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Article *in* Physics of Particles and Nuclei Letters · September 2023

DOI: 10.1134/S1547477123050102

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ISSN 1547-4771, Physics of Particles and Nuclei Letters, 2023, Vol. 20, No. 5, pp. 1259–1262. © Pleiades Publishing, Ltd., 2023.

PHYSICS OF SOLID STATE
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Investigation of the Radiation Resistance and Optical Properties of New Composite Thermal Barrier Coatings

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Received November 14, 2022; revised April 25, 2023; accepted May 2, 2023

Abstract—Improved thermal barrier coatings (TBCs) will enable future gas turbines to operate at higher gas temperatures. Considerable effort is being invested, in identifying new materials with even better performance than the current industry standard, yttrium stabilized zirconia (YSZ). TBCs are also supposed to be applied in spacecrafts as protective layer against heat. The operation of spacecrafts in cosmic conditions in turn suggests continuous irradiation with cosmic rays, particularly with MeV energy protons, electrons and neutrons. Therefore, it is very important to investigate the behavior of such barrier coatings under irradiation conditions. In this work, we investigate the radiation resistance of TBCs based on silicate compounds obtained by a hydrothermal microwave method by using proton and neutron beam irradiation. For this purpose, zinc silicates and cerium-doped zinc silicates were irradiated with 18 MeV protons with doses 10^{13}–10^{15} p/cm^2 and neutrons with doses 10^{13}, 10^{15} n/cm^2. The diffuse reflectance measurements and X-ray diffraction analysis (XRD) of materials before and after irradiation indicated that the samples have high radiation resistance, and the samples maintain the crystalline structure.

Keywords: zinc orthosilicate, proton irradiation, neutron irradiation, diffuse-reflection, radiation resistance, thermal barrier coatings

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INTRODUCTION

Insufficient radiation resistance of spacecraft structural materials is one of the main obstacles for the development of modern astronomy. Minimizing the impact of cosmic radiation (CR) or increasing the durability of structural materials is one of the most important challenges for state of the art science. The CR can affect not only astronomers, but also cosmic stations, artificial satellites and equipment therein. If in case of astronauts the exposure of CR can be minimized by limiting their working period in the space, in case of space stations, this problem is much deeper, as the equipment are continuously operating at space conditions for decades. Most sensitive to radiation damages are semiconductor and polymer materials, which are an indispensable part of spacecrafts [1]. Considerable effort is being invested in identifying new materials with even better performance than the current industry standards. Silicate solution- based powders (Zn_2SiO_4, $ZrSiO_4$, Na_4SiO_4, Y_2SiO_5), pyrochlore oxides ($La_2Zr_2O_7$), perovskites, aluminates as well as YSZ obtained by plasma spraying (PS), electron beam physical vapor deposition (EB—PVD), sol-gel or laser chemical vapor deposition [2, 3], are among the most known materials used as a TBC [4–9].

A typical TBC is yttria-stabilized zirconia, made of ~6–8 wt % Y_2O_3-stabilized ZrO_2 ceramics. During last few decades nanostructured zirconia TBCs have become a hot research field due to their low thermal conductivity, high coefficient of thermal expansion and excellent mechanical properties. The application of TBCs, of which the thickness traditionally ranges from 100–500 μm, along with internal cooling of the underlying superalloy component, offers 100–300°C reduction in the surface temperature of the superalloy. This allows the superalloy-made workpieces to be operated at environmental temperatures above the melting temperature of the superalloy (~1300°C), thereby improving the engine efficiency and performance. This technology has been regarded as one of the most important and effective developments in efforts to improve the propulsion efficiency of advanced aero engines, as the efficiency and power are

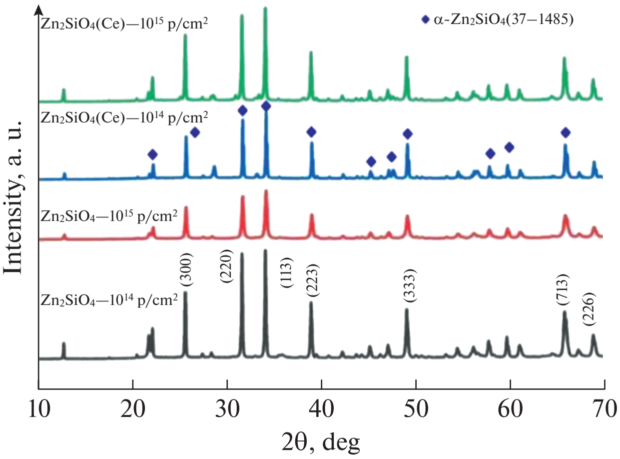

Fig. 1. XRD patterns of Zn_2SiO_4 and Ce–Zn_2SiO_4 samples subjected to 18 MeV proton irradiation with 10^{14} and 10^{15} p/cm^2 doses.

