Feasibility study of non-destructive defect analysis by positron annihilation lifetime measurement

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We examined the feasibility of on-site non-destructive defect analysis using positron annihilation lifetime measurement. This analysis requires a short measurement time and lattice defect information has to be extracted from positron lifetime data that involve high statistical uncertainties. In this study, we simulated positron lifetime spectra for a short measurement of approximately 1 min, using experimental positron lifetimes of shot-peened stainless steel, and studied the variations of the center-of-gravity positron lifetime, the “mean” positron lifetime and the shift in the starting time (ΔT₀) with the positron trapping rate into defects, which is a physical quantity proportional to the lattice defect density. As a result, distinguishing the positron lifetime spectra of different trapping rates in a range of up to 1.1 × 10¹¹ s⁻¹ was possible by creating a 2D plot of the “mean” positron lifetime and ΔT₀. © 2020 The Japan Society of Applied Physics

1. Introduction

Positron annihilation lifetime measurements¹–⁴ have high sensitivity to open-volume defects in metals and free volumes of polymers. For this reason, they are used for analysis of atomic vacancies and nanometer-scale holes in various structural and functional materials. The required measurement time depends on the type of the positron lifetime spectrometer and the radiation activity of the positron source. Usually, it is approximately 3 h and during this time, more than 10⁸ counts of positron annihilation events are accumulated. By using a multi-component analysis of the positron lifetime spectrum of a metal or semiconductor containing lattice defects and applying the trapping model⁵ to the analysis result, one can obtain the lifetime of the positron in the defects (correlated with the defect size), and the trapping rate of positrons into defects (proportional to the defect density).

However, in many cases, multi-component analysis of the positron lifetime spectrum is difficult. For example, an on-site non-destructive defect analysis using positron annihilation lifetime measurements requires a measurement time of approximately 1 min;⁶⁻⁷ however, directly obtaining the lifetime of positrons in the defects and the trapping rate from the positron lifetime data by multi-component analysis is extremely difficult.

Often used for evaluating positron lifetimes from lifetime data obtained in a short time are the center-of-gravity positron lifetime⁸,⁹ and the “mean” positron lifetime¹⁰,¹¹ obtained by a single-component analysis to a multi-component spectrum. The center-of-gravity positron lifetime and the “mean” positron lifetime increase with increasing not too high defect density; therefore, they can be used as parameters that reflect the relatively low-defect density. For example, Oshima et al. succeeded¹² in obtaining imaging data (approximately 8000 pixels) of the lattice defect distribution in metal samples by calculating the center-of-gravity positron lifetime based on the positron lifetime data obtained from a pulsed energy-variable high intensity positron microbeam.¹² However, if a large number of positrons are annihilated in the defects, the center-of-gravity positron lifetime and the “mean” positron lifetime do not accurately reflect the defect density. Using simulations, we found that the shift in the starting time T₀ (ΔT₀) of the positron lifetime spectrum obtained in the single-component analysis changed with the defect density even in the case where most positrons were annihilated in the defects.¹³,¹⁴ Additionally, we validated the utility of ΔT₀ in the defect analysis by using an anti-coincidence method¹⁵–¹⁸ that can minimize the fluctuation of T₀ from one spectrum to another for shot-peened stainless steel with defects (dislocations and atomic vacancies). Furthermore, we reported that the drift in the positron lifetime spectrum, caused by temporal instability of photomultiplier and high-voltage power supply can be corrected using dual start stop data acquisition.¹⁹,²⁰

On comparing the behaviors of the centre-of-gravity positron lifetime, the “mean” positron lifetime and ΔT₀, we observed that the first two markedly change only in low-defect-density regions, while the third showed a considerable change in the high defect densities but with complicated dependency on defect density. Based on this fact, obtaining information about the defects in a wide-density range from the positron lifetime data may be possible by combining multiple parameters with varying dependencies on the defect density. In this study, we simulated some positron lifetime spectra for a short measurement and studied the variations of the centre-of-gravity positron lifetime, the “mean” positron lifetime and ΔT₀ with the positron trapping rate into defects, which is a physical quantity proportional to the lattice defect density. As a result, distinguishing the positron lifetime spectra of different trapping rates up to a range of 1 × 10¹¹ s⁻¹ was possible by creating a 2D plot of the “mean” positron lifetime and ΔT₀.

