Drift compensation in dual start/stop data acquisition positron lifetime measurements

Masato Yamawaki¹, Naoya Uesugi², Hirokazu Ando³, and Yoshinori Kobayashi⁴*

¹National Institute of Advanced Industrial Science and Technology, Tsukuba, Ibaraki 305–8565, Japan
²TOYO SEIKO Co., Ltd., Yatomi, Aichi 490–1412, Japan
³Osaka Prefecture University College of Technology, Neyagawa, Osaka 572-8572, Japan
⁴Waseda Research Institute for Science and Engineering, Waseda University, Shinjuku, Tokyo 169-8555, Japan

E-mail: yoshi-kobayashi@aoni.waseda.jp

Received November 28, 2019; revised January 9, 2020; accepted January 22, 2020; published online February 14, 2020

When analyzing positron annihilation lifetime spectra with multiple components using an exponential function with a single component, there is a difference between the experimental data and the fitted spectrum. Compensating for this difference causes a shift in the starting time ($T_0$) in the fitted spectrum. Previously, we applied single-component analysis to the positron lifetime spectrum of a defect-containing metal and confirmed the effectiveness of the shift in $T_0$ ($\Delta T_0$) for defect analysis. To optimally use the $T_0$ variation for defect analysis, however, it is necessary to minimize the drift in the positron lifetime spectrum due to the temporal instability of the photomultiplier tube and high-voltage power supply. In this study, we report significant suppression of the drift by using dual start/stop data acquisition (dual acquisition), which employs each of the two $\gamma$-ray detectors to acquire start and stop signals. © 2020 The Japan Society of Applied Physics

1. Background of research

Positron (annihilation) lifetime measurements¹–⁴ have high sensitivity to open-volume defects in metals and free volumes of polymers. They are used for analysis of atomic vacancies and nanometer-scale holes in various structural and functional materials.

Application of a trapping model⁵ to the defect analysis of a metal sample enables the derivation of the lifetime, which is correlated with the defect size, and the trapping rate, which is proportional to the defect concentration. However, multicomponent analyses of positron lifetime spectra are often difficult. For example, various kinds of defects in the sample, such as dislocations, vacancies or voids, can produce several positron lifetime components of only slightly different lifetimes and high defect concentrations can lead to the shortest lifetime component with an unresolvably short lifetime. To tackle this issue, we have devised a simple method⁶ for defect state analysis by analyzing the lifetime spectrum with a single component to obtain the “mean” positron lifetime and shift $\Delta T_0$ at the starting time $T_0$ of the lifetime spectrum. This shift $\Delta T_0$ is particularly effective in cases of high defect concentrations.⁷ Although the shift $\Delta T_0$ due to defects is as small as 10 ps, we have demonstrated the usefulness of $\Delta T_0$ using the anti-coincidence method,⁸–¹⁰ which allows $T_0$ to be determined with reduced fluctuations from one measurement to another. However, to make the best use of the $T_0$ variation for defect analysis, it is necessary to minimize the drift of the positron lifetime spectrum due to the temporal instability of the photomultiplier tube and high-voltage power supply.

For bulk samples, the positron lifetime is measured from the time difference between the 1.27 MeV decay $\gamma$-ray of $^{22}\text{Na}$ and one of the 0.511 MeV annihilation $\gamma$-rays. The conventional method uses two $\gamma$-ray detectors (Detector_1 and Detector_2), as shown in Fig. 1: Detector_1 measures the 1.27 MeV decay $\gamma$-ray as the start signal, and Detector_2 measures the 0.511 MeV annihilation $\gamma$-ray as the stop signal.

Different energy windows are set for the two $\gamma$-ray detectors, so that Detector_1 does not measure the 0.511 MeV annihilation $\gamma$-ray, and Detector_2 does not measure the 1.27 MeV decay $\gamma$-ray.¹¹–¹³ If lifetime measurements from the 1.27 MeV decay $\gamma$-ray detected by Detector_1 and the 0.511 MeV $\gamma$-ray detected by Detector_2 are made simultaneously with lifetime measurement from the 0.511 MeV annihilation $\gamma$-ray by Detector_1 and the 1.27 MeV annihilation $\gamma$-ray by Detector_2 (dual start/stop data acquisition), two positron lifetime spectra can be obtained simultaneously. In this paper, we call this measurement method dual acquisition and the conventional method single acquisition. Various factors contribute to the drift in positron lifetime spectra, but those resulting in a relative time difference between the two signals from the detectors would be corrected by taking the average of the two values of $T_0$ obtained by the dual acquisition method.¹⁴

In this study, we developed a dual acquisition system, to compare the temporal drifts in the positron lifetime spectrum between single and dual acquisition.

2. Experimental method

In the conventional positron lifetime measurement for bulk samples, Lifetime_1 is measured as the time difference between the start signal from Detector_1 and the stop signal from Detector_2 as shown in Fig. 2(a). In the dual acquisition method, Lifetime_2 is measured in parallel, as shown in Fig. 2(b). This yields two simultaneous positron lifetime spectra and effectively doubles the count.

