Technical Specification

# ISO/TS 23878

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# Nanotechnologies — Positron annihilation lifetime measurement for nanopore evaluation in materials

*Nanotechnologies – Mesure d'annihilation de la durée de vie de positrons pour l'évaluation de nanopores dans des matériaux*



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# Contents

Page

# Foreword

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This document was prepared by Technical Committee ISO/TC 229, *Nanotechnologies*.

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# Introduction

This document describes a method for measuring and reporting the lifetime of *ortho*-positronium utilizing the positron annihilation lifetime technique. Some of the positrons introduced into insulating materials, like oxides and organic polymers, can form the spin parallel positron-electron bound state *ortho*-positronium, which tends to localize in voids. In its trapped state, *ortho*-positronium annihilates with a lifetime that is less than its intrinsic lifetime of 142 ns in vacuum via a two-gamma annihilation process, where this lifetime component is well correlated with the void dimension. Based on this principle, one can evaluate the average porosity originating from nanometer size voids, such as free volumes in polymers. It is well documented that the positron annihilation lifetime technique is a powerful tool for characterizing the nanopores of various functional materials. Increased demands on the reliable evaluation of nanopores using this technique have emerged for various industrial applications.

This document describes a method for performing positron annihilation lifetime measurements to analyse the lifetime of the *ortho*-positronium ranging from 1 ns to 10 ns (ascribed to a pore size from approximately 0,3 nm to 1,3 nm in diameter), observed for polymeric materials. It also contains measurement procedures, data analysis, and reporting sections. In the annexes, the results of an interlaboratory comparison using two types of reference materials conducted by eight participating institutions, are described, followed by details of measurement systems that are based on the available analogue and digital methods, and a list of parameters and measurement conditions provided as a guide to the user.

# Nanotechnologies — Positron annihilation lifetime measurement for nanopore evaluation in materials

## 1 Scope

This document describes a method for performing positron annihilation lifetime measurements using a $^{22}Na$ positron source that decays with $\beta^+$ emission. The $\beta^+$ (positron) lifetime is determined from a measurement of the lifetime of the *ortho*-positronium which ranges from 1 ns to 10 ns (ascribed to a pore size from approximately 0,3 nm to 1,3 nm in diameter), as observed for polymeric materials in which the positronium atoms mostly annihilate via a two-gamma annihilation process.

This document is not applicable to thin surface layers (that are less than several micrometers).

This document does not apply to measuring:

— non-positronium forming materials;

— positronium-forming materials that induce a spin conversion reaction;

— positronium-forming materials that contain chemicals influencing the annihilation process of *ortho*-positronium by chemical reactions;

— positronium-forming materials that contain mesoporous silica gels with a large contribution from the three-gamma annihilation process.

## 2 Normative references

There are no normative references in this document.

## 3 Terms and definitions

For the purposes of this document, the following terms and definitions apply.

ISO and IEC maintain terminology databases for use in standardization at the following addresses:

— ISO Online browsing platform: available at https://www.iso.org/obp

— IEC Electropedia: available at https://www.electropedia.org

### 3.1 General

**3.1.1**
**positronium**
bound state of a positron and an electron

**3.1.2**
**positron lifetime**
component lifetime corresponding to the annihilation of a large number of positrons or positroniums, and extracted from a measured lifetime spectrum

## 3.2 Experimental set-up

**3.2.1**
**scintillator**
material that luminesces when excited by radiation, wherein the luminescent energy is related to the energy deposited by the injected radiation

**3.2.2**
**photomultiplier tube**
vacuum phototube that converts incident light to an electronic signal, the magnitude of which is based on the energy and number of the incident photons, and subsequently amplifies that signal to provide an electrical output

**3.2.3**
**counting rate**
measured number of events per unit time where an event is the coincident detection of the photons generated during the production and annihilation of positrons

**3.2.4**
**positron source**
emitter of positrons due to nuclear transmutation via $\beta^+$ decay

Note 1 to entry: Sodium-22 ($^{22}$Na) transmuting to $^{22}$Ne is a positron source, for example.

**3.2.5**
**amplifier**
module that increases the amplitude of a signal

**3.2.6**
**gate and delay generator**
module that generates a logic pulse of a desired duration (gate width) and with the desired delay relative to a reference event

# 4 Symbols and abbreviated terms

**ADC** analogue-to-digital converter

**CFDD** constant fraction differential discriminator

**CRM** certified reference material

**DCFD** differential constant fraction discriminator

**GDG** gate and delay generator

**I** relative intensity of lifetime component

**LLD** lower level discriminator

**MCA** multichannel analyzer

**NIM** nuclear instrument modules

**PMT** photomultiplier tube

**Ps** positronium

**RI** radioisotope

**TAC** time-to-amplitude converter

**ULD** upper level discriminator

***o*-Ps** *ortho*-positronium (triplet state)

***p*-Ps** para-positronium (singlet state)

**τ** positron or positronium component lifetime extracted from the measured lifetime spectrum

**$^{22}$Na** sodium with an atomic mass number of 22

**$^{22}$Ne** neon with an atomic mass number of 22

**$^{22}$Ne*** excited state of neon with an atomic mass number of 22

## 5 Principle

A fraction of the total number of positrons injected into an insulating polymer, such as polyolefins, can form positronium (Ps), the bound state of a positron and an electron. Ps forms either in:

— the singlet state, i.e. with antiparallel spins (called *p*-Ps);

— or in the triplet state with parallel spins (called *o*-Ps);

The positrons that do not form Ps will annihilate with the surrounding electrons with a mean lifetime of several hundred ps. The annihilation of *p*-Ps and *o*-Ps in vacuum have intrinsic lifetimes of 125 ps and 142 ns, respectively.

