Effect of Alpha-Particle Energies on CR-39 Line-Shape Parameters using Positron Annihilation Technique

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Polyally diglycol carbonate "CR-39" is widely used as etched track type particle detector. Doppler broadening positron annihilation (DBPAT) provides direct information about core and valance electrons in (CR-39) due to radiation effects. It provides a non-destructive and non-interfering probe having a detecting efficiency. This paper reports the effect of irradiation α-particle intensity emitted from ^{241}Am (5.486 MeV) source on the line shape S- and W-parameters for CR-39 samples. Modification of the CR-39 samples due to irradiation were studied using X-ray diffraction (XRD) and scanning electron microscopy (SEM) techniques.

1 Introduction

Polyallyl diglycol carbonate ($C_{12}H_{18}O_7$, $\rho = 1310$ kg/m^3) is a thermoset polymer [1]. Polyallyl diglycol carbonate, CR-39, has been used in heavy ion research such as composition of cosmic rays, heavy ion nuclear reactions, radiation dose due to heavy ions, exploration of extra heavy elements etc. Its availability in excellent quality from different manufactures is also an advantage for further applications [1].

Swift heavy ions (SHI) produce permanent damage in polymeric materials as latent tracks along their path due to dissociation of valence bonds, cross linking and formation of free radicals [2, 3].

Positron Annihilation Technique (PAT) has been employed for the investigating Polymorphism in several organic materials [4] and it has emerged as a unique and potent probe for characterizing the properties of polymers [5]. In PAT, the positron is used as a nuclear probe which is repelled by the ion cores and preferentially localized in the atomic size free-volume holes [6] of the polymeric material. The motion of the electron-positron pair causes a Doppler shift on the energy of the annihilation radiation. As a consequence, the line-shape gives the distribution of the longitudinal momentum component of the annihilating pair. Positron Annihilation Doppler Broadening Spectroscopy (PADBS) is a well established tool to characterize defects [7]. The 0.511 MeV peak is Doppler broadened by the longitudinal momentum of the annihilating pairs. Since the positrons are thermalized, the Doppler broadening measurements provide information about the momentum distributions of electrons at the annihilation site.

Essentially all prior Doppler broadening measurements [8, 9] have been performed using either slow positron beams or wide-energy-spectrum positron beams from radioactive sources. Two parameters S (for shape), and W (for wings) [10] are usually used to characterize the annihilation peak. The S-parameter is more sensitive to the annihilation with low momentum valence and unbound electrons. The S-parameter defined by Mackenzie et al. [11] as the ratio of the integration over the central part of the annihilation line to the total integration. Diffraction peaks are analyzed through common fitting procedures, which result in parameters like the center of gravity and the width of the distribution. The W-parameter is more sensitive to the annihilation with high momentum core electrons and is defined as the ratio of counts in the wing regions of the peak to the total counts in the peak.

Fig. 1 shows Doppler broadening line-shape from which the S- and W-parameters are calculated using the following equations:

$$S = \frac{\displaystyle\int_{xc-g1}^{xc+g1} y(x)dx}{area}\,,$$

$$W = \frac{\displaystyle\int_{xc-g3}^{xc-g2} y(x)dx + \int_{xc-g2}^{xc-g3} y(x)dx}{area}\,,$$

where $area = \int_{g\,\min}^{g\,\max} y(x)dx$, and xc is the center of the peak.

In this regard, the main goal of the positron annihilation technique experiments is to point out the CR-39 line-shape parameters resulting from the effect of α-particle energies.

2 Experimental technique

Track detectors "CR-39" were normally irradiated in air by different α-particle energies with different fluxes from 1476.42 particles/cm^2 at 1.13 MeV to 48130.25 particles/cm^2 at 4.95 MeV from 0.1 μCi ^{241}Am source. Collimators of different thickness were used to change the α-particle energy.