directly related to the gas temperature entering the turbine section. State of the art TBCs are complex, multifunctional thick films (typically 100 μm to 2 mm thick) of a refractory material that protect the metal part from the extreme temperatures in the gas. The TBC components must withstand the harsh environment coupled with high temperature, large temperature gradient, complex stress condition and corrosive atmosphere. No single coating component is able to satisfy these multifunctional requirements. As a result, a complex TBC structure has been developed. TBCs undergo recurrent changes in their composition, microstructure and crystalline phases during its service life. The effectiveness of TBCs is largely dependent on their thermal properties, however insulating effects can also be modified by applying different mixtures and thicknesses. Traditionally the TBCs are composed of metallic bond-coat and ceramic top-coat. The bond-coat is usually composed of MCrAlY (M = Ni and/or Co), the ceramic top-coat is usually composed of 6–8 wt % YSZ. Such structures are resistant against oxidation, against impact of charged particles, as well as to the effects of solar and space radiation [10]. However, in the case of long-term orbital flights, even such materials are subject to structural changes and lose their initial properties. Large-amount of defects and absorption centers appear under the influence of high energy particle radiation resulting in reduced reflection in a wide spectral range and hence increase in absorption [11, 12]. Generally, silicates have their applications in different areas of electronics and other fields due to high chemical resistance. The performance of any device based on such materials will significantly depend on defects available in materials due to synthesis method as well as defects, created during its operation lifetime.

Structural changes in the materials under the influence of electron beam have been previously studied in other oxide compounds (Al_2O_3, Al_2O_3:Cr, $Y_3Al_5O_{12}$) [13–15] and it has been shown, that in the case of high energies structural defects appear in the materials. In contrast to electrons and protons, ultraviolet radiation changes the charge state of defects in materials.

EXPERIMENTAL

In our previous work, we show that microwave-assisted hydrothermal synthesis methods allow the production of pure and Ce-doped Zn_2SiO_4. Also, UV-Vis-NIR absorption/reflection measurements of the electron-irradiated materials indicate that Ce–Zn_2SiO_4 exhibits better radiation resistance compared to pure Zn_2SiO_4 with a high crystalline structure [16].

Irradiation with proton was performed at Radioisotope Production Center (Yerevan, Armenia) by using Cyclotron C-18 (beam current 0.6 μA). Proton beam of 18 MeV energies was used. The average flux density of protons was kept at approximately 10^{12} cm^{-2} s^{-1} to avoid heating the samples during irradiation. To understand the dose dependence of proton irradiation, the samples were irradiated with three different doses: 10^{13}, 10^{14}, and 10^{15} p/cm^2. Neutron irradiation of samples was performed at IBR-2, JINR, FLNP with 10^{13}, 10^{15} n/cm^2 doses. The crystallinity and phase compositions of the materials were investigated by X-ray diffraction (XRD) analysis with Ni-filtered Cu *K*α radiation (D8 Advance, Bruker), operated at 40 kV and 40 mA with a step size of 0.025 and a counting time of 5 s for the angular range of 10°–70° (2θ). Diffuse reflection measurements of the samples were performed in the wavelength range from 190 to 1000 nm using a Cary-60 UV-Vis-NIR spectrophotometer with a Video-Barrelino diffuse reflection attachment.

RESULTS AND DISCUSSION

The structure and phase composition of the synthesized compounds were determined by X-ray diffraction analysis (XRD), according to which, even after irradiation with a dose of 10^{15} p/cm^2, the samples maintained their crystal structure. Figure 1 shows the XRD patterns for pure and Ce-Zn_2SiO_4 samples after proton irradiation. All the diffraction lines (Fig. 1) are indexed to the pure phase of α-Zn_2SiO_4, and no peaks from other phases can be detected.

The dependence of the diffuse reflection coefficient of pure and cerium-doped samples irradiated with protons on various doses of radiation was also studied; the results are presented in Fig. 2.