2. Positron lifetime spectrum simulation

We simulated some positron lifetime spectra with Monte Carlo simulation¹⁴ that reproduced the trapping model in which a positron was annihilated in the 2 states of lattice...
(bulk) or defect. According to Ref. 20, 1 min measurement corresponds to approximately 10⁴ counts in positron lifetime spectrum. By considering on-site non-destructive defect analysis, we fixed the total number of positron annihilation events to 10⁴ and generated positron lifetime spectra using the trapping rate, product of specific trapping rate and the defect density, in a range from 1 × 10⁶ s⁻¹ to 1 × 10¹¹ s⁻¹. For the positron lifetimes in bulk and in defects, we used experimental positron lifetime of shot-peened stainless steel⁴,2¹ that were 100 ps and 160 ps (a lifetime accounting for multiple defects), respectively. By considering an experiment using a commercial anti-coincidence system,²¹,²² we used a source component with a 380 ps lifetime (the positron lifetime in the source container, Kapton) and 25% intensity, a time resolution (FWHM) of 180 ps and a channel width of 10 ps/ch. Finally, we added a background of 0.1 counts/ch. The somewhat high intensity of this source component is due to that positrons annihilated in the source are not eliminated by anti-coincidence processing.¹⁶ There are cases where positrons annihilate from 3 states (bulk, dislocations, vacancies); this happens as a result of shot-peening and metal fatigue. In some cases positrons annihilate from even more different states, making it appropriate to consider positron lifetime distribution. Our previous simulation¹⁴ showed that the tendency of ΔTc variation depends on the number of the states and the lifetime distribution. In the present study, we restrict ourselves to the case of 2-state positron trapping simply as a first step towards more complicated cases. Figure 1 shows an example of the positron lifetime spectrum simulation, and Table I shows the parameters of each positron lifetime spectrum group generated by the simulation.

3. Results

3.1. Center-of-gravity positron lifetime

Based on the report by Oshima et al.⁹ we obtained the centre-of-gravity positron lifetime (Tc) from the simulated positron lifetime spectrum using the following equation:

\[ T_c = \sum_{\tau = 700}^{1200} \tau(i) \times \text{count}(i) \]

\[ \sum_{\tau = 700}^{1200} \text{count}(i) \]

Here, \( \tau(i) \) and \( \text{count}(i) \) are the time and count for channel \( i \), respectively. The origin of the temporal axis corresponds to the simulation time 0, and we obtain \( T_c \) by integrating over the range from -1 to 7 ns. We used Microsoft Excel for the computation.

Figure 2 shows the relationship between the trapping rate and the centre-of-gravity positron lifetime. With an increasing trapping rate, the centre-of-gravity positron lifetime gradually increased from approximately 190 ps and reached approximately 230 ps with a trapping rate of 5 × 10¹⁰ s⁻¹ before becoming constant. A constant centre-of-gravity positron lifetime indicates the annihilation of most positrons in defects.

The centre-of-gravity positron lifetime is approximately 90 ps longer than the bulk positron lifetime (100 ps) set by the simulation. Moreover, the width of change in the centre-of-gravity positron lifetime by the trapping rate is approximately 40 ps, which is smaller than the difference between the bulk lifetime and defect lifetime (60 ps) set by the simulation. This is likely caused by the Kapton lifetime component of 25% set as the source component. In fact, without the Kapton component, the positron lifetime spectrum simulation with 1.2 × 10⁵ counts resulted, with the trapping rate ranging from 1 × 10⁶ s⁻¹ to 1 × 10¹¹ s⁻¹, in a width of change of 55 ps (from 121.7 ps to 176.7 ps) in the centre-of-gravity positron lifetime that was close to the difference between the bulk lifetime and the defect lifetime.