The *o*-Ps that is trapped in a pore will annihilate via the pick-off process with an electron at the pore walls and will have its lifetime shortened from the intrinsic lifetime of 142 ns. The annihilation lifetime of *o*-Ps decreases as the pore volume decreases, accordingly. An *o*-Ps lifetime of between 1 ns and 10 ns is correlated with the size of the pores. Based on this principle, the pore sizes can be estimated from the measured lifetime of *o*-Ps.

In this document, radioactive $^{22}$Na with a $\beta^+$ decay is used for the positron source. The $^{22}$Na nucleus that emits a positron transmutes to $^{22}$Ne* within a short time, which subsequently relaxes by emitting a gamma ray photon with an energy of 1,275 MeV (see Figure 1).

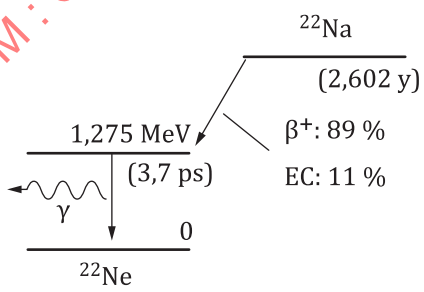



**Figure 1 — Decay process of $^{22}$Na**

The lifetime of a single positron annihilated in a target specimen, placed in close proximity to the $^{22}$Na positron source, is determined by measuring the time difference between the detection of the 1,275 MeV gamma ray photon and the 511 keV annihilation gamma ray photon in the specimen. The 1,275 MeV gamma ray photon is emitted almost simultaneously with the β+ decay. A lifetime histogram (also known as a positron annihilation lifetime spectrum) is obtained by accumulating the time differences over a large number of annihilation events, so that the mean lifetime of *o*-Ps annihilated in the sub-nanometer and nanometer-sized pores can be determined subsequently by data analysis.

The mean lifetime ($\tau = 1/\lambda$, where $\lambda$ is the annihilation rate) and the respective fraction (relative intensity $I$) of each process can be determined by using a model function in the analysis and assuming a proper number of decay functions ascribed to the number of expected positron and Ps annihilation processes.

The number of surviving (unannihilated) positrons in the specimen at a laboratory time frame $t \geq 0$, is given by:

$$N(t) = N_0 \sum_{j=1}^{J} I_j \, exp\left(-\frac{t}{\tau_j}\right) \quad (1)$$

where $N_0$ is the initial number of the positrons, and $J$ is the number of the annihilation components. Thus, the time dependence of the observed annihilation events, that is the positron annihilation lifetime spectrum [$C(t)$], is proportional to the rate of reduction of the positron number at a given time. It can be expressed as:

$$C(t) = \sum_{j=1}^{J} \frac{I_j}{\tau_j} exp\left(-\frac{t}{\tau_j}\right) \quad (2)$$

The $o$-Ps lifetime is obtained as the long-lived component (with a lifetime greater than 1 ns) in the experimental distribution (see Clause 10).

## 6 Overview of the positron annihilation lifetime measurement

A simplified schematic overview of a typical positron annihilation lifetime measurement system is shown in Figure 2.

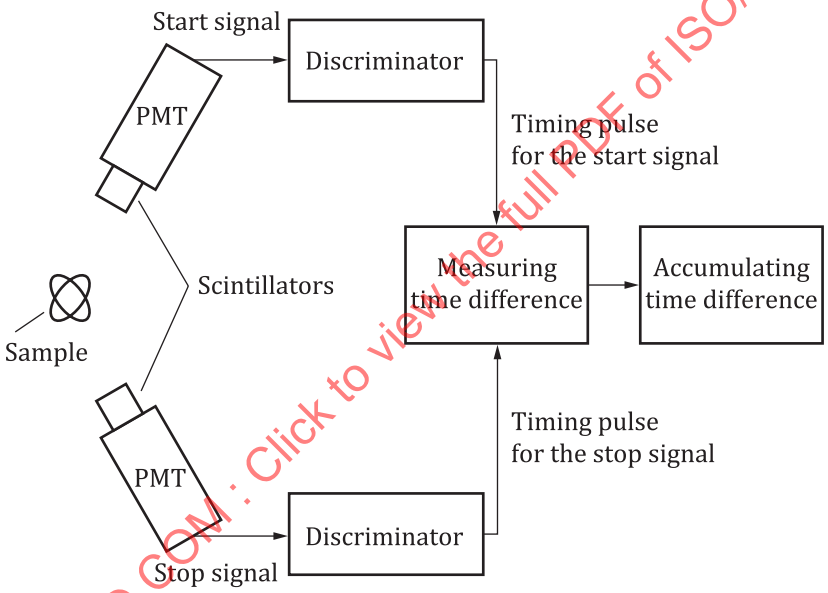



**Figure 2 — Schematic overview of a positron lifetime measurement system**

Start and stop signals, corresponding to the detection of the gamma ray photons from the production and the annihilation of a positron, are generated by a set of gamma-ray photon detectors. Each detector consists of a scintillator and a photomultiplier tube (PMT) that are placed on either side of the sample. Refer to Clause B.1 for guidance on detector placement. The detected signals are analysed to measure the time delay for each positron production and annihilation event.

