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Optimization of the Scintillator Size for Positron Lifetime Measurements

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Abstract. Optimization of the measurement condition for positron annihilation lifetime spectroscopy (PALS) is discussed from the viewpoint of non-destructive "on-site" material inspection. Numerical analysis based on a least-squares estimation and experiments with various sizes of BaF₂ scintillators by conventional PALS suggested that the use of relatively large BaF_2 scintillators enables on-site material inspection by PALS with reasonable accumulation time and time resolution.

1. Introduction

On-site inspection of building structures, in-line flow products, etc. by positron annihilation lifetime spectroscopy (PALS) requires reliable measurements in a reasonably short time [1], preferably, several minutes or less. In conventional positron lifetime measurements it is necessary to sandwich a sodium-22 positron source with two specimens cut out from the material to be inspected and several hours are generally required to accumulate annihilation events for deducing positron lifetimes of the materials. Recently we have developed a positron lifetime measurement system based on an anti-coincidence method [2]. This system enables measurements to be performed without taking out sample pieces from the target materials; it is, in principle, applicable to non-destructive "on-site" inspection of various structures. The use of smaller scintillators in the anti-coincidence system provides a better time resolution but reduces the count rate, as in the conventional positron lifetime measurements.

We discuss analytical relations among the count rate, time resolution, and lifetime evaluation accuracy based on the least-squares estimation method. Using the results of the numerical analysis and the PALS experiments performed with different sizes of BaF₂ scintillators, it is shown that the use of larger scintillators can shorten the time required for the positron lifetime measurements, aimed for non-destructive on-site material inspection.

2. Mathematical modeling and analytical consideration

The effect of the deviations of different parameters on the deviation of the fitting function can be written as the following equation.

$$\delta y(\tau_i) = K_{N1} \delta N_1 + K_{\lambda 1} \delta \lambda_1 + K_{N2} \delta N_2 + K_{\lambda 2} \delta \lambda_2 + K_{\sigma} \delta \sigma + K_{t_0} \delta t_0 + \delta n_b \quad i = 1, \cdots, m$$
(1)

Here $y(\tau_i)$ is the mathematical model of the lifetime histogram which consists of two components having different lifetimes $1/\lambda_1$ and $1/\lambda_2$

$$y(\tau_{i}) = \frac{N_{1}\lambda_{1}\Delta\tau}{\sqrt{2\pi\sigma}} e^{-\lambda_{i}\tau} \int_{-\infty}^{\tau_{i}} e^{-\frac{(t-t_{0})^{2}}{2\sigma^{2}}} e^{\lambda_{i}t} dt + \frac{N_{2}\lambda_{2}\Delta\tau}{\sqrt{2\pi\sigma}} e^{-\lambda_{2}\tau} \int_{-\infty}^{\tau_{i}} e^{-\frac{(t-t_{0})^{2}}{2\sigma^{2}}} e^{\lambda_{2}t} dt + n_{b} \quad i = 1, \cdots, m$$
(2)

where τ_i is the time corresponding to the *i*-th channel and n_b is the background noise. The coefficients in the right hand side of differential equation (1) are given by the following equations:

$$K_{Nk} = \frac{\lambda_k \Delta \tau}{\sqrt{2\pi\sigma}} J_{0k}(\tau_i) \qquad k = 1,2$$
(3)

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$$K_{\lambda k} = \frac{N_k \Delta \tau}{\sqrt{2\pi\sigma}} [(1 - \lambda_k \tau_i) J_{0k}(\tau_i) + \lambda_k J_{1k}(\tau_i)] \qquad k = 1,2$$
(4)

$$K_{\sigma} = \sum_{k=1}^{2} \frac{N_{k} \lambda_{k} \Delta \tau}{\sqrt{2\pi} \sigma^{2}} \left[-(1 - \frac{t_{0}^{2}}{\sigma^{2}}) J_{0k}(\tau_{i}) - \frac{2t_{0}}{\sigma^{2}} J_{1k}(\tau_{i}) + \frac{1}{\sigma^{2}} J_{2k}(\tau_{i}) \right]$$
(5)

$$K_{t_0} = \sum_{k=1}^{2} \frac{N_k \lambda_k \Delta \tau}{\sqrt{2\pi\sigma^3}} [-t_0 J_{0k}(\tau_i) + J_{1k}(\tau_i)]$$
(6)

where N_k is the count of the k-th component. $J_{0k}(\tau_i)$, $J_{1k}(\tau_i)$ and $J_{2k}(\tau_i)$ are defined as follows.

