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Effects of 80 KeV proton radiation on the optical properties and microstructure of type-GG17 glass as rubidium lamp envelope

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ABSTRACT

Effect of 80 KeV proton radiation with fluences of 10^{14}–2×10^{16} protons/cm^2 on the optical properties and microstructure of type-GG17 borosilicate glass as a candidate material for rubidium spectra lamp envelope was investigated. The change in microstructure before and after proton radiation was evaluated by means of UV–vis–NIR spectroscopy, X-ray photoelectron spectroscopy (XPS) and positron annihilation lifetime spectrometer (PALS). The experimental results show that under radiation of 80 KeV some broken Si–O bonds were induced and thus the number of non-bridge oxygen (NBO) increased. These irradiation-induced defects caused the degeneration of optical property and loosened glass structure which accelerates rubidium diffusion into lamp envelop.

1. Introduction

Borosilicate glass has been widely used in industry, especially as various nonpolar lamp-envelops. GG17 which is a type of model of borosilicate glass analogous to Pyrex in America is broadly manufactured in China. When this type of glass is used as the envelope material of rubidium spectral lamps [1] which are the light sources of rubidium atomic clocks for space application, it will be subjected to particle irradiation or ion implantation under space environment. It is thus of importance to study irradiation effects on the type-GG17 borosilicate glass.

The irradiation-induced defects in glassy materials have been extensively studied by various analytical methods. Degradation of spectral properties of glass under irradiation is well investigated by using UV–vis–NIR spectrophotometer [2–4] and the electron state of each atom is reported by XPS [5–8]. Positrons are sensitively trapped at defects and give us very useful information about their type, size and concentration. Positron annihilation in various materials has been extensively studied and positronium (Ps) formation and positron trapping at radiation-induced defects have been reported [9–13]. In addition, Ps related to network structure has been proved to be especially powerful for studies of microvoids in materials. Borosilicate glass has lower density (2.23 g/cm^3) than quartz and is considered to have structurally intrinsic microvoids. Ps is thus expected to be restricted in the microvoids and to convey useful information about the microvoids. Hence in the present work, radiation-induced damage and intrinsic defects in glasses are investigated by the combined measurements of UV–vis–NIR, XPS and PALS after the type-GG17 glass specimens are irradiated with protons of the energy 80 KeV at room temperature.

2. Experimental techniques

2.1. Glass composition

A commercially available type-GG17 glass in the form of slide was used in this work. The main composition of the glass (in

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wt%) as given by the manufacturer from China is as follows: SiO_2 (80.5), B_2O_3 (12.6%), R_2O (4.4%, R represents alkali elements, sodium and potassium) and Al_2O_3 (2%). Each glass slide has the dimension $20 \times 20 \times 2$ mm^3. The glass transition temperature is 560 °C and the density is 2.23 g/cm^3.

2.2. Irradiation procedure

The glass specimens were irradiated using a proton source simulator for the space proton radiation at the Space Materials and Environment Engineering Laboratory, Harbin Institute of Technology, China. Each sample was subjected to the same proton flux with 1.5×10^{12}/cm^2 s. The simulator type used was SCRM/HIT. Four proton fluences of 10^{14}, 10^{15}, 10^{16} and 2×10^{16}/cm^2 were used. The proton energy was chosen as 80 KeV. The specimen holder was placed in a vacuum clamber with the pressure of 10^{-4} Pa, and the temperature was kept at ~298 K.

2.3. Analysis methods

UV–vis–NIR spectra were used to characterize the glasses and to observe the irradiation-induced defects. A double beam spectrophotometer (Lamda 950 type, American Perkin Elmer) covering the wave range from 200 nm to 3200 nm was used. The optical source is deuterium. Percentage transmission spectra were recorded in the wavelength range 200–820 nm, using air as the reference. All measurements in this work were taken immediately after irradiation due to the thermal fading of the glass spectra.

XPS (PHI5700 ESCA) was used to analyze chemical bond structure of the glasses. Spectra were recorded using Al K_α source, operated at a power of 250 W and at constant pass energy of 187.85 eV for wide spectra and 29.35 eV for fine spectral. The vacuum of the sample chamber was under 10^{-6} Pa. A thin layer contamination carbon was smeared on the sample surface for the correction because of charging effect of the glass as a nonconductor. Peak fitting was performed by using a mixed Gaussian–Lorentzian function for obtaining peak position and FWHM. Peak areas were determined following Shirley's background subtraction method.