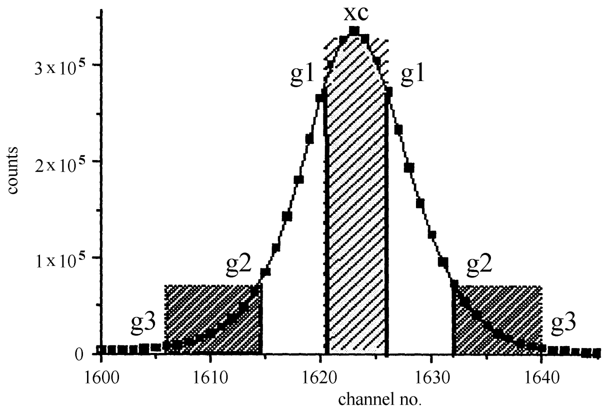


Fig. 1: Definition of the S- and W-parameters [12] (note, that the limits $g1$, $g2$, $g3$ are arbitrary to a certain degree, but have to be the same for all annihilation lines analyzed).

After irradiations, the samples were etched in 6.25 M NaOH solution at 70°C for 6 hr.

The simplest way to guide the positrons into the samples is to use a sandwich configuration as shown in Fig. 2. ^{22}Na is the radioactive isotope used in our experiment.

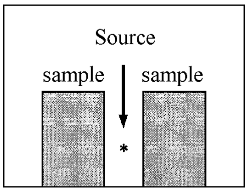


Fig. 2: Sandwich configuration of the positron source respect to a pair of specimen.

The positron source of 1mCi free carrier ^{22}NaCl was evaporated from an aqueous solution of sodium chloride and deposited on a thin Kapton foil of 7.5 μm in thickness. The ^{22}Na decays by positron emission and electron capture (E. C.) to the first excited state (at 1.274 MeV) of ^{22}Na. This excited state de-excites to the ground state by the emission of a 1.274 MeV gamma ray with half life $T_{1/2}$ of $3{\times}10^{-12}$ sec. The positron emission is almost simultaneous with the emission of the 1.274 MeV gamma ray while the positron annihilation is accompanied by two 0.511 MeV gamma rays. The measurements of the time interval between the emission of 1.274 MeV and 0.511 MeV gamma rays can yield the lifetime τ of positrons. The source has to be very thin so that only small fractions of the positron annihilate in the source.

The system which has been used to determine the Doppler broadening S-and W-parameters consists of an Ortec HPGe detector with an energy resolution of 1.95 keV for 1.33 MeV line of ^{60}Co, an Ortec 5 kV bais supply 659, Ortec amplifier 575 and trump 8 k MCA. Fig. 3 shows a schematic diagram of the experimental setup. Doppler broadening is caused by the distribution of the velocity of the annihilating electrons in the directions of gamma ray emission. The signal coming from the detector enters the input of the preamplifier and the output from the preamplifier is fed to the amplifier. The input signal is a negative signal. The output signal from the amplifier is fed to a computerized MCA. All sample spectra are acquired for 30 min.

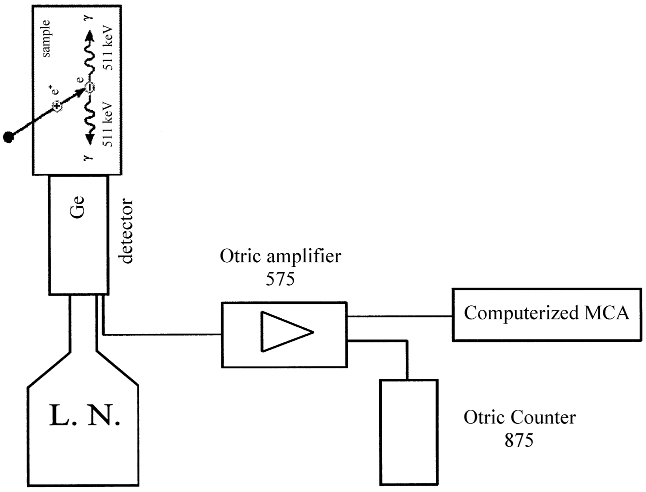


Fig. 3: Block diagram of HPGe-detector and electronics for Doppler broadening line-shape measurements.

