IOP Conf. Series: Materials Science and Engineering **731** (2020) 012015 doi:10.1088/1757-899X/731/1/012015

Improving the Mechanical Properties of SiC-ceramics by means of Vacuum Electron-ion-plasma Alloying with Titanium

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Abstract. The investigation results of elemental and phase composition, state of defective substructure and microhardness of the surface layer of «film (Ti)/substrate (SiC-ceramics)» system (Ti film 0.5 μm thick was deposited on the surface of SiC-ceramics) subjected to treatment with an intense pulsed low-energy electron beam (15 J/cm^2, 200 μs, 0.3 s^{-1}, 20 pulses) are presented. It is shown that irradiation of the «film (Ti)/substrate (SiC-ceramics)» system with an electron beam is accompanied by the formation of multielement multiphase (SiC; TiC; Ti_5Si_3) surface layer having submicro- and nanocrystalline structure. Microhardness of the irradiated surface layer reaches a value of 74 GPa, that is twice the value of microhardness of SiC-ceramics (36 GPa).

1. Introduction

Silicon carbide (SiC) ceramic is a substantial structural material applied in fields as aerospace, machinery and electronics due to its superior mechanical properties and chemical stability at high temperature, low density and coefficient of thermal expansion (CTE) [1-4]. In particular, this property mixture makes them promising candidates as lightweight alternative for several aircraft turbine components instead of super alloys. Recently, the application of first SiC parts was demonstrated in the hot section of new jet engines [5, 6]. However, the fabrication of complex SiC ceramic shaped components utilized in practical engineering was limited by its inherent nature of high brittleness, low malleability and poor machinability [7, 8]. Many strategies have been proposed to tackle this problem and adding reinforcing materials as a second phase in the form of particles [9-12], platelets/flakes [13-15], nanotubes [16-20], whiskers/fibers [21-25] is a useful method. But in most cases, in order to increase the mechanical properties of ceramics, it is only necessary to modify the surface layer [26-29]. The modifying method of the material surface with intense pulsed low-energy (to 30 keV) electron beams of submillisecond duration is the modern approach of the controlling microstructure and properties of parts and products. Impact of such beams on metals, alloys, cermet and ceramic materials is accompanied with the formation of an amorphous, nano and submicrocrystalline structure in the surface layer of the product, contributing to significant improvement the physicochemical, electrophysical, strength and many other properties [30-33].

The aim of work – revealing the regularities in the formation of the structure and properties of the surface layer of SiC-ceramics subjected to titanium alloying by irradiating the «Ti/SiC-ceramic» system with an intense pulsed electron beam.

2. Materials and methods

SiC powder with particle sizes (0.9 – 4.0) μm and nanopowder of the same composition (3 wt.%, particle size 60 nm) were used to fabrication ceramic samples. Samples with a diameter of 14 mm and thickness of 3 mm were obtained by SPS-sintering (SPS-515S (SPS SYNNEX), NR TPU, Tomsk). Sintering mode: sintering temperature 2100 °C, pressure up to 70 MPa and sintering time 10 min. The deposition of a 0.5 μm thick titanium film was carried out on a «KVINTA» unit (IHCE SB RAS) by vacuum electric arc spraying with plasma assistance («PINK» plasma generator) of a target of technically pure titanium of the VT1-0 grade. The «film (Ti)/substrate (SiC-ceramics)» system was irradiated with an intense pulsed electron beam at the «SOLO» device (IHCE SB RAS) with beam parameters: the accelerated electron energy is 18 keV, energy density of the electron beam is 15 J/cm^2, pulse duration 200 μs, quantity of pulses 20 at a repetition rate of 0.3 s^{-1}; the pressure of the residual gas (argon) in the working chamber is 10^{-2} Pa. Investigations of the structural-phase state of the modified surface layer of ceramics before and after the effect of the electron beam were carried out using the equipment of the Nano-Center TPU and the Center for Collective Use of the ISPMS SB RAS «NANOTECH» using scanning electron microscopy (JSM-7500FA, JEOL) and transmission diffraction electron microscopy (JEM-2100F, JEOL), X-ray diffractometer (XRD-7000S, Shimadzu). Microhardness of the modified layer was determined on the «PMT-3M» device.