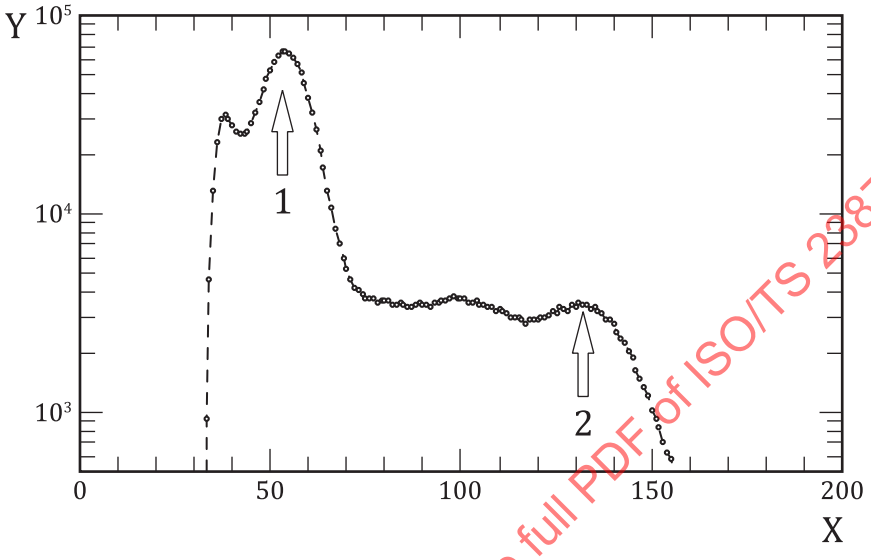
The signals output from the PMT have amplitudes that range between a few mV to V, and are proportional to the energy of each gamma-ray photon (see Figure 3) and the scintillation and detection efficiency of that photon. The start and stop signals shall be selected by discrimination, that is, by processing only those signals with pulse amplitudes greater than the lower level discriminator (LLD) but less than the upper level discriminator (ULD), where the LLD and ULD are set differently for the start and stop signals.

The LLD and ULD assigned for each detector shall be set so that noise signals, as well as mismatch signals, are excluded from acquisition (see Figure 4). The timing pulses, which act as timing surrogates for the gamma-ray photons, can be produced by processing the PMT output signals according to several methods, such as a constant fraction discrimination (see Clause B.2). These timing surrogates are often necessary because:

— the amplitude of the signals from the PMTs is variable;

— stable timing fiducials are needed for subsequent computation of time delays.

The time delay, $\Delta t$, between the timing pulses from the start and stop signals is measured by a time-to-amplitude converter (TAC) module in the analogue method (see Clause B.2) or by direct processing of the digitized waveforms of the detected signals in the digital method (see Clause B.3). Consequently, by accumulating the set of $\Delta t$, one for each positron production and annihilation event, a lifetime histogram of the annihilated positrons is obtained (see Figure 5). The horizontal scale of the measurement system, generally the bin width of the lifetime histogram, shall be calibrated to the unit of second.




**Key**

X channel

Y counts/channel

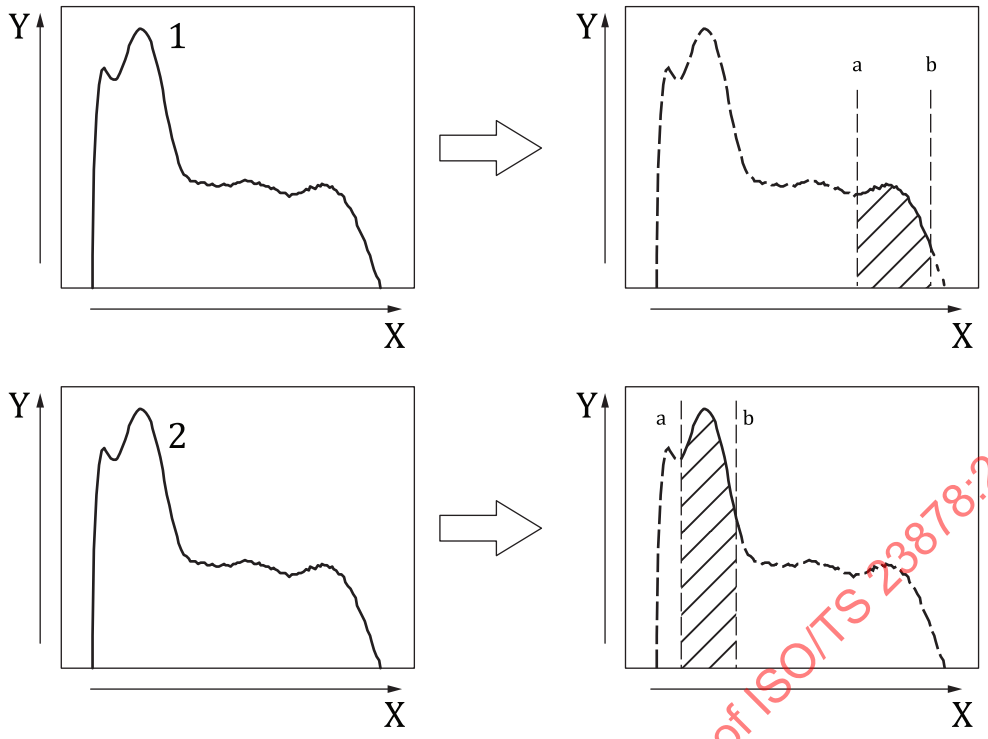
1 peak originating from the detection of 511 keV annihilation gamma ray photon

2 peak originating from the detection of 1,275 MeV gamma ray photon

NOTE 1 Figure 3 shows the typical amplitude distribution of the detected signals from a PMT with $BaF_2$ scintillator.

