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Positron annihilation lifetime spectroscopy study of Kapton thin foils

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Abstract
Variable energy positron annihilation lifetime spectroscopy (VE-PALS) experiments on polyimide material Kapton are reported. Thin Kapton foils are widely used in a variety of mechanical, electronic applications. PALS provides a sensitive probe of vacancy-related defects in a wide range of materials, including open volume in polymers. Varying the positron implantation energy enables direct measurement of thin foils. Thin Kapton foils are also commonly used to enclose the positron source material in conventional PALS measurements performed with unmoderated radionuclide sources. The results of depth-profiled positron lifetime measurements on 7.6 μm and 25 μm Kapton foils are reported and determine a dominant 385(1) ps lifetime component. The absence of significant nanosecond lifetime component due to positronium formation is confirmed.

Keywords: positron annihilation, polymers, positron annihilation lifetime spectroscopy

(Some figures may appear in colour only in the online journal)

1. Introduction

The polyimide material Kapton belongs to a group of polymers that exhibit excellent chemical resistance, mechanical, thermal and dielectric properties, and are used in a diverse range of applications including dielectrics, nonlinear optical materials, and membranes [1]. The low dielectric constant, high dielectric strength, and high resistivity make polyimide materials suitable for a range of insulation applications [2, 3]. Positron annihilation lifetime spectroscopy (PALS) is capable of providing information on the size distribution of open volume in polymeric systems, and on vacancy-related point defects in inorganic materials and metals. Free volume in polymers is normally characterised by detecting the lifetimes and intensities of 'pick-off' annihilation events caused by the interaction of positronium (Ps), formed by positron implantation within open volume sites, with the cavity walls. This process can be analysed using a simple quantum mechanical model and information of cavity sizes determined [4, 5]. However, in contrast to the majority of polymers the Ps pick-off lifetime component, typically in the range ~1–30 ns, is absent in Kapton. This absence, combined with its excellent chemical and mechanical properties, contribute to make thin Kapton foils ideal supporting substrates for the deposition of radionuclide sources for various positron annihilation experiments on materials, in particular PALS measurements on bulk samples.

The most commonly used radionuclide is ^{22}Na which emits positrons with a β-spectrum of energies, up to a maximum of 0.546 MeV. In consequence these are implanted to range of depths within the material under study. PALS measurements performed using a Kapton foil enclosed ^{22}Na source require detailed knowledge of the positron lifetime spectrum resulting from the small fraction of positrons implanted into the foil [6, 7]. The PALS spectrum is dominated by the contributions from positrons annihilating within the material under study. However, the inverse problem decomposition of these lifetime components due to the material is sensitive to the subtraction of the extrinsic annihilation events, the source correction terms. Here we report the direct measurement of the positron

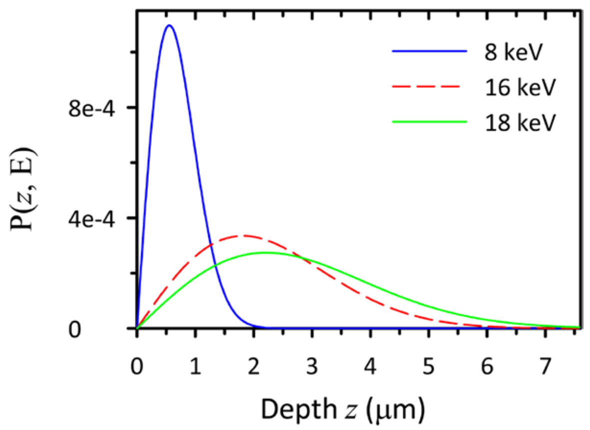

Figure 1. Makhovian positron implantation profiles for Kapton determined using the empirical parameters $A = 2.81\ \mu g \cdot cm^{-2}\ keV^{-n}$, $m = 2$, and $n = 1.71$, and the density of $1.42\ g \cdot cm^{-3}$.

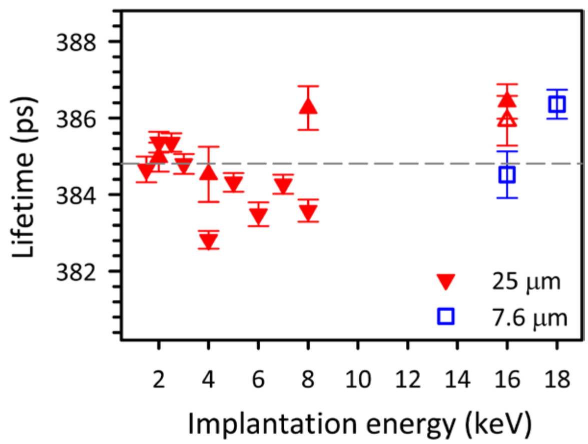

Figure 2. The positron lifetime value for the dominant component of VE-PALS measurements on 7.6 μm (open squares) and 25 μm (triangles) Kapton HN foils. Individual sets of measurement on 25 μm Kapton HN foils are denoted by different triangle symbols.

annihilation lifetime spectra obtained using monochromatic positrons implanted exclusively within thin Kapton foils.

Previous measurements of the positron lifetime spectrum of Kapton have been performed using thick Kapton [8], or on stacks of Kapton foils [9–13]. Recently measurements have been reported on stacks approximately 40 pieces of 125 μm Kapton using an 8 μm Kapton supported ^{22}Na positron source [13]. The aim of the study was to investigate the effect temperature and of high dose electron beam irradiation. The single lifetime decompostion were reported to give the best fits. The as-received lifetime was 385(3) ps, this reduced to ~378 ps after a ~66 MGy absorbed electron beam dose, and remained approximately constant up to 200 MGy. The lifetime also increased with increasing temperature reaching ~397 ps at ~360 °C.

