The adhesive effect of the positron source for positron annihilation lifetime spectroscopy (PALS)

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1. Introduction

Positron annihilation lifetime spectroscopy (PALS) is a powerful tool for analyzing material defects and the free volume of a high molecular substance. The unsealed source must be considered to surrounding contamination and handling convenience because the positron source of the PALS is liquid. During PALS, positrons annihilate in the samples and source supporting foil. For accurate sample analysis, the ingredient from the positron source should be corrected. The source correction will perform only correction for the supporting foil. The supporting foil of PALS is used as metallic material (Ni, Al, and Ti). In the metallic material, the positron lifetime appears in two parameters. The first component comprises positron annihilated probability in the bulk state and the annihilated probability at the lattice where positrons are trapped. The first component is called the short lifetime (τ_1). The intensity of the first lifetime indicated the symbol I_1. The second component is the probability that annihilates after a long time compared to the bulk state because the lifetime of the positron increases within the defect of materials. The second component is called a long lifetime (τ_2). The intensity of the long lifetime indicated the symbol I_2.

The positron source may be necessary to correct the adhesive using the adhesive when manufacturing the positron source. There are adhesive commonly used in the manufacturing process of the positron sources. Typically, glue uses ethyl cyanoacrylate (ECA). The ECA hardens by causing a polymerization with surrounding moisture. The ECA is a high molecular substance, and it have a free volume. If the material has a free volume, a lifetime of ortho-positronium (*o*-Ps) will have a lifetime longer than the τ_2. It is called *o*-Ps lifetime (τ_3). The intensity of the τ_3 indicated the symbol I_3. The τ_3 component can occur due to the surroundings of the detector or signal errors.

If the component of the source is not accurately identified when the source correction is perform, an error in analyzing the component detected from the source can occur in the sample, so it is very important to check the component from the source. We studied the positron lifetime components of adhesives.

2. Materials and Methods

2.1 Positron Annihilation Lifetime Spectroscopy

PALS system consists of a high voltage power supply (HVPS), fast plastic scintillators, photomultiplier tubes (PMTs), constant fraction differential discriminators (CFDDs), nanosecond delay, multichannel analyzer (MCA), and time to amplitude converter (TAC). Fig 1 shows the structure of the installed devices. Two HVPSs connected to the PMT base and a high voltage of -1.9 kV applied for the gamma energy of the start signal (1.27-MeV) and -2.5 kV for the gamma energy of the stop signal (0.511-MeV). The PMT base's output signal (start and stop) has received two CFDDs. One of the generated two output signals (start and stop signals) at PMT base was transmitted in the nanosecond delay to optimize the time interval of each output signal, and the output signal in CFDD is recorded in MCA through time amplification in the TAC. The MCA is integrated with the analog to digital converter (ADC) and sent into 4,096 channels.

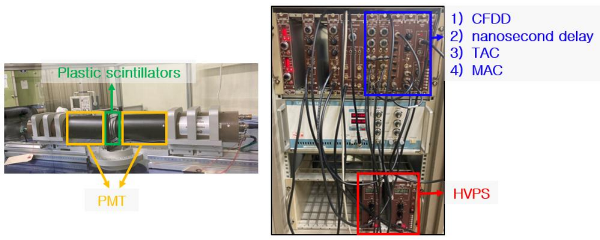

Fig 1. The NIM nodules for positron annihilation lifetime spectroscopy. PMT: photomultiplier tube HVPS: high voltage power supply, CFDD: constant fraction differential discriminator, nanosecond delay, TAC: time to amplitude converter, MCA: multi-channel analyzer.

A 30-μCi $^{22}NaCl$ was dried on both sides of nickel (Ni) foil (thickness: 2.5-μm). We covered the Ni plate (thickness: 50-μm). The Ni plates are punched at central because it needs to fix the positron source in a supporting holder. The positron source is located between the nickel plate, and the enclosed positron source appears in Fig 2. Each positron lifetime spectra of the samples was obtained more than 1×10^6 counts. The geometry of the detector and samples appears in Fig 3.

