



New system for a pulsed slow-positron beam using a radioisotope

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Abstract

To analyze the surface of polymers using positron-annihilation lifetime spectroscopy (PALS), a pulsed slow-positron beam system having both a high pulsing efficiency and a good time resolution is now under development. The time resolution, which is defined by the full width at half maximum (FWHM), and the pulsing efficiency of this system were achieved to be 0.54 ns and 50%, respectively. The lifetime of *ortho*-positronium (*o*-Ps) in low-density polyethylene (LDPE), obtained by PALS using our new pulsing system, agreed with that obtained by a conventional PALS. © 2000 Elsevier Science Ltd. All rights reserved.

1. Introduction

In order to study the depth-dependent characteristics near to the surface of polymers using PALS, a system for pulsing slow positrons has been developed (Oshima et al., 1997a). In our system, positrons randomly emitted from a moderator are injected into a target periodically by adjusting the time of flight between the moderator and the target using a time-varying electric field, which is applied between the moderator and a ground mesh-electrode. This method brings about two advantages: the system can be constructed on a small scale by using a radioisotope as a positron source; this

system can achieve a high pulsing efficiency by using an ideal pulsing bias.

The time resolution of PALS using a previous system was achieved to be ~0.8 ns (FWHM; Hamada et al., 1999), and the lifetimes of *o*-Ps in several polymers were obtained (Hamada et al., 1998, 1999; Oshima et al., 1997a, 1997b, 1999). However, the lifetime spectrum contained a pseudo-lifetime component, which was formed by accelerated positrons in the decreasing part of the pulsing bias.

Since the pseudo-lifetime was close to the lifetime of the *o*-Ps in polymers, it was difficult to evaluate the lifetimes accurately.

In order to eliminate the pseudo-lifetime component and to achieve a better time resolution while maintaining the advantages of the previous method, a new pulsing system has been constructed. In this report, the status of our new system and the preliminary results of

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PALS for aluminum and LDPE using this system will be shown.

2. The new pulsing system

The new pulsing system is shown schematically in Fig. 1.

Slow positrons were produced using ^{22}Na and a tungsten polycrystalline moderator of 6 μm . Since the deviation in the injection time of pulsed positrons at the target was influenced by the energy distribution of reemitted positrons from the moderator (Hamada et al., 1998), an annealing chamber was attached to the pulsed slow-positron beam-line in order to obtain a well-conditioned moderator, which supplied positrons with a narrow energy distribution.

In our new pulsing system, we combined two pulsing methods: a pre-buncher using the time-varying moderator bias method (previous method) and a main buncher using a radio-frequency pulsing method. In other facilities Schödlbauer et al., 1988; Suzuki et al., 1991), the time resolution of the pulsed slow-positron beam system has been achieved to be ~ 0.2 ns (FWHM) by using the radio-frequency pulsing method. Therefore, this new system was expected to maintain a high pulsing efficiency ($\sim 90\%$) by the pre-buncher and to achieve a good time resolution of 0.5 ns (FWHM) by the main buncher.

The pulse-repetition rate of the pre-buncher was 25 MHz (a third of the frequency of the main buncher). In order to focus the injection time of positrons at an accelerating gap of the main buncher located 1.93 m from the moderator, the moderator bias, $V(t) = (m_e L^2 / (2et^2))(t < 0)$ [m_e , e , and L are the

positron mass (kg), charge (C) and flight length (m), respectively], was increased from 167 to 227 V during 36 ns ($-252 < t < -216$ ns), and then decreased to the initial voltage within 4 ns for the next pulsing. The accelerated positrons during the effective time period (36 ns) were expected to be injected into the accelerating gap periodically. In the remaining 4 ns, the positrons formed a pseudo-lifetime component. The pulsing bias was formed by an arbitrary waveform generator (AG5100, Yokogawa) and a 10 W post amplifier (T142-4029A, Thamway).

We used a $\lambda/4$ coaxial resonator as the main buncher, made of stainless steel and copper tube with a quality factor of about 650. The radio-frequency voltage from a stabilized 75 MHz signal generator (RFSG, ESG-1000A, Hewlett Packard) is adjusted to the appropriate voltage by using an attenuator and amplifier, and is then fed to a resonator. The phase difference between arbitrary waveform generator of the pre-buncher and RFSG of the main buncher was controlled by using a common time-base signal with 10 MHz, which was produced by RFSG.

