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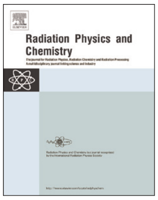

Effects of proton pre-irradiation on radiation induced conductivity of polyimide

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HIGHLIGHTS

- Radiation induced conductivity (RIC) of radiation damaged Kapton-H was studied.
- Proton pre-irradiation causes obvious decreasing of the degradation of RIC.
- Proton induced Displacement damage is the dominate effect on the RIC degradation.
- The degradation of pyromellitimide is the main factor to degrade the RIC.

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ABSTRACT

As an important parameter to evaluate the charge/discharge behaviors, radiation induced conductive behaviors of proton pre-irradiated polyimide films were investigated. The results indicate that the proton pre-irradiation results in decrease down to three orders in radiation induced conductivity (RIC) of polyimide as the proton (60–110 keV) fluence is just 1×10^{14} p/cm^2. Further analysis implies that, the dominate effect on the RIC degradation is proton pre-irradiation induced displacement damage, which results in the decomposition of pyromellitimide in polyimide.

1. Introduction

As a dielectric material, polyimide is widely used in space industry due to its excellent electric properties and thermal stability. However, the orbital proton and electron irradiations could result in ionization and displacement effects, inducing structural damage and property degradation in polymeric materials (Li et al., 2007). It is known that during the irradiation, ionization effect could also cause the increase of conductivity of dielectric material as the in-situ bias is on. This phenomenon is defined as Radiation Induced Conductivity (RIC). For the insulative materials, RIC is one of important parameters for charging/discharging analysis under the orbital particle irradiation environments (Rodgers et al., 1998; Robinson and Coakley,1992).

For RIC behaviors of polymers, Rose and Fowler proposed a model (named as R–F model) based on quasi-band theory to expose the dependence of transient and steady state of RIC on the radiation dose rate. In this model, the steady state RIC could be expressed as a power law to the irradiation dose rate in polymer (Rose, 1955; Fowler, 1956; Meyer et al., 1956; Gross et al., 1980; Faria, 1992; Gross et al., 1981). Based on RIC data of many kinds of polymers, Tyutnev et al. further developed a power dynamic mode to show the relationship of transient RIC with irradiation time under pulse/short irradiation (Tyutnev et al., 1979, 1984, 2001, 2004, 2006a, 2006b; Arkhipov et al., 1984; Siddiqui, 1984; Khatipov, 2001). Following R–F model, Faria found that, the dynamic RIC of polymer is a double exponential function of irradiation time under long time irradiation (Faria, 1992). However, these models provided just some experimental rules for the RIC evolutions without possibility to show a deep mechanism on the corresponding effects of polymer structure damage during irradiations. Moreover, some of results seem to be confused. In order to explain the dependence of the RIC on the polyimide structure, Kafafi et al. (1990) suggested the two-step photoconduction mechanism. Firstly, once the light was on, the electrons would be

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excited from the diphenyl ether moiety, which acts as a donor. As the electrons diffuse and react with the pyromellitimide moiety, which acts as an acceptor, the radical anions could be formed. These carriers may be recombined in the polyimide after irradiation. However, if there is a bias on the sample, the induced carriers would be transported in hopping model between the acceptor groups in the materials. In this case, the structural change could exert important effects on the RIC behaviors. It is known that, energetic particles irradiation could induce structural change in the polymer such as chain session and crosslinks. Thus, it is necessary to investigate the effects of proton irradiation on the structural damage and on the RIC behaviors in the polyimide. However, few literatures could be found to report RIC behavior in the damaged polymers so far.

In this paper, the RIC behaviors were studied for the damaged polyimide after 60, 80 and 110 keV proton pre-irradiation. The corresponding radiation-induced currents in proton pre-irradiated samples were measured under 140 keV electron irradiation in vacuum. The chemical state and structural change of the proton pre-irradiated polyimide were analyzed using X-ray photoelectron spectroscopy (XPS) measurements in order to discuss the mechanisms of RIC behaviors.

2. Experimental

2.1. Sample preparation and XPS measurement

Polyimide film (Kapton-H, Du-pont, Inc., USA) with a thickness of 25 μm was cut as specimens with a diameter of Φ 30 mm. The chemical structure was shown in Fig. 1. In order to investigate the effects of irradiation damage, the specimens were firstly pre-irradiated by proton in a ground-based complex irradiation simulator (Ukraine, Physics Institute of Cryogenic, Ukraine Academy of Science). The proton energy and flux were set at 60 keV, 80 keV, 110 keV and $2.5 \times 10^{11}/\text{cm}^2$ s, respectively. The maximum proton fluence was applied up to $1 \times 10^{16}/\text{cm}^2$ in this study. It should be noted that both sides of the samples were irradiated. Afterwards, for the conductivity measurements, the polyimide specimens were double sided deposited with 100 nm thick and 20 mm diameter of Au film as contacts using vacuum e-gun deposition technology. During the depositing process, vacuum is kept at 2×10^{-3} Pa, while the deposition rate was controlled as low as 1.5 nm/s in order to prevent sample heating.