NOTE 2 The values of the horizontal axis refer directly to the amplitude of the signals from the PMT and indirectly to the energy of those gamma ray photons.

**Figure 3 — Typical amplitude distribution with $BaF_2$ scintillator**

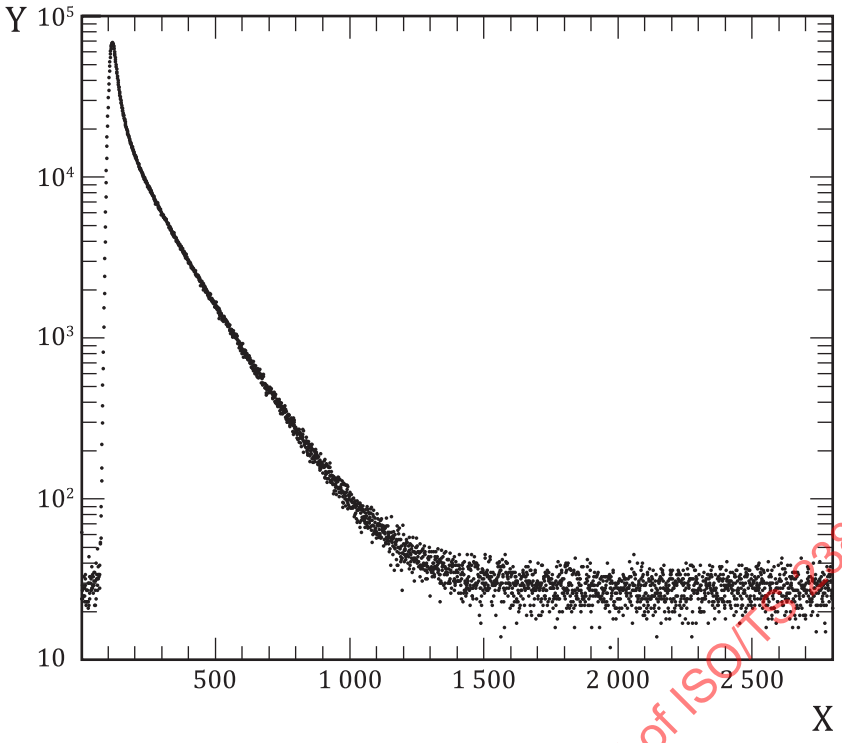



**Key**

X amplitude
Y counts/amplitude
1 for the start signals
2 for the stop signals
a LLD.
b ULD.

NOTE Figure 4 shows the typical setting of the ULD and the LLD for the threshold voltage of the DCFD.

**Figure 4 — Typical setting of the ULD and the LLD**




**Key**

X channel

Y counts/channel

NOTE The values of the horizontal axis correspond to the time-bins.

**Figure 5 — Typical lifetime histogram of positron annihilations**



## 7 Apparatus

### 7.1 Specification of the detector

The scintillation detector comprises a scintillator and a photon detector such as a PMT. A transparent crystal or plastic containing aromatic compounds, which has a fast response as well as a short luminescence decay, is typically used as the scintillating material. The time resolution of the measurement system, conventionally given by the full width at half maximum of the prompt coincidence peak of the lifetime spectrum, is improved by reducing the volume of the scintillator, but with a loss of detection efficiency. Typically, the scintillator is in the form of a cylinder or frustum with a diameter and thickness from 1 cm to 3 cm and may be used in combination with a suitable reflector. Table 1 lists examples of available scintillators.

**Table 1 — Examples of scintillators**

| Material | Max. luminescence wavelength | Rise time | Decay time | Note |
|---|---|---|---|---|
| $BaF_2$ | 220 nm | — | 0,8 ns | — |
| Plastic [EJ-228 and BC-418[a)]] | 391 nm | 0,5 ns | 1,3 ns | Base: Polyvinyl toluene |

[a)] EJ-228 and BC-418 are examples of suitable products available commercially. This information is given for the convenience of users of this document and does not constitute an endorsement by ISO of these products. See References [1] and [2].

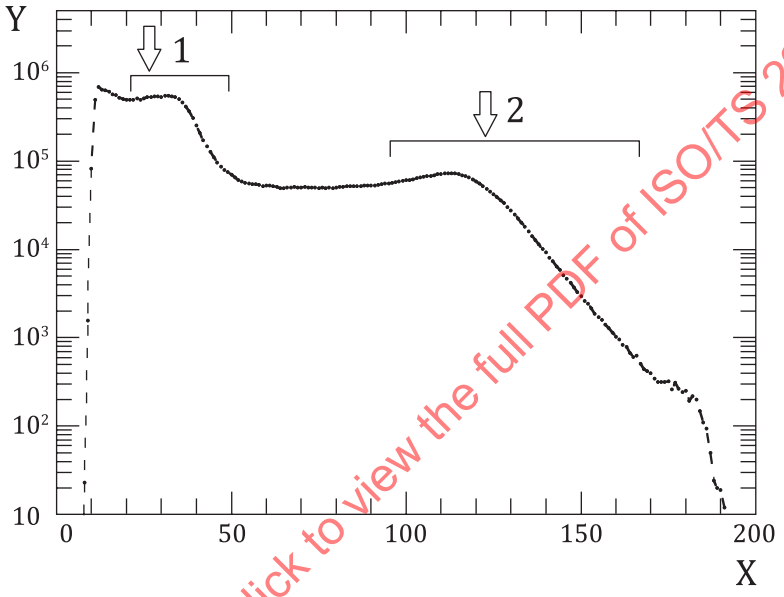
The PMT should have an output gain great enough to attain signals with appropriate amplitudes for processing. The impulse response of the PMT should be of as short a duration as possible (typically, with a transition duration of less than 1 ns). The window material of the PMT shall be selected in order to minimize the absorption of the luminescent light from the scintillator. In the case of $BaF_2$, which emits luminescent light

in the UV range, fused silica is useful as the window material of the PMT. The diameter of the PMT window (typically 25 mm to 50 mm) should be the correct size to enable optical coupling with the scintillator. This coupling can be improved by using an appropriate optical coupling agent, such as silicone oil, grease, etc.

