STUDY ON APPLICATION OF POSITRON LIFETIME SPECTROSCOPY IN INVESTIGATION OF RADIATION INFLUENCES ON NUCLEAR TRACK DETECTOR POLYMER

Khuong Thanh Tuan, Tran Dai Nghiep, Nguyen Manh Hung, Nguyen Duc Thanh and Le Anh Tuyen

Institute for Nuclear Science and Technology

ABSTRACT: Study on determination of micro porosity of materials using positron lifetime technique is an advanced and promoted tendency in physics and material science presently. In Vietnam, studies in this field have been carried out by some projects funded by VAEC in recent few years, with their object is determination of porosity of oil-contained rock and polymers. This project is established to study the relationship between characteristic of positron lifetime spectra and micro porosity of polymer material, which a type of nuclear track detector made from, after irradiation. The experimental result shows a linear relation of a particular characteristic of lifetime spectra - intensity of lifetime components above 1 ns - and the density of track in polymer created by irradiation.

Part I - INTRODUCTION

Positron techniques presently is an advanced method for materials science [1, 2], especially positron lifetime measurement technique. Particularly, in determination of radiation effects on polymers, this technique seems to be a strong and advanced.

Up to now, there have been some projects carried out in this field in VAEC, such as application of positron technique in determination of porosity of oil-contained rock (Dr. Nguyen Duc Thanh), project for determination of track density in polymers (Khuong Thanh Tuan).

However, the number of projects of this field is still limited, so this project is established to continue the researches of application of positron lifetime technique for a kind of polymer that nuclear track detectors made from.

Aim of the project:

Study the applicability of positron lifetime measurement for determination of track density created in polymer by.

Particularly, the object of studies is the relationship between track density caused by irradiation and intensity of lifetime component characterized for orhto-positronium state in polymer, I_3.

Part II - THEORY AND REVIEWS

Studies on positron lifetime in macro-pore materials is accquiring interests in the world, especially about ortho-positronium formation in cluster of pores. In polymer, studies on determination of porosity of the material after irradation get much of interests recently.

1.1. Positron lifetime

For a positron, lifetime can be considered as the time since positron exist from positron source until it annihilates with an electron. Distribution of lifetime in materials reflects how annihilation occurs, which process is dominant; this distribution depends on density, electron density, effective charge, micro and macro porosity of the medium, etc.

There are three order of magnitude of positron lifetime: the first value is about 100 ps for free annihilation; the second one is about 125 ps, characterize for annihilation of para-positronium state of positron and electron; the last value, which is high in comparison to others, is the result of annihilation of ortho-positronium state. In practice, it can reach few ns, typically.

Different annihilation processes cause different lifetime distribution in lifetime spectrum. Positron lifetime techniques are among advanced techniques in material science.

1.2. Positron lifetime spectroscopy

The conventional positron lifetime measurement is possible since a γ-quantum with energy of 1.27 MeV is emitted almost simultaneously with the positron in the ^{22}Na source. The positron energy, which extends up to 540 keV, decreases in the sample within a few picoseconds by non-elastic interactions. The mean positron penetration depth of this so-called thermalization process is of the order of 100 μm. The thermalization time usually amounts to a few picoseconds. It is thus small compared with the positron lifetime and can be neglected. On reaching thermal energies, the positron diffuses in the periodic lattice potential before it is possibly trapped in a lattice defect. The diffusion length is in of order of 100 nm. This distance determines the number of atoms to be probed for positron traps during the positron lifetime. Hence, the diffusion length strongly determines the sensitivity of the positron methods to detect defects.

The positron lifetime of a single event can be measured by detecting the time difference between the birth γ-quantum of the γ-decay in the source and one of the annihilation γ-quanta of energy of 511 keV. The activity of the source must be sufficiently low in order to ensure that on average only one positron is in the sample. This avoids the intermixing of start and stop quanta originating from different annihilation events. A special “sandwich” arrangement of foil source, samples, and detectors guarantees that all positrons emitted from the source are penetrating the sample material. The γ-rays are converted by scintillator–photomultiplier detectors into analog electrical pulses. The pulses are processed by discriminators. Their output pulses start and stop a time-to-amplitude converter as an “electronic stopwatch”. The amplitude of the output pulse is proportional to the time difference between the birth and the annihilation γ-quanta and, thus, represents a measure of the positron lifetime. The single annihilation event is stored after analog-digital conversion in the memory of a multi-channel analyzer. The channel numbers represent the time scale. In order to obtain the complete lifetime spectrum, more than 106 annihilation events must be recorded.

