



# Irradiation effect on optical properties and structure of sapphire single crystal



Rui Ke\*, Yumin Zhang, Yufeng Zhou, Zhongwei Yin

Science and Technology on Advanced Composites in Special Environments Laboratory, Harbin Institute of Technology, Harbin 150080, China

## ARTICLE INFO

### Article history:

Received 16 April 2013

Accepted 1 September 2013

### Keywords:

Sapphire  
Irradiation  
Surface roughness  
Transmittance

## ABSTRACT

Sapphire single crystal for optical window material was irradiated by rays and particles when served in the space environment, thus, the optical properties and structure would be changed even the window lost its effectiveness. The change of the surface roughness and transmittance caused by electron, proton and integrated currents was discussed through the ground simulation experiment. The AFM test indicated that the space particle irradiation led to roughness decreased with dose effect and energy effect, meanwhile, the integrated current caused the maximum effect and the electron minimum; different range between electron and proton caused the distinguishing effect on roughness of back side. According to the UV-Vis-VIR and FTIR testing result, the particle irradiation gave rise to decrease of transmittance accompanied by dose effect and energy effect, especially in ultraviolet band, different kinds of particle caused different effect in each band.

© 2013 Elsevier GmbH. All rights reserved.

## 1. Introduction

With the rapid development of electro-optical technology, sapphire single crystal ( $\alpha\text{-Al}_2\text{O}_3$ ) was widely used for various space optical windows material in aerospace field because of its high hardness [1], corrosion resistance [2] and excellent optical performance [3,4]. However, the sapphire optical window would inevitably be irradiated by different rays [5,6] (such as X-ray,  $\gamma$ -rays) and ion currents [7,8] (protons electrons and other particles) service in space environment, therefore, the optical properties and structure would be changed and affected the performance. Thus, the research of properties variation because of the irradiation would provide effective help for improving the work performance of optical windows, extending the service life and developing the new windows.

In this paper, the surface roughness and the transmittance before and after the particle irradiation were characterized through series of detection devices. Finally, the optical properties and structure change of sapphire single crystal for optical windows caused by electron, proton and integrative irradiation were discussed.

## 2. Experimental

### 2.1. Experimental content

We used the proton current, electron current, integrated (proton and electron) current radiated by the ground simulation experiment for simulating the space radiation environment which sapphire windows served in, the experimental content was shown in Table 1. The change of optical properties and structural including surface roughness and transmittance caused by irradiation were characterized.

### 2.2. Specimen preparation

The large-size sapphire single crystal material which was self-developed through SAPMAC (Sapphire Growth Technique with Micro-pulling and Shoulder at Cooled Center) process [9] by Science and Technology on Advanced Composites in Special Environments Laboratory of Harbin Institute of Technology was used in this paper. The foursquare flaky specimens shown in Fig. 1 with size of  $10\text{ mm} \times 10\text{ mm} \times 0.43\text{ mm}$  were cut from the same position of the crystal through ultraviolet laser processing technology, precision mechanical and chemical double-sided polishing made the surface roughness reach Ra 0.3–0.4 nm.

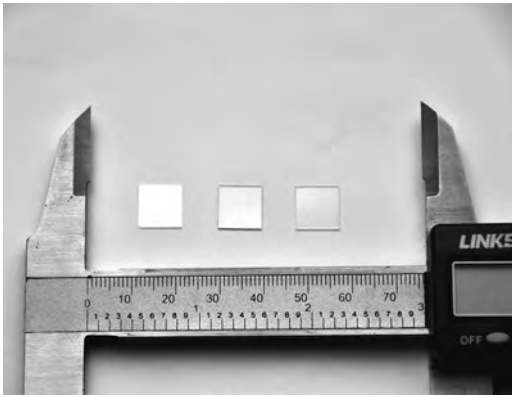
### 2.3. Experimental installation

КИФК Combined Irradiation Analog Device made in Ukraine was used for ground simulation experiment. The device included a vacuum system, proton and electron accelerators and control

\* Corresponding author. Tel.: +86 451 86412236; fax: +86 451 86412236.  
E-mail address: [kerui@hit.edu.cn](mailto:kerui@hit.edu.cn) (R. Ke).

