Development of a commercial positron annihilation lifetime measurement system

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Abstract. In order to realize a commercial system with a user-friendly interface for positron annihilation lifetime (PAL) measurements, we have applied our previously developed anti-coincidence method to a compact system, controlled by dedicated software with a data analysis module. The functionality of the data-analysis code was confirmed by examining the reproducibility of input average lifetimes for calculated PAL histograms. A prototype for the commercial system was constructed and the validity of the analysis using the system was ensured by measuring a reference material.

1. Introduction
The positron annihilation lifetime (PAL) technique is known as a powerful method to detect open spaces, such as lattice defects and free-volume holes in materials. However, for non-experts of this technique there is some difficulty in adjusting to practical applications due to the complicated data recording and analysis process with a conventional system for bulk-sample measurements. In this work, in order to achieve a commercial digital PAL measurement system with a user-friendly interface, we have applied the anti-coincidence (AC) method [1–3] to a compact measurement system, controlled by dedicated software with a data analysis module.

2. Technical elements for the PAL measurement system
2.1. Anti-coincidence (AC) method
Previously our group developed the AC method for PAL measurements [1–3], in order to overcome the disadvantage of the conventional “sandwich” method [4, 5] using a Na-22 radioisotope as a positron emitter, which requires two specimens so that the positron source is sandwiched by these specimens. A block diagram of PAL measurement based on the AC method is shown in Figure. 1(a).

In this technique, a Na-22 positron source with an activity of 1 MBq is placed on a plastic scintillator (Pilot-U) combined with a photomultiplier tube (PMT; R7400U, HAMAMATSU JPN) so as to construct a positron detector (Figure. 1(b)). The positron source, sealed with 7.5-µm-thick Kapton films, was provided by the Japan Radioisotope Association. The lifetime of a single positron is determined from the time interval $\Delta T$ between the start signal due to the emission of near-simultaneous $\gamma$-ray from the Na-22 nucleus and the corresponding stop signal due to its annihilation in the sample. A lifetime histogram for the sample is obtained by accumulating the annihilation events
with adequate counting-statistics. Annihilation events in the positron detector, corresponding to events where the positron is not annihilated in the sample, are rejected through the AC method. During actual measurements the positron source side of the positron detector is brought into contact with the sample surface so that emitted positrons are introduced into the sample, meaning the AC method enables us to analyze a single specimen without cutting or preparing a pair of samples from the object under study.

2.2. Dedicated software for the measurement system

Dedicated software was developed for a PAL measurement system equipped with a digital storage oscilloscope (DSO; HD4024, Teledyne LeCroy). Up to 200 signals from the $\gamma$-ray and positron detectors are stored in the DSO memory with a sampling rate of 2 GS/s and those are processed to obtain $\Delta T$, followed by the accumulation of a PAL histogram. To determine the detected times of the start and stop signals from the two $\gamma$-ray detectors, consisting of BaF$_2$ scintillator with truncated conical shape of $\phi 30 \text{ mm} \times \phi 40 \text{ mm} \times h 40 \text{ mm}$ and PMT (H3378-51, HAMAMATSU Japan), the constant fraction $[6]$ was fixed to 30% of the signal height, and the energy windows for the signals were assigned to the corresponding photo peaks of 1.27 MeV and 0.511 MeV, respectively (Figure 2). In order to set the upper- and lower-level discriminators (ULD and LLD) for both the windows, the pulse-height histograms of the signals are accumulated, followed by fixing the trigger level for LLD and the maximum level for ULD of the amplitude ranges of the DSO. Prior to

![Figure 1](image1.png)

**Figure 1.** (a) Block diagram of the AC method. (b) Positron source and detector.

![Figure 2](image2.png)

**Figure 2.** (a) Assignment of lower-level discriminator (LLD) and upper-level discriminator (ULD) for the detected $\gamma$-ray signals. (b) Software dialog for adjusting LLD and ULD.
starting measurements, the software can automatically adjust these amplitude ranges.

The software also has a data analysis module, which can evaluate mean positron lifetimes through a least-square fit to the accumulated PAL histogram. In order to ensure the validity of the developed code for the fitting, the reproducibility of input positron lifetimes was examined by analyzing lifetime histograms, mathematically generated with a variation of average positron lifetimes, which simulate a data set for various materials such as metals, alloys, semiconductors, polymers and porous oxides. In the calculations for two kinds of the lifetime histograms with 2 and 3 lifetime components (see Table 1), the time resolution in FWHM, the background level with the Gaussian error function due to random \( \gamma \)-rays coincidence, and the total counting statistics were fixed to 150 ps, 2 counts/ch, and 100 k counts, respectively. Here, spectra with low counting statistics were examined in order to confirm that the analysis program is appropriate for onsite short-time measurements. A typical calculated histogram is shown in Figure 3. Figure 4 shows the relationship between the input average lifetime and the output average lifetime.

Table 1. Input average lifetime and intensity for calculated PAL histograms.

| Number of components | \( \tau_1 \) [ps] | \( I_1 \) [%] | \( \tau_2 \) [ps] | \( I_2 \) [%] | \( \tau_3 \) [ns] | \( I_3 \) [%] |
|----------------------|------------------|--------------|------------------|--------------|----------------|--------------|
| 2                    | 100–350          | 85           | 380              | 15           | N/A            | N/A          |
| 3                    | 150              | 30           | 400              | 30           | 1.5–100.0      | 40           |

3. Prototype for the commercial measurement system

Figure 5 shows the analysis chamber, equipped with the detectors and a high voltage bias, for the prototype of the commercial measurement system. The geometry of the detectors is comparable to that reported previously [1]. We confirmed that the system can measure PAL spectra with a counting rate of \(~75\) cps and a time resolution of \(~220\) ps. Figure 6 displays a PAL histogram obtained for fused silica reference material (NMIJ CRM 5601a) at room temperature. The obtained average lifetime for ortho-positronium (\(\alpha\)-Ps) was \((1.605 \pm 0.007)\) ns, in agreement with the certified value of CRM, \((1.62 \pm 0.05)\) ns, indicating the validity of the measurement system. The geometry of the detectors is now under optimization so that the target values for counting rate (100 cps) and time resolution (\(~180\) ps) are attained.
4. Summary

We applied the AC method to a compact PAL measurement system, controlled by newly developed software with a data analysis module, in order to achieve a commercial digital measurement system with a user-friendly interface. The software can automatically adjust the amplitude ranges of the equipped DSO to set properly the energy windows for the $\gamma$-ray detectors. Testing the calculated lifetime histograms using the developed program signified that the analysis code is able to evaluate properly positron and positronium lifetimes for practical materials. A prototype for the commercial system was constructed, and it was confirmed that the average $\alpha$-Ps lifetime for fused silica reference material observed by the prototype system agrees well with the certified value of the reference material.

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