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Kinetics of Radiation Damage of Quartz Glass by Low-Energy Protons

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In a ground-based simulation of the space environment of high vacuum, heat sink, and low-energy protons (140 keV), the change in optical transmittance of JGS3 optical quartz glass was studied, and a kinetic model for the evolution of color centers in the process of radiation damage was explored. Experimental results show that radiation damage effects obviously occur in the surface layer of quartz glass under high-flux and low-energy proton radiation. The optical density change increases rapidly and then a saturation trend appears with increasing absorption dose. Based on experimental results, a kinetic model for the evolution of color centers in quartz glass irradiated with protons is proposed, and the change in optical density is given. The fitted curve is similar to experimental ones. It is believed that the proposed kinetic model can be used in the quantitative description of the change in optical properties of quartz glass increasing absorption dose under low-energy proton radiation.

Nomenclature

A	=	optical density
D	=	radiation absorption dose
d	=	thickness of the concentrated layer of color centers
E	=	radiation energy
E^*	=	photon energy
e	=	unit charge
f	=	conversion fraction from potential trap defects to color centers
J	=	radiation dose rate
m^*	=	electronic reduced mass of color center
N	=	concentration of potential trap defects
n	=	concentration of color centers
n_Z	=	refractive index
q	=	annihilation fraction of color centers during radiation
R	=	incident particle penetration depth
T	=	transmittance
W	=	full width at half height
α	=	absorption coefficient
α_m	=	absorption coefficient of absorption peak
ρ	=	material density
σ	=	average electron trapped section
Φ	=	radiation fluence

I. Introduction

AS the most appropriate optical material for highly stable optical systems in spacecraft, quartz glass has a series of excellent properties such as optical, mechanical, thermal, and antiradiation stability and is extensively used as optical windows and optical lenses in spacecraft.[1,2] The optical elements or materials are degraded in properties[3–6] as the spacecraft flies in orbits subjected to the radiation of protons and electrons with a wide energy spectrum in the Van Allen radiation belts.

In the past, the study of radiation effects of space-charged particles on optical materials focused on particles with energies over 1 MeV. But the flux of particles increases with their energy decrease in the Van Allen radiation belt, which consists principally of charged protons and electrons. Protons and electrons with energies below 200 keV have large fluxes, up to 10^8 particles/cm^2·s. The lower the particle energy is, the relatively shorter the penetration depth of particles into materials is. Therefore, the energy absorbed and deposition of particles in materials mainly concentrate in their surface layers. Consequently, the effects of proton radiation with such low energies on optical materials are important, especially for long-term spacecraft. The present study investigates the change of spectrum properties of JGS3 quartz glass induced by radiation of protons with 140 keV in an environment of vacuum and a heat sink and analyze the color center evolution kinetics.

II. Experiment

The experimental material is JGS3 optical quartz glass with impurity less than 5×10^{-3}%, in which OH groups are removed. The sample dimension is $20 \times 20 \times 2$ mm, and the surfaces are polished. The average transmittance of the samples in the region of 200–3200 nm is 93%.

The ground-based simulation of proton irradiation was performed in a high-atmosphere-ion simulator composed of a positive ion accelerator, an evacuated vacuum sample chamber, and a control and measuring system, which is shown in Fig. 1. The equipment can simulate irradiation with protons at 30–200 keV under a 10^{-4}-Pa vacuum and 77-K heat sink environment.[7]

In the simulation of proton space radiation, a proton energy of 140 keV is chosen, the beam density is 0.2 μA/cm^2, and the highest proton fluence is 2×10^{16} cm^{-2}, which is equal to the 7-year fluence in the Earth's radiation belts in the geomagnetic equator region, in which the proton spectra are shown in Fig. 2 (Refs. 8 and 9). The transmittance of the samples was analyzed using spectrophotometer UV-3101PC made by SHIMDZU before and after irradiation.

III. Results and Discussion

The change in transmittance spectrum of the JGS3 optical quartz glass irradiated with the protons is shown in Fig. 3. Curve 1 was the transmittance before radiation, whereas curves 2–5 showed the transmittance after radiation with different fluences. With increasing fluence, the transmittance of the quartz glass begins to decrease, and this effect gradually spread from the near ultraviolet to the visible region, but little influence is observed in the infrared region, as shown in Fig. 3a. Figure 3b shows that the obvious transmittance change occurs in the region of 220–250 nm.