3 Results and discussion

3.1 Positron annihilation measurements

Fig. 4 shows the Doppler broadening line shape parameters measured for unirradiated and irradiated CR-39 samples at α-particle energies of 2.86 and 4.86 MeV. The measured line-shape profiles reveal similar line-shape counts for samples (unirradiated and irradiated with α-particle energy, i. e. 4.86 MeV). A minimum line-shape counts are obtained at 2.86 MeV. The other observation is that the Full Width at Half Maximum (FWHM) for 2.86 MeV irradiated sample is more broadening than others. From such behavior it is clear that either something happened during irradiation with 2.86 MeV and it recovers again at higher energies or some kind of transition occurs at 2.86 MeV of α-particle energy.

The Doppler broadening line-shape S- and W-parameters are calculated using SP ver. 1.0 program [13] which designed to automatically analyze of the positron annihilation line in a fully automated fashion.

The S- and W-parameters calculated using the previous program were correlated as a function of α-particle energy with different fluxes deposit into CR-39 detector, the results are illustrated in Fig. 5. The S-parameters has values around 46% while values of about 15% are obtained for W-parameters. An abrupt change definitely observed at irradiation energy 2.86 MeV of α-particles for both S- and W-parameters. At this energy a drastically decrease in the S-parameter comparable with a drastically increase in the W-parameter. Values of about 35% and 28% were observed for the S- and W-parameters respectively at 2.86 MeV of α-particle energy.

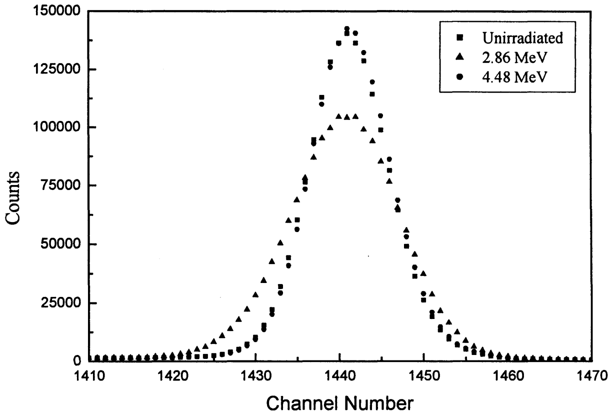


Fig. 4: The line shape spectra of the unirradiated sample and irradiated with α-particle energies of 2.86 and 4.84 MeV.

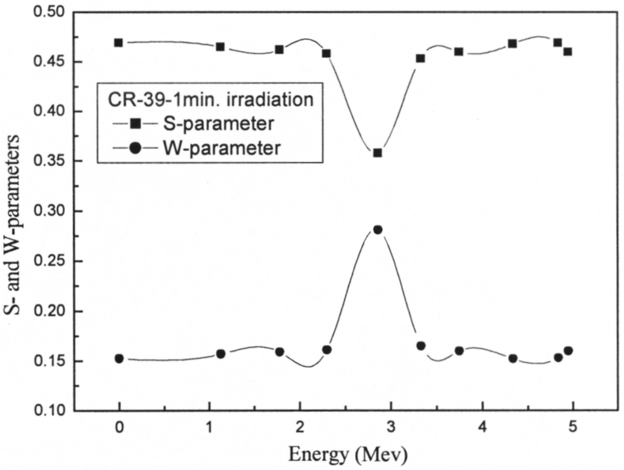


Fig. 5: The behavior of the S- and W-parameters as a function of α-particle energies.

A high concentration of defects, or an increase in the mean size of defects, leads to a larger contribution of annihilation photons from low momentum electrons because positrons are trapped at defects [14]. This is reflected in Doppler broadening measurements by an increase in S-parameter and a decrease in W-parameter. The behavior of S- and W-parameters reveal an abrupt change at the position of the transition. The behavior of the line-shape S- and W-parameters can be related to the different phases. Like many others molecular materials, the use of PAT also proven a very valuable in the study of phase transition in polymers. The same results have been obtained by Schiltz et al. [15]. Walker's et al. [4] measurements have indicated the conversion of one polymorphism to another. Srivastana et al. [16] have investigated polymorphic transitions in DL-norlevcine and hexamethyl benzene.

The transitions in the crystalline phase are related to the lattice transformation from monoclinic to hexagonal and setting in of torsional oscillations in the polymer chain.