An age momentum correlation (AMOC) study of Kapton, performed using a high energy positron beam, fitted the resulting spectra using two positron lifetime components and two associated Doppler broadening spectroscopy (DBS) *S*-parameter values [14]. The lifetime components were reported to be 229 ps and 378 ps, the latter with an intensity of 87 %; the nature of the Kapton studied was not described. Bulk lifetime measurements on stacked 125 μm Kapton HN foils using an 8 μm Kapton HN foil supported source reported two lifetime components, at ~277 ps and at ~410 ps, the latter with an intensity of ~70%, giving an average lifetime of 369(3) ps [12]. Measurements on bulk 1 mm plates of Kapton polyimide (PI2540), pyromellitic dianhdride-oxydianiline (PMDA-ODA), report a single lifetime value of 354 ps [8], earlier measurements by the same author were performed on a large number of stacked 3μm Kapton (PMDA-ODO) foils and reported two components, one at 220 ps and a second at 376 ps with intensity of 86 % giving an average lifetime of 354 ps [11]. The deconvolution into two components is attributed to an artefact of the spectrum analysis that arises when the positron lifetime distributions are particularly broad [14, 15].

The adoption of Kapton foils for use in PALS followed a study by MacKenzie and Fabian [16], that reported a single lifetime component of 382 ps and a weak temperature dependence. Subsequently studies on stacked Kapton foils reported lifetime values of 386(7) ps [9], and of 382(3) ps [10]. The latter measurement used a stack of fifty 25 μm Kapton foils, measurements were also performed with some of the foils replaced by 8 μm Kapton with no change in the results.

The depth distribution of implanted positrons can be described by a Makhovian profile, with the median depth obtained using a power law expression, $z_{1/2} = (A/\rho)E^n$, were A is normally given in units of $\mu g \cdot cm^{-2}$ and E is in keV. Measurements using three different polymers obtained values $A = 2.81(20)\ \mu g \cdot cm^{-2}\ keV^{-n}$ and $n = 1.71(5)$ [17]. The density of Kapton is $1.42\ g \cdot cm^{-3}$, and the resulting implantation profiles are shown in figure 1. The mean implantation depth increases with increasing implantation energy, for 10 keV monoenergetic positrons it is approximately 1 μm.

2. Experiment

Variable energy (VE) PALS measurements were performed on a 7.6 μm and 25 μm Kapton® HN foils using the PLEPS instrument on the neutron induced positron source (NEPOMUC) at the Heinz Maier–Leibnitz Zentrum (MLZ) Munich research reactor (FRMII) [18–20]. The foils were carefully mounted with double-sided conducting tape on to the sample holders. Measurements on the 25 μm foils were performed using positron beam energies between 1 to 18 keV, and each lifetime spectra contained greater 3×10^6 counts. Measurements on the 7.6 μm foil were made at 16 keV and 18 keV, the spectra contained 4.0×10^6 counts. The instrument timing resolution function is described by a sum of Gaussian functions and the mean width is energy dependent. The value, averaged over all implantation energies, was 254(26) ps.

3. Results and discussion

The measurements on the 25 μm and 7.6 μm Kapton exhibit a single dominant positron lifetime component with an intensity of greater than 99.3%, and a second negligible intensity nanosecond component. The lifetimes obtained for the dominant first component are shown in figure 2, the average value is

385(1) ps. Figure 1 shows representative Markovian implantation profiles demonstrating that the positrons were confined to the Kapton foils for all implantation energies used. Implantation energies of 16 keV and 18 keV where selected for the 7.6 μm Kapton as the resulting depth distributions optimally sample the foil and provide lifetime values of direct relevance to conventional PALS experiments using Kapton supported positron sources.

These results provide clear evidence for a dominant single positron lifetime component with a value of 385(1) ps in Kapton HN for both foil thickness studied. This is in agreement with the early bulk PALS reports of a single lifetime value of 382 ps [16], and 382(3) ps [10], and with the most recent study giving 385(3) ps [13]. The measurements provide further evidence that previously reported two component fits result from an artefact of the fitting process [11, 12, 14]. The AMOC results require the existence of at least two positron states [14], but are complicated by in-flight annihilation and by the high (3.5 MeV) positron energy and hence the requirement for thick samples. The low values reported for Kapton PI2540 of ~354 ps suggest that this is different to Kapton HN [8]. The low average lifetime for Kapton HN of ~370 ps reported by McGuire and Keeble [12], resulted from incorrect ^{22}NaCl source correction, the value obtained by applying a systematic source correction procedure was reported to be 385(4) ps [6].

These measurements demonstrate that conventional, bulk, positron lifetime measurements using radionuclide positron sources supported thin Kapton HN foils can be analysed by fixing the lifetime of the foil source correction component to a value of 385(1) ps. The results of Hirade *et al* [13], suggest that this value may decrease with time for high activity sources.

4. Conclusions

Positron lifetime spectra were directly measured from 7.6 μm and 25 μm Kapton HN foils with VE-PALS using implantation energies for which all the positrons annihilate from within the foil. A single dominant lifetime component with an intensity greater than 99% was obtained from all measurements with a lifetime value of 385(1) ps.

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