## 7.2 Discrimination of the detected signals

Typical pulse amplitude distributions using $BaF_2$ for the scintillators are shown in Figure 3. The peaks in the signal amplitude distribution shown in Figure 3 corresponding to the 1,275 MeV and 511 keV gamma ray photons exhibit a broad distribution, which is due to the poor time and energy resolution of the detectors. Generally, fluctuations in the amplitude of the detected signals lead to the lower time resolution of the measurement system, as the difference between the LLD and ULD is increased.

When a plastic scintillator is used for the detector, no clear photoelectric peak is observed in the energy distribution as shown in Figure 6. In this case, to determine the respective LLD and ULD for the start and stop signals, energy levels for the Compton edges can be used.




**Key**

X channel

Y counts/channel

1 peak originating from the detection of 511 keV annihilation gamma ray photon

2 peak originating from the detection of 1,275 MeV gamma ray photon

NOTE 1 Figure 6 shows the typical amplitude distribution of the detected signals from a PMT with a plastic scintillator.

NOTE 2 The values of the horizontal axis refer directly to the amplitude of the signals from the PMT and indirectly to the energy of those gamma ray photons.

**Figure 6 — Typical amplitude distribution with plastic scintillator**

## 7.3 Measurement conditions

The time intervals (width per channel) in the lifetime histogram should be set in the range from 10 ps to 50 ps. The epoch (duration of observation period) for the histogram shall be adequately long so that the background levels of the histogram are included. For instance, the epoch should be set to longer than 50 ns for typical polymers such as polycarbonate. More than $10^6$ coincidence events should be accumulated for each measurement in order to obtain sufficient counting statistics and to avoid a count of zero in any time interval. For that portion of the histogram corresponding to the background signal, it is preferable that the average number of counts at each time interval is greater than 15 counts. For currently available analogue

systems, the counting rate should be no more than 200 counts per second. For digital systems, the count rate should not be so great as to introduce detector pile-up errors exceeding about 0,001 % of the total number of production and annihilation events or to prevent the acquisition of one waveform per production or annihilation event.

## 8 Preparation of the positron source

An appropriate amount of $^{22}$NaCl aqueous solution (approximately 3,7 MBq/100 µL) is placed using a dropper on a several-µm thick foil of corrosion-proof metal (such as titanium) or polymer, followed by drying and sealing with a foil of the same material. Polyimide films are useful for this purpose because some of them do not form Ps when exposed to positrons. It is preferable to keep the spot size of the droplet as small as possible, so as to act as a point source, but also as thin as possible, to reduce annihilations within the source.

The foils are adhered together using a heat-sealing mechanism or by using a commercial adhesive to prevent the migration of the $^{22}$NaCl salt from the foils or films. If adhesive is used for sealing, the adhesive shall not make contact with the $^{22}$NaCl source. When a source with a long-lived component is used, an appropriate correction for this component shall be taken into account in the data analysis.

Generally, a positron source with an activity in the range from 0,1 MBq to 2,0 MBq is used. Increased positron emission rates can be achieved by using a source of higher activity, however, this also leads to a higher fraction of background signals due to random coincidence of annihilating positrons.

## 9 Preparation of the measurement specimen

To avoid contamination of the specimen with a broken radioisotope (RI) source, the specimen surface in contact with the RI source shall be kept as smooth as possible. The specimen and the RI source shall be in close contact with each other, so that as many positrons as possible, emitted from the RI source, annihilate within the specimen.

The specimen shall be of a sufficient size so that as many positrons as possible, emitted from the RI source in the direction of 4π steradians, annihilate in the specimen. The depth range $L$ [kg/m$^2$] of the injected positrons with an incident energy $E$ [MeV] from 0,15 MeV to 0,8 MeV is estimated based on:

$$L = 4{,}07E^{1{,}38} \tag{3}$$

Here, the range of $L$ is approximately 0,3 kg/m$^2$ to 3,0 kg/m$^2$ for $E$ in the corresponding range of about 0,15 MeV < $E$ < 0,8 MeV. Using this formula, the maximum penetration depth $Z$ (mm) in a material with density $\rho$ (kg/cm$^3$) for positrons from $^{22}$Na with a maximum energy of 0,546 MeV is calculated as:

$$Z = \frac{1{,}77 \times 10^3}{\rho} \tag{4}$$

For instance, the specimen thickness for a polymer sheet with a density of 1,0 × 10$^3$ (kg/m$^3$) should be more than approximately 1,8 mm. Therefore, the method is not suitable for thin polymer films. Further information on the measurement of positron lifetimes in polymer materials can be found in, for example, Reference [12].

## 10 Data analysis

The positron and Ps lifetime parameters, i.e. $\tau$ and $I$ of Formula (1), are determined by applying the model function described in Clause 10 using the recorded positron annihilation lifetime data by the nonlinear least-squares method. For some materials, a lifetime spectrum is obtained which can be best fitted with three lifetime components, corresponding to:

— a $p$-Ps component;

— free positrons;

— an *o*-Ps component.