XPS measurements were performed using PHI5700-type x-ray photoelectron spectroscopy with Al Kα excitation source (energy = 1486.6 eV), operated at a power of 250 W, and the vacuum of sample chamber is kept at 10^{-6} Pa.

2.2. In-situ measurements of radiation induced conductivity

For the radiation-induced current measurements, the proton pre-irradiated samples were electron irradiated in the simulator with an in-situ current measurement unit. The electron energy and flux were set at 140 keV and $6.25 \times 10^{11}/\text{cm}^2$ s, respectively. Hence, the tracking process and range of the incident electrons was calculated using a commercial Geant-4 code. The results indicate that the electron with energy of 140 keV could penetrate through the 25 μm Kapton specimens including the Au contacts, thus, the induced currents could be in-situ measured during the electron irradiation. In this study, a Keithley 2612-type double channel digital meter was used for measuring the sample currents under a bias of 200 V.

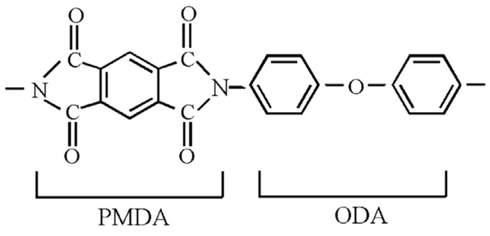

Fig. 1. Chemical structure of Kapton-H.

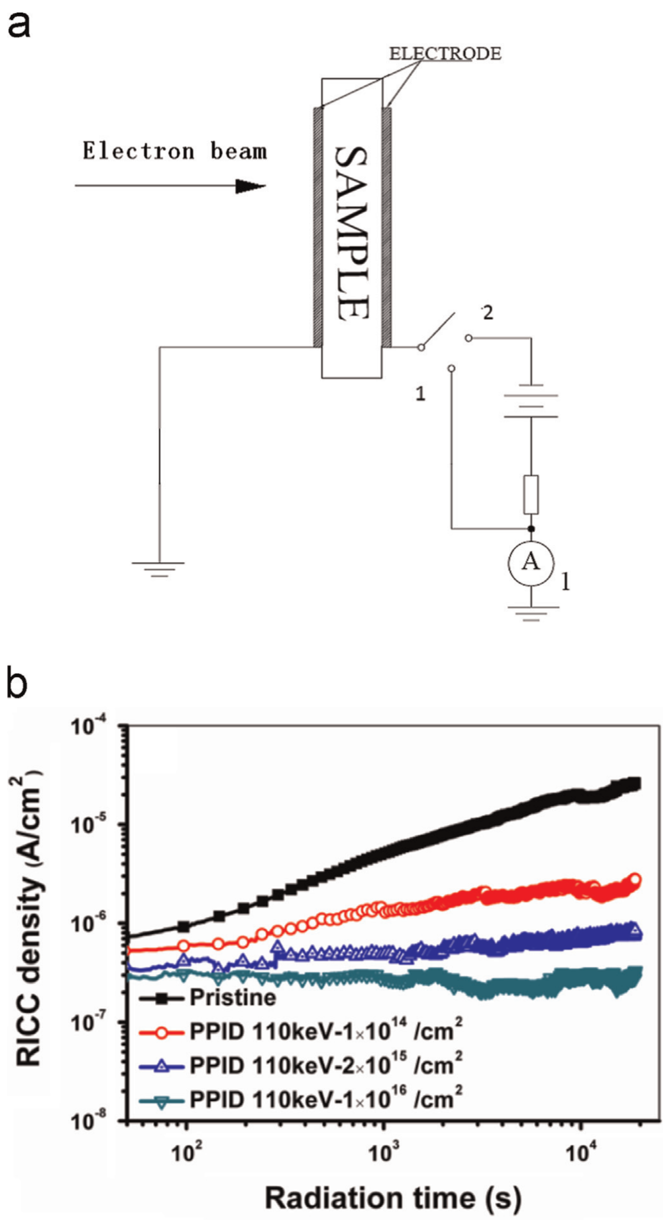

Fig. 2. (a) Schematic diagram of experimental setup for in-situ RIC measurement of polyimide [21]; (b) Evolution of Radiation Induced Conductive Current (RICC) density of 110 keV proton pre-irradiated polyimide flims.

Fig. 2(a) shows the schematic diagram of experimental setup for the radiation-induced current measurement in polyimide specimens (Yue et al., 2012). Wherein, the switches 1 and 2 were designed to obtain the precise radiation-induced currents through removing the transparent and scattering currents of the incident electrons. As the 1 tip switches on, the bias voltage between the two contacts on the sample is 0, but one could measure current (I_{st}) from the transparent and scattering electron during the irradiation. When the 2 tip switches on, there is a 200 V bias between the sample poles, one could get the total current $I_{measure}$ during