Because the pulse-repetition rate of the main buncher was three times that of pre-buncher, the background positrons, which were accelerated in the decreasing part of the pulsing bias of the pre-buncher, were also bunched by the main buncher, and then two satellite peaks in addition to the main peak were formed. Since the two satellite peaks made it difficult to evaluate the lifetimes accurately, we had to install a chopper system between the pre-buncher and main buncher so as to eliminate any background positrons. Since this new system has combined the two pulsing methods, we need not use a high-power post amplifier for the pre-buncher. Therefore, the chopper system

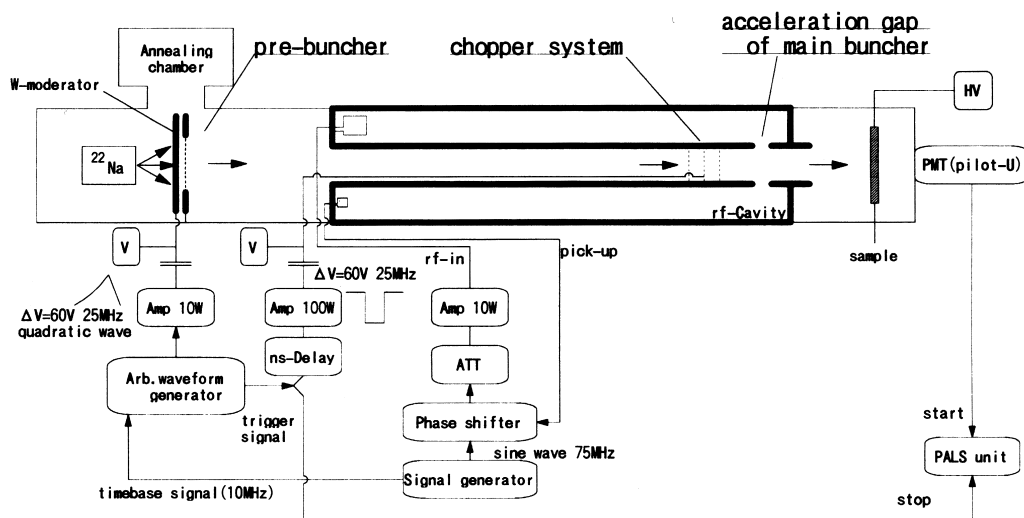


Fig. 1. Schematic of the new pulsing system.

was installed between the pre-buncher and main buncher without any difficulty. The chopper was made of three grids, which were placed at equal intervals of 2 mm. This system could eliminate any unnecessary positrons by a pulsed electric field longitudinal to the beam direction (Suzuki et al.,

1991). In order to avoid velocity modulation by the chopper system, the outside grids were grounded. Furthermore, the chopper was located about 20 mm in front of the acceleration gap of the main buncher. The chopper bias was produced by amplifying an output pulsing trigger signal from arbitrary waveform