| Table I. Main parameters of each positron lifetime spectrum group generated by the simulation. |
|------------------------------------------|
| Trapping rate (s⁻¹) | Total counts | Number of simulated spectra |
| 1 × 10⁶ | 10⁴ | 12 |
| 5 × 10⁶ | 10⁴ | 12 |
| 1 × 10¹⁰ | 10⁴ | 12 |
| 2 × 10¹⁰ | 10⁴ | 12 |
| 5 × 10¹⁰ | 10⁴ | 12 |
| 1 × 10¹¹ | 10⁴ | 12 |

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**Fig. 1.** (Color online) Example of a 2-state positron lifetime spectrum generated by simulation. By considering short on-site non-destructive defect analysis, we set the total number of positron annihilation events to 10⁴, of which 25% were caused due to the source component with a lifetime 380 ps. ■ represents the component due to positron disappearing from the bulk, ○ represents the component of positron annihilation in the defect and □ is the sum of ■, ○, background (BG) and source component (Kapton).

**Fig. 2.** Relationship between the centre-of-gravity positron lifetime (Tc), calculated from the simulated positron lifetime spectrum (10⁴ counts), and the trapping rate.
from the simulated positron lifetime spectrum (104 counts), as a function of trapping rate. Because Kapton intensity Fix (25%) Kapton lifetime Fix (380 ps) sample. The width of change is larger than that of the centre-of-gravity positron lifetime; resulting in a similar value to the positron lifetime in the saturated at approximately 160 ps. The trend of change is lifetime gradually increased from approximately 100 ps and (60 ps). The remainder is likely due to the effect of the background integrated from −1 ns to 7 ns.

3.2. “Mean” positron lifetime and ΔT0 deduced by single-component analysis, taking account of the source component

We conducted a single-component analysis, taking account of the source component, of the positron lifetime spectrum generated by the simulation. We used the positron lifetime measurement software IPALM by Toyo Seiko Co., Ltd.22) Table II shows the fitting conditions of the analysis. We conducted the analysis with a fixed lifetime and a fixed source component intensity.

Figure 3 shows variations of “mean” positron lifetime (τm) and ΔT0 deduced by the single-component analysis, taking account of the source component, with trapping rate of positrons into defects. When approximating a 2-component positron lifetime spectrum with a single-component exponential function, a discrepancy arises between the lifetime spectrum and the fitting. T0 shifts to compensate for the discrepancy.14) When no positrons are trapped by defects or all of them are trapped by a single type of defect, the positron lifetime spectrum would have one component, thereby leading to ΔT0 of 0 ps.14)

With an increasing trapping rate, the “mean” positron lifetime gradually increased from approximately 100 ps and saturated at approximately 160 ps. The trend of change is similar to that of the centre-of-gravity positron lifetime; however, it is not affected by the source component, thereby resulting in a similar value to the positron lifetime in the sample. The width of change is larger than that of the centre-of-gravity positron lifetime, and its value is almost the same as the difference between the bulk lifetime and defect lifetime (60 ps). Conversely, −ΔT0 increased once, peaking to $1 \times 10^{10}$ s$^{-1}$−$2 \times 10^{10}$ s$^{-1}$, before decreasing. The change in −ΔT0 with the trapping rate is of similar magnitude to the fluctuation in −ΔT0 at the same trapping rate, thereby distinguishing the defect conditions solely by ΔT0 becomes difficult. For example, when the results for trapping rates $5 \times 10^{10}$ s$^{-1}$ and $1 \times 10^{11}$ s$^{-1}$ are compared, the change in −ΔT0 (from 7.4 to 4.3 ps) with the change in the trapping rate is 3.1 ps, while the fluctuation in −ΔT0 is 1.9 and 1.7 ps (3.6 ps in total).

3.3. Correlation between ΔT0 and the centre-of-gravity positron lifetime and the “mean” positron lifetime

Because distinguishing the positron lifetime spectra obtained in a short measurement time solely by ΔT0 is difficult, we examined approaches that consider ΔT0 together with either the centre-of-gravity positron lifetime or the “mean” positron lifetime.