**Table 1**  
Experimental content of the simulate space irradiation.

Specimen number	Irradiation content	Irradiation energy (keV)	Irradiation dose (cm <sup>-2</sup> )	Irradiation time (s)
0#	No irradiation	0 (for contrast)	0 (for contrast)	0 (for contrast)
1-1#	Irradiated by electron	100	$2 \times 10^{15}$	2000
1-2#			$5 \times 10^{15}$	5640
1-3#			$2 \times 10^{16}$	16,400
2-1#	Irradiated by proton	150	$2 \times 10^{15}$	2000
2-2#			$5 \times 10^{15}$	5640
2-3#			$2 \times 10^{16}$	16,400
3-1#	Irradiated by proton and electron ( $p^+ = e^-$ )	100	$2 \times 10^{15}$	2000
3-2#			$5 \times 10^{15}$	5640
3-3#			$2 \times 10^{16}$	16,400
4-1#	Irradiated by proton and electron ( $p^+ = e^-$ )	150	$2 \times 10^{15}$	2000
4-2#			$5 \times 10^{15}$	5640
4-3#			$2 \times 10^{16}$	16,400
5-1#	Irradiated by proton and electron ( $p^+ = e^-$ )	100	$2 \times 10^{16}$	16,400



**Fig. 1.** Sapphire specimens for the ground simulation experiment of space irradiation.

systems to make sure that the device could get the proton beam, electron beam or integrated beam with energy range between 30 keV and 200 keV, the device could ensure the cleanliness of the specimens chamber and the reliability of the experimental results. The flaky specimens were placed in the specimen chamber with the vacuum degree of  $4.2 \times 10^{-4}$  Pa.

Surface roughness of the sapphire single crystal before and after the irradiation was gauged by the CSPM5550 AFM (Atomic Force Microscopy) manufactured by Being Nano-Instruments Ltd., the surface roughness was gauged from seven different regions with

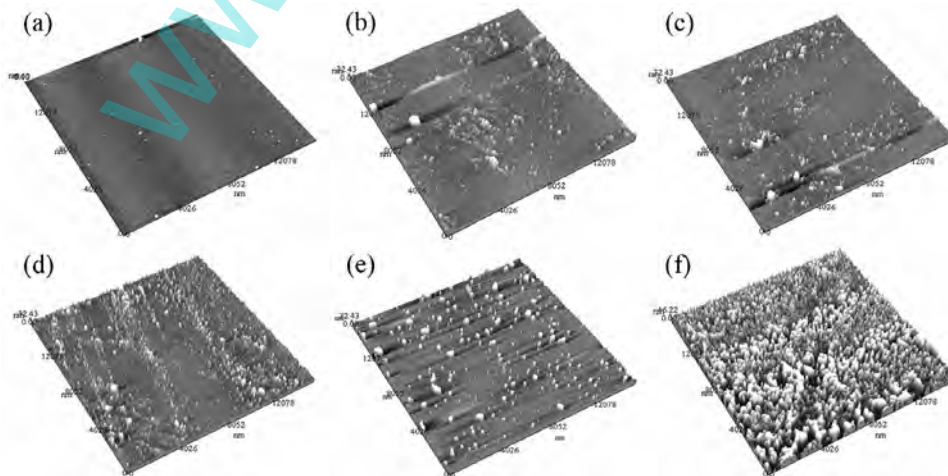
the area of  $16 \mu\text{m} \times 16 \mu\text{m}$  on the specimens and the average value was adopted in the paper finally. Lambda 950 UV-Vis-NIR Spectrophotometers manufactured by PerkinElmer Inc. was used for testing of transmittance in UV-vis-NIR (ultraviolet-visible-near infrared) band (wavelength during 200–2500 nm) with step of 4 nm, meanwhile, the Spectrum One B FT-IR spectrometer was used for testing transmittance in MIR (middle infrared) band (wavelength during 2800–5000 nm) with step of 1 wave number (wave number = 10 nm/wavelength).

