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Free Volume Measurements through the Positron Annihilation Techniques for Cable Materials of Nuclear Power Plants

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1. Introduction

Cables in nuclear power plants are used in various areas such like high temperature, humidity, and high radiation environment. NPP cable, which are practically not easy to be replaced during operation, show the degradation as it is used for a long time in these severe conditions. However it is difficult to assess its aging degradation.

The positron annihilation lifetime (PAL) techniques have been widely used for studying defects of materials during the last two decades. The PAL can be used as a powerful probe for the measurement of the existence free –volume holes in polymer. For the characterization of polymers, the determination of the free volume plays an important role [1]. When positrons are injected to a polymer, they form positronium (Ps) in the two states: ortho- and para-positronium (o-Ps, p-Ps). In this study, positron annihilation technique was used to measure the lifetime of a Ps atom.

2. Experimental and Methods

2.1 Cables in nuclear power plants

Cables for nuclear power plants must be better in quality, and with a higher reliability than ordinary cables. The function of electrical cable is to provide a medium for transmitting electrical energy (power control or signals) between two points in a common electrical circuit, while simultaneously maintaining the electrical isolation of the transmission path

Among various polymers, we measured the PAL of ethylene propylene diene M-class (EPDM). EPDM is widely used for cable jacketing and insulation. This material has relatively high durability and radiation resistance. Fig. 1 shows the chemical structure of EPDM. To check the validity of PAL system, irradiation process with gamma rays was performed at KAERI-ARTI in Jeongup. The samples were irradiated with the following doses: 100, 500 Gy.

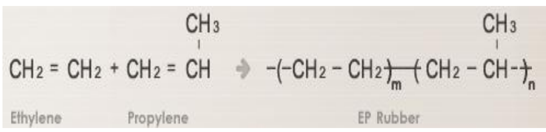

Fig. 1. Chemical structure of EPDM

2.2 Experimental

A conventional fast-fast coincidence system in KAERI has been used to measure the lifetime spectra, incorporating two BaF_2 scintillation detectors, two constant fraction differential discriminators, a time to amplitude converter and multi-channel analyzer. The instrumental time resolution of the system is 250 ps of the full width at half maximum (FWHM). The positron sources used for the measurement consist of ^{22}Na sandwiched between two Ni foils. The equipment is kept in a room at a constant room temperature to reduce the electronic drift. Through this PAL system, we can measure the time difference between the appearance of two γ-quanta (start and stop gamma) as like Fig. 1.

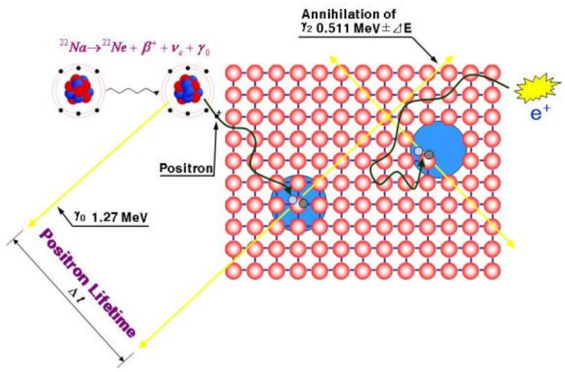

Fig. 1. Scheme of positron annihilation. Positrons(start signal with the 1.27MeV birth-γ) from Na-22 source.

2.3 Positronium in polymers

Upon implantation into a polymer sample, they lose their energy through ionization of molecules and formation of excess electrons in a few picoseconds. Some of positrons combine with an electron to form the hydrogen-like bound state. In spin-allowed annihilation of p-Ps occur with a lifetime of the order of 0.125 ns and decay with emissions of two photons. The spin-forbidden annihilation of the o-Ps decays after approximately ~140ns. 75% The o-Ps annihilation may be enhanced if it is able to interact with other electron and the spin exchange or pick-off annihilation occurs reducing the lifetime to around 1-5 ns. Positrons not forming positronium have a lifetime of ~ 0.4 ns through interaction with the outer of molecules with which they collide [2,3].

