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Sealed ^{22}Na sources for positron annihilation lifetime spectroscopy

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Abstract. Sealed radioactive sources of ^{22}Na for positron annihilation lifetime spectroscopy (PALS), free from the legal regulations, are commercially available from an American company. However, thick foils are used to seal ^{22}Na in these sources and large fractions of the positrons annihilate in the sealing materials. Further, it is pointed out that a long lifetime component over 1 ns appears in the positron lifetime spectrum acquired with the Kapton sealed source (POSK-22, IPL Inc.). In this research, attempts were made to develop high quality sealed positron sources, potentially free from regulations, for ubiquitous application of PALS. The sources prepared in the present work are of high quality and applicable to our new PALS, which does not require sample cutting and is potentially applicable to truly nondestructive, onsite inspection of various materials.

Introduction

Positron annihilation lifetime spectroscopy (PALS) is certainly a powerful means for monitoring and inspecting the fatigue and damage to various materials, but traditional PALS requires a pair of specimens, typically of 20 × 20 × 1 mm^3 size. The two specimens have to be cut out from the sample material and it is not possible to perform in situ measurements of various materials such as those used in nuclear reactors by traditional PALS. We have developed a new system, which does not require sample cutting and is potentially applicable to truly nondestructive onsite inspection of various materials [1].

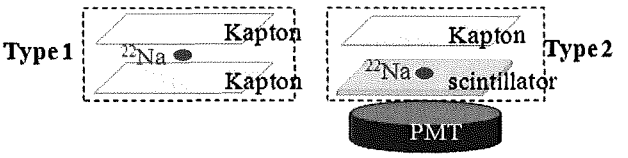
Apart from the requirement of two specimens in traditional PALS, there is a problem concerning the radiation source. For example, in Japan the use of a hand-made radiation source is allowed, in most cases, only in the radiation-controlled area for unsealed radioactive isotopes, legally approved by the Government. On the other hand, the use of a sealed ^{22}Na source produced by an authorized supplier is not limited to the radiation-controlled area, as far as their activity is equal to or less than 1 MBq. A sealed ^{22}Na radiation source for PALS (POSK-22, IPL Inc.), free from regulations, is commercially available. However, there appears a long lifetime component in the positron lifetime spectrum, although ^{22}Na is sealed between two Katpon films without long positron lifetimes [2]. Perhaps the long lifetime component is due to positron annihilation in adhesive used for gluing Kapton films together. Therefore, for ubiquitous application of PALS, sealed ^{22}Na radiation sources with better quality are highly desired.

In the present study, we produced sealed ^{22}Na radiation sources and tested their quality by performing PAL measurments for single crystal Si. The data were collected with a traditional PAL system using two specimens sandwiching the positron source as well as with our new PAL system using one specimen.

Experiment

1. source preperation

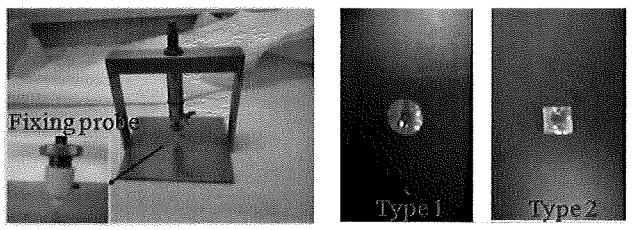
We produced two types of sealed ^{22}Na radiation sources (Fig. 1). Type 1 is 60 kBq and 850 kBq ^{22}Na sealed between two 7.5 μm thick Kapton sheets. Type 2 is 850 kBq ^{22}Na sealed between 7.5 μm thick Kapton and a plastic scintillator of 15 × 15 × 5 mm^3 size. The Type 1 source can be used for traditional PALS as well as for our novel PALS. The Type 2 source is for our novel PALS.



Arrangement of Kapton films in radiation sealing

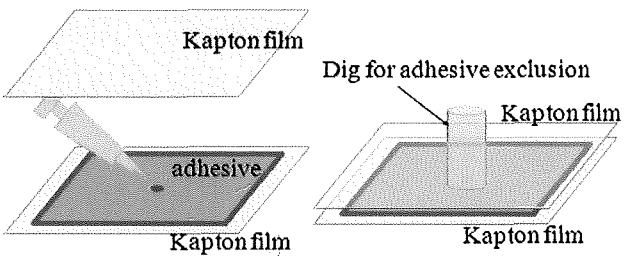
Fig. 1 Sealed radiation sources produced. Type 1 is 60 kBq and 850 kBq ^{22}Na sealed between two 7.5 μm thick Kapton sheets. Type 2 is 850 kBq ^{22}Na sealed between 7.5 μm thick Kapton and a plastic scintillator of 15 × 15 × 5 mm^3 size. .