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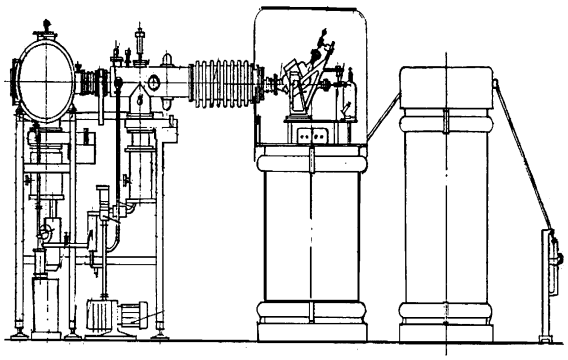


Fig. 1 Schematic diagram of positive ion accelerator.

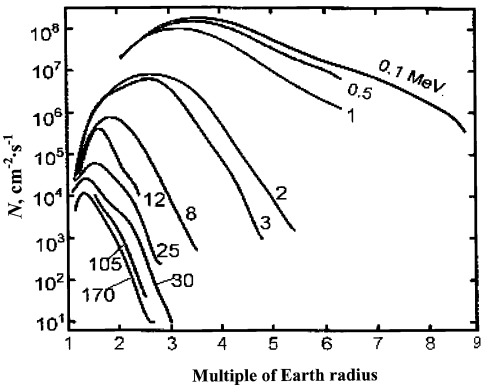
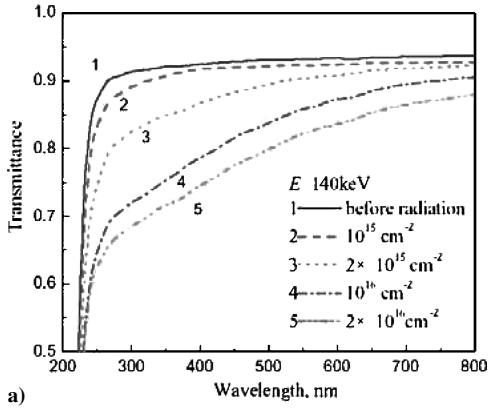


Fig. 2 Proton spectra of the Earth's radiation belts in geomagnetic equator region.[9]



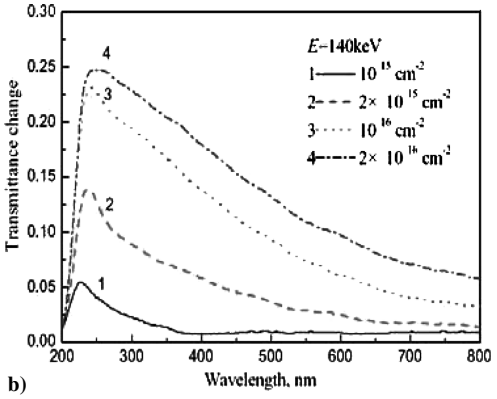


Fig. 3 JGS3 quartz glass before and after proton irradiation at 140 keV: a) spectral transmittance and b) transmittance change.

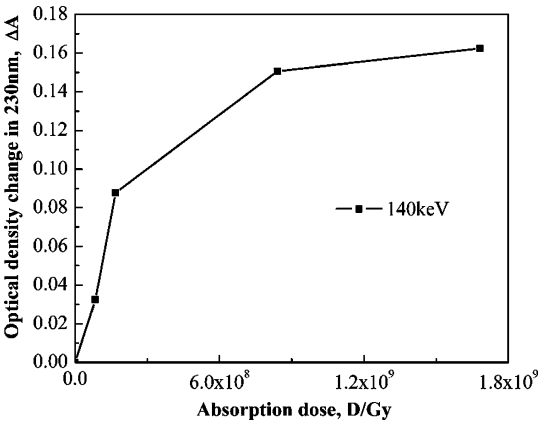


Fig. 4 Change in optical density at 230 nm with absorption dose in quartz glass after 140-keV proton radiation.