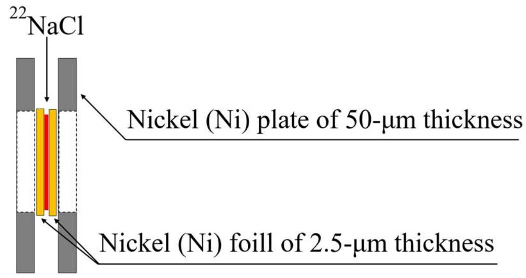

Fig 2. A structure map of a positron source

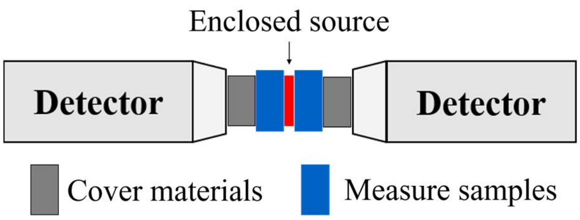

Fig 3. Structure of the detector's setting.

The time resolution of the PALS system was about 230 ps in full width at a half maximum (FWHM). The spectra were analyzed by the *PALSFit3* software [2].

We used the Tao-Eldrup model [3] to calculate the radius of the free volume and the fractional free volume (F_f) using the τ_3. The expression of the Tao-Eldrup model is shown as follows:

$$\lambda_3 = \frac{1}{\tau_3} = 2\left[1 - \frac{R}{R+1.66} + \frac{1}{2\pi}\sin\left(\frac{2\pi R}{R+1.66}\right)\right], \quad (1)$$

where the symbol λ_3 is the annihilation fraction of the o-Ps. The symbol R is the average hole radius (Å). The free volume size (V_f) and fractional free volume (F_f) are, in turn, given by:

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

and

$$F_f = R \times I_3. \quad (3)$$

2.2 Samples

Three materials were measured to analyze the effect of adhesive. Additionally, we estimated the impact that positron annihilates at surrounding air of detectors. The detail of the materials is described in Table 1. In the case of Loctite 401 and Scotch, the adhesive were dried at room temperature for six day. The liquid glue has dried about 2.5-mm thickness, but we added the cover plates next to the enclosed source. All cover plates used pure nickel (1-mm).

Table 1. Sample information

Samples	Thickness (mm)	Cover plates
Ni	1	Ni
Loctite 401	2.5	Ni
Scotch	2	Ni

3. Results

For pure nickel measured in PALS, the τ_1-value is observed at about 110 ps [3]. The τ_1-value will be the component of positrons that have been annihilated from the foil, cover, and sample. The τ_3 is the component that has been annihilated from the free volume. As a result of the nickel sample, The τ_1-value was observed as 114.6 ps. We unfolded the τ_3-value by fixing the τ_3-value measured from the nickel sample when measuring the adhesive. The lifetime for a high molecular substance has been reported as about 1.0-5.0 ns [4]. The results are shown in Fig 4. When the adhesive sample was measured, the I_3 increased compared to the nickel sample. The τ_3 and I_3 of the adhesive samples were similar to each other within the error range.

The V_f, F_f, and R appear in Table 2. All of the V_f-values were similar within the error range, and we estimate that the V_f-values are identical because they are free volumes from adhesives.

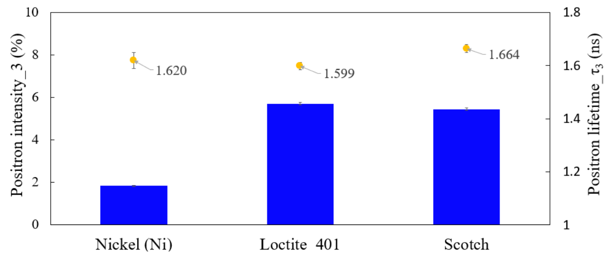

Fig 4. The blue bar is the ortho-positronium (o-Ps) intensity (I_3) and the yellow circle is the o-Ps lifetime (τ_3).

Table 2. The results of average hole radius (R), fractional free volume (F_f), and free volume size (V_f).

Samples	R (Å)	F_f (%)	V_f (nm^3)
Ni	2.47 ± 0.03	0.21 ± 0.01	6.32 ± 0.10
Loctite 401	2.45 ± 0.02	0.63 ± 0.02	6.15 ± 0.05
Scotch	2.52 ± 0.02	0.66 ± 0.02	6.69 ± 0.05

3. Conclusions

We identified the positron lifetime of the adhesive (Loctite 401 and Scotch). The ingredient of the adhesive appears in the positron source. If the positron source included many adhesives, the I_3 will be increased in the PALS spectrum. Therefore, the adhesive component should be removed for the accurate analysis of PALS.

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