The scheme of the positron lifetime measurement is shown in Fig 1. NaI(Tl), BaF2 or common plastic scintillators and photomultipliers with a short pulse rise-time are used to obtain a high time resolution. The discriminators suppress noise and generate standard timing pulses by the constant-fraction discrimination principle. This principle

is favored over leading-edge discrimination in order to ensure stable time markers independent of the pulse height. Another task is to guarantee that the 1.27-MeV and 0.51-MeV quanta are accepted only in the appropriate channels. The discriminators are of differential type (single-channel analyzer) and accept input pulses within an adjustable energy window.

The timing pulses are used to start and stop the charging of a capacitor in the time-to-amplitude converter (TAC). The time linearity is ensured there by constant-current charging that is stopped at the arrival of the stop pulse originating from the annihilation γ-quantum. The stop pulse is coax-cable delayed in order to shift the time spectrum into a linear region of the TAC. The spectrum is stored in a multi-channel analyzer.

This experimental arrangement is called "fast-fast coincidence" setup. The term is related to the fact that the time measurement as well as the energy selection is performed in a fast channel. A slow channel was used for energy selection when fast differential discriminators were not available at the beginning of positron lifetime experiments. This arrangement is called a fast-slow setup. Inexpensive multi-channel plug-in boards for personal computers with about 2000 channels are sufficient for storing the spectra. The time resolution of the spectrometer is determined mainly by the scintillator-multiplier part and ranges between 180 and 280 ps. The practical consequence of this relatively poor resolution is the limitation of the determination of positron lifetime components larger than about 50 ps. The determination of positron lifetimes can, however, be carried out with an accuracy of about 1 ps.

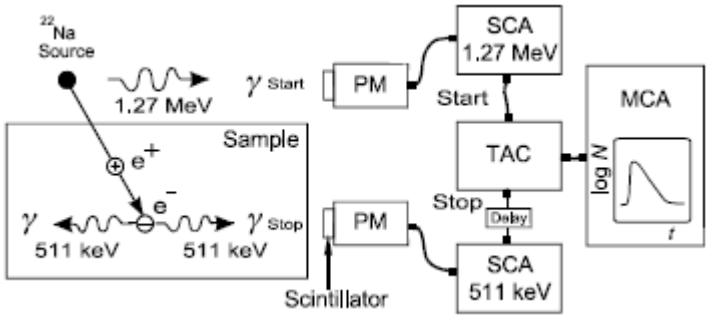


Fig. 1. Principle of basic lifetime spectroscopy

1.3. Major properties of annihilation in polymers

Annihilation in polymers has interesting characteristics. The density of free electrons in polymers is small in comparison to other solids, so free annihilation is hard to occur. Instead of that, positronium formation and annihilation become dominant. Otherwise, if there are free volumes in polymers, positron lifetime can be about ns in value. When trapped in these free volumes, positrons can form ortho-positronium atoms with electrons and this process has the highest probability. The lifetime of positronium state is in range of few ns, assigned by I_3, causes lifetime component τ_3 in the distribution.

Part III - EXPERIMENT

This experiment aims to determination of positron lifetime characterization in CR-39 polymer and to obtain the dependence of parameters of lifetime spectra on track density in polymer after irradiation.

3.1. Samples preparation

Nuclear track detectors were cut into samples with dimension of 2x2 cm^2. There are 29 samples and divided into three sets A, B and C, noted from A1 to A9, B1 to B9 and C1 to C9. Three sets were irradiated with the same condition for samples with corresponding index, which means Ai, Bi and Ci samples were irradiated with the same condition. After irradiation, set A was prepared for etching to obtain track density values (in table 1) for each sample. Remained two sets, B and C, were prepared for lifetime measurement.

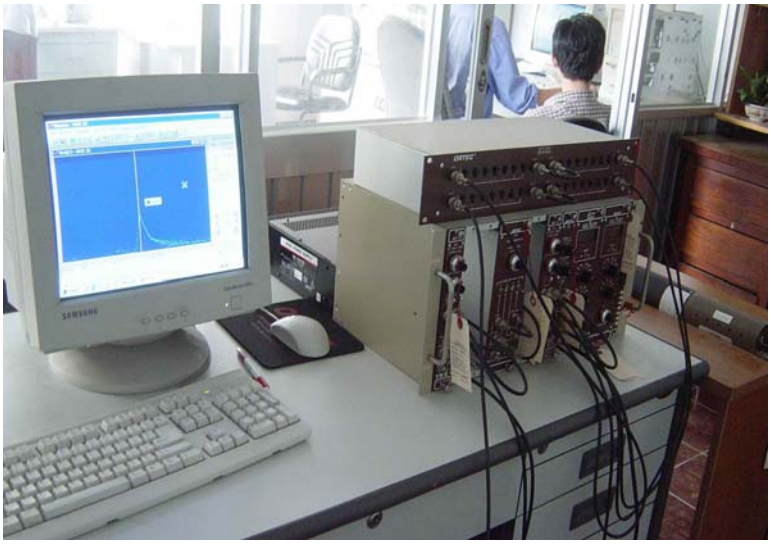
Tab 1. Track densities obtained by etching process

