3.52   | 1.689      |
|   |        | After     | 0.427 | 8.6    | 2.76   | 2.665      |

Geometrically, the sample grows smaller as the isothermal exposition temperature grows (figure 3), which testifies that the sintering process has actually occurred. It should be noted that the surfaces of all samples after the electron-beam treatment become darker as compared with the original color.

Figure 3. Sample pictures: a) – before sintering; b–f) – after sintering, 1400 °C (b), 1500 °C (c), 1600 °C (d), 1700 °C (e), 1900 °C (f).

Such color change can be explained by the change of the elemental distribution over the volume of the irradiated sample. Figure 4 shows the elemental distribution map over the sample section near its boarder. The samples were irradiated from above.
Elemental analysis has shown that aluminum and oxygen, in contrast to titanium, are distributed uniformly over the sample volume (figure 4a). On the other hand, titanium turned out to be uniformly distributed over volume, while forming a thin titanium layer (about 10 µm) on the surface (figure 4b). The observed effect can be explained that the melted titanium made its way to the surface on account of capillary forces. This formation of the titanium layer may also explain the change of color.

![Figure 4.](image)

Thus, the experiments have shown that the most optimal temperature of isothermal exposition in terms of obtaining a dense material is its heating to 1700 °C. In the process, the density of the sintered sample reaches 2.35 g⋅cm⁻³. At higher temperatures, the sample surface layers are observed to melt (figure 3f). The formation of the titanium surface layer after sintering begs for further investigations.

4. Conclusion
The electron-beam sintering of metal-ceramic mixtures using a forevacuum plasma electron source enables fabrication of metal-ceramic materials. Thus, sintering of metal-ceramic containing fine powders of Al₂O₃ (80 mass %) and Ti (20 mass %) produces a layer of the material with a uniform distributions of alumina and oxygen over depth and the layer density 2.35 g⋅cm⁻³. Increasing the isothermal exposition temperature increases the density of the sintered sample. The optimal temperature for sintering that still occurs without melting is 1700 °C.

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References
[1] Besisa D H A and Ewais E M M 2016 Advances in Functionally Graded Ceramics – Processing, Sintering Properties and Applications. Advances in Functionally Graded Materials and Structures (London: IntechOpen Ltd.)
[2] Moustafa S F, Abdel-Hamid Z and Abd-Elhay A M 2002 Materials Letters 53 244.
[3] Shahrjerdi A, Mustapha F, Bayat M, Sapuan S M and Majid D L A 2011 Int. J. Phys. Sci. 6 2258
[4] Jin X, Wu L, Guo L, Yu H and Sun Y 2009 Eng. Fracture Mechanics 76 1800
[5] El-Wazery M S and El-Desouky A R 2015 Journal of Materials and Environmental Science 6 1369
[6] Gonzalez P, Schwarzer E, Scheithauer U, Kooijmans N and Moritz T 2019 J. Vis. Exp. 143 e57943
[7] Watanabe Y, Yamanaka N and Fukui Y 1998 Composites Part A: Applied Science and Manufacturing 29 595
[8] Watanabe Y, Murase M, Sato H and Tsukamoto H. 2018 Materials Science Forum 941 1978
[9] Yeo J G; Jung Y G; Choi S C 1998 J Eur Ceram Soc. 18 1281
[10] Belmonte M, Gonzalez-Julian J, Miranzo P and Osendi M I 2009 Acta. Mater. 57 2607
[11] Jamaludin S N S, Mustapha F, Nuruzzaman D M and Basri S N 2013 Scientific Research and Essays 8 828
[12] Watari F, Kondo H, Matsuo S, Miyao R, Yokoyama A, Omori M and Kawasaki T 2003 Materials science forum 423 321
[13] Kazakov A V, Klimov A S and Zenin A A. 2012 Doklady Tomskogo gosudarstvennogo universiteta sistem upravleniya i radioelektroniki 2-2 186 (in Russian)
[14] Klimov A S, Burdovitsin V A, Zenin A A, Oks E M, Khasanov O L, Dvilis E S and Khasanov A O. 2015 Technical Physics Letters 41 747
[15] Goreev A K, Burdovitsin V A, Klimov A S and Oks E M 2012 Inorganic Materials: Applied Research 3 446
[16] Burdovitsin V, Zenin A, Klimov A, Oks E, Dvilis E S, Sokolov V, Kachaev A A and Khasanov O L 2014 Advanced Materials Research 872 150
[17] Klimov A, Bakeev I, Oks E and Zenin A 2019 Laser and Particle Beams 37 203
[18] Dvilis E S, Burdovicin V A, Hasanov A O, Oks E M, Klimov A S, Zenin A A and Hasano O L 2016 Fundamental'nye issledovaniya 10 270 (in Russian)