3. Results and discussion

Vacuum electric arc spraying with plasma assistance («PINK» plasma generator) of a target of technically pure titanium of the VT1-0 grade is accompanied by the formation of a titanium film containing particles of a drop fraction on the surface of SiC ceramic (Figure 1a). The sizes of drops are changing from tens nanometers to units of micrometers. Irradiation of the «film (Ti)/substrate (SiC-ceramics)» system with an intense pulsed electron beam leads to melting of the titanium film (Figure 1b). On the surface of samples, regions with sizes up to 100 μm having dendritic crystallization structure are formed (Figure 2). Apparently, these regions were formed as a result of melting and high-speed crystallization of the droplet fraction particles.

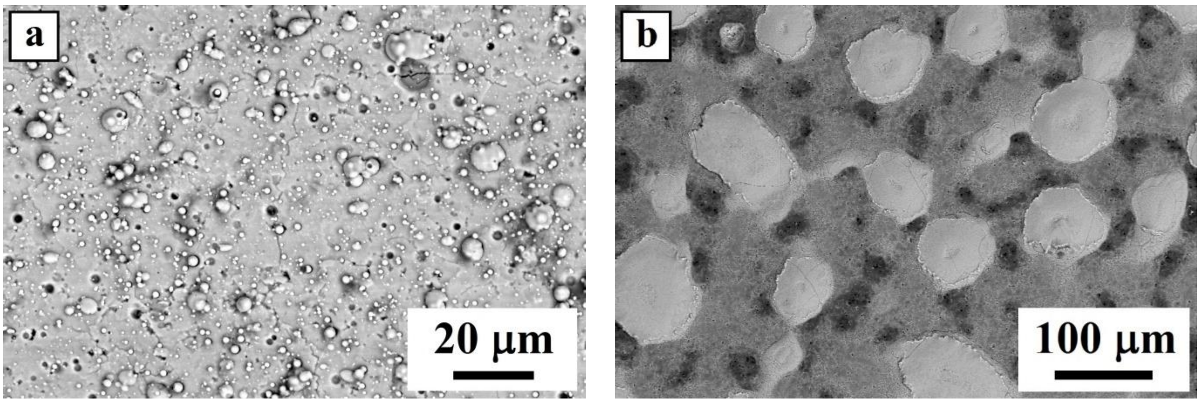

Figure 1. SEM images of the surface of SiC-ceramics samples with a deposited titanium film before (a) and after (b) irradiation with an intense pulsed electron beam.

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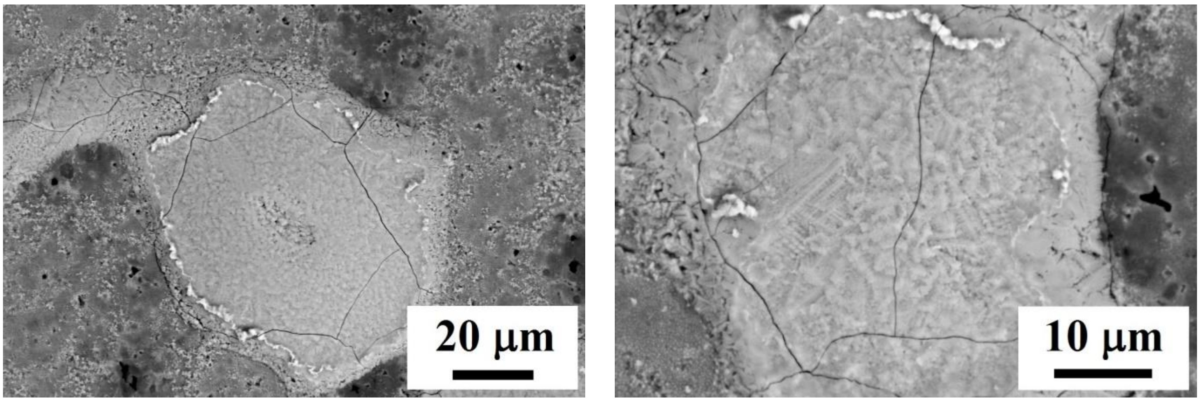
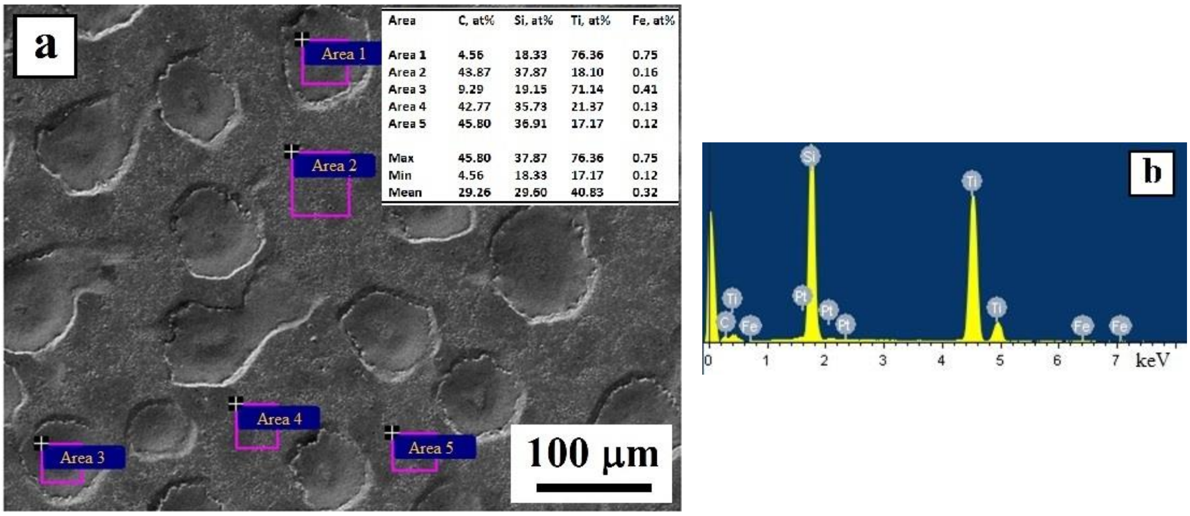