Figures 4 and 5 show the centre-of-gravity positron lifetime plot and the “mean” positron lifetime plot, respectively,
with respect to $-\Delta T_0$. In both the plots, the data group with the constant trapping rate moves to the right (both positron lifetimes increase) with an increasing trapping rate and upward ($-\Delta T_0$ increases) before moving downward ($-\Delta T_0$ decreases). In Fig. 4, the fluctuations within the same trapping rate show no correlation and the data overlap with those in groups with other trapping rates. In contrast, the fluctuations show a liner correlation and the data little overlap with those in other groups in Fig. 5. Moreover, this correlation, shown in the figure, is effective in distinguishing the positron lifetime spectra with trapping rates higher than $1 \times 10^{11}$ s$^{-1}$. Table III shows the slopes of approximate straight lines and the correlation coefficients between $-\Delta T_0$ and the centre-of-gravity positron lifetime as well as the “mean” positron lifetime obtained from the positron lifetime spectra group with same trapping rate. Even though the statistical precision is low, all the groups of the “mean” positron lifetime have a correlation with the positive slopes.

Little overlap between data with different trapping rates, as shown in Fig. 5, suggests estimating the trapping rate based on the “mean” positron lifetime and $-\Delta T_0$ of the positron lifetime spectrum obtained in short measurement time is possible provided that the master curve relating the “mean” positron lifetime and $-\Delta T_0$ with high statistical accuracy to the trapping rate is available. Although the “mean” positron lifetime and $-\Delta T_0$ in the data from short measurements would deviate from the master curve because of statistical uncertainties, identifying the location on the master curve (i.e. estimating the trapping rate) would be possible based on a single measurement data using correlation for the data group with the same trapping rate.

### 4. Summary

In this study, we simulated a positron lifetime spectrum for a short measurement of approximately 1 min and studied the variations of the centre-of-gravity positron lifetime, “mean” positron lifetime and $-\Delta T_0$ with the positron trapping rate into defects. The latter two quantities ($\tau_m$ and $-\Delta T_0$) were obtained by single-component analysis, taking account of the source component. As a result, distinguishing positron lifetime spectra of different trapping rates in a range of up to $10^{11}$ s$^{-1}$ was possible by creating a 2D plot of the “mean” positron lifetime and $-\Delta T_0$. For constant trapping rate, the “mean” positron lifetime shows a characteristic correlation with $-\Delta T_0$, there by estimating the trapping rate, which is physical quantity proportional to the defect density, becomes possible by using the “mean” positron lifetime together with $-\Delta T_0$.

The 2D plot of the “mean” positron lifetime and $-\Delta T_0$ obtained from single-component analysis by taking the source component into account can be used in inspection for infrastructures and quality management in shot-peening processes. In the future, we would like to work on simulation and experiments that focus on establishing the industrial use of positron annihilation lifetime measurements using a commercial positron annihilation lifetime measurement system$^{21}$ for non-destructive analysis.

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### Table III

| Trapping rate (s$^{-1}$) | Center-of-gravity positron lifetime, $\tau_m$ | “Mean” positron lifetime, $\bar{\tau}_m$ |
|-------------------------|---------------------------------|---------------------------------|
| $1 \times 10^6$         | $-1.1 \times 10^{-4}$          | $-0.4 \times 10^{-4}$          |
| $1 \times 10^9$         | $-1.1 \times 10^{-4}$          | $-0.4 \times 10^{-4}$          |
| $2 \times 10^9$         | $0.6 \times 10^{-4}$           | $0.10$                          |
| $3 \times 10^9$         | $2.1 \times 10^{-4}$           | $0.35$                          |
| $5 \times 10^9$         | $2.1 \times 10^{-4}$           | $0.35$                          |
| $1 \times 10^{10}$      | $2.0 \times 10^{-4}$           | $0.26$                          |
| $5 \times 10^{10}$      | $2.0 \times 10^{-4}$           | $0.26$                          |
| $1 \times 10^{11}$      | $2.0 \times 10^{-4}$           | $0.26$                          |

The slopes of approximate straight lines and the correlation coefficients between $-\Delta T_0$ and the centre-of-gravity positron lifetime ($\bar{\tau}_m$) as well as the “mean” positron lifetime ($\tau_m$) obtained from the positron lifetime spectra group (10$^4$ counts) with the same trapping rate.

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