$$J_{0k}(\tau_i) = \int_{-\infty}^{\tau_i} e^{\frac{(t-t_0)^2}{2\sigma^2}} e^{-\lambda_k(\tau_i-t)} dt \qquad k = 1,2$$
(7)

$$J_{1k}(\tau_i) = \int_{-\infty}^{\tau_i} t e^{\frac{(t-t_0)^2}{2\sigma^2}} e^{-\lambda_k(\tau_i - t)} dt \qquad k = 1,2$$
(8)

$$J_{2k}(\tau_i) = \int_{-\infty}^{\tau_i} t^2 e^{-\frac{(t-t_0)^2}{2\sigma^2}} e^{-\lambda_k(\tau_i - t)} dt \qquad k = 1,2$$
(9)

 σ is the standard deviation of the system time resolution, t_0 is the time when the start signal is generated, n_b is the background noise and $\Delta \tau$ is the time width of the frequency channel.

According to the least-squares method, standard deviations s_{N1} , s_{N2} , $s_{\lambda 1}$, $s_{\lambda 2}$, s_{σ} , s_{t0} and s_{nb} of parameters N_1 , N_2 , λ_1 , λ_2 , σ , t_0 and n_b are obtained from the element $[(\boldsymbol{M}^T\boldsymbol{M})^{-1}]_{ii}$ of the diagonal matrix $(\boldsymbol{M}^T\boldsymbol{M})^{-1}$. Here \boldsymbol{M} is a matrix defined by the following equations:

$$\boldsymbol{M} = \boldsymbol{W}\boldsymbol{H} \tag{10}$$

$$\boldsymbol{W} = \begin{bmatrix} 1/\delta n_1 & 0 & \cdots & 0 \\ 0 & 1/\delta n_2 & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \vdots & 1/\delta n_m \end{bmatrix}$$
(11)

$$\boldsymbol{H} = \begin{bmatrix} \boldsymbol{h}_1 & \boldsymbol{h}_2 & \cdots & \boldsymbol{h}_m \end{bmatrix}^T$$
(12)

$$\boldsymbol{h}_{i} = \begin{bmatrix} K_{N1}(\tau_{i}) & K_{N2}(\tau_{i}) & K_{\lambda1}(\tau_{i}) & K_{\lambda2}(\tau_{i}) & K_{\sigma}(\tau_{i}) & K_{\sigma}(\tau_{i}) & 1 \end{bmatrix} \quad i = 1, 2, \cdots, m$$
(13)

 δn_i is the weight used in the least-squares fitting and is described as shown below, considering that positron annihilation is random events which follow the Poisson process.

$$\delta n_i = [y(\tau_i) + \delta n_w^2]^{1/2} \quad i = 1, 2, \cdots, m$$
(14)

where $y(\tau_i)$ is the term equivalent to the square of the standard deviation of count errors which occurred in Poisson process and δn_w is the standard deviation of white noise assumed to be contained uniformly in every channel.

The standard deviation of λ_1 is given as follows:

$$s_{\lambda 1} = ([(\boldsymbol{M}^T \boldsymbol{M})^{-1}]_{22})^{1/2}$$
(15)

The standard deviation $s_{\tau 1}$ of the averaged lifetime τ_1 is denoted by the following formula:

$$s_{\tau 1} = s_{\lambda 1} / \lambda_1^2 \tag{16}$$



Figure 1 Variations of relative standard deviations of positron lifetime for various time resolutions as a function of the average lifetime



Figure 2 Variations of relative standard deviations of positron lifetime for various total counts as a function of the average lifetime

The above-mentioned mathematical model is useful to obtain the numerical relation among the standard deviation of the lifetime, the system time resolution and the total counts of lifetime data. It is also useful to seek the possibility of the existence of additional lifetime components. Considering the typical condition of the application to metallic material inspection, it was assumed in the present numerical analysis that the lifetime spectrum consists of two components: the first component with the average lifetime ranging from 100 ps to 250 ps and the second component with an average lifetime of 380 ps

(the positron lifetime of Kapton to sandwich sodium-22) and 17% frequency (relative intensity). The total number of counts and the time resolution were fixed to 2×10^4 and 150 ps, respectively.