Figure 2. SEM images of surface of the «film (Ti)/substrate (SiC-ceramics)» system irradiated with an intense pulsed electron beam.

Concentration of elements in the surface layer of the «film (Ti)/substrate (SiC-ceramics)» system was determined by the energy-dispersive X-ray spectroscopy (EDS). It is established that the regions of dendritic crystallization (areas 1 and 3 in Figure 3a) are enriched in titanium atoms. Space separating these regions (areas 2, 4 and 5 in Figure 3a) is enriched with silicon and carbon and also contain titanium. On the average over the surface area of sample (Figure 3b), the concentration of carbon, silicon and titanium atoms is 30.4 at.%, 33.3 at.% and 36.0 at.%, respectively.



Area	C, at%	Si, at%	Ti, at%	Fe, at%
Area 1	4.56	18.33	76.36	0.75
Area 2	43.87	37.87	18.10	0.16
Area 3	9.29	19.15	71.14	0.41
Area 4	42.77	35.73	21.37	0.13
Area 5	45.80	36.91	17.17	0.12
Max	45.80	37.87	76.36	0.75
Min	4.56	18.33	17.17	0.12
Mean	29.26	29.60	40.83	0.32

Figure 3. Structure of the «film (Ti)/substrate (SiC-ceramics)» system irradiated with an intense pulsed electron beam (a). Areas of analysis of elemental composition are indicated. The table shows the elemental composition of these areas; (b) energy spectra obtained from the sample surface area given in (a).

The phase composition of surface layer of the «film (Ti)/substrate (SiC-ceramics)» system, irradiated with an intense pulsed electron beam was studied by X-ray diffraction analysis. As a result of performed studies, the presence of following phases was revealed: SiC – 18.5 %, TiC – 36.6 %, Ti_5Si_3 – 44.9 % (Figure 4). Therefore, electron beam treatment of the «film (Ti)/substrate (SiC-ceramics)» system is accompanied by melting of titanium film, doping the melt with silicon and carbon atoms, formation of new phases on the high-speed crystallization step and subsequent cooling.

Defective substructure of surface layer of the «film (Ti)/substrate (SiC-ceramics)» system irradiated with an intense pulsed electron beam was analyzed by transmission electron microscopy of thin foils. The foils were prepared by ionic thinning of plates cut perpendicular to the irradiation surface. This method of preparing foils (transverse foil method) allows sighting study the elemental and phase composition, defective substructure of material depending on the distance from the irradiation surface (Figure 5a).

