



Depth-dependent positron annihilation in different polymers



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ABSTRACT

Depth-dependent positron annihilation Doppler broadening measurements were conducted for polymers with different chemical compositions. Variations of the S parameter with respect to incident positron energy were observed. For pure hydrocarbons PP, HDPE and oxygen-containing polymer PC, S parameter rises with increasing positron implantation depth. While for PI and fluoropolymers like PTFE, ETFE and PVF, S parameter decreases with higher positron energy. For chlorine-containing polymer PVDC, S parameter remains nearly constant at all incident positron energies. It is suggested that these three variation trends are resulted from a competitive effect between the depth-dependent positronium formation and the influence of highly electronegative atoms on positron annihilation characteristics.

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1. Introduction

Doppler broadening energy spectroscopy on a variable-energy positron beam has been widely applied to study damage and defect depth-profiling of polymers. In most cases, Doppler broadening linear parameter S increases with respect to incident positron energy [1–3], which is widely accepted as a result of increased Ps formation in the bulk of polymers. However, exceptions occur in some cases. For example, in 1995, Kobayashi [4] found S parameter of high dose irradiated PEEK films decrease with increasing positron energy at low incident positron energy levels. Later, S parameter of PVC [5] and PTFE [6] were reported to show similar variation. These researchers tried to attribute the variation trend to active species formed by irradiation, lower number of free-volume defects or lower concentration fluorine atoms in surface area. Due to complicated factors involved in irradiated polymers and lack of comprehensive study of polymers with different chemical composition, more general explanation seems impossible.

It has been known that the physical properties of polymers near the surface or interfaces are different from those in the bulk, this is the so-called surface or interface effect [7–9]. Due to this effect, positron annihilation near polymer surface might be different from that in the bulk. A fully study of the depth distribution of positron annihilation in polymers is necessary for understanding positron annihilation characteristics and will be helpful to study the

damage or defect depth distribution. In this work, we systematically studied the depth distribution of positron annihilation in eight different virgin polymers with the aid of a positron beam coupled with positron annihilation Doppler broadening, and try to give a general explanation to different variation behaviors of S parameter.

2. Experimental

2.1. Samples

Polymer samples used in this study were purchased from Goodfellow (Cambridge, UK), including high-density polyethylene (HDPE), polycarbonates (PC), polypropylene (PP), polytetrafluoroethylene (PTFE), ethylene tetrafluoroethylene (ETFE), polyvinyl fluoride (PVF), polyimide (PI), polyvinylidene chloride (PVDC). PI used here was a kind of kapton polymers containing alumina particles. Among them, PI, PVDC and fluoropolymers were semi-crystalline polymers. All polymer samples were used as received. The structure of polymers under investigation is listed in Table 1.

2.2. Positron annihilation measurements

Positron annihilation Doppler broadening measurements were conducted with a magnetically guided variable-energy positron beam (0–10 keV) at IHEP, Beijing. The Doppler broadening energy spectra (DBES) were recorded using a high-purity Ge detector. The obtained DBES spectra were expressed in S parameter, which was defined as a ratio of integrated counts in the central parts

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Table 1
The structure of polymers under investigation.

Group	Sample	Thickness (mm)	Structure	Fraction of C&H
Polyolefin	HDPE	1	$\left[\text{CH}_2 \right]_n$	1
	PP	0.5	$\left[\text{CH}_2 - \underset{\text{CH}_3}{\text{CH}} \right]_n$	1
Oxygen-containing polymers	PC	0.5	$\left[\text{O} - \text{C}_6\text{H}_4 - \underset{\text{CH}_3}{\text{C}} - \text{C}_6\text{H}_4 - \text{O} - \text{CO} \right]_n$	0.9
	PI	0.075	$\left[\text{C}_6\text{H}_4 - \text{O} - \text{C}_6\text{H}_4 - \text{N} - \text{C}_5\text{H}_2 - \text{N} \right]_n$	0.82
Chlorine-containing polymer	PVDC	0.05	$\left[\text{CH}_2 - \text{CCl}_2 \right]_n$	0.66
Fluoropolymers	PVF	0.05	$\left[\text{CH}_2 - \text{CHF} \right]_n$	0.83
	ETFE	0.125	$\left[\text{CH}_2 - \text{CF}_2 \right]_n$	0.66
	PTFE	0.5	$\left[\text{CF}_2 - \text{CF}_2 \right]_n$	0.33

(510.2–511.8 keV) to the total counts of the 511 keV annihilation peak after proper background subtraction.

Conventional positron annihilation lifetime measurements were also performed. All the spectra were de-convoluted using LT 9.0 software. Well annealed nickel platelets were measured as a reference material for estimating the positron source correction.

2.3. X-ray diffraction measurements

In order to characterize surface and bulk crystallinity of the samples, grazing incidence X-ray diffraction (GIXRD) and wide-angle X-ray diffraction (WAXRD) experiments were carried out using synchrotron radiation with a wavelength 1.5404 Å at the Beijing Synchrotron Radiation Facility. Diffraction intensity at different angles starting from 10° to 35° was taken. Two incidence angles, $\alpha=0.2^\circ$ and 0.5° for GIXRD, $\alpha=2^\circ$ and $\alpha=5^\circ$ for WAXRD, were used respectively. Penetration depth of XRD and corresponding incidence positron energy are given in Table 2 for PI and PVDC.

3. Results and discussion

Depth profile of S parameter for eight different virgin polymers is shown in Fig. 1. Evidently, three categories of S parameter dependence with respect to incident positron energy can be observed. For polymers like PP, HDPE and PC, S parameter increases with increasing incident positron energy until 4 keV, and then reaches leveling-off at higher energies. For PI and fluoropolymers including PTFE, ETFE and PVF, S parameter decreases from polymer

surface to bulk. And for chlorine-containing polymer PVDC, S parameter is independent of incident positron energy and remains nearly constant. In bulk, S parameter for polymers with different chemical composition are quite different, e.g. at 10.18 keV, the difference of S value between PP and PTFE is as large as 0.055. In contrast, positron annihilation S parameters for different polymer surface are somewhat similar to each other, e.g. only 0.015 in difference at 0.18 keV between PP and PTFE.

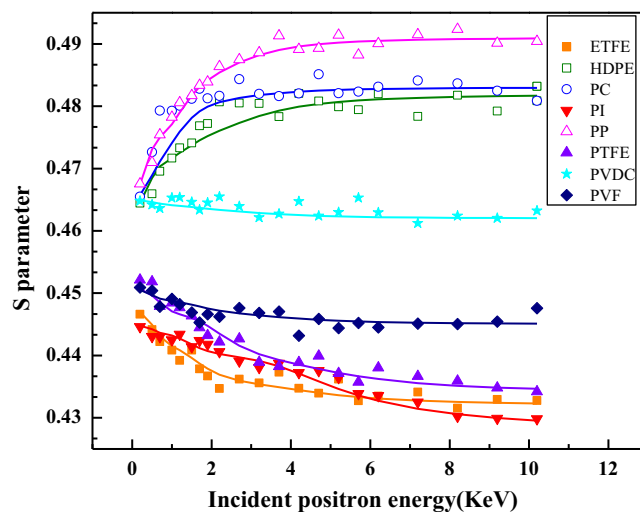


Fig. 1. Depth profile of S parameter for different polymers (the lines are just guide for eyes).

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