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Application of XAD4 water solutions for positronium imaging phantom

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Abstract

Positron Emission Tomography (PET) is a non-invasive method of medical examination, that allows for early diagnosis of diseases (such as cancer) and therapeutic process monitoring. At the heart of this method is registration of gamma quanta sourcing from the direct annihilation of positron originating from β^+ decay and electron from surrounding matter.

Positrons from β^+ decay can also bond with electrons and create a meta-stable atom called positronium. Positronium exists for a limited period of time dependent on medium properties. Measurement of lifetime of positronium is the subject matter of Positronium Annihilation Lifetime Spectroscopy (PALS).

Positronium Annihilation Lifetime Spectroscopy (PALS) is a research method that uses dependency of the lifetime of positronium on free volume sizes for examining properties and structure of the matter. New PET imaging method developed at Jagiellonian University enables measurement and imaging of the lifetime of ortho-positronium. Research conducted at Jagiellonian University shows that the ortho-positronium lifetime measurement can be applied for medical diagnostics purposes to improve specificity of PET imaging. To assess the accuracy of this new method it is essential to develop a phantom that will allow for measurement of the lifetime of ortho-positronium alongside activity concentration.

Presented in this thesis is a method for assessing mean lifetime of ortho-positronium dependency on concentration of XAD4 and water mixture using PALS technique. Within this dissertation hypothesis was tested, that adding water to XAD4 polymer will result in gradual filling of pores, with increase of water concentration in a mixture and with electron free volume decrease, mean lifetime of ortho-positronium is expected to shorten accordingly. A total of 6 samples were measured: dry XAD4, distilled water and 4 mixtures of XAD4 and water in different concentrations. Acquired lifetime spectra were analysed using PALS Avalanche program. The results of the measurements show dependency of mean lifetime of ortho-positronium on XAD4 mass percentage in water.

The presented method of controlling mean lifetime of ortho-positronium in XAD4 pores will serve in the future to prepare a phantom for assessment of precision of the positronium imaging method developed at Jagiellonian University.

Streszczenie

Pozytonowa Tomografia Emisyjna (PET) jest nieinwazyjną techniką badań medycznych, która pozwala na wczesne wykrywanie chorób (np. nowotworowych) oraz monitorowanie przebiegu procesu leczenia. Opiera się ona na rejestracji kwantów gamma pochodzących z anihilacji bezpośredniej pozytonu powstałego w wyniku rozpadu β^+ radioizotopu z elektronem z otaczającej materii. Pozytony, pochodzące z rozpadu β^+ , mogą także wiązać się z elektronami tworząc meta-stabilny stan związany nazywany pozytonium. Pozytonium istnieje w materii ograniczony czas, zależny od właściwości danego ośrodka. Pomiar tego czasu może być wykorzystany do badania struktury za pomocą techniki Spektroskopii Czasu Życia Pozytonium (PALS).

Spektroskopia Czasu Życia Pozytonium (ang. Positronium Annihilation Lifetime Spectroscopy PALS) jest metodą badawczą wykorzystującą zależność czasu życia pozytonium od wielkości wolnych przestrzeni do badań właściwości i budowy materii. Nowa metoda obrazowania PET stworzona na Uniwersytecie Jagiellońskim pozwala na pomiar czasu życia pozytonium. Badania przeprowadzone na Uniwersytecie Jagiellońskim wskazują na to, że pomiar czasu życia pozytonium można wykorzystać w celach diagnostyki medycznej do polepszenia swoistości obrazowania PET. Aby ocenić precyzję nowej metody obrazowania należy skonstruować fantom, który pozwoli na pomiar zarówno czasu życia orto-pozytonium jak i koncentracji aktywności.

W tej pracy przedstawiona została metoda określenia zależności średniego czasu życia orto-pozytonium w porach polimeru XAD4 od stężenia mieszaniny XAD4 i wody za pomocą techniki (PALS). W ramach pracy sprawdzano hipotezę, że dodanie wody do polimeru XAD4 będzie skutkowało stopniowym wypełnianiem porów wraz ze wzrostem zawartości wody w mieszaninie, a w wyniku tego stopniowym zmniejszaniem się przestrzeni wolnych od elektronów i skróceniem średniego czasu życia orto-pozytonium. Dokonano pomiaru 6 próbek: wysuszonego XAD4, wody destylowanej oraz 4 mieszanin XAD4 i wody w różnych koncentracjach. Uzyskane widma czasów życia pozytonium analizowano za pomocą programu PALS Avalanche. Wyniki pomiarów wskazują na istnienie zależności między czasem życia orto-pozytonium a stężeniem procentowym XAD4 w wodzie.

Opracowana metoda określenia zależności średniego czasu życia orto-pozytonium w mieszaninie XAD4 i wody posłuży w przyszłości do przygotowania fantomu do badania precyzji nowej metody obrazowania opracowanej na Uniwersytecie Jagiellońskim.

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Acronyms

PET Positron Emission Tomography. 4, 5

PALS Positron Annihilation Lifetime Spectroscopy. 4, 5

PET Positron Emission Tomography. 8

PALS Positron Annihilation Lifetime Spectroscopy. 8, 9, 28, 29

CFD Constant Fraction Discriminator. 16

PTFE Polytetrafluoroethylene. 16, 22, 28, 32

1. Introduction

The goal of this thesis is to devise a phantom for quantitative assessment of precision of the Positron Emission Tomography (PET) scanner in terms of positronium imaging.

Positronium imaging was invented and developed at Jagiellonian University [1]. It utilizes positronium bound states lifetime information to provide additional insight of internal structure of studied objects and is promising as tool for raising specificity of medical diagnostics. This method can be applied in PET diagnostics, but unlike a standard PET procedure, it involves registration of more than two annihilation gamma quanta. To perform that, it is necessary to adjust a scanner for multiphoton detection and image reconstruction. Therefore it was not considered as feasible until recently. The first scanner with that ability, J-PET, was constructed at Jagiellonian University [1, 2]. J-PET detector is build from plastic scintillators in contrast to standard PET scintillators, which are build from crystals. Moreover, scintillators are placed along the patient's body and signals are collected by the photomultipliers at both ends of the detector [3]. Due to the fact that plastic scintillators have long attenuation, the light produced in the interaction point is transferred to both ends in similar fashion as in a lightguide. Additionally plastic scintillators have better timing resolution than crystals, but due to their lower density suffer from worse gamma quanta detection efficiency [2].

The feasibility of positronium imaging was first studied using simulations [4]. The main interest of this study was concentrated on imaging the positronium decay into three gamma quanta. It was shown that it is possible to reconstruct the density distribution of annihilation points of positronium and to reconstruct an image of mean lifetime of ortho-positronium for each voxel using the J-PET detector. It was also proven experimentally that it is possible to perform reconstruction of three-photon positronium annihilations to reproduce an image of a cylindrical positronium production chamber [5].

In PET, inside the patient's body, about 40% of positron annihilation occurs through the production of positronium and most of the ortho-positronia annihilate into two photons by the pick-off process, in which a positron in a positronium encounters an electron from surrounding matter and they annihilate predominantly with production of two gamma quanta. Probability of pick-off is dependent on tissue structure and metabolism and could provide information about disease progression [6]. Feasibility of employing this phenomena to determine positronium lifetime was evaluated [7]. It was proven that one can extract the mean ortho-positronium lifetime image by detection 2+1 photons: two corresponding to positronium annihilation into two gamma quanta and one prompt deexcitation gamma quantum. It was presented that reconstruction of the mean ortho-positronium lifetime image based on annihilation into two photons is beneficial in relation to imaging using three photons and may be more useful as a mean of diagnosis.

Positron Annihilation Lifetime Spectroscopy (PALS) is a technique that also applies the phenomenon of positron and electron annihilation. It is based on dependency of sizes of electron free volumes on lifetime of ortho-positronium [8]. Studies performed using PALS technique have shown differences between lifetime of ortho-positronium in healthy tissues and cancerous tissues [9]. Later it was proven that the data obtained with J-PET detector in positronium imaging technique in first ex vivo study using phantom with cardiac myxoma and healthy adipose tissues allows for discerning between cancerous and healthy tissues on the grounds of measuring lifetime of ortho-positronium for specific voxel of activity [10, 11].

Although this type of imaging requires 3 gamma quanta, two annihilation from pick-off and one prompt, total-body PET performance makes it adequate for hospital use [12]. But before applying the method it is essential to assess the method precision for which a phantom is needed. In this study I present the proposition of first such phantom and devise the method of quantitative study of the results.

First step of this study was development of a replicable method for measuring lifetime of ortho-positronium in mixtures of water and XAD4 using Positron Annihilation Lifetime Spectroscopy (PALS) technique. This was done to estimate a dependency of lifetime of ortho-positronium on concentration of XAD4 in water. In this dissertation hypothesis is formulated that addition of the water to XAD4 will result in partial filling up of larger crevices with water, and consequently decrease of size of electron-free volumes and mean lifetime of ortho-positronium. The results of this measurements will be later used for preparation of NEMA-like [13] phantom for quantitative assessment of precision of the new imaging method developed by J-PET collaboration. The proposition of such phantom is presented in Figure 1.1.

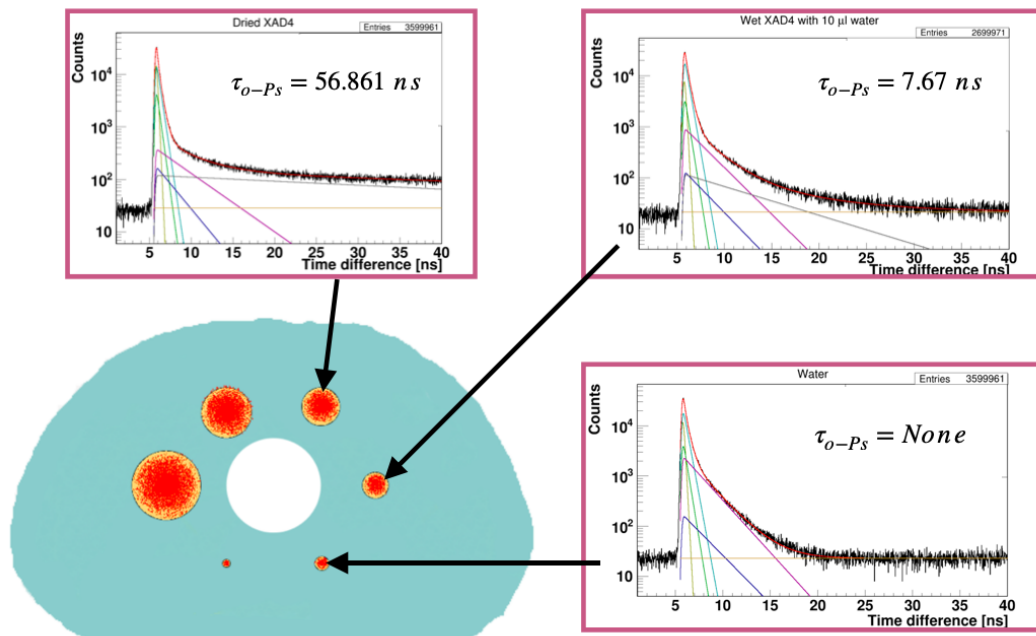


Figure 1.1: The proposition of NEMA-like [13] phantom for assessment of the precision of J-PET positronium imaging method. For each of the volumes there is same activity concentration (depicted as red areas) but different mean lifetimes of ortho-positronium (arrows pointing to different lifetime spectra). τ_{o-Ps} in the upper right corner of the spectra graphs is exemplary lifetime of ortho-positronium for second (longer) component found for XAD4 pores.