### 3. Results and discussions

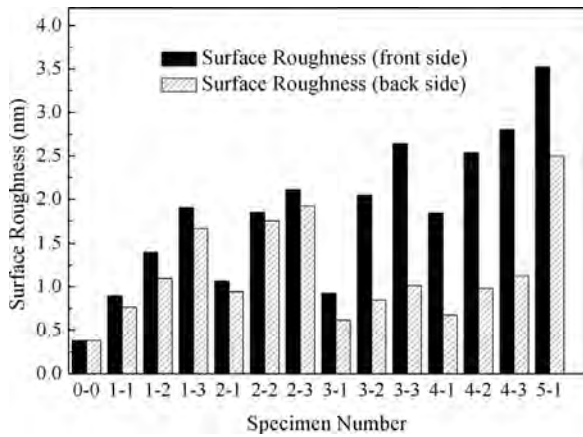
#### 3.1. Surface roughness analysis

The front sides of the specimens were irradiated by particles in the experiments, afterwards, we got clear surface stereogram of both sides by Atomic Force Microscopy, a part of stereogram was shown in Fig. 2.

We found that the specimens irradiated by the particles were rougher than specimens which were not irradiated through contrasting Fig. 2(a)–(f), meanwhile, the surface roughness of the back side also increased after the irradiation through contrasting Fig. 2(a)–(c) and (e). The causation of this phenomenon was the surface charge effect [10] was triggered when the particles reached the front superficies of the specimens, that is to say, the charged particles led to the relative potential difference in different region of superficies, the superficies of sapphire will be sparked over and part of them melted. In order to analyze and contrast the change



**Fig. 2.** The surface stereogram of the specimens which were irradiated by electron, proton and integrative currents respectively. (a) 0#, no irradiation; (b) 1-3#, front side; (c) 1-3#, back side; (d) 3-3#, front side; (e) 3-3#, back side and (f) 5-1#, front side (the irradiation dose, energy and time were shown in Table 1).



**Fig. 3.** Histogram of the surface roughness before and after irradiation. (For each specimen, the irradiation dose, irradiation energy and irradiation time were shown in Table 1.)

of the surface roughness after irradiation, we displayed the surface roughness in form of histogram which was showed in Fig. 3.

According to Fig. 3 we could figure out that the dose effect and energy effect was accompanied by the space particles irradiation, in other words, with the increase of the irradiation dose and energy the surface roughness of front and back sides increased. Compared to the irradiation by the low dose of particles, the energy effect on the specimens which were irradiated by high dose was not so obvious, thus, we hypothesized that the energy effect would disappeared when the irradiation dosage increased to the critical value  $\Phi_c$ , meanwhile, the dose effect would dominated in the sort of situation.

