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Radiation damage in polyvinyltoluene (PVT)

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Abstract

Polyvinyltoluene (PVT) is an organic polymer which is luminescent when it is irradiated by ionizing radiation. The luminescence yield is proportional to the radiation stopping power and to the absorbed dose. High stopping power radiations, such as ion beams, induce radiation damage in PVT with the production of C−H bond break, dehydrogenation and free radical formation, as investigated by the mass quadrupole spectrometry. The damage reduces the luminescence yield significantly. A reduction of about 50% is obtained with 300 keV proton and argon ions, having 300 keV energy, at a dose of about 10¹⁴ and 10¹³ ions/cm², respectively. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Polyvinyltoluene; Luminescence; Quenching; Scintillator

1. Introduction

Polyvinyltoluene (PVT) is an organic polymer with excellent properties of light scintillation. The monomer contains a benzenic ring bonded to a methylic group $-CH_3$ and to a vinylic group CH_2-CH- . It is employed in nuclear physics as a detector of ionizing radiation. Its luminescence is a function of the radiation stopping power and absorbed dose. At high stopping powers, quenching effects due to non-radiative deexcitations, produce a luminescence saturation (Torrisi et al., 2000). Recently, thin PVT films are used in radiotherapy as relative dosimeter for light particles at high-energy (Beddar, 1994).

The radiation energy released to the PVT not only excites the luminescence centers but also breaks chemical bonds modifying drastically the polymer properties at high incident doses. The C-H bond break produces free radicals and hydrogen degassing from the polymer. The residual material becomes rich in carbon content, as a typical trend of many hydrogenated polymers (Venkatesan et al., 1987).

In this work, the radiation damage in PVT induced by particles at different stopping powers and ion doses

*Tel.: +39-090-6765052; fax: +39-090-395004. *E-mail address:* lorenzo.torrisi@unime.it (L. Torrisi). is investigated. Obtained results for ion beams are presented and discussed.

2. Experimental

The investigated organic polymer is chemically represented by $(C_9H_{10})_n$ monomer chains, has a density of $1.032\,\mathrm{g/cm^3}$, shows an emission light in the visible region with a maximum at 423 nm wavelength, has a luminescence decay time of 2.5 ns and an attenuation length of 250 cm. Table 1 reports the main physical properties of the polymer.

Radiation damage in PVT samples has been obtained by irradiation with different ions at energies ranging between 50 and 400 keV. Protons and helium beams have been employed for their low stopping powers, and argon and xenon beams for their high stopping powers. The irradiation has been performed at the Ion Implanter Laboratory (IIL) of the Physics Department of Catania University. Irradiation occurs in vacuum (10⁻⁷ Torr) in a chamber provided by a mass quadrupole spectrometer (MQS) to detect "in situ" the species desorbed from the polymer.

Fig. 1 shows a scheme of the experimental set-up. The MQS spectrometer detects in the range 1–100 amu.

Table 1 Physical characteristics of the polyvinyltoluene scintillator

Monomer	C ₉ H ₁₀
Effective mass number	0.542
Density (g/cm ³)	1.032
Maximum light emission (nm)	423
Refraction index	1.58
Attenuation length (cm)	250
Specific heat (J/g °C)	1.7
Softening temperature (°C)	70

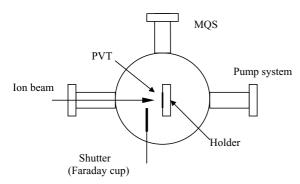


Fig. 1. Experimental setup.

In order to investigate the radiation damage produced in PVT as a function of the stopping power and ion dose, the luminescence yield induced by proton beams has been detected. Proton beams have been accelerated up to 60 MeV at PSI Laboratory of Zurich, at 24 MeV at INFN-LNS of Catania and within 50–350 keV at IIL of Catania.

The luminescence yield has been detected coupling the scintillator to a photo-multiplier (Hamamatsu H5783) having the maximum sensitivity at 430 nm and monitoring the output signal with a fast amplifier, ADC converter and MCA multi-channel analyzer, as discussed in the previous paper (Torrisi et al., 1997a).

The PVT emission spectra have been investigated as a function of the irradiation stopping power and an ion dose using a spectrophotometer excited at 300 nm wavelength and recording the luminescence in the wavelength region 300–600 nm.

Stopping powers and ion ranges have been calculated using the computerized TRIM simulation programs (Ziegler et al., 1985).