Figure 3 shows the energy spectrum of $^{22}\text{Na}$ and the energy windows used in the single acquisition and dual acquisition methods. In the single acquisition method, the energy windows are set as shown by the broken lines, and DSO Channel_1 is used to provide the start signal (the 1.27 MeV total-absorption peak), and Channel_2 to provide the stop signal (the 0.511 MeV total-absorption peak). In the dual acquisition method, a wide energy window is set to

---

© 2020 The Japan Society of Applied Physics
include both the start and stop signals. The recorded signals are separated into start and stop signals in the program. In this way, it is possible to obtain two positron lifetime spectra simultaneously with start and stop signals from both Detector_1 and Detector_2.

We used PSA-TypeL-II\textsuperscript{15}) from Toyo Seiko Co. Ltd. for the positron lifetime measurement. This system uses the anticoincidence method, which allows the measurement of a single sample. For the \( \gamma \)-ray detector, we employed a photomultiplier tube and a BaF\textsubscript{2} scintillator (size: \( \phi 30 \text{ mm} \times \phi 40 \text{ mm} \times 40 \text{ mm} \)), which were optically connected with each other with optical grease. The photomultiplier tube was model H3378-51 by Hamamatsu Photonics K. K., the BaF\textsubscript{2} scintillator was from Ohyo Koken Kyogo Co., Ltd., the high-voltage power supply module was HPMR-3N from Matusada Precision Inc., the sealed \( ^{22}\text{Na} \) positron source was NA351S by Japan Radioisotope Association and the DSO was model HDO4024 from Teledyne LeCroy, Inc.

We performed measurements of a sample of stainless steel SUS304, from which defects had been removed by “bright annealing” for approximately 2 h (about 500,000 counts), repeating the measurements continuously for 45 h. For signal processing and analysis, we used the positron lifetime measurement program IPALM by Toyo Seiko Co., Ltd. For the positron lifetime spectra, we performed single-component analysis taking into account the source component, whose lifetime was fixed at 380 ps, the positron lifetime in Kapton\textsuperscript{®}.

We used a single Gaussian function for the time resolution, setting the width (i.e. the FWHM) as a free parameter.

3. Results

Figure 4 shows an example of two positron lifetime spectra obtained simultaneously with dual acquisition. DSO Channel_1 and Channel_2 had a wide energy window, as shown in Fig. 3(b). After recording the waveform within this energy window, we separated the start and stop signals using a software program. Figure 4(a) shows the positron lifetime spectrum measured using the method shown in Fig. 2(a), while Fig. 4(b) shows the positron lifetime spectrum measured using the method shown in Fig. 2(b).

The lifetime spectra were successfully analyzed into a single component due to positron annihilation in the bulk. The positron lifetime we obtained from the spectrum in Fig. 4(a) was 105.9 ± 0.7 ps and the positron lifetime from the spectrum in Fig. 4(b) was 105.0 ± 0.7 ps. An analysis of the positron lifetime spectrum obtained with energy windows by single acquisition, as shown in Fig. 3(a), yielded a positron lifetime of 105.2 ± 0.7 ps. The vertical resolution of many DSOs is 8 bits (256 ch), but the vertical resolution of HDO4024 is 12 bits (4096 ch). This makes it possible to use the dual acquisition method, with a wide energy window as shown in Fig. 3(b). Comparing the time resolution for reference, when dual acquisition was performed with the settings shown in Fig. 3(b), the time resolution (FWHM) in Fig. 4(a) is 177.6 ps, and that (FWHM) in Fig. 4(b) is 180.8 ps. The time resolution (FWHM) was almost equivalent to 175.6 ps when the single acquisition in Fig. 3(a) was set.

We used the two positron lifetimes \( \tau (a) \) and \( \tau (b) \) and their standard deviations \( \sigma (a) \) and \( \sigma (b) \) obtained by dual acquisition to evaluate the combined positron lifetime \( \tau \text{sum} \) and standard
deviation $\sigma_{\text{sum}}$ using the following equations:

$$\tau_{\text{sum}} = \frac{\tau(a) \times \sigma(b)^2 + \tau(b) \times \sigma(a)^2}{\sigma(a)^2 + \sigma(b)^2}$$

$$\sigma_{\text{sum}} = \frac{\sigma(a) \times \sigma(b)}{\sqrt{\sigma(a)^2 + \sigma(b)^2}}.$$

The results were $\tau_{\text{sum}} = 105.5$ ps and $\sigma_{\text{sum}} = 0.49$ ps.

