Influence of size of metal-ceramic composites on their electron-beam sintering in the forevacuum pressure range

A S Klimov, I Yu Bakeev, V T Tran and A A Zenin

Physics Department, Tomsk state university of control systems and radioelectronics
634050, Tomsk, Russia

Abstract. Ceramic-metal composite materials combine high strength of ceramic and plasticity of metal. The combination of these properties allows the use of cermet in many industries and medicine. Cermet materials are made using powder technology by sintering a compact consisting of a mixture of pressed powders. The properties of the resulting product are determined by both the initial component composition and the sintering method. In the present work, electron beam sintering in the fore-vacuum pressure range was used to obtain cermet. The feature of this method is the possibility of flexible control of the sintering process by changing the parameters of the electron beam. To heat compacts consisting of pressed powders of alumina ceramics and titanium, the surface of compacts was scanned with an electron beam with the diameter of less than 1 mm and the power of 500 W. The surface temperature of the compact during sintering was 1600 °C, and the time of isothermal exposure was 10 minutes. It has been shown that compact thickness has a significant effect on obtaining homogeneous sintered material. Samples with a microhardness of 27 GPa were obtained. The ways of further improving the technology of electron beam sintering of cermet are determined.

1. Introduction

The creation of a new class of materials called composite is a significant achievement in materials science. Such materials consist of two or more components or phases and combine properties that are unattainable for a homogeneous material due to their natural properties. The most widely composites used in technology are cermet composite materials [1]. Ceramics itself is a fairly solid material and has found application in industry. At temperatures above 1000 °C, ceramics are stronger than any alloys, including superalloys, and its creep resistance and heat resistance are higher. The principal disadvantages of ceramics are its fragility and processing complexity. Ceramic materials do not work well under mechanical or thermal shock, as well as under cyclic loading conditions [2]. They are characterized by high sensitivity to incisions. At the same time, ceramic materials have high heat resistance, excellent corrosion resistance and low thermal conductivity, which allows them to be successfully used as thermal protection elements [3]. Filling the ceramic matrix with reinforcing metal can increase the fracture toughness of cermets several times.

There are several ways to create cermet composites. The most common is powder technology. The essence of the technology is as follows: powders of metal and ceramics are mixed in the necessary proportions, subjected to pressing, compacts are obtained, and then sintered. At each stage, it is possible to vary the modes and to some extent control the properties of the obtained cermet. At present, quite a few methods have been described for sintering of cermets: sintering without
pressure [4], pressure [5], spark plasma sintering and its variants [6], selective laser sintering [7], microwave sintering [8], electron beam sintering [9]. Each of the described methods has both advantages and disadvantages, and the choice in favor of one or another method is determined by the requirements for the material obtained. A relatively new method of sintering is electron beam in a protective environment of the forevacuum range. A pressure of tens of pascals (the so-called forevacuum pressure range), on the one hand, serves as an excellent protective medium against oxidation of the metal component of cermet, on the other hand, the plasma created along the path of the beam propagation allows efficient transmission of the beam power to the irradiated surface [10]. In [10–11], the results of the successful application of electron-beam sintering of cermet materials in the fore-vacuum pressure range are presented.

The purpose of this work was to study the influence of electron-beam sintering modes on the properties of metal-ceramic composites.

2. Experimental setup
The experimental setup for electron beam sintering $\text{Al}_2\text{O}_3$-$\text{Ti}$ is shown in figure 1.

![Figure 1. Experimental setup.](image)

Sintering was carried out using powder technology. Alumina and titanium oxide powders were used as starting materials. The average grain size of the alumina powder was 10 $\mu$m, the average grain size of titanium was 30 $\mu$m. The powders were mixed in equal proportions by weight. Samples in the form of disks with a diameter of 10 mm and a thickness of 2.37 mm, 2.77 mm, 4.68 mm, 5.23 mm, 6.68 mm were formed from a mixture of powders by uniaxial pressing. The compacts were located in a graphite crucible, which was installed in a vacuum chamber. For compact sintering, a forevacuum plasma electron source was used, which generated a narrowly focused electron beam [12]. Magnetic coils were used to focus and deflect the electron beam. The minimum diameter of the electron beam was 0.5 mm at an electron energy of 10 kV and a beam current of 50 mA. For uniform heating of the compact surface, an electron beam scan was used. The scan area was a square with a side of 15 mm. Using a scanning beam is much more efficient than using a wide beam with a cross section commensurate with the area of the sample. The fact is that the beam power density, as a rule, has a Gaussian distribution, i.e. the maximum beam power occurs in a small area. To warm the entire sample in this way, it is necessary to increase the diameter of the beam, which leads to an unjustified and inefficient use of its energy. A thin beam scans the surface of the sample at a frequency of 100 Hz,
i.e. 100 times per second hits the same point on the surface, which allows you to evenly heat the sample for sintering. The reduction of heat loss due to heat removal through the fastening elements of the crucible, as well as thermal radiation from its surface, was achieved by placing the crucible on thin rods (not shown in the figure), as well as using a stainless steel heat shield. Part of the thermal radiation from the surface of the sample was reflected by the heat shield and again fell on the sintered sample. The increase in the energy efficiency of the electron beam in this case reached 20%.