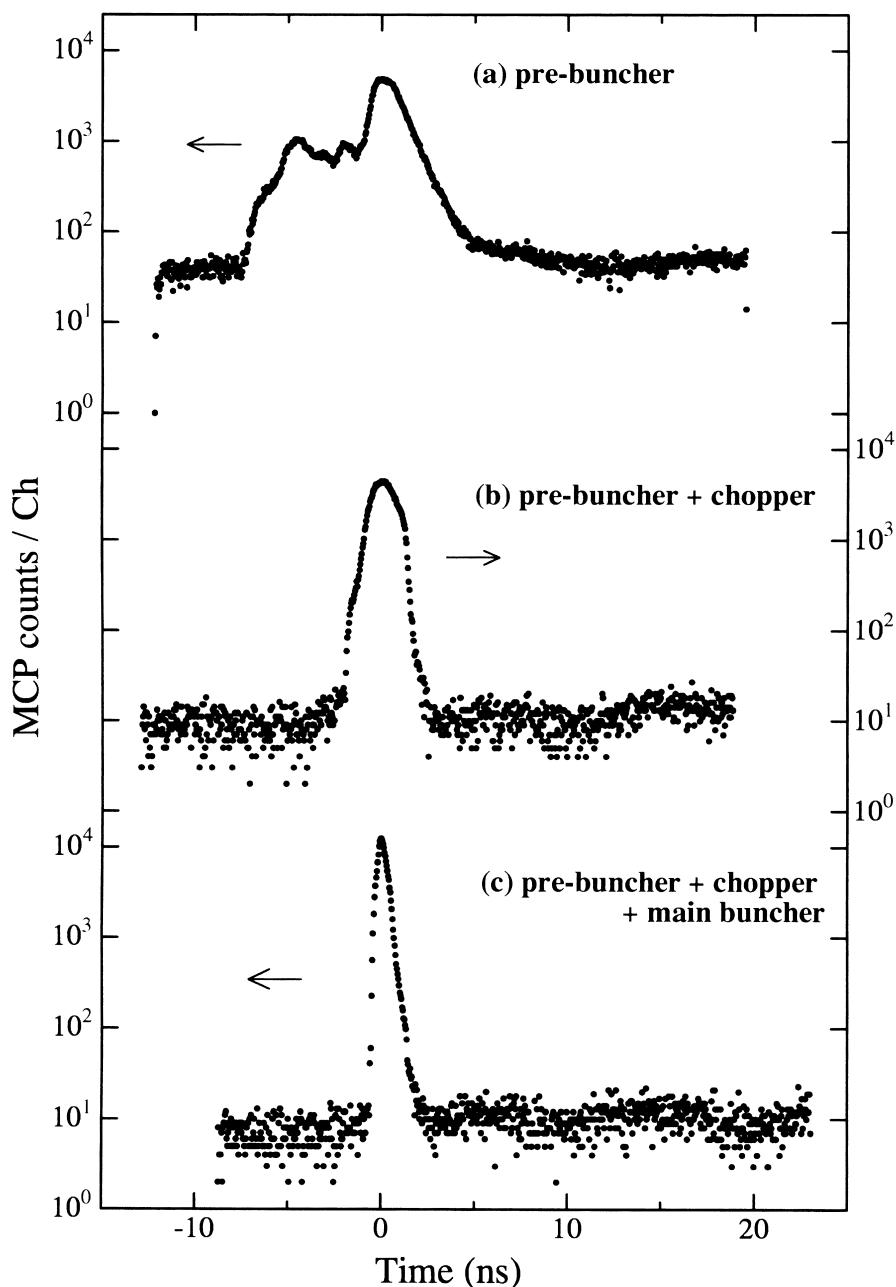


Fig. 2. Injection time profile of pulsed positrons at the target position detected by a micro-channel plate. Each operation condition is as follows: (a) only a pre-buncher, (b) pre-buncher and chopper system, (c) full operation.

generator using a 100 W post amplifier (T142-5059A Thamway). As a result, the pulse-repetition rate and the phase of the chopper system were controlled by arbitrary waveform generator.

In PALS using pulsed slow-positrons, the pulsing trigger-signal was used as a signal of the positron injection-time into a target. The positron lifetime spectra were obtained by a fast-fast coincidence system, determining the time interval between the pulsing trigger-signal and detection of the 0.511 MeV annihilation γ -ray. The γ -rays were detected by a scintillation counter, which comprised a plastic scintillator (Pilot-U) and a photomultiplier (H1949, Hamamatsu).

3. Results and discussions

Fig. 2 shows the time profile of pulsed slow-positrons at the target position using the improved pulsing system. Each spectrum was measured using a micro-channel plate (F4655-12, Hamamatsu) located at the target position.

The time profile of pulsed positrons using only the pre-buncher is as shown in Fig. 2(a). This measured profile contained a main peak and several satellite peaks.

We expected a high pulsing efficiency ($\sim 90\%$) due to the pre-buncher. However, the number of positrons, which formed the main peak with FWHM of 1.7 ns, was 60% of all positrons. The other positrons ($\sim 30\%$), which were accelerated by the non-ideal shape of the pulsing bias, formed several satellite peaks and were widely distributed on the left side of the main peak. The total width of the injection time profile spread about 15 ns.

In the case of our main buncher (frequency 75 MHz; wavelength: 13.33 ns), the effective time width for pulsing the radio-frequency buncher was about 4 ns ($\sim 30\%$ of the wavelength). All positrons, except for the existing positrons in the effective time of 4 ns, had to be eliminated by the chopper system. In order to maintain the pulsing efficiency, we had to operate under the condition with the maximized transmissivity of positrons. After operating under the condition mentioned above, the injection time profile of positrons was as shown in Fig. 2(b), in which the pulsing efficiency decreased to 50%.

By operating all components (the pre-buncher, the chopper system and the main buncher), the injection time profile of pulsed slow-positrons at the target position was as shown in Fig. 2(c). The time width of the pulsed positrons was achieved to be 0.48 ns (FWHM), and the pulsing efficiency was maintained at 50%. This time profile was not symmetrical and already had a decay component with a lifetime of about 0.2 ns. Since this asymmetric profile would affect the lifetime spec-

trum obtained by our new pulsing system, the evaluation of the shorter lifetime than 1 ns was difficult. However, it could be applied to the measurement of the long-lifetime component like the *o*-Ps in the polymer.

