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Source Correction for Positron Annihilation Lifetime Spectroscopy: A Monte Carlo Study

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1. Introduction

Positron annihilation lifetime spectroscopy (PALS) is a non-destructive and defect-sensitive analysis on the surface or inside of a solid. It measures the time difference between positron generation and annihilation inside of the materials [1]. A positron that enters the sample emits two gamma rays that have an energy of 511 keV via an annihilation with an electron. Positron has a positive charge, is repulsed by the nucleus, and is mainly annihilated by defects or free volumes especially in polymer. The unsealed liquid radioisotope ^{22}Na is often used as a positron source after drying it in thin foil due to the short penetration depth of the positron. The maximum positron energy of ^{22}Na is 545 keV so that the positrons usually can penetrate a few millimeters in low-density materials. By this reason, we cannot neglect positron annihilation in the source supporting foil even though the thickness of the foil is only a few micrometers. For accurate PALS, we need a source correction for the amount of positron annihilation in the source-supporting foil before the unfolding process of the positron lifetime spectrum.

In this study, the fraction of positron transmission of the source supporting foils and the source correction for PALS were calculated by Monte Carlo simulations, and the results were compared with measurements in the previous literatures.

2. Materials and Methods

We performed Monte Carlo simulations to calculate a fraction of positrons annihilated in the source foils. MCNP6 code, which is applicable for accurate beta particle simulations, was used for the simulations [2]. The simulation geometry is a sandwich structure with a 'sample-Kapton foil-(^{22}NaCl)-Kapton foil-sample' multilayer. Each size of the source and sample geometry was assumed to be 1 × 1 cm^2. We also assumed that the source has no thickness, and isotropically emits positrons from the square plane. For the calculation of source correction, the F1 tally was applied to the surface between the Kapton foil and sample. The thickness of the samples was 1 mm, which is considered that all the positrons fully stop and annihilate within the sample.

2.1 The Fraction of Positron Transmission

The absorption coefficients α of the positron were calculated using the empirical formula. Schrader et al. [1] suggest for the ^{22}NaCl positron source:

$$\alpha = 31.42\rho Z^{0.0878} \qquad (1)$$

, where Z is the average atomic number of the relevant material (Z_{Kapton} = 4.2) and ρ is the mass density 1.42 g/cm^3.

The fraction of positrons transmitted through the foils can be calculated:

$$T = \mathrm{e}^{-\alpha t} \qquad (2)$$

, where t is Kapton foil thickness.

2.2 Source Correction for PALS

In the PALS experiment, most of positrons transmitted through the source supporting foil, and some of the positrons annihilated in the source supporting foil. The transmitted positrons could be backscattered from the sample. By the reason, both backscattering and annihilation should be considered for the source correction.

Several authors proposed the source correction models for PALS analysis. We compared two source correction models with the Monte Carlo simulations.

Bertolaccini and Zappa [3] suggested an empirical formula source correction for metal foils:

$$I_{\text{Bertolaccini}}(\%) = 0.324\, Z^{0.93} {t_{\text{m}}}^{3.45/Z^{0.41}} \qquad (3)$$

, where t_{m} was mass thickness in mg/cm^2.

Monge and del Rio [4] proposed two formulas based on the experimental results. These equations were the intensity expression for a Kapton foil where thickness was 7 µm, and density was 1.42 g/cm^3.

$$I_{\log} = 88.1 + \frac{11.7(0.35\ln Z - 8.11)}{1 - 0.014(0.35\ln Z - 8.11)} \qquad (4)$$

$$I_{\exp} = 3.5 + \frac{4(1-\exp(-0.117Z)}{1-0.68(1-\exp(-0.117Z))} \qquad (5)$$

3. Results

3.1 The Fraction of Positron Transmission

The positron absorption coefficients and the fraction of positron transmission of the Kapton, nickel, and PET foils were summarized in Table 1. The fraction of positron transmission calculated by the equation (2) (T) and Monte Carlo simulations (T_{MC}) for the Kapton, nickel, and PET foils were within 1.7%.

Table 1. The positron absorption coefficients (α) and The fraction of positron transmission calculated by equation (2) (T) and Monte Carlo simulations (T_{MC})

	Thickness (μm)	α	T	T_{MC}
Kapton	7	50.6	0.950	0.948
Nickel	2.5	375.1	0.905	0.913
PET	7	51.2	0.959	0.943

Fig. 1-3 showed The fraction of positron transmission of the Kapton, nickel, and PET foils in different thickness calculated by Monte Carlo simulations, respectively. The results were log-linearly fitted.

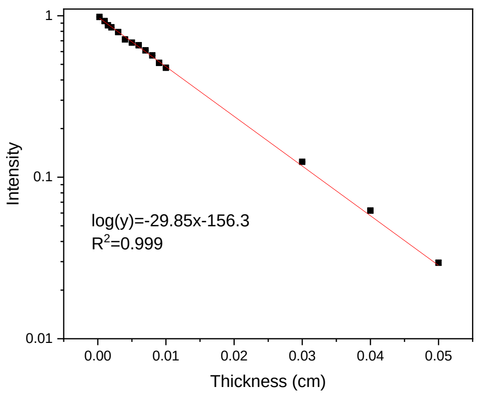

Fig. 1. The fraction of positron transmitted through Kapton foil as a function of thickness. The y-axis is logarithmic scale.