E′ centers are the primary radiation defects in the quartz glass.[9–11] The absorption peaks of the quartz glass at 220–250 nm can be related to the E′ centers and their variants.[12] The change of quartz glass optical density at 230 nm is discussed in this paper. Generally the degree of optical absorption of a material is indicated by the optical density, expressed as the logarithm of the ratio of the transmittance light intensity T to the incident light intensity T_0; that is,

$$A = \log(T/T_0) \tag{1}$$

The additional absorption by quartz glass in some spectra induced by proton radiation was determined by the absorption dose of quartz glass. The absorption dose and the radiation fluence are correlated with each other following the formula[13]

$$D = E\Phi/R\rho \tag{2}$$

Here D is the absorption dose in grays, E the particle energy in megaelectron volts, Φ the radiation fluence in protons per square centimeter, R the incident particle penetration depth in material in centimeters, and ρ the material density in grams per cubic centimeter.

Figure 4 shows the increase in optical density at 230 nm with the absorption dose for the samples irradiated with 140-keV protons. This reflects the kinetics of the change of optical absorption in the process of radiation. In the beginning the curves show a linear increase. When the absorption dose reaches a critical value, the change in optical density tends to level off. This trend might be related to a dynamic balance between the production and annihilation of color centers induced by the proton radiation.

IV. Color Center Formation Kinetics with Radiation Damage

A. Hypotheses

The radiation damage is a complicated physical and chemical process, which depends not only on the radiation dose D, dose rate J, and radiation energy E, but also on material category and defect distribution. According to the above analysis and some research works,[11,14] the following hypotheses can be proposed for discussing the radiation damage kinetics:

1) The potential trap defects of the color center can be inherent in quartz glass or generated by irradiation. The kind of color center is marked as i. Assume that $N_i(x)$ is the concentration of potential trap defects and indicates the distribution of defects along the incidence direction, which does not change during irradiation.

2) During irradiation, there are processes of generation and annihilation of color centers for ionization effect. The quantity dn_i is the number of potential trap defects converted to color centers per unit time dt. A constant fraction of potential trap defects f_i is converted to color centers per unit time, and q_i is the annihilation fraction of color centers during radiation.

3) The concentration of type i color centers $n_{i0} = 0$ when irradiation begins. Before irradiation, the absorption spectrum of quartz glass is regarded as the intrinsic or background absorption.

4) The inelastic ionization effect predominates for low-energy radiation. All of the secondary electrons or vacancies originated from the ionization are trapped by potential trap defects. Assume that σ is average electron trapped section within the concentrated layer that color centers occur in.

B. Deduction of Kinetic Model

Let J be the absorption dose rate and n_i the concentration of color centers at time t. According to the hypotheses, the growth rate of color centers is given by

$$\frac{\mathrm{d}n_i}{\mathrm{d}t} = \sigma J[f_i(N_i(x) - n_i) - n_i q_i] \tag{3}$$

If the concentration of defect traps N_i is constant, the equation can be solved as follows:

$$\begin{aligned} n_i &= n_{i0} + \left(\frac{f_i N_i(x)}{f_i + q_i} - n_{i0}\right)\{1 - \exp[-(f_i + q_i)\sigma J t]\} \\ &= n_{i0} + \left(\frac{f_i N_i(x)}{f_i + q_i} - n_{i0}\right)\{1 - \exp[-(f_i + q_i)\sigma D]\} \end{aligned} \tag{4}$$

Based on assumption (3), $n_i = n_{i0} = 0$ at $t = 0$, the concentration of color centers can be given as

$$\begin{aligned} n_i &= \frac{f_i N_i(x)}{f_i + q_i}\{1 - \exp[-(f_i + q_i)\sigma J t]\} \\ &= \frac{f_i N_i(x)}{f_i + q_i}\{1 - \exp[-(f_i + q_i)\sigma D]\} \end{aligned} \tag{5}$$

Equation (5) presents a relation between the absorption dose and the average concentration of color centers per unit area of surface and reflects the accumulation kinetics of color centers.