Figure 5 shows the temporal stability of $T_0$ in the positron lifetime spectra when measured repeatedly by the dual acquisition method. The symbol $\triangle$ shows the values of $T_0$ in the positron lifetime spectra obtained from the positron lifetime spectra measured using the method shown in Fig. 2(a) (Spectrum_1), the symbol $\square$ shows the values of $T_0$ in the positron lifetime spectra measured using the method shown in Fig. 2(b) (Spectrum_2), and the symbol $\bullet$ shows the average of the two. The standard deviations were 2.20 ps for $\triangle$, 2.30 ps for $\square$ and 0.35 ps for $\bullet$.

4. Discussion

Figure 6 shows the relationship between the values of $T_0$ obtained from Spectrum_1 and Spectrum_2: there was a negative correlation between the two. The slope of the correlation was almost $-1$, with a correlation coefficient $R = -0.9527$. The significant reduction in the variation of $T_0$ obtained by averaging, shown in Fig. 5, was likely due to this strong negative correlation.

We believe that the drift in the positron lifetime spectrum was mainly due to the fluctuation in the output of the high-voltage power supply. The transit time of electrons in the photomultiplier tube became short when the applied voltage was high and long when the applied voltage was low. Because the data represented by the symbols $\triangle$ and $\square$ showed relatively large drifts, as shown in Fig. 5, there were likely fluctuations in the high-voltage power supply on the time scale of a few hours.

Comparison of the standard deviations of the values of $T_0$ obtained in the experiment (Fig. 5) with those obtained from the fitting analysis of the lifetime spectra shows that the standard deviations of $\triangle$ ($T_0$ of Spectrum_1) and $\square$ ($T_0$ of Spectrum_2) of 2.20 ps and 2.30 ps, respectively, are far greater than the standard deviation of $T_0$ from the fitting analysis, 0.44 ps. This indicates the large instability of the measurement apparatus. However, the standard deviation of $\bullet$ (the average of $\triangle$ and $\square$) was 0.35 ps, which is similar to the standard deviation (0.44 ps) by fitting analysis. This demonstrates that averaging the values of $T_0$ from Spectrum_1 and Spectrum_2 obtained by the dual acquisition method suppresses the drift due to the instability of the measurement apparatus.

5. Summary

In this study, we developed a dual acquisition system and evaluated the temporal drift in the positron lifetime spectrum. The dual acquisition method not only doubles the count rate.
but also corrects the drift in the positron lifetime spectrum by averaging the values of $T_0$ obtained from the two positron lifetime spectra.

The shift $\Delta T_0$ obtained by analyzing the positron lifetime spectra of metals with defects is useful for defect analysis, but the evaluation of high defect concentrations requires a precision of 1 ps or lower.\(^1\) By correcting the drift in the positron lifetime spectrum using the dual acquisition method, it is possible to maximize the accuracy of defect analysis using $\Delta T_0$. In the future, we would like to examine the applications of this technique to other measurement methods.

### Acknowledgments

We gratefully acknowledge comments by Dr. Tetsuya Hirade of JAEA and Dr. Brian O’Rourke of AIST. This work was supported by JSPS KAKENHI Grant No. 17K05123.

---

1. S. J. Tao, J. Chem. Phys. 56, 5499 (1972).
2. Y. Kobayashi, W. Zheng, E. F. Meyer, J. D. McGervey, A. M. Jamieson, and R. Simha, Macromolecules 22, 2302 (1989).
3. Y. C. Imam, Microchem. J. 42, 72 (1990).
4. J. Kansy, Nucl. Instrum. Methods Phys. Res., Sect. A 374, 235 (1996).
5. D. C. Conners and R. N. West, Phys. Lett. 30A, 24 (1969).
6. M. Yamawaki and N. Uesugi, Japanese Patent Application No. 2018-228103.
7. M. Yamawaki, N. Uesugi, and Y. Kobayashi, Jpn. J. Appl. Phys. 58, 126501 (2019).
8. C. Hugenschmidt, U. Holzwarth, M. Jansen, S. Kohn, and K. Maier, J. Radioanal. Nucl. Chem. 210, 583 (1996).
9. M. Yamawaki, Y. Kobayashi, K. Hattori, and Y. Watanabe, Jpn. J. Appl. Phys. 50, 086301 (2011).
10. M. Yamawaki, Y. Kobayashi, K. Hattori, and Y. Watanabe, Patent No. 8785875 (US) Patent No. 5843315 (JP).
11. L. Van Hoorebeke, A. Fabry, E. van Walle, J. Van de Velde, D. Segers, and L. Dorkens-Vanpraet, Nucl. Instrum. Methods Phys. Res., Sect. A 371, 566 (1996).
12. H. Saito and T. Hyodo, Phys. Rev. Lett. 90, 193401 (2003).
13. T. Hirade, H. Ando, K. Manabe, and D. Ueda, Nucl. Instrum. Methods Phys. Res., Sect. A 931, 100 (2019).
14. M. Yamawaki, Japanese Patent Application No. 2019-214447.
15. M. Yamawaki, K. Ito, K. Hattori, and N. Uesugi, J. Phys.: Conf. Ser. 791, 012038 (2017).