In chapter 2 the positronium system is discussed. Presented in this chapter are description of the source of positrons that was used in a course of this experiment, a short explanation of what is positronium, a brief description of Tao-Eldrup model, which is used to explain dependency of ortho-positronium on size of regions of lower electron density in the matter, and a characterization of the material used in this study. This chapter is based on literature studies.

In chapter 3 experimental methods used to gather data containing information on the ortho-positronium lifetime are shown. Firstly measurement setup is described, then the method of calculating proportions of XAD4 material and water while preparing a sample is presented. This chapter also contains measurement protocol, description of conducted measurements, method of analysing acquired signal that leads to producing a lifetime spectra of positronium, data analysis of acquired lifetime spectra. Additionally the description of benchmark measurements and their results are presented at the end of this chapter.

In chapter 4 results along with conclusions are shown. Acquired results indicate a dependency of the mean lifetime of ortho-positronium on mass percentage of XAD4 in water for both components that were corresponding to ortho-positronium lifetime in XAD4 pores. For small values of mass percentage of XAD4 in water, mean lifetime of ortho-positronium is shorter than for bigger values of mass percentage, which means that with filling up of crevices with water the mean lifetime shortens, which confirms the hypothesis. Intensity of positronium production for shorter-living component decreases with increase of mass percentage and intensity of positronium production for longer-living component increases with mass percentage.

In chapter 5 summary and perspectives of this study are presented.

Additionally there is chapter 6 containing lifetime spectra with fitted functions and tables with the results of the fits for all of the measurements.

2. Positronium system

2.1 Source of positrons

The source of positrons described in this study is ^{22}Na isotope. It has a half-life of 2.6 years. In most likely case it decays in β^+ decay, in which a positron and an electron neutrino are emitted and sodium atom transforms into neon in excited state. In the process of neon deexcitation gamma quantum with energy of 1274 keV is emitted [14]. The scheme of ^{22}Na decay is shown in Figure 2.1.

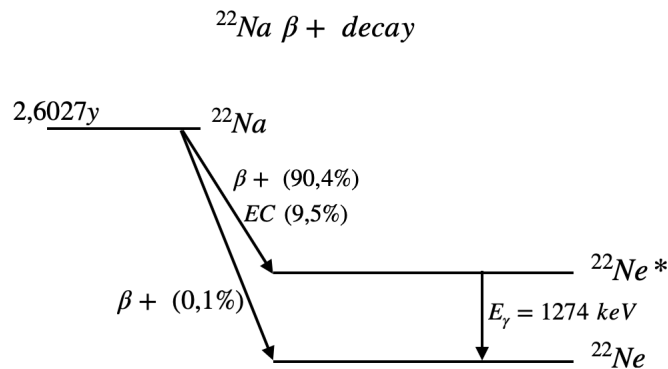


Figure 2.1: Decay scheme of ^{22}Na . In most likely case this isotope undergoes β^+ decays with emission of positron and becomes ^{22}Ne in excited state. Deexciting neon emits gamma quantum.

In Figure 2.2 the sequence of events in measuring lifetime of positronium is depicted. After emission of positron, due to ^{22}Na decay, positron undergoes thermalization in the sample. At the same time neon deexcites and deexcitacional gamma quantum (with energy of 1274 keV) is registered by measurement setup. This is a START signal for lifetime measurement. At similar time positron creates positronium with electron from surrounding matter. Those processes happen in order of picoseconds. After that ortho-positronium exists for a short period of time, in the order of nanoseconds, until it encounters an electron and annihilates in a pick-off process with emission of two gamma quanta with energy 511 keV. Registering annihilation gamma quantum is a STOP signal for measuring lifetime of positronium [15]. The source used in PALS is in form of drop of NaCl sealed between a kapton foil. Additionally it was sealed in parafilm to prevent water from the sample to cause leaking. The source with foil was attached to a ring. Schematic depiction of source and a picture of a ring with source is presented in Figure 2.3.

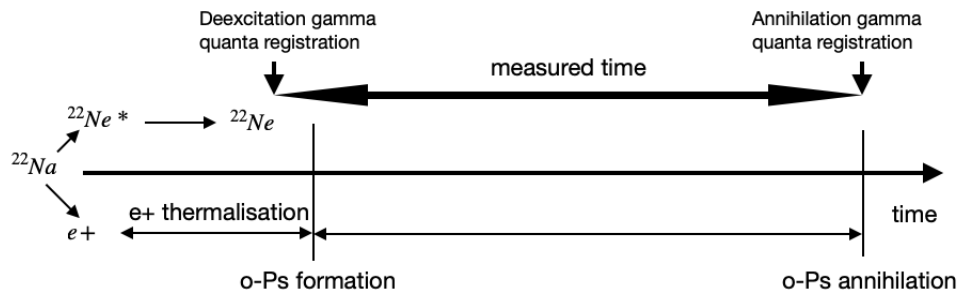


Figure 2.2: The order of events in measuring lifetime of positronium. The timeline is not to scale.

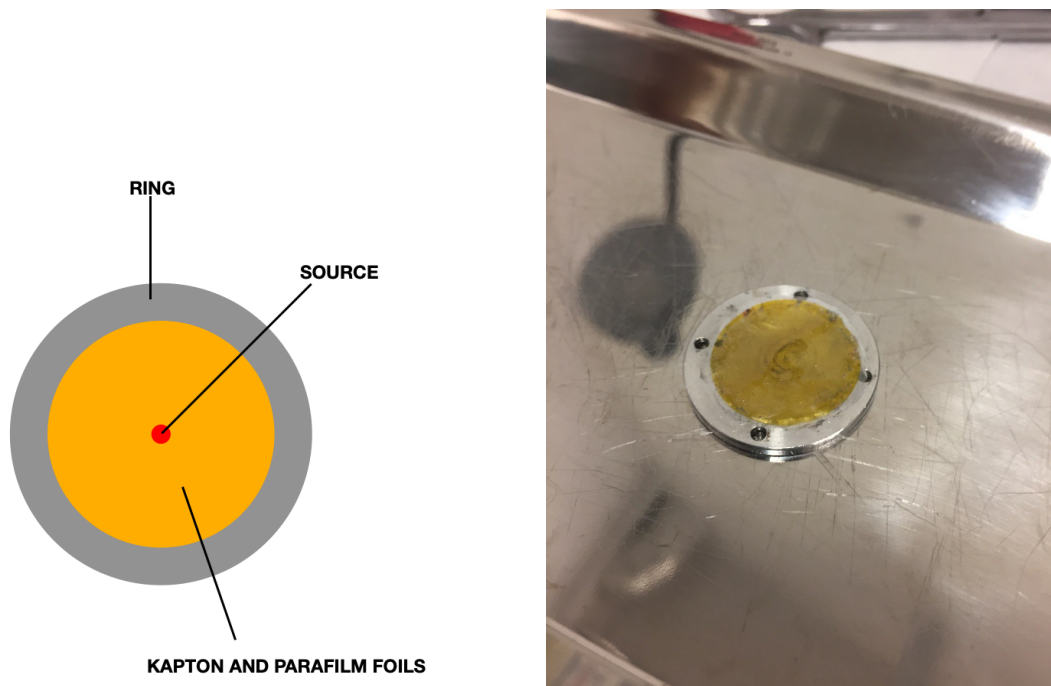


Figure 2.3: *Left:* Schematic depiction of a source in a ring and foil. *Right:* Picture of a source sealed in kapton and parafilm in a metal ring.

2.2 Positronium bound state

Positronium is a meta-stable atom, consisting of one electron and one positron orbiting around their mutual center of mass. The source of positrons can be a β^+ decay, which decays according to equation (2.1). An example of an isotope undergoing β^+ decays is ^{22}Na .

$${}^A_Z X \rightarrow {}^A_{Z-1} Y + e^+ + \nu_e, \quad (2.1)$$

where A denotes mass number and Z atomic number of the atom, X is a parent nuclide, Y is daughter nuclide, e^+ is a positron, ν_e is electron neutrino.

Positronium occurs in two states: para-positronium and ortho-positronium. Ortho-positronium is characterized by decay into three or larger odd number of gamma quanta.

Para-positronium decays to even number of gamma quanta, most often two, each with energy of 511 keV. In both cases the sum of energy of gamma quanta is equal to invariant mass of e-e+ system [16]. Ortho- and para-positronium are schematically depicted in Figure 2.4.



Figure 2.4: Schematic depiction of positronium decay into (*left*) three and (*right*) two gamma quanta.

The lifetime of para-positronium in vacuum is equal to 0.125 ns, which is three orders of magnitude less than the lifetime of ortho-positronium in vacuum. The lifetime of ortho-positronium strongly depends on electron-free volumes in medium in which it exists. In vacuum its lifetime is equal to 142 ns [17], while in water it is equal to 1.9 ns. Shortening of the lifetime corresponds to the possibility of encountering an electron and decaying due to a pick-off process, shown schematically in Figure 2.5 in which two gamma quanta are emitted, both with energy of 511 keV [18]. This phenomenon may be used for example in examination of biological tissues [19].

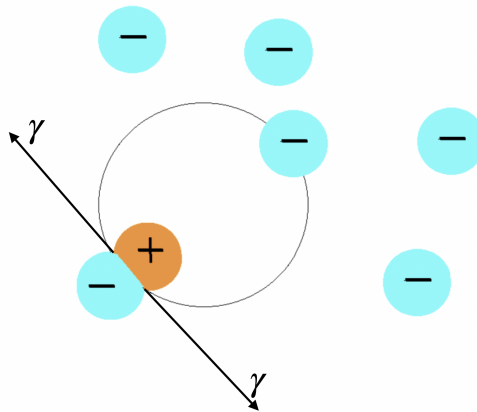


Figure 2.5: Schematic representation of a pick-off process. The blue circles represent electrons and the orange circle represent positron. In the process of annihilation two gamma quanta are emitted with energy of 511 keV at an angle of almost 180°.

2.3 Tao-Eldrup model

The behaviour of positronium bound state in matter is theoretically explained by Tao-Eldrup model in which positronium is trapped inside free volumes in the material structure. Free volume is a term coined to describe a region of lower electron density explained by

Wigner-Seitz theory. In their approximation free volumes may be understood as a finite potential wells represented in Figure 2.6 with the potentials:

$$\begin{aligned} U &= U_0, \quad r < r_0, \\ U &= 0, \quad r_0 < r < r_1, \end{aligned} \quad (2.2)$$

with boundary conditions:

$$\begin{aligned} \Psi_{Ps,out}(r_0) &= \Psi_{Ps,in}(r_0), \\ \Psi'_{Ps,out}(r_0) &= \Psi'_{Ps,in}(r_0), \\ \Psi'_{Ps,in}(r_1) &= 0, \end{aligned} \quad (2.3)$$

where U_0 is initial value of the potential, r_0 is the radius of barrier representing the molecular forces in the medium, r_1 is summary radius of the well and barrier, $\Psi_{Ps,out}$ and $\Psi_{Ps,in}$ are positronium wave functions outside and inside the well [20].