Figure 1 shows an example of the results numerical analysis obtained by under constraint of τ_2 =380 ps. The number of total counts was fixed to 2×10^4 . It is seen that the measurement error of the 150 ps lifetime is 1-2% when the time resolution is 150 ps (FWHM) and the improvement of the time resolution reduces the lifetime estimation errors whatever the lifetime is, but it is not so effective unless the lifetime of the first component is less than 100 ps. Also, it is clear that the lifetime measurement near 50 ps or less will bring about the rapid increase of lifetime estimation errors with time resolution.

Figure 2 shows the variations of the estimation errors of lifetime $1/\lambda_1 = 150$ ps as a function of the average lifetime for total

counts raging from 10K to 1000K. Despite the value of the lifetime $(1/\lambda_1)$ the lifetime estimation error decreases in proportion to the reciprocal of the square root of the total counts of the lifetime data. For example, the error of $1/\lambda_1 \cong 150$ ps is about 2% for the total counts of 10K but it is as low as 0.2% for 1000K counts.

Moreover, another analysis indicates that the use of the second lifetime as a known parameter is effective for the improvement in accuracy. Under the fixed second lifetime of 380 ps and intensity of 17%, the first lifetime of around 150 ps can be determined with several picoseconds errors if the total counts are larger than 2×10^4 .

3. Experiment and consideration

We performed positron lifetime measurements of Si single crystal to study the influence of the scintillator size on time resolution and count rate. A sodium-22 (850 kBq) source was sandwiched between two pieces of a Si wafer. The PAL measurement system used in this experiment was mainly composed of BaF₂ scintillators made by Ohyo Koken Kogyo Co.,LTD, photomultiplier tubes H3378-



Figure 3 Deviation of FWHM and Relative count rate versus thickness of scintillator's

51 produced by Hamamatsu Photonics K.K. and a digital oscilloscope, Wave surfer 44MXs-B, produced by LeCroy corporation. Positron lifetimes were measured by determining the time intervals between 1.27 MeV start γ -rays and 0.511 MeV stop γ -rays. BaF₂ scintillators used for the measurements were 30 mm in diameter on top, 40 mm in diameter on bottom and 10, 20, 30 and 40 mm in thickness. Figure 3 shows variations of time resolution in FWHM and relative count rate

with thickness of the scintillators. The relative count rate is expressed as percentage to the count rate $(2.0 \times 10^4/5 \text{min})$ obtained with two

10 mm thick scintillators. We see that with increase of the size of scintillators the count rate rapidly increases but the time resolution deteriorates rather slowly. With the help of the mathematical model in section 2 and eq.(16) we evaluated standard deviations of lifetime $1/\lambda_1=150$ ps (a typical positron lifetime for a metal) based on the experimentally determined time resolutions and count rates in Fig. 3 (the positron lifetime in Si is ~220 ps and much longer than 150 ps). The evaluated standard deviations of the 150 ps lifetime for 5 min accumulation are listed in table 1. The standard deviation decreases continuously from 2.31% for the 10 mm thick scintillators to 1.39% for the 40 mm thick scintillators. This result indicates that the increased count rate obtainable with a pair of larger scintillators effectively improves the accuracy of the positron lifetime measurements in spite of some worsening of the time resolution. The use of large scintillators can shorten the accumulation time as required by onsite material inspection by PALS.

Table 1 Estimated standard deviations of the 150 pslifetime for 10, 20, 30 and 40 mm thick (5 min accumulation)							
nickness of scintillators [mm]	10	20	30	40			

Thickness of scintillators [mm]	10	20	30	40
Standard deviation of 150 ps lifetime [ps]	2.31	1.55	1.48	1.39

4. Conclusion

With the help of the least-squares estimation theory, which uses the fitting function derived from the linearization of the mathematical model of the positron lifetime histogram, standard deviations of the 150 ps lifetime were evaluated from experimental data of Si obtained with various sizes of BaF_2 scintillators. Our conclusion is that the use of larger scintillators can improve the accuracy of the positron lifetime determination and shorten the time required for the positron lifetime measurements, in particular, those aimed for non-destructive on-site material inspection.

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