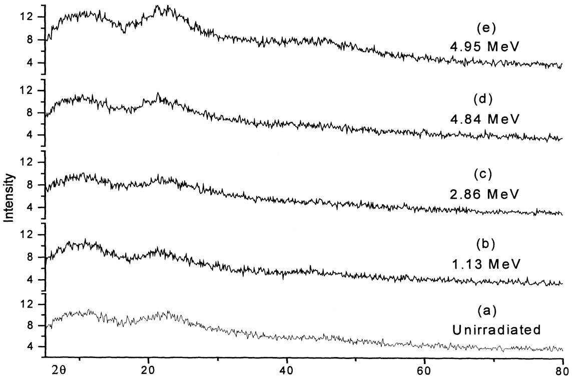


Fig. 6: X-ray diffraction pattern of "CR-39" Polyallyl diglycol carbonate.

3.2 X-ray diffraction pattern (XRD) and Scanning Electron Microscopy (SEM)

The X-ray diffraction analysis was used to obtain information about the transformation as a result of change in α-irradiation intensity. The XRD intensity measurements as a function of diffraction angle (2θ) for unirradiated sample and samples irradiated at different α-particle energies are shown in Fig. 6.

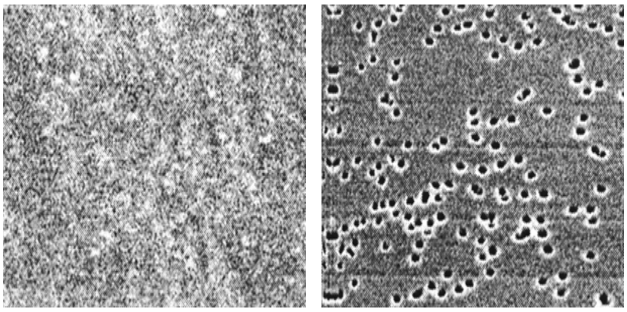
From the X-ray charts it is observed that, an increase in the intensity is obtained at higher α-irradiation intensity 4.84 and 4.95 MeV. At these energies, the XRD chart reveals a new peak that start to appear at 2.86 MeV α-particle energy. The one prominent X-ray peak is located at $2\theta = 21.5^\circ$ and it grows up with increasing α-particle energy. The appearance of this peak might be related to phase transition.

A number of papers on the study of polymer show that the amorphous state is altered by structural relaxation and crystallization processes. Positron annihilation behavior in the amorphous state has been described both in terms of topological short range ordering (TSRO) and chemical short-range ordering (CSRO) at the basis of the structural relaxation mechanisms [11, 15, 17–19]. During crystallization the positron behavior is determined by the phase diagram of the amorphous and crystallized system. On our X-ray diffraction patterns might be the first sign of the crystallization onset appears at 2.86 MeV. This sign is increased at higher α-particle energies as shown in the Fig. 6.

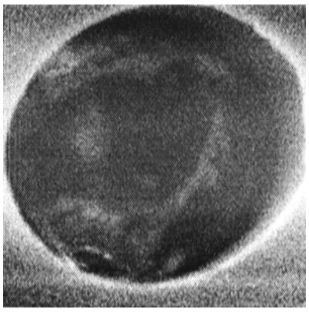
The SEM images taken for unirradiated and irradiated CR-39 samples at 4.84 MeV with magnification of 500 are shown in Fig. 7a and b. Tracks are obtained as a result of exposure of α-particle energy. A different magnified (15000) image for one track is shown at Fig. 7c. Cumbrera et al. [19] showed that rings of the structure (metastable structure) were already present in the scanning electron micrographs.

4 Conclusion

Doppler broadening positron annihilation (DBPAT) provides direct information about core and valance electrons in CR-



(a) Unirradiated ($M = 500$) (b) 4.84 MeV ($M = 500$)



(c) 4.84 MeV ($M = 15000$)

Fig. 7: SEM for unirradiated sample and irradiated sample at 4.84 MeV α-particle energies for different magnifications

39 due to radiation effects. The behavior of the S- and W-parameters supports the idea that positrons are trapped by defects and inhomogeneities inherently present in the as-received CR-39 polycarbonate. The annihilation characteristics of positrons are very sensitive to phase transitions. The phase transition in the CR-39 polycarbonate remain complex. XRD pattern and SEM technique of polymers studied in the present work clearly show crystalline and amorphous regions in the samples.

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