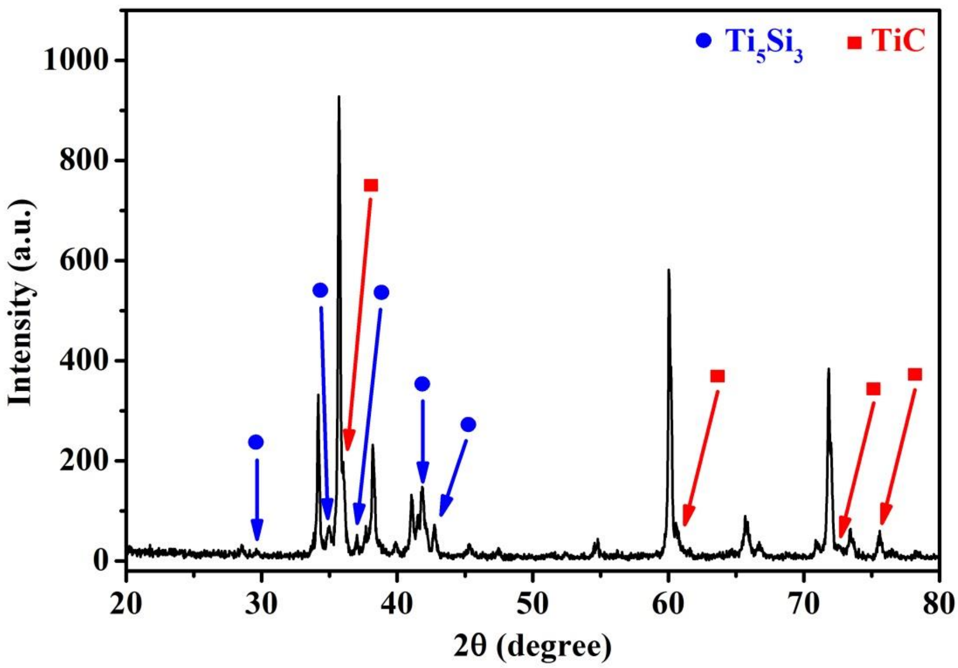


Figure 4. XRD pattern obtained from the surface layer of the «film (Ti)/substrate (SiC-ceramics)» system irradiated with an intense pulsed electron beam.

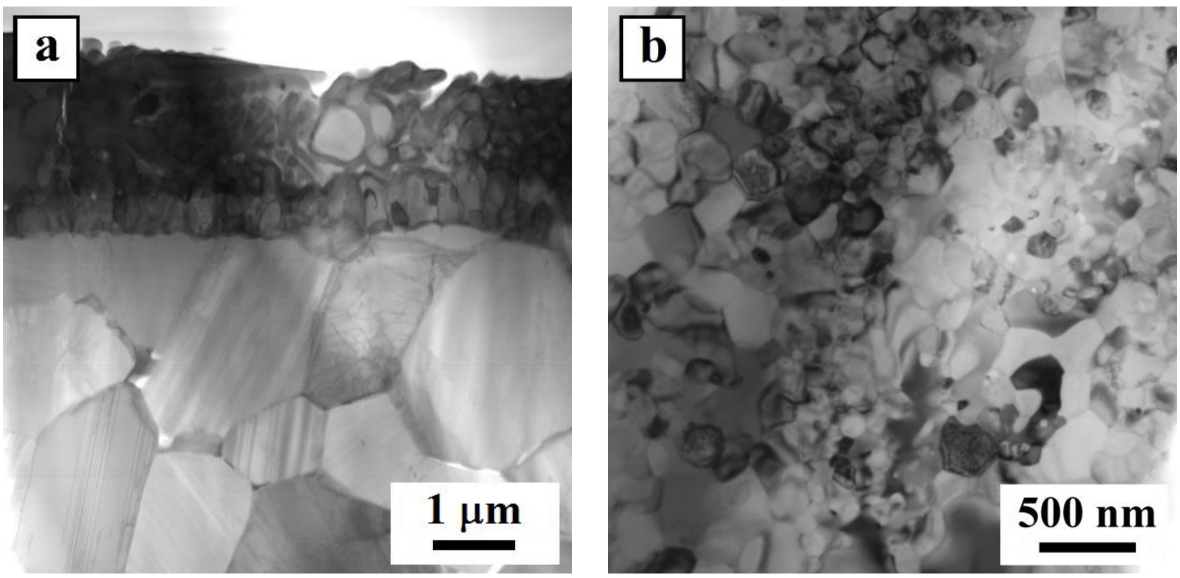


Figure 5. The regions of the structure formed during melting and high-speed crystallization, enriched with titanium atoms (regions containing the droplet fraction particle).

