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Degradation of polyimide under irradiation with swift heavy ions

D. Severin [a,*], W. Ensinger [a], R. Neumann [b], C. Trautmann [b], G. Walter [b], I. Alig [c], S. Dudkin [c]

[a] *Department of Chemistry, Philipps-University Marburg, Hans Meerwein Street, D-35043 Marburg, Germany*
[b] *Gesellschaft für Schwerionenforschung (GSI), Planckstreet 1, D-64291 Darmstadt, Germany*
[c] *Deutsches Kunststoff-Institut (DKI), Schlossgartenstreet 6, D-64289 Darmstadt, Germany*

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Abstract

Stacks of polyimide foils were irradiated with different swift heavy ions (Ti, Mo, Au) of 11.1 MeV/nucleon energy and fluences between 1×10^{10} and 2×10^{12} ions/cm^2. Beam-induced degradation of the imide group was analyzed by Fourier-transform infrared spectroscopy studying the absorption band at 725 cm^{-1} as a function of dose. In the UV–Vis spectral range, the absorption edge is shifted to larger wavelengths indicating carbonization. Such modifications are linked to the deposition of a critical dose of 2.7 MGy (Ti) and 1 MGy (Mo, Au). In addition, irradiation-induced changes of the electrical conductivity were studied by means of dielectric spectroscopy.

Keywords: Polyimide; Polymer degradation; Swift heavy ion irradiation

1. Introduction

The interaction of energetic ion beams and polymers has been studied by numerous groups investigating radiation-induced degradation and track formation by means of spectroscopy (e.g. infrared, UV–Vis, Raman), high-resolution microscopy (e.g. scanning force and transmission electron microscopy) and others techniques [1–9]. Also chemical etching of ion tracks has been analyzed in great detail, in order to better control the fabrication of nanoporous ion-track membranes and thus enabling new applications [10–12].

In this study we concentrate on radiation-induced ageing effects of polymers when used as insulating material. The experimental data presented here are the first results of a larger research

* Corresponding author. Tel.: +49 6421 2825596; fax: +49 6421 2822124.
E-mail address: severin@chemie.uni-marburg.de (D. Severin).

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project which is motivated by the future plans of GSI (Darmstadt, Germany) for an International Facility for Antiproton and Ion Research (FAIR). The facility will consist of a 100/200-Tm double-ring synchrotron SIS 100/200 and a system of associated storage rings. The present GSI accelerators (UNILAC and SIS 18) will be used as injectors and particle beams from protons and antiprotons, radioactive ions, up to uranium projectiles of extremely high intensities (10^{12} ions/s) and energies (up to ~20 GeV/nucleon for U^{92+}) will be provided [13]. The synchrotron rings will consist of novel, rapidly cycling superconducting magnets that are presently being designed and developed [14,15]. During long-term operation of the accelerators, some of the insulating parts of these new magnets will be exposed to high-dose radiation of X-rays, gammas, neutrons and charged particles at low temperatures (5 K). In a first step, we are investigating radiation-induced ageing of insulator films of polyimide by exposing them to heavy ions at room temperature. The experiments were performed with Ti, Mo and Au of several MeV per nucleon (MeV/u), an energy regime where the stopping powers are close to the Bragg maximum. The modifications induced were characterized by infrared, UV–Vis and dielectric spectroscopy.

2. Experimental

2.1. Material and irradiation

The polyimide samples were commercial Kapton™ foils from DuPont of thickness 12 μm, 25 μm and 50 μm. We composed stacks of three foils (size 5×5 cm^2, sequence 25, 50, 12 μm) and irradiated them under vacuum with ^{50}Ti, ^{98}Mo and ^{197}Au ions of initial energy 11.4 MeV/u at the UNILAC linear accelerator of GSI. The applied fluence varied between 1×10^{10} and 2×10^{12} ions/cm^2 with the beam flux limited to 2×10^8 ions/cm^2 s to avoid sample heating. The fluence was measured by monitoring the signal from a secondary-electron emitting Al-foil detector placed in front of the samples and calibrated via a Faraday cup (precision ~20%). The energy loss (dE/dx) of the ions when entering the foil stack varies between 2.2, 6.6 and 14.4 keV/nm for Ti, Mo and Au, respectively [16]. The projected range of the different projectiles in Kapton is between 140 and 170 μm, and thus exceeds by far the foil thickness. We therefore regard the mean dE/dx in a given sample to be homogeneous, the corresponding dose D deposited is determined by:

$$D = 1.6 \times 10^{-10} \times E \times \Phi/(\rho \times d),$$

where the dose D is given in Gy, E is the total ion energy in MeV, Φ denotes the fluence in ions/cm^2, ρ = 1.43 g/cm^3 is the mass density, and d is the foil thickness in cm.

2.2. Infrared and UV–Vis spectroscopy

After irradiation, the 12 μm thick Kapton foils (third position in stack) were characterized by Fourier transform infrared (FTIR) and UV–Vis spectroscopy. The FTIR spectra were recorded with a Nicolet Magna-IR 550 spectrometer (spectral resolution of 2 cm^{-1}) in the transmission mode. The absorption bands were analysed with the baseline method determining the absorbance of the centroid height.

2.3. Dielectric spectroscopy

To check the insulating properties of irradiated Kapton foils, dielectric spectroscopy was performed by recording the electrical conductivity [17–19]. To assure good electrical contact, the two surfaces of 50 μm thick foils (diameter 2 cm) were sputter-coated with gold. The spectra were measured at room temperature in a frequency range from 1 MHz to 0.1 Hz by using a frequency response analysis system consisting of a computer-controlled Solartron SI 1260 Impedance/Gain-Phase Analyzer and a Novo-control broadband dielectric converter.

3. Results

3.1. Infrared spectroscopy

The infrared spectra of Kapton before and after the irradiation with Mo ions (2×10^{12} ions/cm^2) is