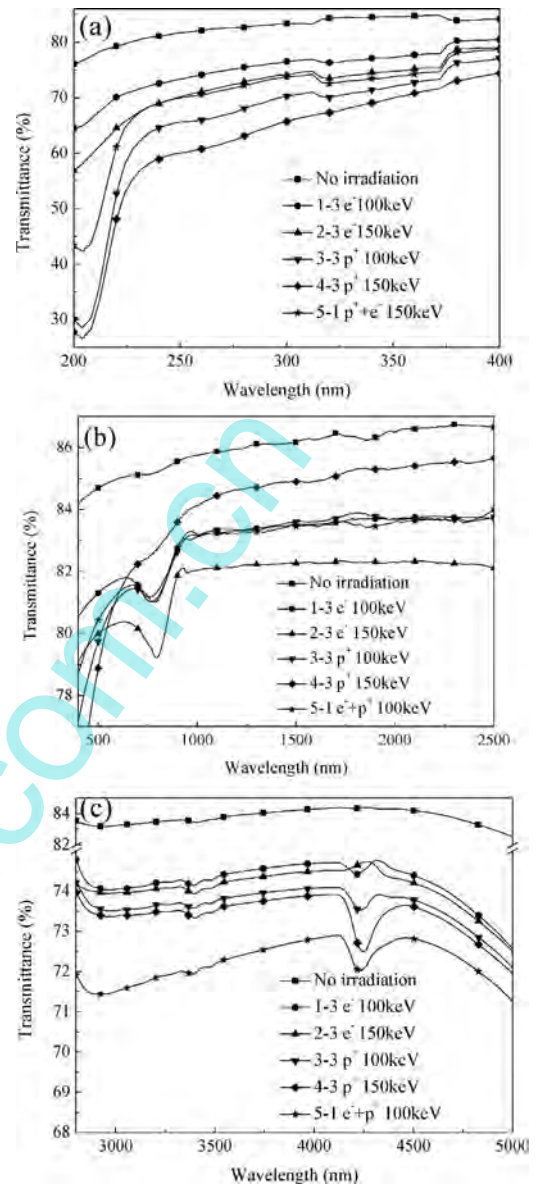
Contrasted the surface roughness of specimens irradiated by different kinds of particles from Fig. 3, we can come to the conclusion that the proton current led to more damage than electron current with same dose, because the mass of the proton was 1800 times to the electron although they carried the same quantities of electric charge, therefore, the proton lost more energy than electron when they dashed against the surface and then the Coulomb collision occurred. We could also figure out that the integrated currents caused the maximum damage to the sapphire both than the proton and electron currents according to Fig. 3, the reason leading to this phenomenon was not only the surface charge effect but also the bulk charge effect which was caused by the different internal range between proton and electron in material, thus, the voltage generated by the internal electric field rose with increasing of irradiation dose until the material was sparked over at the critical value  $U_c$ .

When the specimens were irradiated by proton, the surface roughness of back side was distinguishable from electron irradiation which caused the obvious change of surface roughness on back side. The empirical equation of electronic maximum range Eq. (1) and empirical equation of protonic maximum range Eq. (2) [11] could interpret this phenomenon adequately.

$$R_{e-max} = 0.407E_{e-max}^{1.38} \quad (0.10 \text{ MeV} < E_{e-max} < 0.8 \text{ MeV}) \quad (1)$$

$$-\frac{1}{\rho} \frac{dE}{dx} = \frac{4\pi N_0 z^2 e^4 Z}{m_e c^2 \beta^2 A} \left[ \ln \frac{2m_e c^2 \beta^2}{I(1-\beta^2)} - \beta^2 - \frac{C}{Z} - \frac{\delta}{2} \right] \quad (2)$$

Calculated the range of particles according to Eqs. (1) and (2) we found that the range of electron was 100 times than the proton in the sapphire single crystal, therefore, the electron current affected the sapphire deeper than the proton.