As a result of the investigations carried out, it was established that after irradiation with the electron beam of the «film (Ti)/substrate (SiC-ceramics)» system, surface layer is formed, the crystallite sizes of which range from 20 nm to 500 nm (Figure 5). Layer adjacent to the surface of SiC ceramic has a columnar structure (Figure 5a). Structure of the main volume of surface layer is formed by crystallites having a globular shape (Figure 5b).

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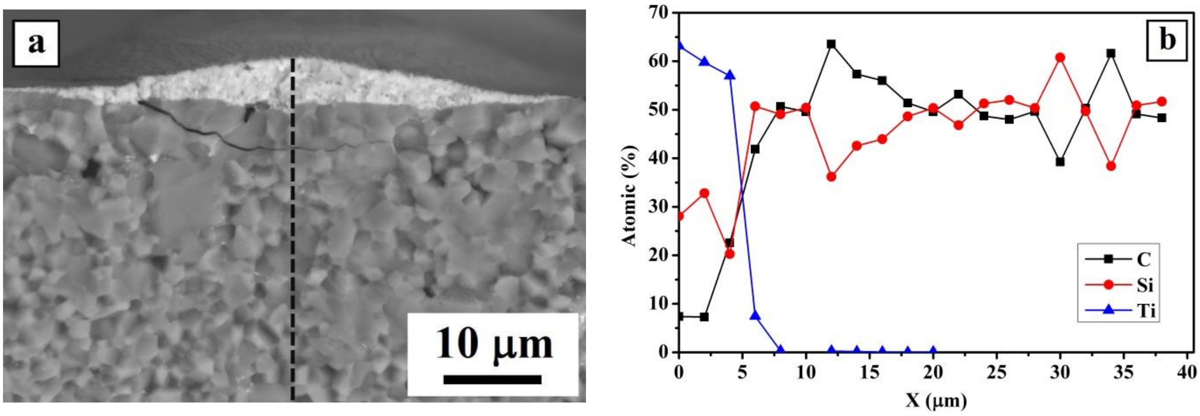

Figure 6. Structure of cross-section of the «film (Ti)/substrate (SiC-ceramics)» system (a); (b) the concentration profile of the main elements along the line indicated in (a).

Distribution of basic elements of ceramics modified layer was studied by the EDS analysis (Figure 6). It is established that high-speed melting of a titanium film leads to the formation of a multielement state: in the surface layer, there are of titanium, carbon and silicon atoms. Therein, titanium is the major element in the drop fraction particle (Figure 6b). When going across the «droplet/substrate» interface, the titanium concentration swiftly decreases. The thickness of titanium-doped layer of ceramic reaches 3 μm. Consequently, irradiation of the «film (Ti)/substrate (SiC-ceramics)» system with an intense electron beam is accompanied by the diffusion of ceramic elements into the deposited coating. Therefore, coating has a multiphase composition (titanium and silicon carbides, silicides and silicocarbides of titanium).

The formation of a nanocrystalline multielement multiphase surface layer as a result of irradiation of the «film (Ti)/substrate (SiC-ceramics)» system with a pulsed electron beam is accompanied by a significant increase of microhardness, the value of which reaches 74 GPa (microhardness of SiC ceramics is 36 GPa). The hardness maximum values are revealed near the boundary separating the regions enriched with titanium (droplets) from the main volume of the coating.

4. Conclusions

It is shown that irradiation of the «film (Ti)/substrate (SiC-ceramics)» system with an intense pulsed electron beam (15 J/cm^2, 200 μs, 0.3 s^{-1}, 20 pulses) leads to the formation of a surface layer, having a multielement multiphase structure of the submicro-nanocrystalline range. The mutual diffusion the atoms of film and substrate have been revealed. The thickness of titanium-doped layer of ceramic reaches 3 μm. It has been established that the microhardness of the «film (Ti)/substrate (SiC-ceramics)» system after irradiation with an electron beam reaches a value of 74 GPa, that is twice the value of microhardness of SiC-ceramics (36 GPa). The hardness maximum values are revealed near the boundary separating the regions enriched with titanium (droplets) from the main volume of the coating. It is obvious that such high values of microhardness are due to the formation of a nanocrystalline, multielement, multiphase state formed as a result of mutual diffusion of titanium, carbon and silicon atoms during irradiation with an intense pulsed electron beam.

Acknowledgment
This work was supported by the program of fundamental scientific research for 2019-2021. (Subject № 0291-2019-0002).

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