Order	**Sample**	**Track density,** (tracks/cm^2)
1	A1	0
2	A2	10 ± 1
3	A3	180 ± 6
4	A4	316 ± 5
5	A5	710 ± 8
6	A6	1544 ± 11
7	A7	4752 ± 17
8	A8	2800 ± 12
9	A9	3307 ± 10

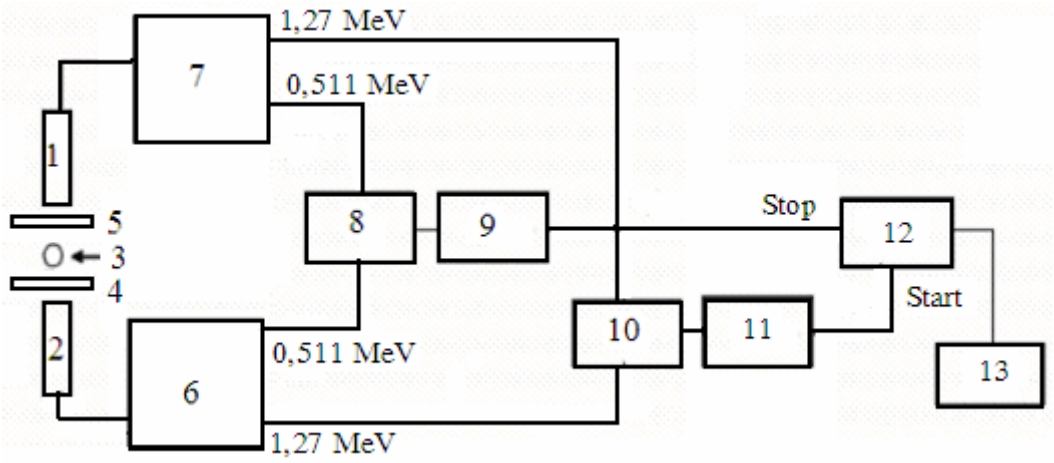
3.2. Experiment arrangement

In fig. 2, the lifetime spectroscopy system is shown with following components:

- Two scintillators NaI BS-148 with fast photomultipliers 8850 (Canberra);
- Two fast Constant Fraction Discriminators (CFD) ORTEC 583;
- Two delay units ORTEC DB463;
- Fast coincidence unit ORTEC 414A;
- Time to Amplitude converter (TAC) ORTEC 566;
- Multichannel Analyser (MCA) with 8192 channels, equipped with MAESTRO software run on PC.



(a)



(b)

Fig. 2. Photo (a) and scheme (b) of positron lifetime spectroscopy system

In this configuration, samples were kept in location 4 and 5, and positron source Na^{22} was sandwiched between samples.

Part IV- RESULT AND DISCUSSION

4.1. Experimental result

In Fig. 3 shows some typical lifetime spectrum obtained. It can be clearly to see the porosity effect in the lengthen in tails of spectra.

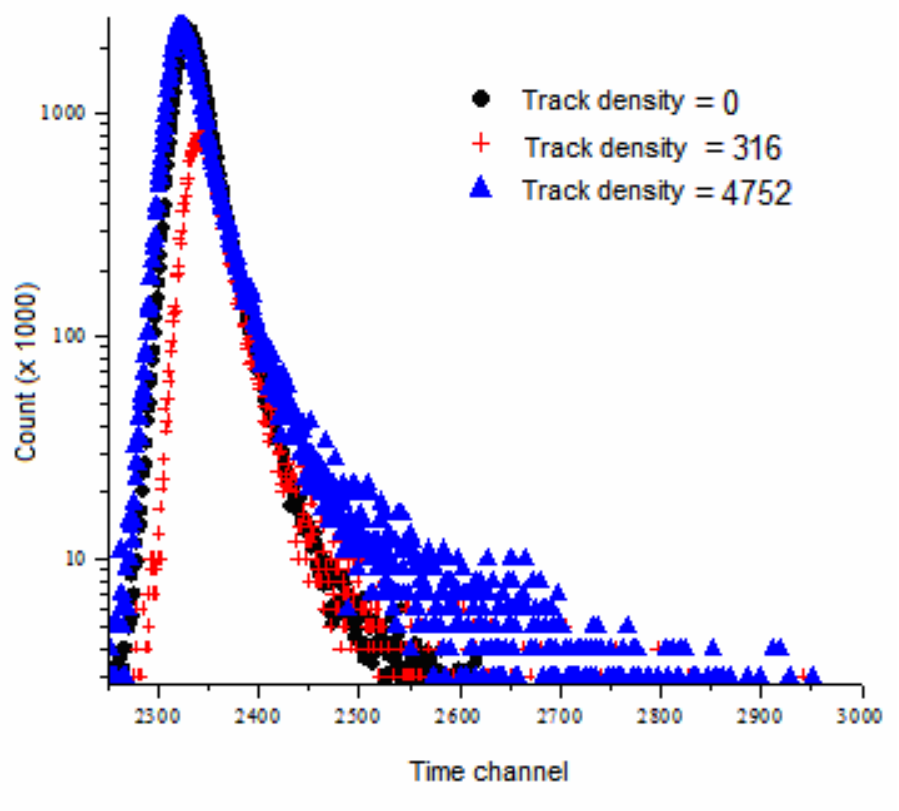


Fig. 3. Typical lifetime spectra for CR-39

Tab 2. Values of lifetime components and their intensities for polymer samples in measurements