3. Results and discussion

Fig. 2 reports the experimental results about the luminescence yield as a function of proton stopping

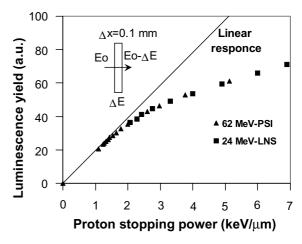


Fig. 2. Luminescence yields as a function of the proton stopping powers.

powers. At about $2.5 \, keV/\mu m$ stopping, saturation occurs, as a result of significant quenching phenomena due to non-radiative de-excitations (Torrisi, 1998).

PVT shows a strong dehydrogenation and an emission of different $C_x H_y$ groups with the absorbed dose. Fig. 3 reports a mass spectra comparison for the main ejection occurring at masses 2, 26 and 41, corresponding to the emission of H_2 , C_2H_2 and C_3H_5 , respectively. The ejection of mass 2 is about one order of magnitude higher than mass 26 and about 25 times higher than mass 41. The kinetics of molecular emission of masses 26 and 41 is slower and it appears very different with respect to the mass 2 emission, which is very fast. From the transient signal, obtained just after the beam switch off, it is possible to observe that by increasing the detected mass the MQS delay time increases.

These results demonstrate that a high rate of formation and recombination of free radicals is produced along the ion path in the polymer and a diffusion process of the recombined species occurs towards the polymer surface.

Using the MQS calibration procedure, as given in a previous paper (Torrisi, 1999), has been possible to evaluate the ejection yield in terms of the number of ejected molecules per 100 eV incident energy (chemical yield). The experimental chemical yields are 1, 0.1 and 0.03 molecules/100 eV, for masses 2, 26 and 41, respectively.

The residual polymer contains more and more carbon with the irradiation dose. Generally, a dose of $10^{14} \, \text{ions/cm}^2$ of $100 \, \text{keV}$ helium beam produces a black color of the polymer. In terms of the absorbed dose this value corresponds to about $10 \, \text{MGy}$.

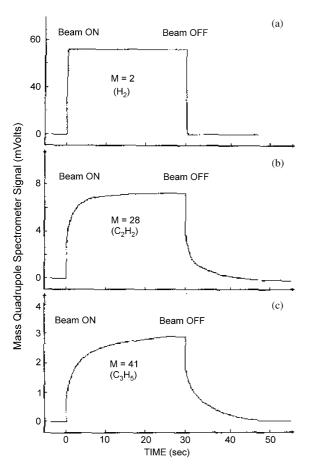


Fig. 3. MQS spectra relative to the kinetics of masses 2, 26 and 41, at the beam ON and beam OFF irradiation switches.

The radiation damage effect increases with the stopping power of the incident particle and with the ion dose. The damage produces a decrease in luminescence. For 60 MeV proton irradiation (stopping power $1.1\,\text{keV}/\mu\text{m}$), this reduction is about 15% at a dose of $1\,\text{kGy}$. The same decrement is obtained with $300\,\text{keV}$ protons (stopping power $65.8\,\text{keV}/\mu\text{m}$) at about $60\,\text{kGy}$ dose and with $300\,\text{keV}$ argon (stopping power $705\,\text{keV}/\mu\text{m}$) at about $500\,\text{kGy}$ dose.

Fig. 4 shows a comparison between the emission spectra from PVT as a function of the irradiation dose for 300 keV protons and 300 keV argon beams. All spectra are induced by a 300 nm excitation wavelength.

In conclusion, ion irradiation of PVT drastically produces polymer modifications. Damage appears at doses of the order of $10^{12} \, \text{ions/cm}^2$, at which a low hydrogen desorption occurs. It becomes predominant at doses of about $10^{13} \, \text{ions/cm}^2$, at which hydrogen and $C_x H_y$ groups are ejected. At higher values, corresponding to MGy absorber doses, the polymer transforms in hydrogenated amorphous carbon and loosens its lumi-

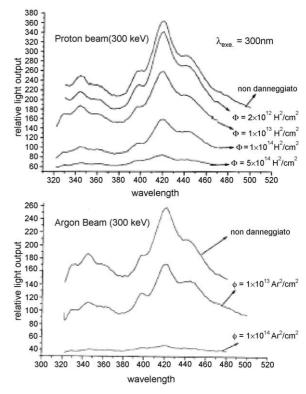


Fig. 4. Spectra comparison of PVT wavelength emission as a function of the implanted dose for proton and argon ions.

nescence characteristics, according to the literature (Torrisi et al., 1997b).

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