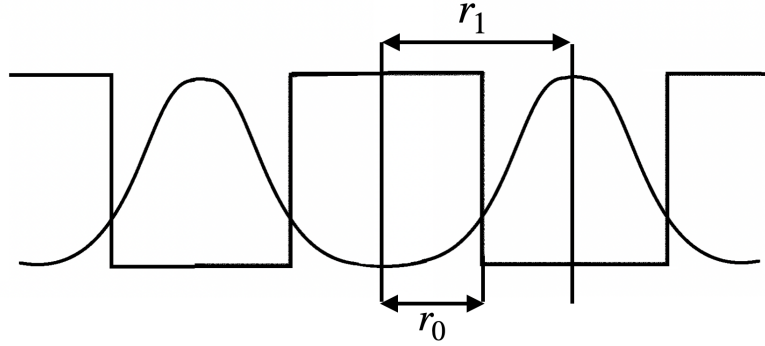


Figure 2.6: Finite periodical square well potentials. r_1 is the radius of both barrier and the well. r_0 represents the barrier radius (medium).

Correlating mean lifetime of ortho-positronium to the size of a free volume would require complicated calculations, which can be omitted if applying Tao's simplification, in which free volume is described as infinite spherical potential well. The probability of finding ortho-positronium outside of the well is described by:

$$P = 4\pi \int_R^\infty \Psi_{out}^2(r) r^2 dr, \quad (2.4)$$

where R is a radius of a free volume, Ψ is wave function of positronium outside the well.

Tao also introduced a ΔR parameter, which represents the overlap of the positronium wave function with the medium wave functions and fulfills the condition:

$$\int_R^\infty \Psi_{out}^2(r) r^2 dr = \int_R^{R+\Delta R} \Psi_{in}^2(r) r^2 dr, \quad (2.5)$$

where Ψ_{out} is wave function of positronium outside the well, Ψ_{in} is a wave function of a particle inside the well and is approximated by $\sin(kr)/r$, R is the volume radius.

Physically ΔR represents the thickness of the layer of electrons on the walls of the rigid sphere (a pore) with radius R in which the positronium is confined [20].

Eldrup formulated the dependency of the lifetime of ortho-positronium for a volume radius:

$$\tau_{Ps} = \frac{1}{2} \left(1 - \frac{R}{R+R_0} + 2\pi \sin \left(\frac{2\pi R}{R+R_0} \right) \right)^{-1}, \quad (2.6)$$

where τ_{ps} is lifetime of ortho-positronium, R is volume radius, R_0 is an empirical value of 0.166 nm [21].

The given formula applies best to medium with spherical volumes with radius from 0.2 nm up to 2 nm [21].

To produce larger fraction of o-Ps in the experiment a highly porous material is necessary.

2.4 Porous materials

Previously described dependency of mean lifetime of ortho-positronium on volume of electron-free spaces can be observed measuring lifetime in pores of XAD4. Though the average size of a pore is said to be 50 Å some pores may connect and create long crevices through which positronium may wander longer before annihilation. It is acknowledged that in porous materials it is possible to attain three components for lifetime of ortho-positronium [22]. As the water penetrates into the XAD4 pores it should result in decrease in size of electron-free volumes and mean lifetime of ortho-positronium. Schematic structure of XAD4 pores and ortho-positronium path is shown in Figure 2.7 on the left.

The porous material used in this thesis was XAD4 polymer shown in Figure 2.7 on the right, produced by Sigma-Aldrich company and shipped in a form of small beads. Its mean pore diameter is 50 Å, which allows water molecules to penetrate it. It is used to remove small hydrophobic compounds from polar solvents. The polymer sold by Sigma-Aldrich company was shipped wet, it can absorb water up to 60% of its mass [23].



Figure 2.7: *Left:* Schematic representation of different sized pores in XAD4 and ortho-positronium paths. *Right:* Sample of dried XAD4 used in the measurements in a beaker.

3. Experimental methods

3.1 Measurement setup

Measurement setup, shown schematically in Figure 3.1, consisted of two detectors, each containing a scintillation crystal BaF_2 manufactured by Scionix and H3378-51 Hamamatsu photomultiplier. The detectors were powered by CAEN SY4527 high voltage power supply. Detectors were connected to LeCroy 608C constant fraction discriminator (CFD), where thresholds were applied to signals from the detectors. The bottom detector, that was registering deexcitation gamma quanta (with energy 1274 keV), had threshold of over 70 mV applied. For the top detector, that was registering annihilation gamma quanta (511 keV), the threshold was set to over 14.5 mV.

Signal was passed on to LeCroy662 coincidence module with coincidence window of 110 ns. Acquired data was sent over to the computer with DRS4 digitizer evaluation board, and saved in binary format.

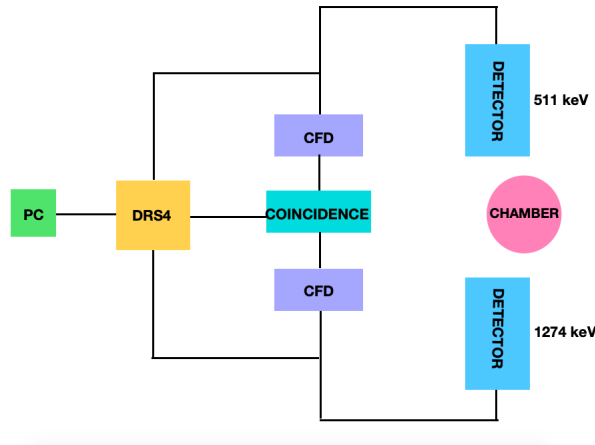


Figure 3.1: Scheme of the measurement setup. The source and the sample were placed in chamber (pink), off the center of detectors axis. The detectors (depicted in blue color) are registering gamma quanta with respective energies: upper detector registers gamma quanta with energy of 511 keV, the bottom detector registers gamma quanta with energy of 1274 keV. The signal is split and passed on to constant fraction discriminator (CFD) (violet) as well as DRS4 evaluation board (yellow). From CFD the signals are transmitted to coincidence module (turquoise) and from there to DRS4 evaluation board. From DRS4 the signals are passed on to the computer (captioned as PC, depicted in green).

The chamber with the sample was placed in the holder in between the detectors. The detectors are mounted 180° towards each other and the chamber is placed off center to avoid registering both 511 keV gamma quanta. The ^{22}Na source used for the measurements had activity of 1.3 MBq and was produced on 02.2021. One of the used chambers (PTFE chamber) is shown in Figure 3.2. It consists of smaller upper part, bigger bottom part and ring with a source. In both parts of the chamber there are half-spherical spaces 1cm in diameter for holding the sample. The ring with source is mounted in between the chamber parts and during the measurements the source is surrounded by the sample. Picture of

sample inside the chamber is shown in Figure 3.3. In Figure 3.4 are pictures of measurement setup and chamber in the holder.

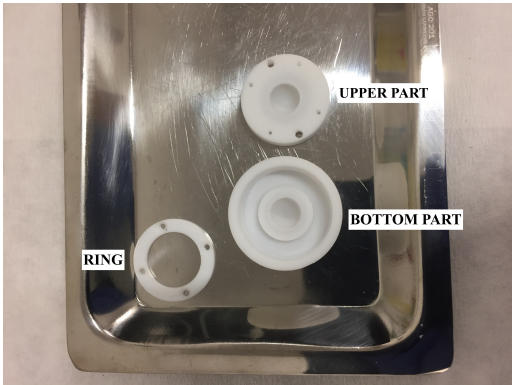


Figure 3.2: Picture of disassembled Teflon chamber. *Left:* Ring, to which the source is mounted. *Top:* Upper part of the chamber. *Bottom, on the right:* Bottom part of the chamber.

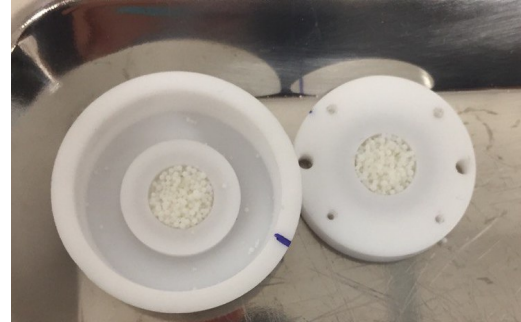


Figure 3.3: Both halves of Teflon chamber filled with dry XAD4. The chamber has a marking to assure repeatability of chamber assembly.

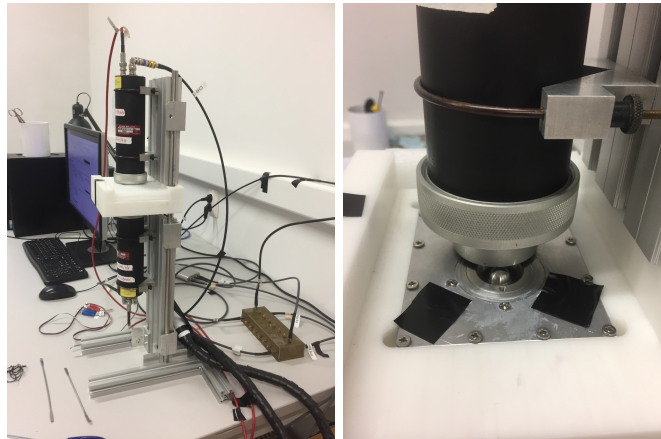


Figure 3.4: *Left:* Picture of measurement setup. *Right:* Picture of chamber in the holder.

3.2 Sample preparation

3.2.1 Mass percentage

The weight of the substance inside the chamber was measured with a scale. To calculate proportions, in which it was mixed with water, the weight of XAD4 in upper and bottom parts of the chamber were added and divided by two. Weight of added water was calculated using approximation that density of water is 1 g/cm^3 . Mass percentage was used as a way of describing proportions of used polymer and water. Mass percentage was calculated using formula:

$$C_p = \frac{m_s}{m_s + m_r} \cdot 100\%, \quad (3.1)$$

where m_s is mass of the substance (in case of this experiment XAD4) and m_r is mass of the solvent (in this case water).

3.3 Measurement protocol

Before the proper measurements presented in this dissertation, there was a set of measurements performed to assess the right method of measuring lifetime of ortho-positronium in XAD4. As a result it was concluded, that for allowing measurement of component corresponding to medium mean lifetime for XAD4, data for 3,6 million events must be gathered in one measurement. But for allowing visibility of the longest component in XAD4 the measurements would need to be elongated to achieve even better statistic. Having source with activity less then 1.3 MBq (this value references to source used and it is not a result of calculations) it would require more time then 4 hours.

Prior to the measurements the temperature of $22^{\circ}C$ was set on the thermostat providing constant temperature of fluid flowing through chamber holder. The chamber, sample and ring with source were handled using disposable gloves and pliers. Upper and bottom parts of the chamber were weighted with a precise Radwag scale [24] and noted. Then the final weights of chamber parts with sample inside were assessed and added such as the weight of added polymer would be the the same in both parts, around 0,08g of dried substance. Afterwards the source was mounted on the upper part of the chamber taking care to always position the source the same way, for this the markings were done on the ring and both parts of the chamber. Upper part of the chamber was placed into the bottom part as it is shown in Figure 3.5 on the left. Then the chamber was mounted into the setup, afterwards hands in gloves were tested for radioactivity leakage with assistance of second person operating radiometer.

Suitable parameters were set using protocol [25]. Prior to measurements timing calibration was performed. Afterwards data acquisition was set to save 120 thousand counts for benchmark measurement (about 10 minutes). Time of start, humidity and temperature inside the laboratory was noted.