However, for other materials, a better fit may be obtained with two *o*-Ps components with a lifetime greater than 1 ns, in which case both components should be reported. The data recorded by the measurement system, $S(t)$, is obtained by the convolution of $C(t)$ as in Formula (2) with the time resolution function of the system $R(t)$:

$$S(t) = R(t) * C(t) \tag{5}$$

where "∗" denotes a convolution operation. $R(t)$ may be approximated by a linear combination of Gaussian functions with one or more components ($n \geq 1$) based on the experimental set-up:

$$R(t) = \sum_{j=1}^{n} w_j \frac{1}{\sqrt{2\pi\sigma_j^2}} exp\left( -\frac{(t-\mu_j)^2}{2\sigma_j^2} \right), \tag{6}$$

where $\mu_j$ and $\sigma_j$ represent the peak position and the standard deviation for the $j$-th component, respectively, and:

$$\sum_{j=1}^{n} w_j = 1 \tag{7}$$

The following function is then obtained from Formula (5) and Formula (6).

$$S(t) = A\int_{-\infty}^{\infty} R(t')\,C(t-t')\,\Theta(t-t')\,dt' + B\ , \tag{8}$$

where $t'$ is the delay used for the fitting. Here $A$, $B$, and $\Theta(t)$ respectively represent the normalized factor, the background, and the Heaviside step function, which is zero at $t < 0$ and unity at $t \geq 0$.

In the analysis, the fitting range shall be determined using the following procedure:

Typically, the fitting range starts from a few channels before the peak (for instance, with counts less than 5 % of the peak counts) to a channel in the background range. It ends in a channel consisting of mainly background annihilations occurring after the peak maximum. The start region for the fit is typically chosen based on prior knowledge of the instrument response function as appropriately determined by the user on a material with known lifetimes. Including a large portion of the instrument response function in the fitting process is ideal but is not always feasible. The stop region for the fit should contain some background region to ensure that the longer lifetimes are fitted correctly. Variations in longer lifetimes can occur if the background channels are not chosen carefully. This can be easily verified by including and excluding background channels during fitting, as well as verifying that the lifetimes are robust. The time-zero ($t_0$) for the specimen is generally determined as part of the fitting process. In all cases, the parameters utilized for analyses shall be specified. The source component should be taken into account in the data fitting, so that more reliable results are attained. Several computing programs are available for the data fitting.

# 11 Reporting

## 11.1 Specific values

The lifetime values of the *o*-Ps component obtained from the fitting procedure are used to determine pore size (see 11.2).

### 11.2 Pore dimension

Pore sizes can be converted from the obtained value of the *o*-Ps lifetime component based on semi-empirical quantum mechanical models. One commonly used model is the Tao-Eldrup model (see References [5] and [6]), where the conversion formula is:

$$\tau_{\mathrm{o-Ps}} = 0{,}5 \times \left[ 1 - \frac{R}{R+0{,}166} + \frac{1}{2\pi} \sin\left( \frac{2\pi R}{R+0{,}166} \right) \right]^{-1} \tag{9}$$

where $\tau_{\mathrm{o\text{-}Ps}}$ is the *o*-Ps lifetime (ns) and $R$ is the pore radius (nm). Using this formula, for example, a 5 ns measured *o*-Ps lifetime corresponds to a pore radius of approximately 0,47 nm.

While the Tao-Eldrup model provides a straightforward approach for estimating pore sizes, researchers should be aware of its limitations, such as assuming homogeneity and a single pore size for the material. Depending on the complexity and specific characteristics of the material, advanced modelling techniques and consideration of other relevant alternative models (such as the extended Tao-Eldrup model) can possibly be necessary for a more comprehensive analysis of the nanopore properties. Several alternative models, widely used in nanotechnology and nanomaterials research, offer valuable insights into different aspects of nanopore behaviour.[13] However, for simplicity, in this protocol, only the standard Tao-Eldrup model described in 11.2 is considered for the determination of pore size from the *o*-Ps component lifetime.

## 12 Reference materials

SI traceable certified reference materials (CRMs), supplied from material producers and national metrology institutes (such as the National Metrology Institute of Japan) are useful in order to ensure the validity of the procedure of the data recording and analysis. See References [7] and [8].

# Annex A
(informative)

# Interlaboratory comparison

To validate this specification, an interlaboratory comparison using two types of reference materials was performed.

Specimens of 1,5-mm-thick fused silica, supplied by the National Institute of Advanced Industrial Science and Technology (AIST) as a certified reference material (NMIJ CRM 5601a[1)]), and commercially available 2,0-mm-thick poly methyl methacrylate (PMMA) were used for this comparison study. Prior to the measurements at each laboratory, the longest-lived lifetime component ($\tau_{o\text{-}Ps}$) for the PMMA specimens were examined at AIST. Repeated measurements and data analysis for PMMA revealed good measurement repeatability of the lifetime value within a relative standard uncertainty of 0,5 %. These unspecified PMMA specimens were provided as a "blind polymer" to the participants with the specification revealed after all the results were disclosed. Separate specimens of each material were distributed to all the laboratories within a specific interval. Measurements at the majority of the participating institutions were carried out from October 22 2019 to February 10 2020, without access to the data of other laboratories.

Figure A.1 shows $\tau_{o\text{-}Ps}$ for:

a) fused silica;

b) PMMA reported from the participant laboratories.