In this work, we attempted to test the new pulsing system by comparing with a conventional PALS, which used fast positrons. Fig. 3 shows the measured lifetime spectra of Aluminium and LDPE by operating all components. In order to avoid the effect of a surface state, which complicates the lifetime spectrum, pulsed slow-positrons were accelerated immediately before entering the target by a potential gap of -10.0 kV. Assuming one component of the time-resolution function, the measured spectra were fitted using PATFIT (Kirkegaard et al., 1989). The time resolution was determined to be 0.54 ns (FWHM). The lifetime spectrum of aluminium comprised of a single lifetime component, 0.43 ns. Although the intrinsic positron lifetime in aluminium merged into the resolution function, the pseudo-lifetime component, which had appeared when using the previous system (Hamada et al., 1998, 1999; Oshima et al., 1999), was not observed. This improvement was quite significant to evaluate the lifetime spectra accurately.

The lifetime spectrum of LDPE (density 0.927 g/cm³; crystallinity 0.3) was also analyzed by PATFIT. For very low-density polyethylene, Uedono et al. (1998) have shown by means of Doppler-broadening measurements and PALS using pulsed slow-positrons that the size of the open spaces in the region 0–3 μm was larger than that in the deeper region. Since the mean depth of the positron with an injection energy of about 10 keV was estimated to be about 1.8 μm , the measured lifetime spectrum might reflect information about the bulk and the surface. The lifetime spectrum of LDPE, measured by the conventional PALS, was analyzed with four-lifetime components (Serna et al., 1989). These extracted components were caused by the annihilation of *p*-Ps (0.12 ns), free positrons (0.37 ns), *o*-Ps in the crystal part (~ 1.1 ns, $\sim 3\%$) and the amorphous part (~ 2.6 ns, $\sim 30\%$) of LDPE. Although we tried to analyze the measured spectrum with three or four components, it was very difficult to separate this spectrum. The lifetime spectrum of LDPE was analyzed with the two-lifetime component. The lifetimes and intensities were determined to be (0.4 ns, 72%) and (2.5 ns, 28%). The first lifetime component was considered to contain the lifetime of *p*-Ps, free positrons and *o*-Ps in the crystal part of LDPE. The second lifetime component agreed with the lifetime of *o*-Ps at the amorphous part in LDPE.

These results indicate that our new pulsing system is useful for measuring the lifetime of *o*-Ps in polymers.

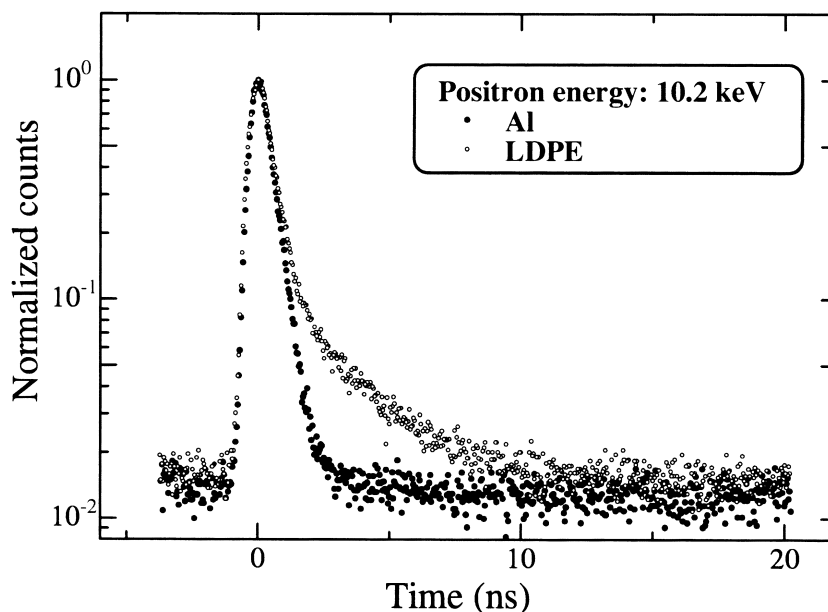


Fig. 3. Positron lifetime spectra of aluminum (Al, ●) and low-density polyethylene (LDPE, ○) measured with a positron incident energy of 10.2 keV.

4. Conclusion

We constructed a new pulsing system, which comprises three components: a pre-buncher using the time-varying moderator bias method; a chopper system; and a main buncher using the radio-frequency pulsing method. The time resolution and pulsing efficiency of this system were achieved to be 0.54 ns (FWHM) and 50%, respectively, and this system was applied to PALS for aluminium and LDPE. From preliminary results, we have shown that our system can measure the lifetime of *o*-Ps in the amorphous part of LDPE.

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