In case of the measurements with water, distilled water was added to clean beaker. After the measurement was finished ending time was noted and read_binary program was used for analysis. During that time the sample was being prepared for proper measurements. Using disposable gloves the chamber was unmounted from the setup and disassembled. Correct amounts of water were added using 'Eppendorf Research plus' single channel pipette with volume range 2-20 μl [26] and 'Discovery Comfort' single channel pipette with volume range 100-1000 μl [27] shown in Figure 3.5 on the right. Same as previously, the chamber was carefully assembled and mounted in the holder of the setup paying attention to retain same alignment of chamber parts and the setup. The gloves were examined with radiometer operated by assisting person. Measurement was set for 3 600 000 counts (about 4 hours and 20 minutes). The time, humidity and temperature inside the laboratory were noted.

3.4 Conducted measurements

In total there were 6 measurements conducted: one measurement of distilled water, one measurement of dried XAD4 and 4 measurements of XAD4 with different amounts of water. One measurement lasted about 4 hours and 20 minutes.

Additionally there were 4 benchmark measurements done of dry XAD4, lasting about 10 minutes each, described in 3.6.1.

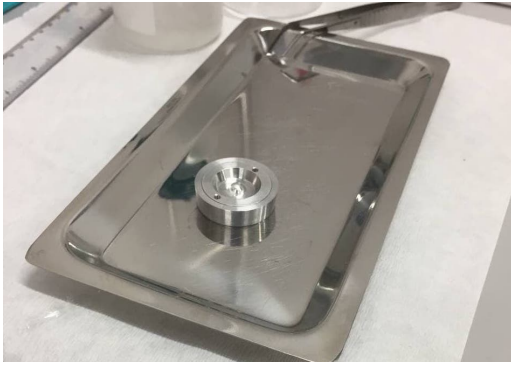


Figure 3.5: *Left*: Assembled aluminium chamber on a tray. *Right*: Picture of the pipettes used for measuring amounts of added water.

In table 3.1 a list of conducted measurements is presented.

Table 3.1: Conducted measurements along with conditions in a laboratory during the measurements. C_p is mass percentage, 'T' and 'H' are temperature and humidity registered in laboratory room near the setup.

Measurement	C_p [%]	Time	T [°C]	H [%]	Chamber
Water	0	4h 25m	23.6	43.6	PTFE
Wet XAD4* with 10 μ l of water	43.6	3h 11m	24.6	26.1	Aluminium
Wet XAD4*	46	3h 56m	22.7	17.2	Aluminium
Dried XAD4 with 40 μ l of water	67.3	4h 19m	24.1	15.3	PTFE
Dried XAD4 with 40 μ l of water	69.2	4h 24m	24.6	25.7	Aluminium
Dried XAD4	100	4h 26m	24.1	35.9	PTFE

Before the measurements some amount of XAD4 was put into a beaker and set aside for the moisture to evaporate from it. Material prepared this way is described in this thesis as 'dried XAD4'.

*Wet XAD4 is term used in this thesis to describe XAD4 used without prior preparation (drying). Those samples were drawn from the original container. For this portion the assumption was made, that the substance in the storage was close to being fully saturated with liquid and its mass percentage of XAD4 in mixture was 46%.

3.5 Signal analysis

Acquired data was stored in binary format. Each signal was probed in time domain and such information was saved for each scintillator counter. The exemplary spectra registered by one of detectors is shown in Figure 3.6. Gathered information about charge and amplitude of the signals allow for distinguishing between deexcitation and annihilation gamma quanta as well as determining the time difference between them.

The amplitude is calculated according to the equation:

$$A = A_{min} - Ped, \quad (3.2)$$

where A_{min} is minimal value of the spectrum, Ped is pedestal value of the signal, that is calculated as a mean value for 10 points after first 20 points from the left side of the spectrum.

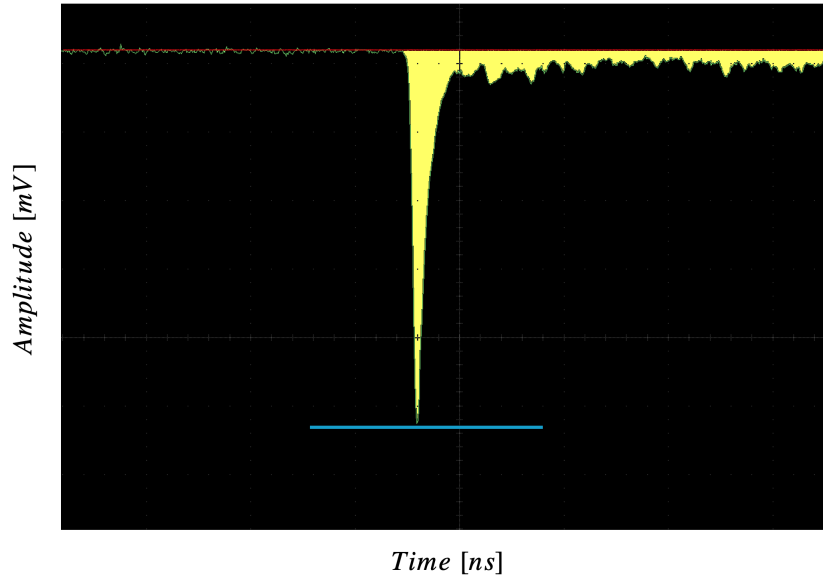


Figure 3.6: Exemplary spectra of signal acquired by one of the detectors. The pedestal of a signal is marked by a red line. The amplitude (marked by a blue line) is calculated as a lowest point of the spectrum (minimal value of the signal), the area of the signal (in yellow) is proportional to the charge.

The charge is calculated with:

$$Q = \int \frac{U(t)dt}{R}, \quad (3.3)$$

where $U(t)$ is signal volage and R is constant value of resistance of given channel on DRS4 evaluation board, equal to 50Ω .

The time difference is calculated for the signals fulfilling the criteria described above and registered in the time window set to 110 ns. The equation for calculating the time difference is:

$$\Delta T = T_{511keV} - T_{1274keV}, \quad (3.4)$$

where T_{511keV} and $T_{1274keV}$ times are calculated as an intersection points on signal spectra between line constructed by connecting the value of signal at 30 % and 80% amplitude and line $y = 0.1 \cdot A$, where A is amplitude.

As the spectra registered by photomultipliers contain regions corresponding to annihilation, deexcitation and also different interactions of gamma quanta with matter (e.g. Compton scattering) appropriate trigger thresholds must be applied to registered signals. For signals coming from annihilation the accepted charge range was 2.5 - 5 pC. For signals coming from deexcitation the accepted charge range was 9 - 12 pC.

As a result at this step of analysis a binary file with time differences was produced. It was used as an input in the next stage of analysis.

3.6 Data analysis

Analysis of obtained positronium lifetime spectra was done using PALS Avalanche program [28].

As it was previously noted, in PALS measurements of polymers such as XAD4, there are three visible components that correspond to value of mean lifetime of ortho-positronium in its pores [22].

PALS Avalanche allows for setting initial values for lifetime parameters and intensities and also fixing the values for lifetime and intensities. Knowing approximate and accurate values of some parameters present in the fit, the only fully free parameters are two components corresponding to mean lifetime of ortho-positronium in XAD4 pores, changing from sample to sample.

The fitted function can be described as:

$$f(t) = y_0 + \sum_{i=0}^{n_\tau} \sum_{j=0}^{n_{Gauss}} P_i \cdot \alpha_j \cdot F(t, \tau_i, t_0^{(j)}, \sigma_j), \quad (3.5)$$

where:

$$F(t, \tau_i, t_0^{(j)}, \sigma_j) = \frac{1}{2\tau} \exp\left(\frac{\sigma^2}{2\tau^2} - \frac{t - t_0}{\tau}\right) \left(\operatorname{erf}\left(\frac{t - t_0 - \frac{\sigma^2}{\tau}}{\sqrt{2}\sigma}\right) - \operatorname{erf}\left(\frac{-t_0 - \frac{\sigma^2}{\tau}}{\sqrt{2}\sigma}\right) \right) \quad (3.6)$$

where y_0 - background level, P_i -intensity of lifetime, α_j - fraction of given Gauss distribution, t_0 - time offset of the detector, σ - resolution of the apparatus, t - time difference between the detectors, τ - mean positron lifetime. The sum of all intensities should be given by $\sum_i^n P_i = 1$ and $\sum_j^{n_{Gauss}} \alpha_j = 1$ [29].

Exemplary fitted spectra is shown in Figure 6.2. The meaning of the components of the spectra is explained in Table 3.2.

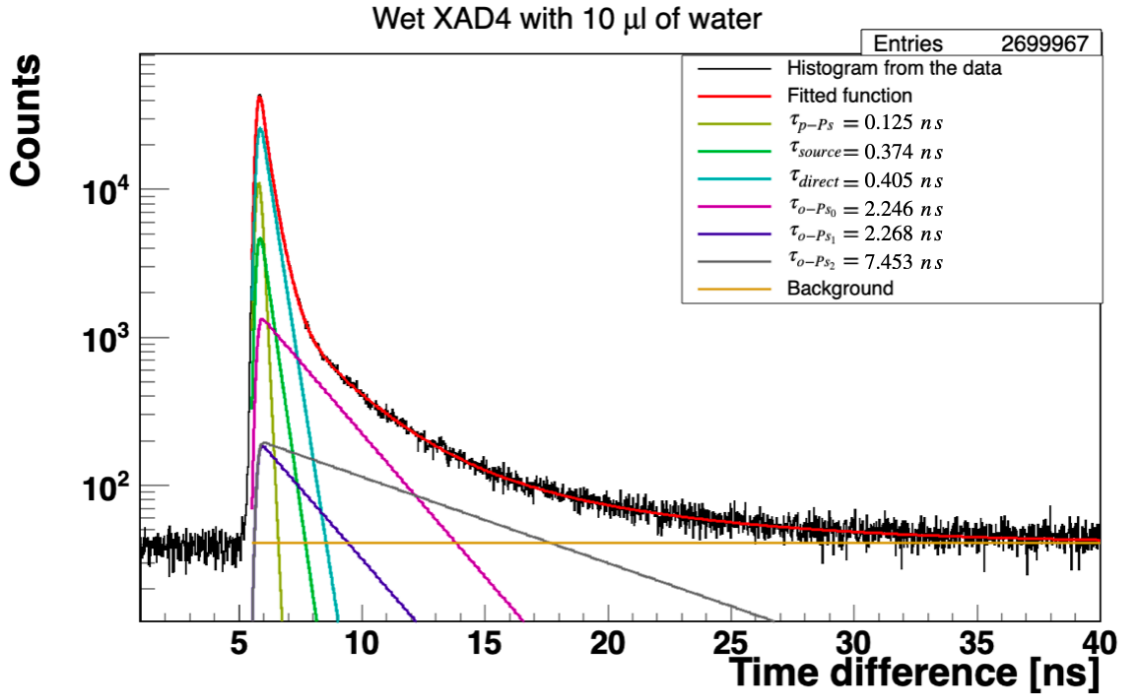


Figure 3.7: Exemplary fitted spectra done using PALS Avalanche program.

Table 3.2: Description of components used in fitted function. 'Type' column states whether the parameter was: lf - lifetime fixed (intensity is free), f - fixed, pf - partially free*, nf - not fixed.

Symbol	Description	Type
τ_{p-Ps}	para-positronium annihilation	lf
τ_{source}	positron annihilation in source	f
τ_{direct}	positron annihilation in sample	pf*
τ_{o-Ps_0}	ortho-positronium annihilation in parafilm	f
τ_{o-Ps_1}	1st component for ortho-positronium annihilation in XAD-4 pores	nf**
τ_{o-Ps_2}	2nd component for ortho-positronium annihilation in XAD-4 pores	nf

The component that corresponds to lifetime of ortho-positronium in parafilm in temperature of $22^{\circ}C$ was ascertained in the previous measurements not covered in this thesis. In those measurements only empty chamber with source wrapped in parafilm was inside the setup. One measurement lasted around 60 minutes collecting 1 million events. In result the component could be added to the function as a fixed parameter to raise accuracy of the fit.