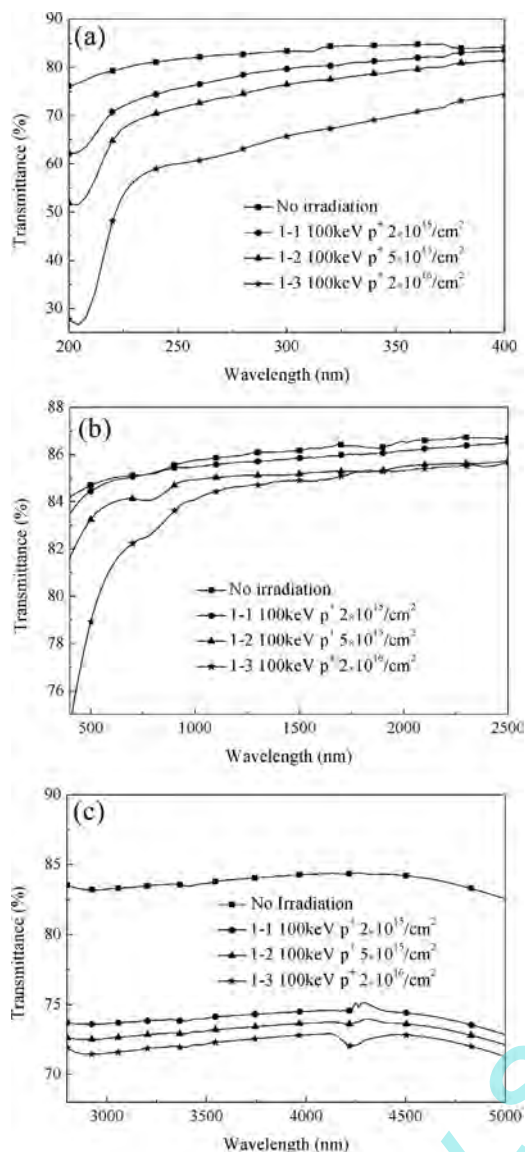


**Fig. 4.** Transmittance–wavelength curves of specimens which were irradiated by kinds of particle and energy with the dose of  $2 \times 10^{16} \text{ cm}^{-2}$ . (a) UV, (b) Vis–NIR and (c) MIR. (The symbols on the curves were only used for distinguishing the type of curves, not meaning that we only tested so few points.)

### 3.2. Transmittance analysis

We discussed the space particles irradiation effect on the transmittance of sapphire single crystal through testing the transmittance in the band of 200–5000 nm by UV-Vis-NIR spectrophotometers and FT-IR spectrometer. Finally, we found that the irradiation caused the decrease of transmittance and each kinds of particle led to different effect in different band. The transmittance–wavelength curves of specimens which were irradiated by kinds of particle and energy with the dose of  $2 \times 10^{16} \text{ cm}^{-2}$  were shown in Fig. 4.

We noticed that the transmittance decreased most fiercely in the ultraviolet band than in Vis–NIR–MIR band through the comprehensive comparison between Fig. 4(a)–(c). We could make out that proton led to the maximum transmittance decrease than electronic and integrative current in the ultraviolet band from Fig. 4(a), the curve with rhombus symbols explained this view ideally. In the Vis band the curve with erect triangle symbols of Fig. 4(b)



**Fig. 5.** Transmittance–wavelength curves of sapphire irradiated by 100 keV proton. (a) UV, (b) Vis–NIR and (c) MIR. (The symbols on the curves were only used for distinguishing the type of curves, not meaning that we only tested so few points.)

pointed out that the electron irradiation had the maximum influence on the transmittance of sapphire single crystal, furthermore, in the Vis–NIR band we could hardly see the exists of the energy effect. When we turned our attention to Fig. 4(c) which were the transmittance–wavelength curves in the MIR band we found the rules of transmittance decrease to follow, the effect on the transmittance adhered to descending order of the integrated current, proton and electron current, contrasted the curve with round dot and curve erect triangle symbols, the curve with handstand triangle symbols and curve with rhombus symbols we found the energy effect existed in the decrease process of transmittance.

We studied the dosage effect on the transmittance of sapphire irradiated by 100 keV proton as a case, the transmittance–wavelength curves were shown in Fig. 5.

Fig. 5 showed that the dose effect really existed in the process of transmittance decrease, in other words, the decline level of

transmittance was related to the dose of irradiation, with the increase of irradiation dose the transmittance decreased seriously.