The data are arranged in increasing order of the lifetime reported by the various institutions. The mean values of $\tau_{o\text{-}Ps}$ including the standard uncertainties are:

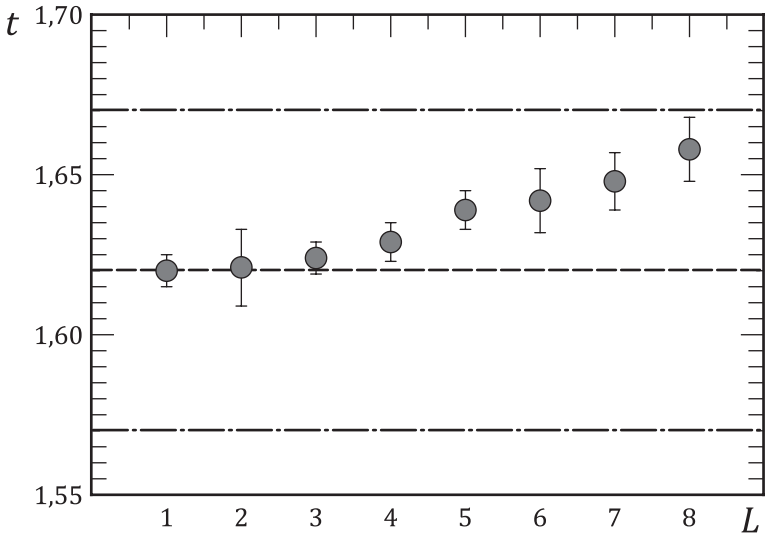
— (1,635 ± 0,014) ns for fused silica;

— (1,923 ± 0,019) ns for PMMA.

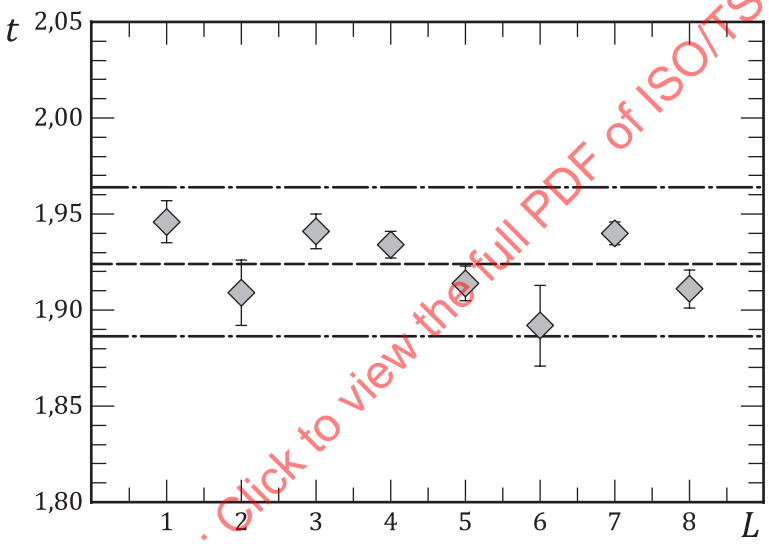
These mean values agree well with the respective reference values, that is, the certified value for fused silica of (1,62 ± 0,05) ns and the mean value for $\tau_{o\text{-}Ps}$ of (1,941 ± 0,011) ns for a number of PMMA specimens measured at AIST.



1) NMIJ CRM 5601a is an example of a suitable products available commercially. This information is given for the convenience of users of this document and does not constitute an endorsement by ISO of these products. See Reference [7].




**a) Fused silica (NMIJ CRM 5601)**




**b) Poly(methyl methacrylate) (PMMA)**



**Key**

*t* lifetime (ns)

*L* laboratory number

NOTE 1 The number ordering of the laboratories was assigned based on the value reported for fused silica (NMIJ CRM 5601).

NOTE 2 The dashed lines represent the certified value for NMIJ CRM 5601 and the average measured value for PMMA respectively. The dashed-dotted lines represent the certified uncertainty for NMIJ CRM 5601 and the combined standard uncertainty (due to the repeatability and the fitting error of the analysis) for PMMA.

NOTE 3 The error bars for each laboratory represent the reported standard deviation for replicate measurements.

**Figure A.1 — $\tau_{o\text{-}Ps}$ for fused silica and PMMA**

The relative standard uncertainties determined from the reported values for the repeated measurements at individual laboratories were 0,85 % for fused silica and 1,0 % for PMMA. These results were considerably smaller than those obtained for fused silica (1,6 %) and organic polymers (3,1 % to 4,2 %) reported in Reference [9] and Reference [10], respectively. The improved lifetime uncertainties validate the applicability

of the current measurement and analysis protocol in evaluating positron annihilation lifetime based on *ortho*-positronium lifetime observed in nanoporous materials.

# Annex B
(informative)

# Configuration of the apparatus

## B.1 Alignment of the detectors

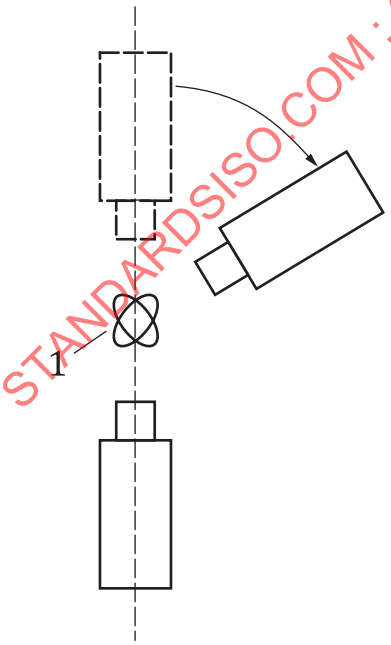
While the detectors need to be placed as close as possible to each other, in order to improve the overall counting rate of the measurements, the following issues should be avoided:

a) Annihilation gamma ray photon detection from the same event on both PMTs.