* τ_{direct} was partially free parameter, which means value of its intensity can change freely and its initial lifetime value can change in range of given variation level. For this certain fitting arrangement variation level was equal to 20%. This means the final values can range from 80% to 120% of the initial value.

**In case of measurement of only water in the chamber τ_{o-Ps_1} component was set as partially free for both value of intensity and value of mean lifetime.

3.6.1 Benchmark measurements

Additionally to the proper measurements 4 benchmark measurements were done. The goal of these measurements was to assess the dispersion of values of mean lifetime obtained from the measurements. The acquired lifetime spectra were fitted using PALS Avalanche program presented in previous chapter, but with five components instead of six (without the component for long-living ortho-positronium in XAD4 pores due to the low statistics of only 10 000 events). The fitted spectra are shown in detail in Appendix A, page 38.

In Table 3.3 the values of the component corresponding to mean lifetime of ortho-positronium and its intensity from fitting the spectra of benchmark measurements are presented.

Table 3.3: Values of component corresponding to mean lifetime of ortho-positronium in XAD4 pores and intensity obtained for benchmark measurements of dry XAD4. χ^2_{ν} is $\chi^2/\text{degrees of freedom}$

Measurement	Lifetime [ns]	Intensity [%]	χ^2_{ν}	Chamber
1	7.36(30)	10.2(7.3)	1.19	Aluminium
2	5.02(18)	11.5(9.1)	1.20	PTFE
3	5.25(20)	10.8(7.7)	1.25	PTFE
4	5.74(22)	10.7(8.3)	1.05	PTFE

In Figure 3.8 the graph with values of mean lifetimes of ortho-positronium in XAD4 pores with uncertainties for each benchmark measurement is presented. The line represents mean value.

Mean value of component corresponding to ortho-positronium lifetime in XAD4 pores is 5.84 ns and mean standard deviation is 0.53 ns.

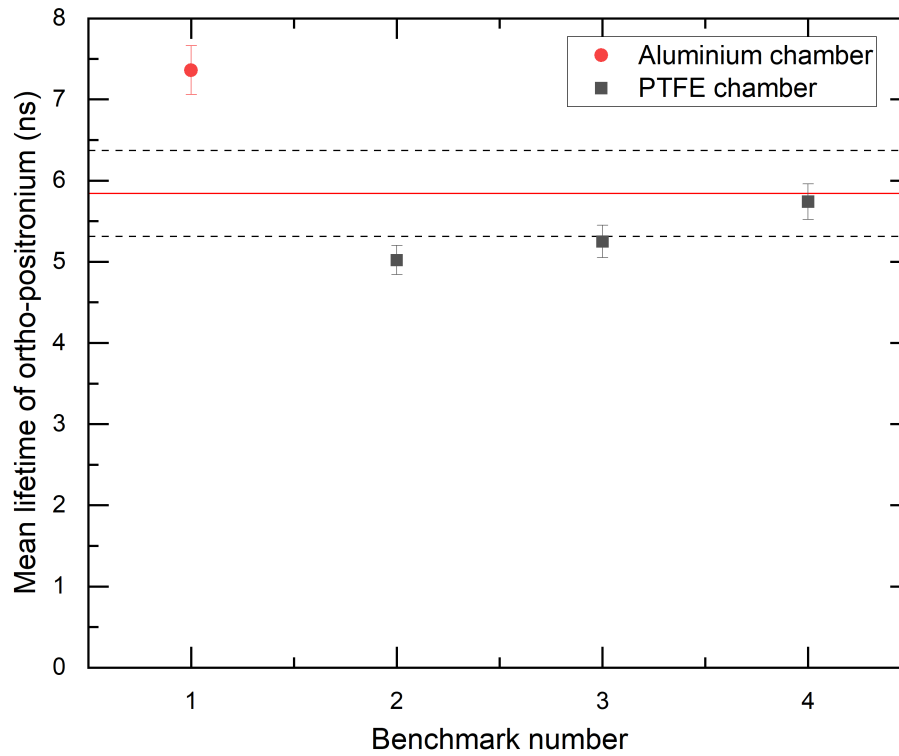


Figure 3.8: Graph of values of mean lifetimes of ortho-positronium in XAD4 pores with uncertainties for each benchmark measurement. The red line represents mean value $y = 5.84$ ns. Two dotted lines above and below the mean represent the mean standard deviation equal to 0.53 ns.

4. Results and conclusions

4.1 Ortho-positronium lifetime dependency

The results for the second component dependency on mass percentage, corresponding to ortho-positronium lifetime in XAD4 pores, are presented in the Table 4.1. The graph showing the dependency of lifetime of ortho-positronium for second component corresponding to XAD4 pores on mass percentage of XAD4 in mixture is presented in Figure 4.1. The graph of the intensity of second component lifetime depending on mass percentage is shown in Figure 4.2.

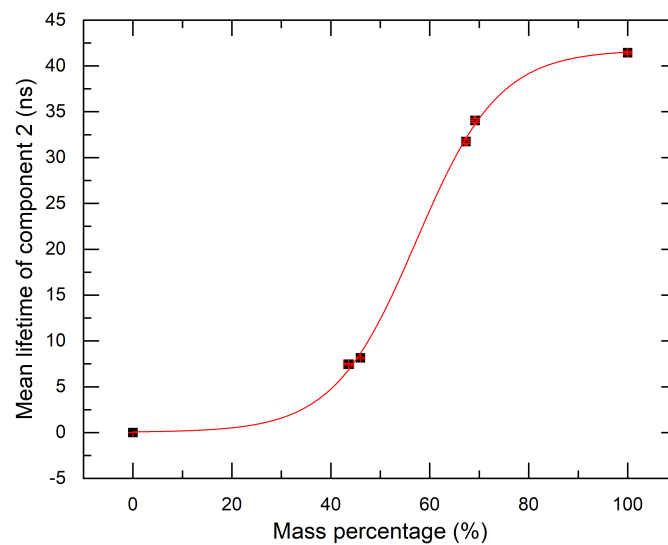


Figure 4.1: Dependency of the lifetime of ortho-positronium for the XAD4 second component on mass percentage of XAD4 in water. Trend line is drawn to guide the eye and it is described by: $y = 20.9 + 20.9 * \tanh(0,06 * (x - 57.27))$.

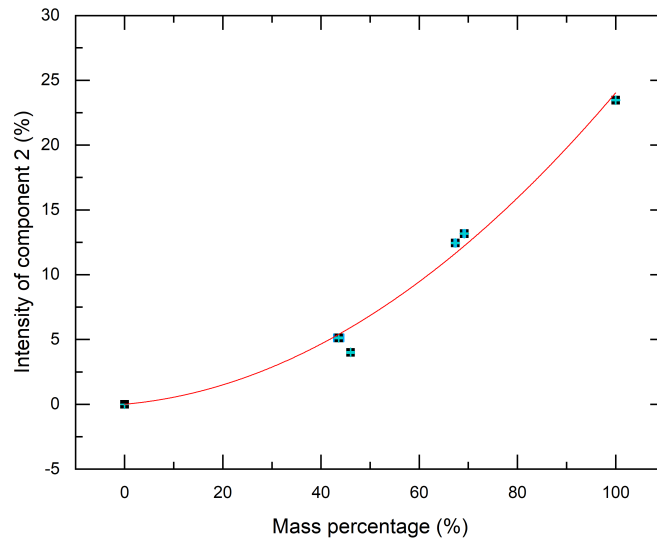


Figure 4.2: Dependency of the intensity of second component on mass percentage of XAD4 in water. Trend line is drawn to guide the eye and it is described by: $y = 0,0337 \cdot x + 0,00207 \cdot x^2$.

The results for the first component dependency on mass percentage, corresponding to ortho-positronium lifetime in XAD4 pores, is presented in Table 4.2. The graph showing the dependency of lifetime of ortho-positronium for first component corresponding to XAD4 pores on mass percentage of XAD4 in mixture is presented in Figure 4.3. The graph of the intensity of first component lifetime depending on mass percentage is shown in Figure 4.4.

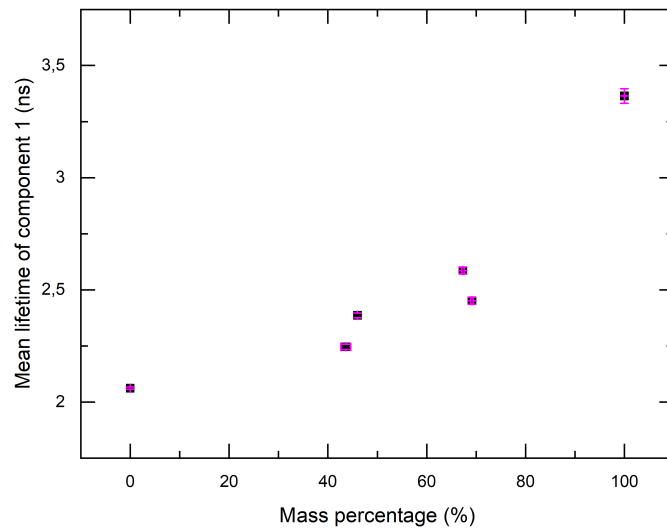


Figure 4.3: Dependency of the lifetime of ortho-positronium for the XAD4 first component on mass percentage of XAD4 in water.

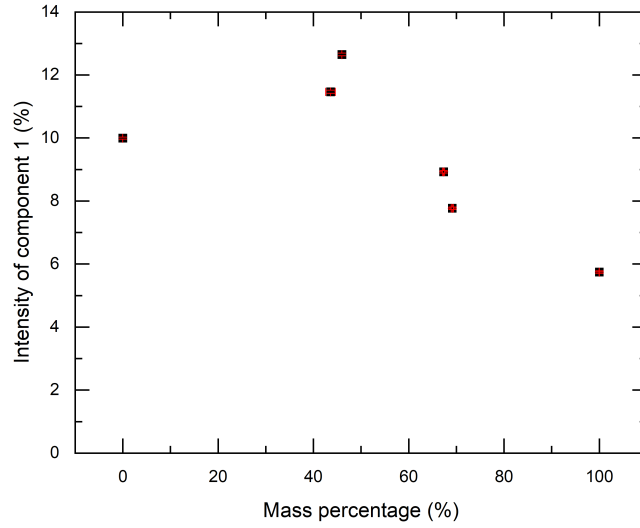


Figure 4.4: Dependency of the intensity of first component on mass percentage of XAD4 in water.

Table 4.1: Results of the PALS measurements of lifetime and intensity of ortho-positronium second component for XAD4. χ^2_ν is the goodness of the fit defined as χ^2 per degrees of freedom.

Measurement	Mass percentage [%]	Lifetime [ns]	Intensity [%]	χ^2_ν
Water	0	0	0	1.44
Wet XAD4 + 10 μ l water	43.6(1.0)	7.453(75)	5.123(50)	1.24
Wet XAD4	46	8.153(99)	4.002(45)	1.26
XAD4 + 40 μ l water	67.33(23)	31.76(22)	12.447(39)	1.38
XAD4 + 40 μ l water	69.16(23)	34.06(22)	13.170(38)	1.34
Dry XAD4	100	41.44(17)	23.467(40)	1.34

Table 4.2: Results of the PALS measurements of lifetime and intensity of ortho-positronium first component for XAD4. χ^2_ν is the goodness of the fit defined as χ^2 per degrees of freedom.