C. The Expression for Optical Properties

The relation between the integral areas of absorption curve and the oscillator strength and concentration of color centers can be expressed as[15]

$$nf^* = \frac{9mc}{2e^2h} \cdot \frac{n_Z}{\left(n_Z^2 + 2\right)^2}\alpha_m W = C \times \frac{10^{17} n_Z}{\left(n_Z^2 + 2\right)^2}\alpha_m W \tag{6}$$

where f^* is the oscillator strength and C is a constant.

According to this equation, when W is constant, the concentration of color centers is proportional to the absorption coefficient; that is, $n = \xi\alpha$. Meanwhile, there is a relation $\alpha = (2.303/d)A$ between the absorption coefficient and optical density, and thus the relation between optical density and concentration of color centers can be given by

$$A = 0.434 d\alpha = (0.434/\xi)dn = gdn \tag{7}$$

where ξ and g are scale factors. Because the effects of proton radiation with low energy mainly focus on the material's surface, the color centers concentrate in a very thin layer on the surface, and d is the thickness of this layer.

According to assumption (3), $n_{i0} = 0$. The concentration of color centers is proportional to the optical density change induced by radiation; that is,

$$\Delta A_i = A_i - A_{i0} = dg(n_i - n_{i0}) = dgn_i = A_i \tag{8}$$

Then, according to Eqs. (5), (7), and (8), the optical density change can be given by

$$\begin{aligned} \Delta A_i &= dg\frac{f_i N_i(x)}{f_i + q_i}\{1 - \exp[-(f_i + q_i)\sigma J t]\} \\ &= dg\frac{f_i N_i(x)}{f_i + q_i}\{1 - \exp[-(f_i + q_i)\sigma D]\} \\ &= \Delta A_{is}[1 - \exp(-\beta D)] \end{aligned} \tag{9}$$

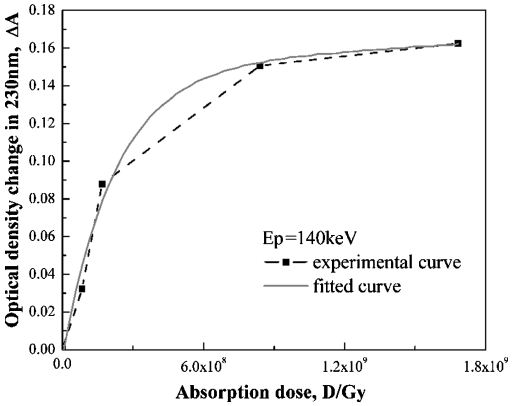


Fig. 5 Fitted curves for the change in optical density of quartz glass at 230 nm under proton radiation.

where $\Delta A_{is} = dg[f_i N_i(x) f_i + q_i]$; $\beta = (f_i + q_i)\sigma$. Because d is the thickness of the concentrated layer of color centers, Eq. (9) reflects the change in average optical density per unit area of sample surface.

According to the preceding analysis, under radiation with 140-keV protons, the optical density change of quartz glass can be fitted by the following function:

$$\Delta A = a[1 - \exp(-bx)] \tag{10}$$

Here the two parameters a and b, respectively, refer to ΔA_{is} and β in Eq. (9).

Figure 5 shows the mathematical fits of experimental curves. The parameters, fitted for the change in optical density at 230 nm, are proton energy, 140 keV; a, 0.16031; and b, 3.9385E-9. Comparing the experimental curve with the fitted one, the result indicates that the functions fitted from the kinetic model of color centers can approximately describe the optical property change for the quartz glass under proton radiation with low energy.

V. Conclusions

Space proton radiation with low energy has a high-flux character and results in obvious radiation damage effects on the surface layer of quartz glass. Initially, the change in optical density increases rapidly, and then a saturation trend appears with increasing absorption dose.

Based on analyzing experimental results, a kinetic model for the evolution of color centers in quartz glass irradiated with protons is proposed, and the change in average optical density per unit area of surface can be given by $\Delta A_i = \Delta A_{is}[1 - \exp(-\beta D)]$. The fitted curve is similar to experimental ones. It might be believed that the proposed kinetic model can be used in the quantitative description of the change in optical properties of quartz glass with increasing absorption dose under proton radiation with low energy.

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