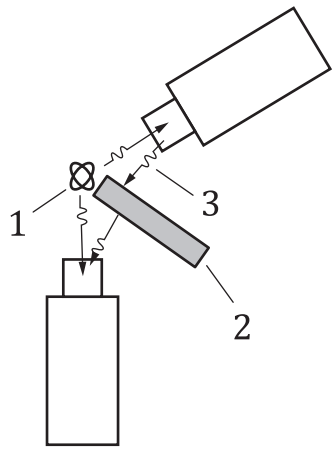
Two annihilation gamma ray photons are emitted in opposite directions (i.e. at 180°) when the positron annihilation occurs via the pick-off reaction with an electron in the material. If both scintillators are located along this line through the positron source, both of the emitted annihilation gamma ray photons can be detected by each detector at the same time, possibly giving rise to a false signal during the measurements. The probability of detecting annihilation gammas in both detectors depends on the scintillator material and is more likely in $BaF_2$ than in plastic scintillators. However, this effect can be suppressed by moving one scintillator away from the above line so that the angle between the detectors is less than 180° [see Figure B.1 a)].

b) False signals due to Compton scattering in the scintillators.

Some of the gamma ray photons incident on the scintillator can be scattered. These gamma ray photons can cause false signals with very short lifetimes, which eventually not only causes a reduction of the relative intensity of the longest-lived Ps component, but also distorts the short-lived positron components (see Reference [3]). Placing a shield of high-Z material, such as lead between the scintillators is useful to suppress this effect (see Figure B.1 b) and Reference [4]).




**a) Suppression of detection**




**b) Suppression of false signals**

**Key**

1 source/specimen assembly

2 gamma ray shield

3 scattered gamma ray

NOTE 1 Figure B.1 a) shows suppression of detection of the same annihilation event in both detectors by moving one detector so that the angle between the detectors is less than 180°.

NOTE 2 Figure B.1 b) shows suppression of false signals due to Compton scattering in the detectors by placement of a suitable shield material, such as lead.

**Figure B.1 — Detector configurations for positron lifetime measurements**

## B.2 Measurement system: Analogue method

The analogue method consists of:

a) Set up: Typical set up based on the analogue method for positron lifetime measurements is shown in Figure B.2.

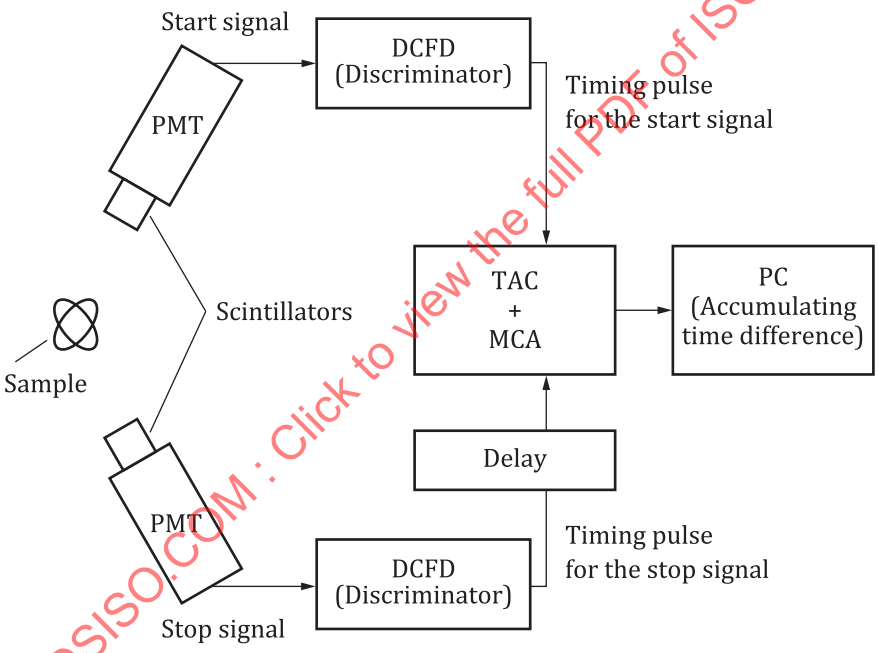



**Figure B.2 — Schematic of positron lifetime measurement system with analogue method**

A set of DCFD, with proper LLD and ULD settings for each DCFD, is employed so that only gamma ray photons associated with the emission of a positron and the corresponding annihilation event are selected, respectively. The DCFD selects the start and stop signals, thereby only allowing signals where the pulse heights are greater than LLD and less than ULD. The DCFDs generate fast logic pulses from the selected input signals according to the constant fraction method.

The time difference between the output pulses arising from the start and stop signals is determined using TAC and multichannel analyser (MCA) modules. The TAC module produces a rectangular output pulse whose amplitude is proportional to the time difference between two input pulses, namely, the start and stop signals. The TAC output pulses are properly stretched in time, then input into the analogue-to-digital converter (ADC) of the MCA. The annihilation events are stored in the channel to obtain a lifetime histogram on a computer. In order to obtain a proper pulse amplitude for operating the MCA, a delay module is placed between the DCFD and TAC modules for the stop signal.