Measurement	Mass percentage [%]	Lifetime [ns]	Intensity [%]	χ^2_ν
Water	0	2.0620(49)	11.97(65)	1.44
Wet XAD4 + 10 μ l water	43.6(1.0)	2.246(12)	11.463(50)	1.24
Wet XAD4	46	2.387(10)	12.646(45)	1.26
XAD4 + 40 μ l water	67.33(23)	2.586(15)	8.924(39)	1.38
XAD4 + 40 μ l water	69.16(23)	2.452(16)	7.767(38)	1.34
Dry XAD4	100	3.364(32)	5.744(40)	1.34

In Table 4.3 the the lifetimes and their intensities for component deriving from direct annihilation in sample and chamber are presented. In the Table 4.4 intensities for component corresponding to para-positronium annihilation are presented. In the most right columns chamber material is noted.

Acquired results indicate that there is a dependency of lifetime of ortho-positronium in XAD4 pores on mass percentage of XAD4 in water. Given statement is true both for

Table 4.3: Results of the PALS measurements of lifetime and intensity of direct annihilation in the sample and chamber. C_p is mass percentage of the XAD4 in mixture.

Measurement	C_p [%]	Lifetime [ns]	Intensity [%]	Chamber
Water	0	0.42606(85)	24.128(39)	PTFE
Wet XAD4 + 10 μ l water	43.6(1.0)	0.40502(66)	57.929(49)	Aluminium
Wet XAD4	46	0.40660(60)	60.248(45)	Aluminium
XAD4 + 40 μ l water	67.33(23)	0.41731(66)	48.856(45)	PTFE
XAD4 + 40 μ l water	69.16(23)	0.40327(63)	48.619(45)	Aluminium
Dry XAD4	100	0.40365(74)	37.879(46)	PTFE

Table 4.4: Results of the PALS measurements of intensity of para-positronium annihilation. C_p is mass percentage of the XAD4 in mixture. Value of the mean lifetime of para-positronium was set to 0.125 ns.

Measurement	C_p [%]	Intensity [%]	Chamber
Water	0	9.992(31)	PTFE
Wet XAD4 + 10 μ l water	43.6(1.0)	13.876(41)	Aluminium
Wet XAD4	46	11.494(38)	Aluminium
XAD4 + 40 μ l water	67.33(23)	18.164(39)	PTFE
XAD4 + 40 μ l water	69.16(23)	18.834(39)	Aluminium
Dry XAD4	100	21.300(40)	PTFE

shorter and for longer component corresponding to ortho-positronium lifetime in XAD4 pores. There is also a dependency of intensity of the longer component on the mass percentage of XAD4 in water.

This study showed that the lifetimes of ortho-positronium in XAD4 with different mass percentage in water range for the first component from about 2 ns in pure water to 3.4 ns in dry XAD4, and for the second component it goes up to about 41 ns. Accurate values are presented in Tables 4.1 and 4.2. More statistics are needed to observe third component corresponding to mean lifetime of ortho-positronium in XAD4 .

Firstly, in the future studies there should be more measurements done for one value of mass percentage to examine the systematical error for this type of measurement. The uncertainties estimated for the points are calculated with propagation of scale and pipette random errors. Those are not the only sources of uncertainty in measurements. It can be seen in the Figures 4.1, 4.3 that dispersion of points does not correspond to the error bars extent. Other sources of uncertainties could be temperature in the lab and especially humidity, as the XAD4 wasn't dried with the vacuum pump and some water molecules were present in polymer pores, it could contribute to imprecision in determining the mass percentage value. Also the mass percentage value of wet XAD4 was based on assumption that the mass percentage of shipped material was 46%. In future studies measurements for this mass percentage should be repeated with proper amount of water added to dry XAD4.

Secondly more measurements for different mass percentage values should be done to better estimate the shape of dependency and matching function.

4.2 Chamber differences

In the course of the research two types of chambers were used: aluminium chamber and PTFE chamber. One of the reason for changing chamber material type was the chemical reactivity of XAD4 and water mixture in contact with aluminium, as the used aluminium chamber exhibited the signs of corrosion.

The difference between results for similar concentrations but for two different chambers was already shown but is placed again in the Tables 4.5 and 4.6 below for convenience. The comparison between intensities for first component shows that it's higher for measurements done using PTFE chamber. Comparison of intensities of the second component shows that it is higher for measurements done using aluminium chamber. The sum of intensities of first and second component are around 22% for both chamber types.

The comparison, in case of benchmark measurements of lifetimes and intensities, shows that of chamber material may affect the lifetime of ortho-positronium and there is an indication of a difference in intensity of the ortho-positronium component, as its value is lowest for aluminium chamber.

Table 4.5: Values of first component corresponding to mean lifetime of ortho-positronium in XAD4 pores and intensity for benchmark measurements and for mass percentages of XAD4 in water of 67.33(23)% and 69.16(23)%. χ^2_ν is $\chi^2/\text{degrees of freedom}$

Measurement	Lifetime [ns]	Intensity [%]	χ^2_ν	Chamber
Benchmark 1	7.36(30)	10.2(7.3)	1.19	Aluminium
Benchmark 2	5.02(18)	11.5(9.1)	1.20	PTFE
Benchmark 3	5.25(20)	10.8(7.7)	1.25	PTFE
Benchmark 4	5.74(22)	10.7(8.3)	1.05	PTFE
67.33(23)% XAD4	2.586(15)	8.924(39)	1.38	PTFE
69.16(23)% XAD4	2.452(16)	7.767(38)	1.34	Aluminium

Table 4.6: Results of the PALS measurements of lifetime and intensity of ortho-positronium second component for XAD4 for mass percentages of XAD4 in water of 67.33(23)% and 69.16(23)%. χ^2_ν is the goodness of the fit defined as χ^2 per degrees of freedom.

Measurement	Lifetime [ns]	Intensity [%]	χ^2_ν	Chamber
67.33(23)% XAD4	31.756(22)	12.447(39)	1.38	PTFE
69.16(23)% XAD4	34.06(22)	13.170(38)	1.34	Aluminium

The comparison of this small number of measurements may be an indication of properties and suitability of both materials in PALS measurements of ortho-positronium lifetime in XAD4 pores.

5. Summary and Perspectives

The goal of this thesis was to develop a replicable method for measuring lifetime of ortho-positronium in mixtures of water and XAD4 using PALS technique. In course of this work six 4-hour-long measurements (4 measurements of different mass percentage of XAD4 in water, 1 of distilled water and 1 of dry XAD4) and additionally four benchmark measurements of dry XAD4 were done. As it is shown in the results of the presented research, the dependence of lifetime of ortho-positronium in XAD4 pores on mass percentage of XAD4 in water was determined.

In the second chapter of this thesis information about source of positrons, positronium formation and ortho-positronium lifetime dependency on free volumes that lays at the heart of PALS technique are presented.

In third chapter experimental setup and protocol were featured as well as the data analysis procedure. At the end of this part results of benchmark measurements were presented, that helped assessing the standard deviation of ortho-positronium mean lifetime values.

In the next part results of the measurements were presented along with conclusions.

In course of this thesis it was shown that addition of water to XAD4 results in partial filling up of larger crevices with water, what consequently decreases of size of electron-free volumes and mean lifetime of ortho-positronium.

The developed method will allow, with some additional measurements, for estimating a concentrations of XAD4 and water mixtures needed to prepare solutions that will, in a course of imaging, exhibit ortho-positronium lifetime values in ranges that are characteristic to the biological healthy and malignant tissues.

The results of this research will be used for preparation of a phantom that can be used for assessing the precision of newly developed method for measuring lifetime of ortho-positronium alongside the activity concentration.

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6. Appendix A - fitted functions

6.1 Ortho-positronium lifetime in XAD and water solution

Fitting was done using PALS Avalanche program written by dr Kamil Dulski.

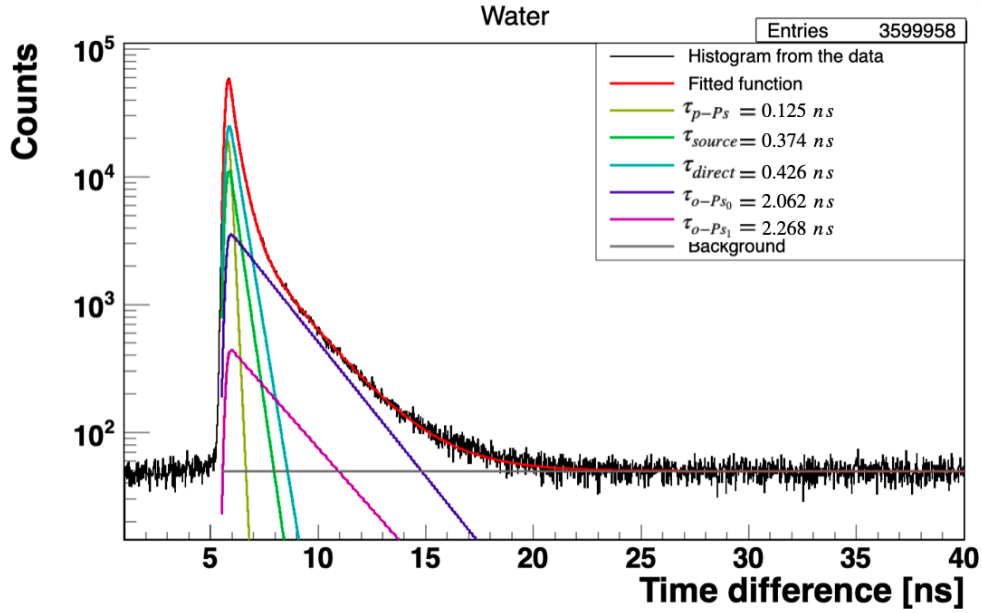


Figure 6.1: Fitted spectra of positronium lifetime in water. Measurement was done using PTFE chamber.

Table 6.1: Lifetimes and intensities for components of the fit. Measurement was done using PTFE chamber. $*\chi^2_{\nu} = 1.44$

Component	Mean lifetime [ns]	Intensity [%]
τ_{p-Pos}	0.125	9.992(31)
τ_{source}	0.347	10
τ_{direct}	0.42606(85)	24.128(39)
τ_{o-Pos_0}	2.268	1.61
τ_{o-Pos_1}	2.0620(49)	11.97(65)

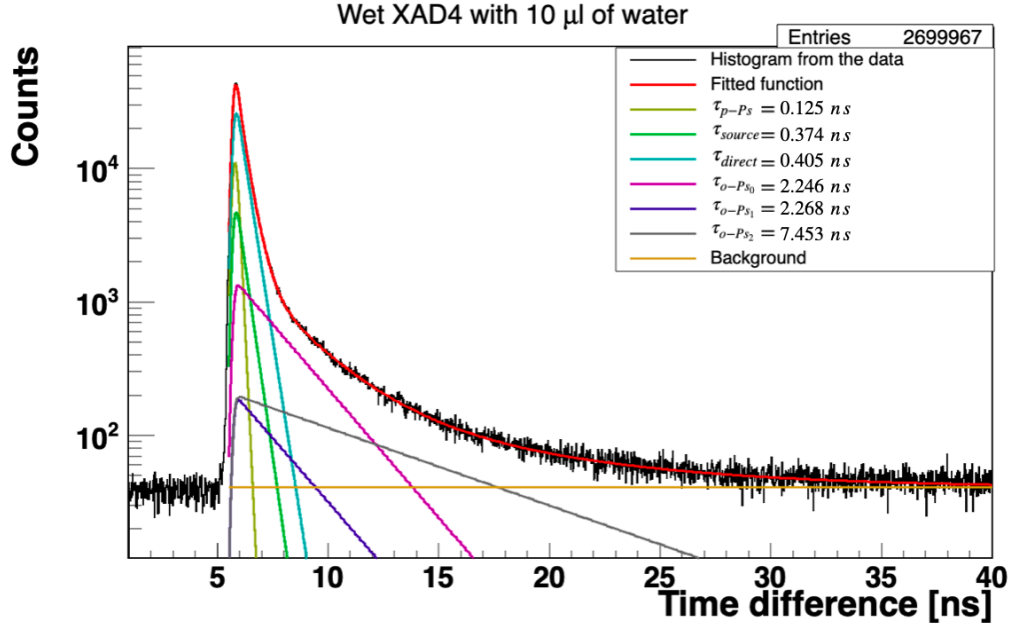


Figure 6.2: Fitted spectra of positronium lifetime in wet XAD4 with 10 ul water added to both parts of the chamber. Mass percentage of XAD4 in water: 43,6(1,0) %. Measurement was done using aluminium chamber.