We used the apparatus shown in Fig. 2 to prepare the sealed ^{22}Na radiation sources. This apparatus was designed to minimize the contamination of the dried spot of aqueous ^{22}Na solution with adhesive for gluing the source sealing materials. The dried spot of ^{22}Na on a Kapton film or a scintillator was covered with another sheet of Kapton. Intrusion of the glue was prevented by covering the area of the ^{22}Na spot with the cylinder of the apparatus as shown in Fig. 3.



Production device Sealed radiation source

Fig. 2 Photographs of the apparatus producing sealed radiation sources (left) and the sources prepared (right). The Type 1 and Type 2 sources were made by Japan Radioisotope Association, an authorized supplier of sealed radioactive sources in Japan.



Conventional bonding method Improved bonding method

Fig. 3 New way of sealed source preparation. The left part shows the traditional way to seal ^{22}Na by Kapton films. The right part shows a new way to seal ^{22}Na by Kapton films Intrusion of the glue is prevented by covering the area of the ^{22}Na spot with the cylinder of the apparatus of Fig. 2.

2. PAL measurements

For evaluating the quality of the prepared sources PAL measurements were conducted with two systems, a traditional PAL spectometer and our novel PAL spectrometer.

The traditional system is a two detector spectrometer. The detectors are two BaF_2 scintillators directly coupled to photomultiplier tubes (PMTs), which detect the birth γ-ray of the β^+-decay from the source and one of the annihilation γ-rays of an energy of 511 keV, respectively. The PAL data were collected using two specimens to sandwich the radioactive source.

The concept of our new PAL system is shown in Figure 4. This system consists of two parts, conventional PAL detectors and a positron detector. The positron detector consists of a thin plastic scintillator, a mirror box and a PMT. The scintillation light generated upon the passage of a positron through the scintillator is guided to the PMT via a mirror box. In PAL measurements, those events, where the signals from the PAL detectors and the positron detector are coincident with each other, are removed from the lifetime spectra by anti-coincidence processing. For the measurments with the Type 1 source the source was sandwiched between the positron detector and Si specimen. For the

measurments with the Type 2 source, the scintillator of the source was used as the sintillator of the positorn detector and the Kapton side of the source is placed on the Si specimen.

Result and discussion

Fig. 5 shows PAL spectra of single crystal Si recorded with the commercially available sealed source (POSK-22, produced by IPL Inc. on 2006.2.1. The original activity was 740 kBq and the current activity is about 190 kBq) and with the Type 1 source (60 kBq) prepared in the present study. The long lifetime component in the former spectrum obtained with POSK-22, likely due to positron annihilation in the adhesive for sealing the two Kapton films, is dramatically reduced in the latter spectrum obtained with the Type 1 source. This reveals that by using the apparatus in Fig. 2 and the method of Fig. 3 the sealed source with little contamination from positron annihilation in the adhesive can be prepared.

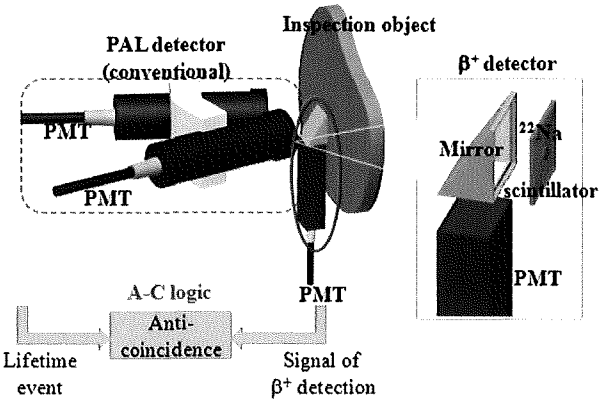


Fig. 4 New PAL system, A-C (Anti-coincidence) logic in the figure is used to exclude events where the signals from the γ-ray detectors and the positron (β^+) detector are coincident with each other.

Figure 6 shows positron lifetime spectra of Si single crystal acquired with the new PAL system in Fig. 5 and the traditional PAL system without the positron detector. The blue and red dotted lines represent the data obtained using the new system with and without A-C processing, respectively. The green dotted line shows the data obtained with the traditional system using sandwich geometry. The Type 1 source (850 kBq) was used for all the measurements. The effect of the A-C processing on the long lifetime component is evident from this figure. The lifetime component of several nanoseconds is clearly weakened in the PAL data acquired with A-C processing (blue dotted line) in comparison with the PAL data acquired without A-C processing (red dotted line). This means that events due to positron annihilation outside the sample material, involving the long lifetime component, can be successfully excluded from the PAL data by using the A-C method. The PAL data recorded with A-C processing are equivalent to the data obtained with the traditional system (green dotted line) except that the background level of the former is slightly lower than the latter. The result presented in Fig.6 demonstrates the potential of new PALS for non-distractive, truly onsite inspection of various materials.