As can be seen, after irradiation with different doses of protons, pure zinc orthosilicate (Fig. 2a) compared to zinc orthosilicate doped with cerium (Fig. 2b) has low diffuse reflectance. In the case of

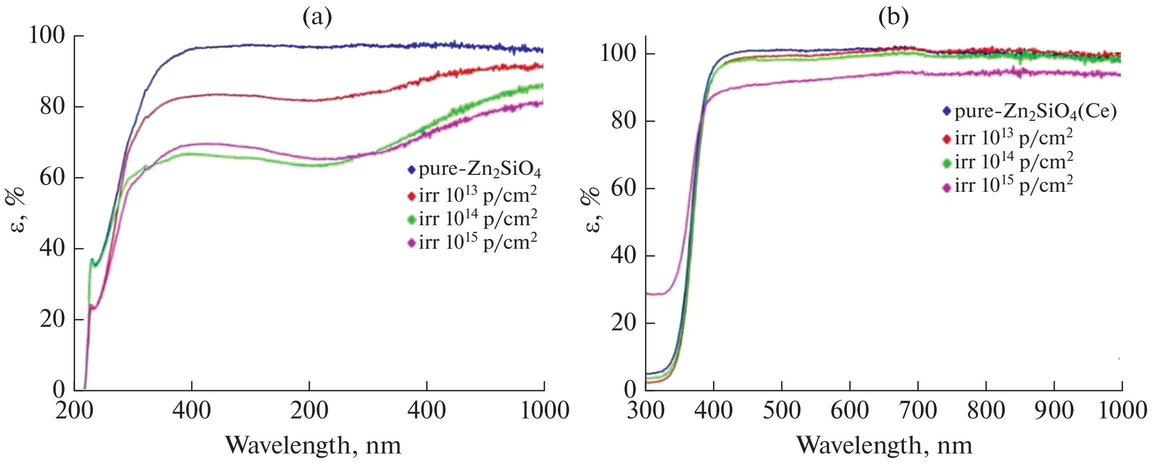

Fig. 2. Diffuse reflectance spectra of Zn_2SiO_4 (a) and Zn_2SiO_4 (Ce_2O_3—5%) (b) samples before and after irradiation with protons with an energy of 18 MeV and doses (10^{13}, 10^{14}, and 10^{15} p/cm^2).

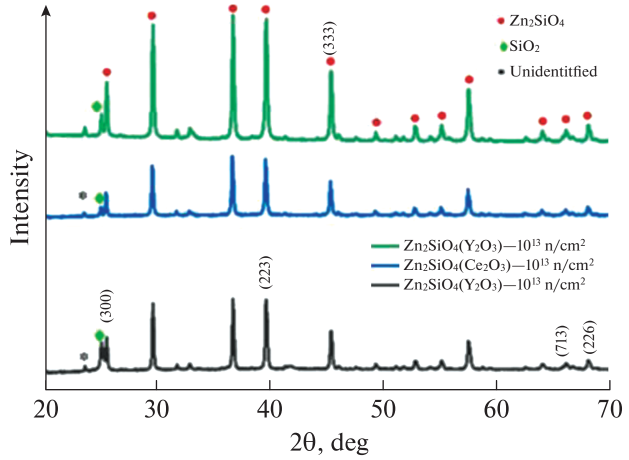

Fig. 3. XRD patterns of Ce/Y–Zn_2SiO_4 samples subjected to neutron irradiation 10^{13}, 10^{15} n/cm^2 doses.

pure zinc orthosilicate, the diffuse reflectance decreases significantly after proton irradiation.

In Fig. 3 we also see that after neutron irradiation the samples maintain their crystalline structure. So we can suggest that these materials have a high radiation resistance and proton and neutron irradiation does not damage the crystalline structure of these materials.

CONCLUSIONS

Pure and Ce/Y-doped Zn_2SiO_4 samples were irradiated with proton and neutron beams. It is found that the change in the values of the diffusion reflection coefficients is due to the formation of new defects in the structure due to changes in the surface of the coating due to the interaction of pigments to the effect of proton irradiation. Measurements of the irradiated materials indicate that Ce–Zn_2SiO_4 exhibits better radiation resistance compared to pure Zn_2SiO_4. The results show that thermal barrier coatings made of cerium-doped zinc silicate can be excellent candidates for use in spacecraft and not only.

FUNDING

The work was supported by the Ministry of Education, Science, Culture and Sports of the Republic of Armenia, Science Committee (grant 21-AA-1C020).

CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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