Table 6.2: Lifetimes and intensities for components of the fit. Measurement was done using aluminium chamber. $*\chi^2_{\nu} = 1.24$

Component	Mean lifetime [ns]	Intensity [%]
τ_{p-Ps}	0.125	13.876(41)
τ_{source}	0.347	10
τ_{direct}	0.40502(66)	57.929(49)
τ_{o-Ps_0}	2.268	1.61
τ_{o-Ps_1}	2.246(12)	11.462(50)
τ_{o-Ps_2}	7.453(75)	5.123(50)

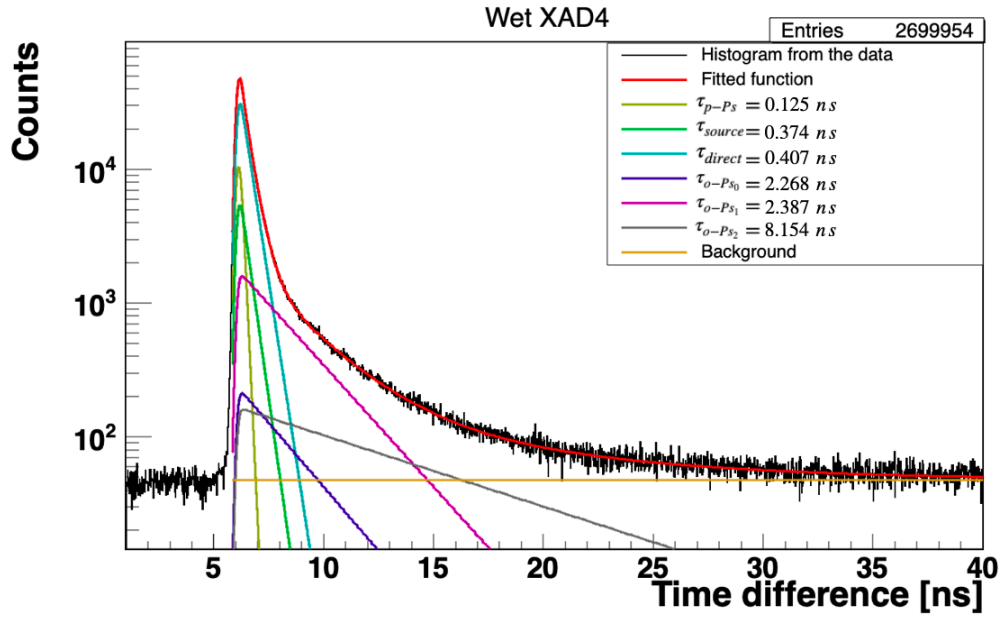


Figure 6.3: Fitted spectra of positronium lifetime in wet XAD4. Mass percentage of XAD4 in water: 46 %. Measurement was done using aluminium chamber.

Table 6.3: Lifetimes and intensities for components of the fit. Measurement was done using aluminium chamber. $*\chi^2_{\nu} = 1.26$

Component	Mean lifetime [ns]	Intensity [%]
τ_{p-Ps}	0.125	11.494(38)
τ_{source}	0.347	10
τ_{direct}	0.40660(60)	59.271(54)
τ_{o-Ps_0}	2.268	1.61
τ_{o-Ps_1}	2.387(10)	12.646(45)
τ_{o-Ps_2}	8.154(99)	4.002(45)

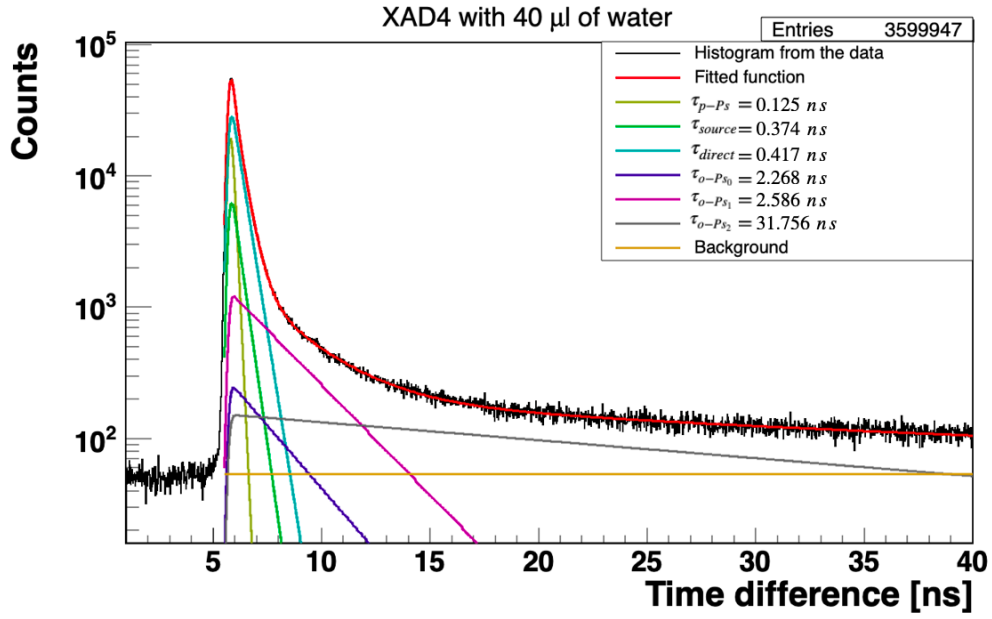


Figure 6.4: Fitted spectra of positronium lifetime in dried XAD4 with 40 μl of water. Mass percentage of XAD4 in water: 67,33(23) %. Measurement was done using PTFE chamber.

Table 6.4: Lifetimes and intensities for components of the fit. Measurement was done using PTFE chamber. $*\chi^2_{\nu} = 1.38$

Component	Mean lifetime [ns]	Intensity [%]
τ_{p-Ps}	0.125	18.164(39)
τ_{source}	0.347	10
τ_{direct}	0.41731(66)	48.856(45)
τ_{o-PS_0}	2.268	1.61
τ_{o-PS_1}	2.586(15)	8.924(39)
τ_{o-PS_2}	31.756(22)	12.447(39)

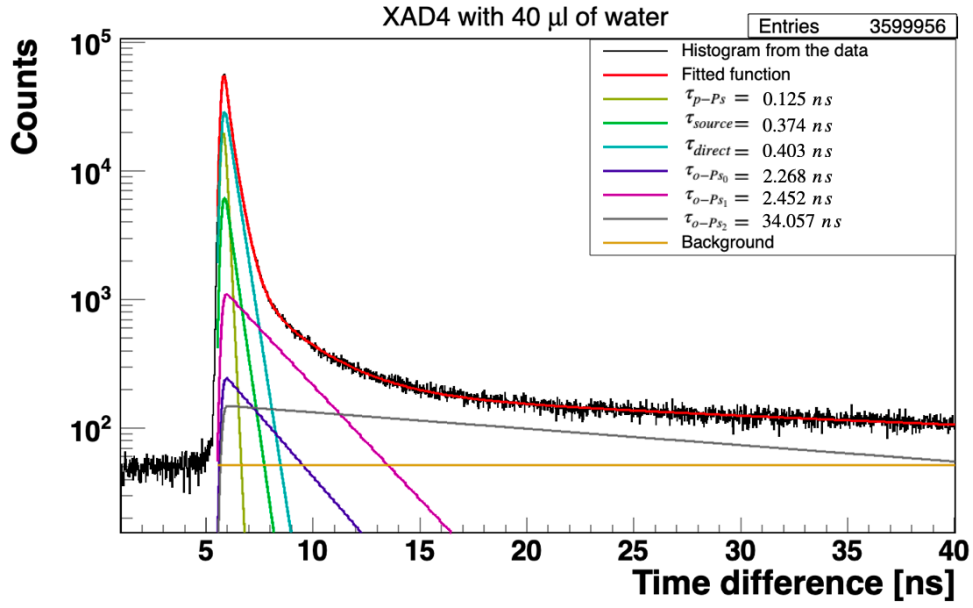


Figure 6.5: Fitted spectra of positronium lifetime in dried XAD4 with 40 μl of water. Mass percentage of XAD4 in water: 69,16(23) %. Measurement was done using aluminum chamber.

Table 6.5: Lifetimes and intensities for components of the fit. Measurement was done using aluminum chamber. $*\chi^2_{\nu} = 1.34$

Component	Mean lifetime [ns]	Intensity [%]
τ_{p-Ps}	0.125	18.834(39)
τ_{source}	0.347	10
τ_{direct}	0.40327(63)	48.619(45)
τ_{o-Ps_0}	2.268	1.61
τ_{o-Ps_1}	2.452(16)	7.767(38)
τ_{o-Ps_2}	34.06(22)	13.170(38)

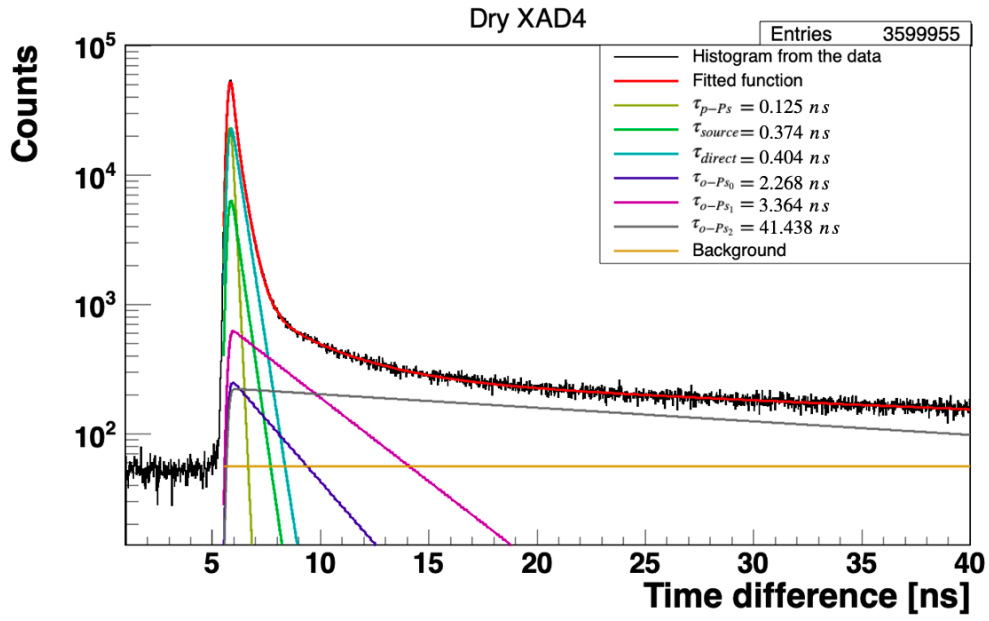


Figure 6.6: Fitted spectra of positronium lifetime in dried XAD4. Mass percentage of XAD4 in water: 100 %. Measurement was done using PTFE chamber.