Sample	**Track density, tracks/cm^2**	I_1 (%)	τ_1 (ns)	I_2 (%)	τ_2 (ns)	I_3 (%)	τ_3 (ns)
A1	0	41.45	0.162	41.61	0.325	16.94	1.89
A2	10	44.1	0.176	36.47	0.399	19.43	2.29
A3	180	43.14	0.184	37.95	0.420	18.91	2.52
A4	316	46.14	0.175	39.59	0.409	14.27	2.56
A5	710	41.8	0.218	35.85	0.520	22.35	2.84
A6	1544	41.93	0.214	37.23	0.493	20.84	2.87
A7	4752	15.92	0.166	38.06	0.400	46.02	2.51
A8	2800	33.92	0.174	37.00	0.392	35.10	2.61
A9	3307	32.07	0.174	37.34	0.394	37.02	2.61

It is interesting that we obtained the linear dependence between I_3 and track density; I_3 is corresponds to lifetime values τ_3 greater than 1 ns (Fig. 4).

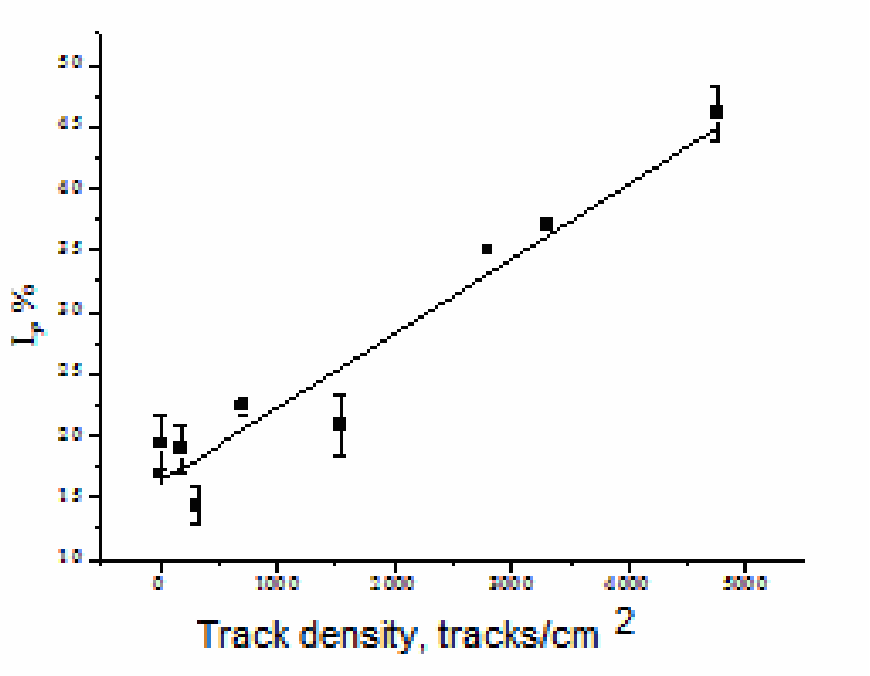


Fig. 4. The linear dependence I_3 – track density for CR-39

This dependence is regressed to:

$Y = (0.00600 \pm 0.00005)x + (16.289 \pm 1.309)$

$R^2 = 0.969$

4.2. Discussion

While I_1 and I_2 are nearly constants with variation of track density, I_3 increases when track density increases. This result is suitable to fact that I_3 reflects annihilation process of ortho-positronium, but I_1 and I_2 does not, as confirmed by other publication [1-3]. This result initially opens the ability of application in determination of track density and porosity in polymers.

REFERENCES

[1]. V. I. Grafutin and E. P. Prokopiev, Positron annihilation spectroscopy in material structure studies, Physics-Uspekhi 45, 1(2002) 59-74.

[2]. Emad A. Badawi, International Journal of Modern Physics B, Vol. 19, 22 (2005).

[3]. Mohamed HF, Abd El-Hady M, Abdel-Hady EE, Radiation effect on positronium formation in solid state nuclear track detector., Appl. Radiat. Isot. 2007 Mar; 65(3):328-34. Epub 2006 Oct 13.