Positron annihilation lifetime (PAL) measurements were made in Institute of High Energy Physics, Beijing, China. The experiment was carried out in air at room temperature using a spectrometer employing fast–slow coincidence technique. The time resolution of the PALS was 194 ps. In the case of each sample, three time spectra were obtained with more than two million counts for each spectrum in order to guarantee good statistical performance. The ^{22}Na isotope, used as a positron source, with intensity about 10 μCi, is placed between two identical samples, forming a 'sandwich' system. The diameter of source spot has the size with about 1 mm. The resulting lifetime spectra were analyzed using a computer program LT9 and processed using three-term lifetime fitting method.

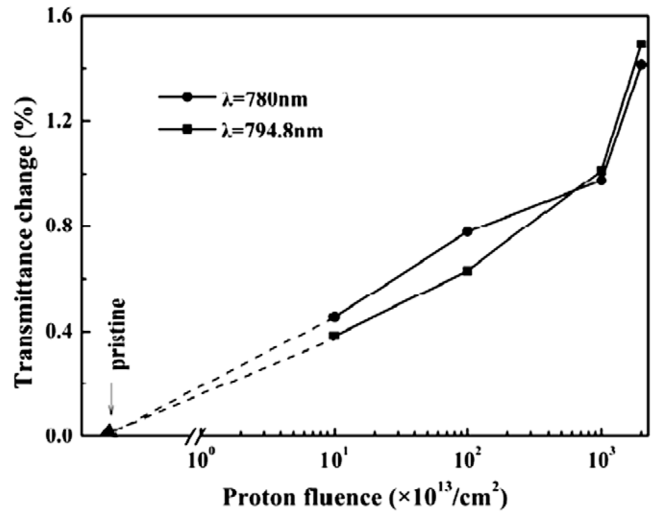

Fig. 2. Transmittance change versus proton fluence at the rubidium spectra D_1 and D_2.

3. Experimental results

3.1. Change in optical properties

For a given proton energy, the effect of different proton fluencies on the spectral transmission is shown in Fig. 1. The wavelength range 250–820 nm was only shown because the optical spectra under 250 nm were cut off for the investigated specimens. It is shown that the optical degradation mainly occurs in the visible region after the 80 KeV proton irradiation. With increasing the proton fluence, the intensity of the absorption band increases and the spectral transmission of glasses decreases. The absorption band shifts to longer wavelength (red shift) and widens due to the proton irradiation-induced weak absorption bands in long wave range. The transmittance decrement at the absorption peak comes to 12% and absorption bands are apparent after irradiation with proton fluence of 2×10^{16}/cm^2. The change in transmittance increases with increasing the fluence, showing that the optical properties are degraded gradually. Correspondingly, the surface color of spec-

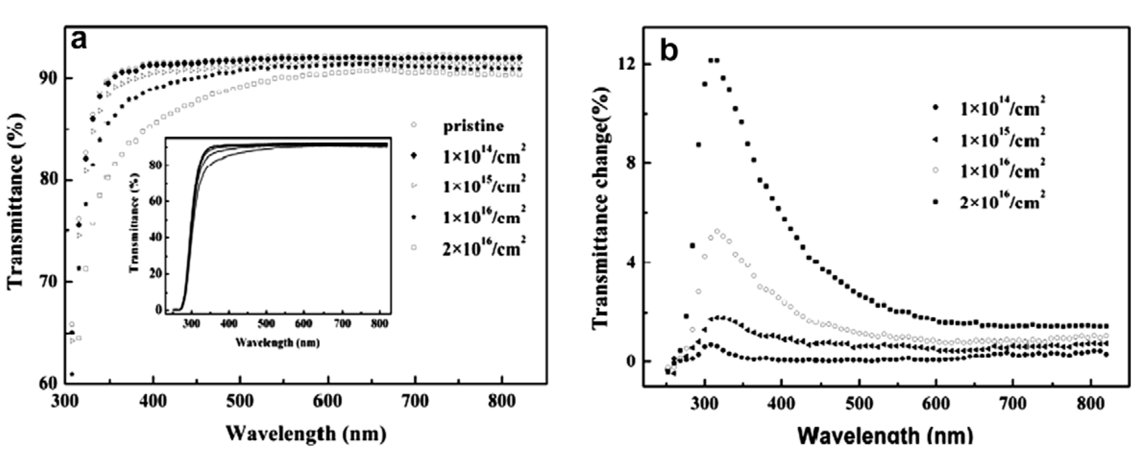

Fig. 1. The spectra transmittance (a) and its change (b) of specimens before and after proton irradiation.