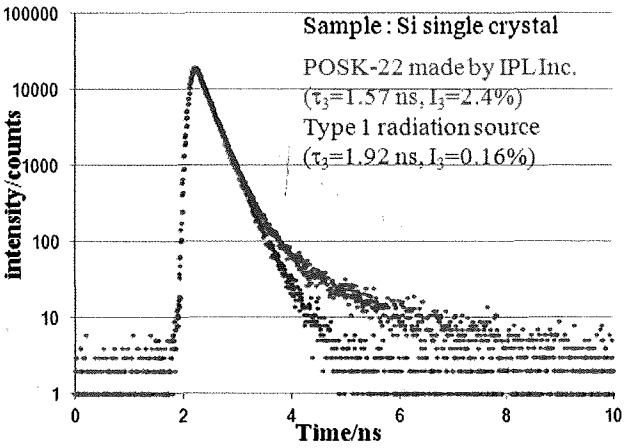


Fig. 5 PAL spectra of single crystal Si recorded with the commercially available sealed source (POSK-22, produced by IPL Inc. on 2006.2.1. The original activity was 740 kBq and the current activity is about 190 kBq) and with the Type 1 source prepared in the present study. Intensity of the long lifetime component is reduced from 2.4 % to 0.16% in the spectrum recorded with the Type 1 source (60 kBq).

Figure 7 shows the comparison of the PAL spectrum of Si single crystal acquired with the new system between the Type 1 and Type 2 sources. The spectra contain two lifetime components, the shorter lifetime component τ_1 being due to positron annihilation in Si and the longer lifetime

component τ_2 being due to positron annihilation in the source. The relative intensity I_2 of the source component in the spectrum recoded with the Type 2 source is reduced to approximately 50% of the spectrum recorded with the Type 1 source. This is obviously due to the reduction of the amount of positron annihilation in Kapton, because the Type 2 source uses only one sheet of Kapton.

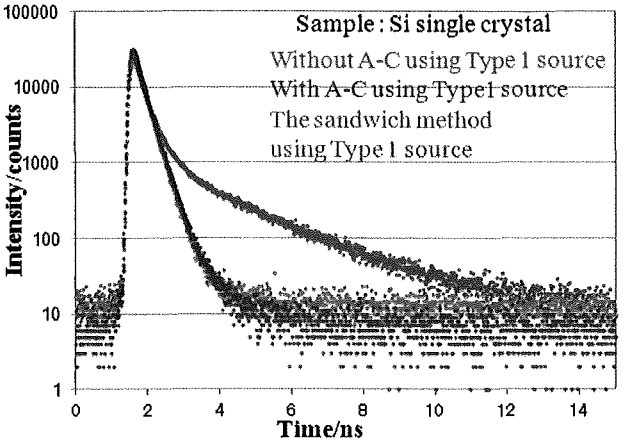


Fig. 6 positron lifetime spectra of Si single crystal acquired with the new PAL system in Fig. 5 and the traditional PAL system without the positron detector. The blue and red dotted lines represent the data obtained using the new system with and without A-C processing, respectively. The green dotted line shows the data obtained with the traditional system using sandwich geometry. The Type 1 source of 850 kBq was used for the experiment.

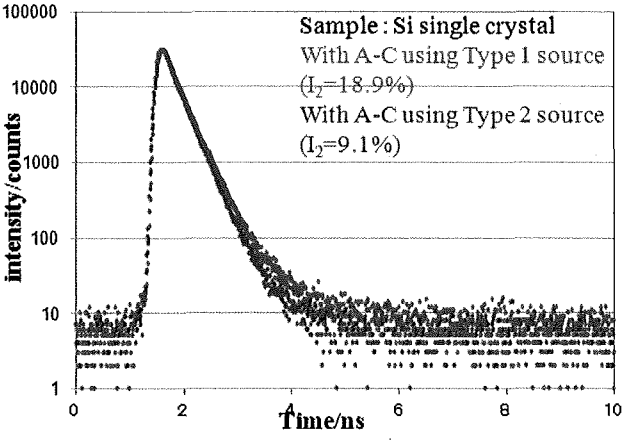


Fig. 7 Experimental results of PAL measurements of Si single crystal for the Type 1 source (850 kBq) and Type 2 source. The long lifetime component originating from Kapton is weakened with the Type 2 source,. I_2 is reduced by about 50%.

Conclusion

^{22}Na sealed radiation sources for PALS, which are possibly free from legal regulations, were prepared and their quality was evaluated. The long lifetime component, likely due to positron annihilation in the adhesive in the commercially available source (POSK-22, IPL Inc.), was substantially suppressed in the PAL data acquired with the Type 1 source. It was confirmed that both the Type 1 and Type 2 sources are of high quality and applicable to our new PALS, which does not require sample cutting and is potentially applicable to truly nondestructive onsite inspection of various materials.

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