#### 4. Conclusions

To research the irradiation of space particles effect on sapphire single crystal for optical windows, a series of ground simulation experiments were carried out through the laboratory equipments. The surface roughness and the transmittance before and after the irradiation under various conditions were discussed. According to the results of AFM testing, we found that the particles irradiation resulted in the increase of the surface roughness, furthermore, the dose effect and energy effect was accompanied by the irradiation process; the energy effect had less of effect even would disappeared with the increase of irradiation dose; the different range between proton and electron in material led to the roughness difference of back side. The UV–Vis–NIR spectrophotometers and FT-IR spectrometer testing of transmittance before and after irradiation showed that the particles irradiation caused the decrease of transmittance and also with dose effect and energy effect, the maximum decrease appeared in the UV band, different kinds of particles had different effect in each band, in the ultraviolet band the effect of proton dominated, the impact of electron was mainly reflected in the Vis–NIR band, meanwhile, the integrated particles resulted in the most decrease of transmittance in MIR band.

#### Acknowledgements

We acknowledge the help of the “Supported by Program for New Century Excellent Talents in University” (Grant No. NCET-10-0069), the “Fundamental Research Funds for the Central Universities” (Grant No. HIT.KLOF.2010024), and the “Key Laboratory Funds for Science and Technology on Advanced Composites in Special Environments Laboratory” (Grant No. 9140C490106130C49001).

#### References

- [1] W.G. Mao, Y.G. Shen, C. Lu, Deformation behavior and mechanical properties of polycrystalline and single crystal alumina during nanoindentation, *Scripta Mater.* 65 (2011) 127–130.
- [2] H. Yin, D.S. Grummon, J. Lucas, Abrasion resistance and strength characteristics of [0001] sapphire fibers modified by 175 keV Mg<sup>+</sup> ion irradiation, *Scripta Mater.* 35 (1996) 749–754.
- [3] W.G. Mao, Y.G. Shen, C. Lu, Nanoscale elastic–plastic deformation and stress distributions of the C plane of sapphire single crystal during nanoindentation, *J. Eur. Ceram. Soc.* 31 (2011) 1865–1871.
- [4] E.J. Haney, G. Subhash, Static and dynamic indentation response of basal and prism plane sapphire, *J. Eur. Ceram. Soc.* 31 (2011) 1713–1721.
- [5] S. Garima, S. Apu, V. Jalaj, et al., Effect of irradiation on the microstructure and mechanical behavior of nanocrystalline nickel, *Scripta Mater.* 65 (2011) 727–730.
- [6] C.L. Chen, K. Arakawa, J.G. Lee, U. Ramamurty, S. Gupta, A. Kumar, J. Chakravarty, Electron-irradiation-induced phase transformation in alumina, *Scripta Mater.* 63 (2010) 1013–1016.
- [7] P.F. Yan, K. Du, M.L. Sui, Alpha-to-gamma-Al<sub>2</sub>O<sub>3</sub> martensitic transformation induced by pulsed laser irradiation, *Acta Mater.* 58 (2010) 3867–3876.
- [8] J.J. Sha, S. Ochiai, H. Okuda, Y. Waku, N. Nakagawa, A. Mitani, M. Sato, T. Ishikawa, Residual stresses in YAG phase in directionally solidified eutectic Al<sub>2</sub>O<sub>3</sub>/YAG ceramic composite estimated by X-ray diffraction, *J. Eur. Ceram. Soc.* 28 (2008) 2319–2324.
- [9] C.H. Xu, M.F. Zhang, S.H. Meng, J.C. Han, G.G. Wang, H.B. Zuo, Temperature field design, process analysis and control of SAPMAC method for the growth of large size sapphire crystals, *Cryst. Res. Technol.* 42 (2007) 751–757.
- [10] S. Tricot, M. Nistor, E. Millon, L.C. Boulmer, N.B. Mandache, J. Perriere, W. Seiler, Epitaxial ZnO thin films grown by pulsed electron beam deposition, *Surf. Sci.* 604 (2010) 2024–2030.
- [11] C.D. Li, D.Z. Yang, S.Y. He, Study of synergistic relation effects of protons and electrons on aluminized Teflon FEP film degradation, *Chin. J. Mater. Res.* 17 (2003) 13–17.