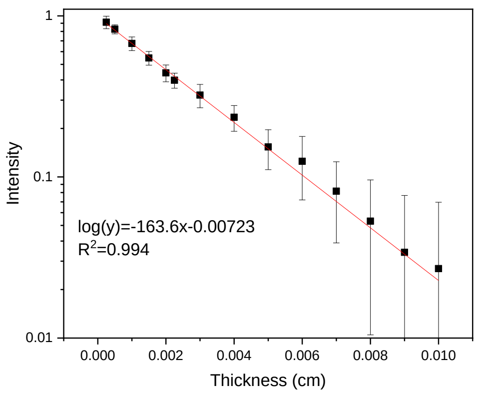

Fig. 2. The fraction of positron transmitted through Ni foil as a function of thickness. The y-scale is logarithmic scale.

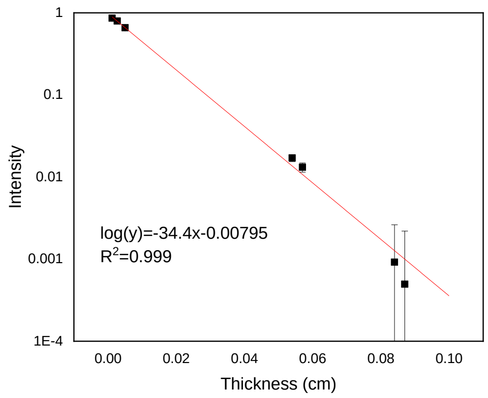

Fig. 3. The fraction of positron transmitted through PET foil as a function of thickness. The y-axis is logarithmic scale.

3.2 Source Correction for PALS

Fig. 4 summarized the source correction for the Kapton foil of 7 μm. The fraction of positron annihilation in the source supporting foil increased when the atomic number Z increased due to the backscattered positrons from the 'source foil-sample' interfaces.

Additionally, the source correction of the nickel foil in 2.5-μm thickness (I_{source}) was calculated for the PALS analysis of the polyethylene terephthalate (PET) samples. The I_{source} for PET was 8.72%. Based on the Bertolaccini and Zappa's model [3], the I_{source} was 8.6%.

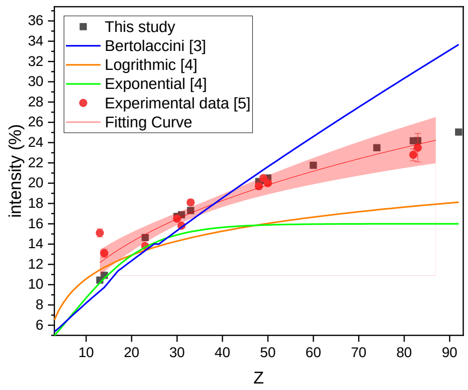

Fig. 4. The fraction of positron annihilated in the 7-μm Kapton source supporting foil in different atomic number, Z. The black dots were Monte Carlo simulation data in this study. The blue line was the modelling data by Bertolaccini and Zappa [3]. The orange and green lines were another modelling data by Monge and del Rio [4]. The red dots and line were the experimental data and fitting curve, respectively [5]. The red shadow was the 95% confidence interval for the red dots.

4. Discussion

The F1 tally in the MCNP code calculated all the number of particles passing through the surface. Without the sample for PALS analysis, the F1 tally results could be directly applied to the fraction of positron transmission because the transmitted positrons were not backscattered. However, in the experimental setup, some of the positrons incident to the sample were backscattered to the source supporting foil. In order to calculate the source correction of the supporting foil from the F1 tally results, the fraction of the backscattered positrons in the sample was eliminated by adding a simple simulation where the source supporting foil was eliminated.

5. Conclusions

The fraction of positron transmission in the source supporting foils and the source correction were calculated by Monte Carlo simulations. The source correction in this study was more compatible with the experimental data than the previous models. The source correction data will be applied for PALS experiments in KAERI.

Acknowledgement

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REFERENCES

[1] D. M. Schrader, S. W. Chin, H. Nakanishi, S. Rochanakij, Positron Annihilation, World Scientific, Singapore, p. 822, 1985
[2] C. J. Werner (editor), MCNP Users Manual - Code Version 6.2, LA-UR-17-29981, 2017.
[3] M. Bertolaccini, L. Zappa, Nuovo Cimento B, Source-supporting foil effect on the shape of positron time annihilation spectra, Vol. 52, pp. 487–494, 1967.
[4] M. A. Monge and J. del Rio, Position annihilation in Kapton source-supporting foils, Journal of Physics: Condensed Matter, Vol. 6, p. 2643, 1994.
[5] N. Djourelov, M misheva, Source correction in positron annihilation lifetime spectroscopy, J. Phys.: Condens.Matter Vol. 8, p. 2081, 1996