In the measurement of polymer by PAL system, there exist three state of positrons characterized by lifetimes τ_1, τ_2, τ_3,: spin antiparallel para-positronium(o-Ps) with τ_1 ~125 ps, a free positron with τ_2 ~450 ps and spin parallel o-Ps with τ_3 ~1-10 ns.

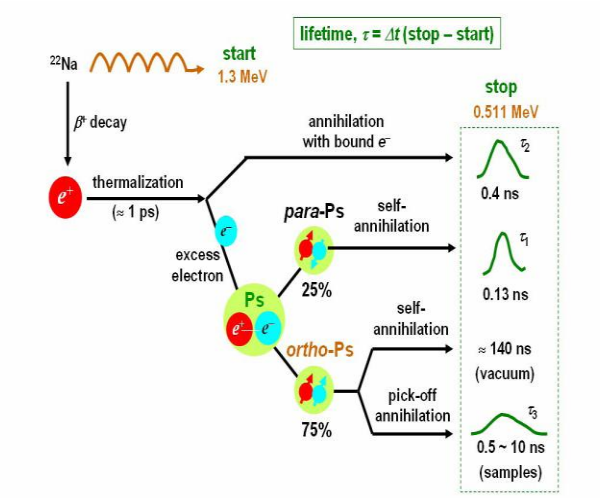

Fig. 1. Scheme of the formation and annihilation of positron and Ps in polymer samples.

3. Results and Discussion

In the PAL measurement, the longest-lived component is of particular importance to the polymer studies, because the lifetime τ_3 is related to the average free volume hole size.

Table 1 lists the o-Ps lifetime, its intensity.

Table I: The o-Ps lifetime, its intensity for gamma-irradiated EPDM.

γ - irradiation	o-Ps lifetime(ns)	Intensity (%)
0	2.4108±0.0138	13.446±0.082
100	2.3040±0.0178	13.549±0.111
500	2.3723±0.0141	13.754±0.088

The free volume holes can be calculated from the lifetime τ_3 in spherical approximation of Tao-Eldrup [2,4] model as

$$\frac{1}{\tau_3} = 2\left[1 - \frac{R}{R_0} + \frac{1}{2\pi}\sin\left(\frac{2\pi R}{R_0}\right)\right] \qquad (1)$$

where $R_0=R+\Delta R$ and ΔR=0.166nm is the thickness of the homogeneous electron layer in which the positron in o-Ps annihilates via the so-called pick-off process [5]. Free volume size V_f in nm^3 is calculated as :

$$V_f = \frac{4\pi R^3}{3} \qquad (2)$$

According to Eqs. (1) and (2), free-volume hole size are calculated as Table II.

Table II: The radius(R) and free volume size V_f for gamma-irradiated EPDM.

γ - irradiation	Radius(nm)	free volume size V_f (nm^3)
0	0.3208±0.0020	0.1383±0.008
100	0.3110±0.0020	0.1260±0.008
500	0.3180±0.0020	0.1347±0.008

These values are familiar with reported papers [6]. However, we could find the effects of gamma irradiation. There is no any noteworthy tendency in τ_3 lifetimes and intensities according to irradiation. That means the irradiation doses are too small to make any defects in polymers.

3. Conclusions

The positron annihilation measurement techniques were applied to measure the free-volume of polymer materials in NPP. From the measurement of ortho--positronium lifetimes, the free-volume size of gamma-irradiated EPDM polymer were calculated. We could find the values of free-volume size are consistent with previous papers. Therefore we identified the feasibility of the PAL system for polymer analysis.

However, we could not found the effects of gamma irradiation. By high dose irradiation, we expect to analyze the properties of irradiation induced defects in polymers. effects of. In this study, we could find the free-volume size in polymers. These data will be used to compare the properties change according to various circumstances.

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