3. Materials and technique of sintering
Finely dispersed alumina ($Al_2O_3$) and titanium ($Ti$) powders as the most common materials for the production of cermet products were used as the basis for the manufacture of composite cermets. The main parameters of alumina ceramic powder: white powder, bulk density from 0.6 to 1.7 g/cm$^3$, contains mainly $\alpha-Al_2O_3$ phase. Particles have a spherical shape. Melting point of $Al_2O_3$ is 2000 °C. The titanium powder has an irregular shape and a developed surface of the particles, due to which it is perfectly formed at relatively low pressing pressures in hard dies, as well as by the method of hydrostatic pressing in elastic shells.

The main parameters of the compacts before sintering are presented in table 1.

| Sample number | 1    | 2    | 3    | 4    | 5    |
|---------------|------|------|------|------|------|
| m [g]         | 0.357| 0.462| 0.703| 0.859| 1.114|
| h [mm]        | 2.37 | 2.77 | 4.68 | 5.23 | 6.68 |
| d [mm]        | 10.15| 10.15| 10.15| 10.15| 10.16|
| $\rho$ [g/cm$^3$] | 1.86 | 2.06 | 1.86 | 2.05 | 2.06 |

The sintering process consisted of three stages - heating to a sintering temperature, holding at a sintering temperature, and cooling. Heating to the sintering temperature was carried out at a speed of 70 deg/min due to a uniform increase in the electron beam power from 50 to 300 watts. Exposure at a temperature of 1600 °C took 10 minutes. The sample was cooled due to a smooth decrease in the power of the electron beam for 10 min. The cooling rate was 100 °C/min.

The surface microstructure, cross section, and elemental composition of the sintered samples were studied using a Hitachi S3400N scanning electron microscope equipped with a Bruker X’Flash 5010 energy dispersive microanalysis attachment. The temperature of the irradiated surface was recorded with an infrared pyrometer 6 (RAYTEK 1MH), with a temperature measurement range of 550-3000 °C.

4. Experimental results and discussion
In earlier works, during sintering of compacts consisting of ceramic powder, we showed that one-sided heating of samples leads to their deformation. It was quite expected to obtain deformed samples in the presented experiments. However, as a result of sintering of compacts 1–5, samples that retained a cylindrical shape were obtained, figure 2. Obviously, the addition of a plastic metal component (titanium powder) does not allow such a strong shrinkage of ceramics, it is also possible to equalize thermal fields due to higher thermal conductivity. In subsequent works, this effect will be considered in more detail.

Deformation was observed in samples more than 4 mm thick. As can be seen in figure 2c, the surface being irradiated and the layer about 2 mm thick shrink. Moreover, the greatest shrinkage was observed in the surface layer. A cross-sectional study of sample 1 on an electron microscope and a map of the volume distribution of aluminum and titanium are presented in figure 3. Three areas can be distinguished in the sample volume, which differ in porosity and elemental composition. Region 1 (see figure 3 a) is a rather thin titanium layer on the surface of the sample and a porous layer with a
thickness of about 50-60 μm, region 2 (see figure 3 b) is the central part of the sample, denser with fewer pores, region 3 (see figure 3 c) is again a porous region, but the pore sizes are much smaller compared to region 1 on the sample surface, figure 3. The distribution of elements by regions is quite uniform, except for region 1. It should be noted that a thin layer of titanium is also present on the side of the sample not exposed to radiation. Moreover, with an increase in the thickness of the samples, the titanium layer is retained only on the exposed side. Table 2 presents the elemental composition of the irradiated and non-irradiated surface of samples 1 and 5. As follows from the analysis of the data in Table 2, the titanium content dominates on the surface of thin samples; in the case of thicker samples (more than 4 mm thick), titanium prevails only on the irradiated surface.

Figure 2. Photos of the cross-section of the samples: a - sample 1, b - sample 4, c - sample 5.

Figure 3. Micrograph of a cross section of sample 1 and the distribution of Al and Ti elements.

The most interesting in our opinion is region 2 of the sample, figure 3. As depth microhardness measurements showed, this portion of the sample volume turned out to be the hardest, figure 4. Microhardness was measured from the surface exposed to radiation. The depth distribution of
microhardness is not uniform due to the heterogeneity of the sample itself. The microhardness was measured in automatic mode and the indenter of the microhardness meter could fall both on flat areas and on pores. However, as can be seen from figure 4, the microhardness in region 2 reached 27 GPa, which is at the microhardness level of alumina ceramics. If we compare the rest of the samples, then such a site with high hardness is present in each of them.

| Table 2. Elemental composition of the irradiated and non-irradiated surface of samples. |
|---|---|---|---|
| Element | Sample 1 Irradiated side [norm. wt.%] | Sample 1 Non-irradiated side [norm. wt.%] | Sample 5 Irradiated side [norm. wt.%] | Sample 5 Non-irradiated side [norm. wt.%] |
| Titanium | 84.23 | 81.69 | 65.89 | 28.66 |
| Aluminium | 6.42 | 9.56 | 12.27 | 59.52 |
| Oxygen | 9.35 | 8.75 | 21.84 | 11.82 |

Further research should be aimed at optimizing sintering conditions to expand the region with high microhardness over the entire volume of the sample.

5. Conclusion
Electron beam sintering of powder compacts consisting of aluminum oxide and titanium was carried out. A feature of the method is the generation and transportation of an electron beam in the fore-vacuum pressure range. Sintering of cermet powders by the electron beam method makes it possible to obtain samples with microhardness at the level of 27 GPa - comparable to the microhardness of alumina ceramics.

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