Table 6.6: Lifetimes and intensities for components of the fit. Measurement was done using PTFE chamber. $*\chi^2_{\nu} = 1.34$

Component	Mean lifetime [ns]	Intensity [%]
τ_{p-PS}	0.125	21.300(40)
τ_{source}	0.347	10
τ_{direct}	0.40365(74)	37.879(46)
τ_{o-PS_0}	2.268	1.61
τ_{o-PS_1}	3.364(32)	5.744(40)
τ_{o-PS_2}	41.44(17)	23.467(40)

6.2 Benchmark measurements

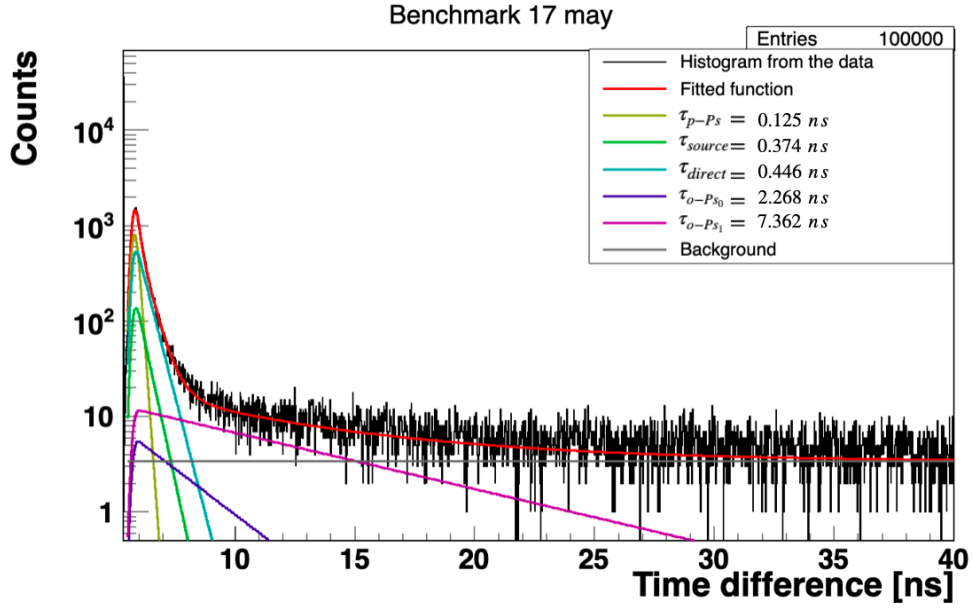


Figure 6.7: Fitted spectra of positronium lifetime in dried XAD4 benchmark measurement. Measurement was done using aluminium chamber.

Table 6.7: Lifetimes and intensities for components of the fit. Measurement was done using PTFE chamber. $*\chi^2_{\nu} = 1.19$

Component	Mean lifetime [ns]	Intensity [%]
τ_{p-Ps}	0.125	34.59(22)
τ_{source}	0.347	10
τ_{direct}	0.4456(51)	43.59(24)
τ_{o-Ps_0}	2.268	1.61
τ_{o-Ps_1}	7.36(30)	10.2(7.3)

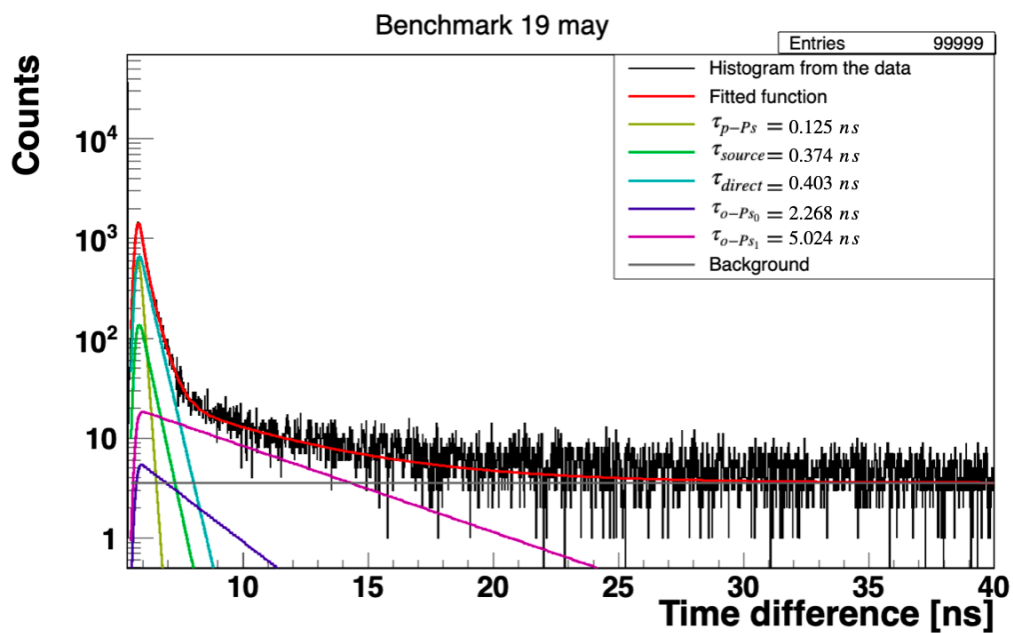


Figure 6.8: Fitted spectra of positronium lifetime in dried XAD4 benchmark measurement. Measurement was done using PTFE chamber.

Table 6.8: Lifetimes and intensities for components of the fit. Measurement was done using PTFE chamber. $*\chi^2_{\nu} = 1.20$

Component	Mean lifetime [ns]	Intensity [%]
τ_{p-Ps}	0.125	25.98(22)
τ_{source}	0.347	10
τ_{direct}	0.4028(42)	50.88(25)
τ_{o-Ps_0}	2.268	1.61
τ_{o-Ps_1}	5.02(18)	11.5(9.1)

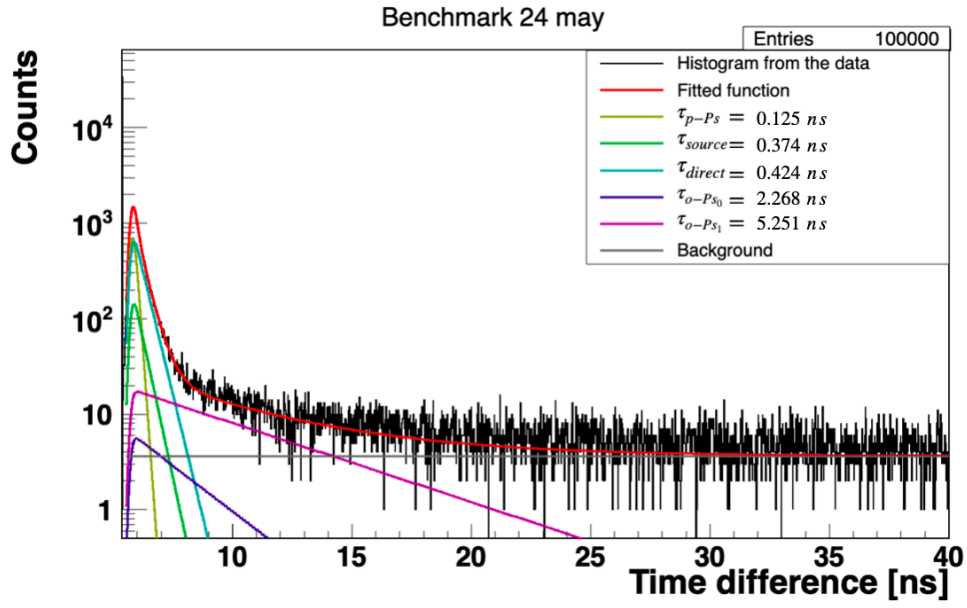


Figure 6.9: Fitted spectra of positronium lifetime in dried XAD4 benchmark measurement. Measurement was done using PTFE chamber.

Table 6.9: Lifetimes and intensities for components of the fit. Measurement was done using PTFE chamber. $*\chi^2_{\nu} = 1.25$

Component	Mean lifetime [ns]	Intensity [%]
τ_{p-Ps}	0.125	28.77(22)
τ_{source}	0.347	10
τ_{direct}	0.4235(44)	48.86(24)
τ_{o-Ps_0}	2.268	1.61
τ_{o-Ps_1}	5.25(20)	10.8(7.7)

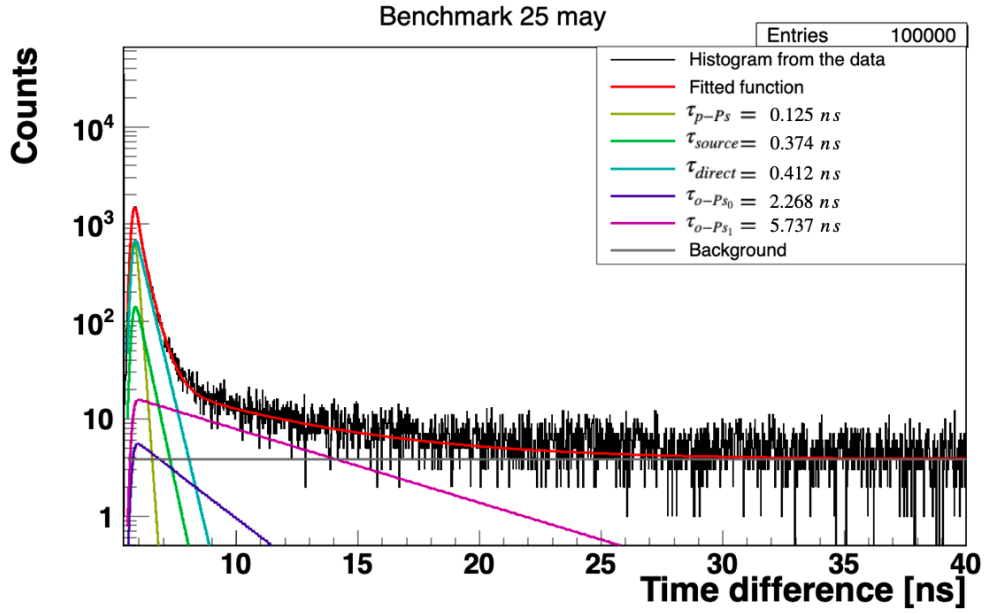


Figure 6.10: Fitted spectra of positronium lifetime in dried XAD4 benchmark measurement. Measurement was done using PTFE chamber.

Table 6.10: Lifetimes and intensities for components of the fit. Measurement was done using PTFE chamber. $*\chi^2_\nu = 1.05$

Component	Mean lifetime [ns]	Intensity [%]
τ_{p-Ps}	0.125	27.26(22)
τ_{source}	0.347	10
τ_{direct}	0.4118(42)	50.41(24)
τ_{o-Ps_0}	2.268	1.61
τ_{o-Ps_1}	5.74(22)	10.7(8.3)

$$*\chi^2_\nu = \chi^2 / \text{Degrees of Freedom}$$