PALS – Setup optimisation and application to macromolecular materials characterisation

Instituto Superior Técnico // Mestrado Integrado em Engenharia Física Tecnológica

Project MEFT

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Introduction

- The free volume structure inside a material greatly influences its physical, chemical and mechanical properties. It can be probed with the ***Positron Annihilation Lifetime Spectroscopy (PALS)*** technique.

- PALS measures the time elapsed between the implantation of positrons into the material being studied and the detection of the γ-rays emitted in a positron-electron annihilation.

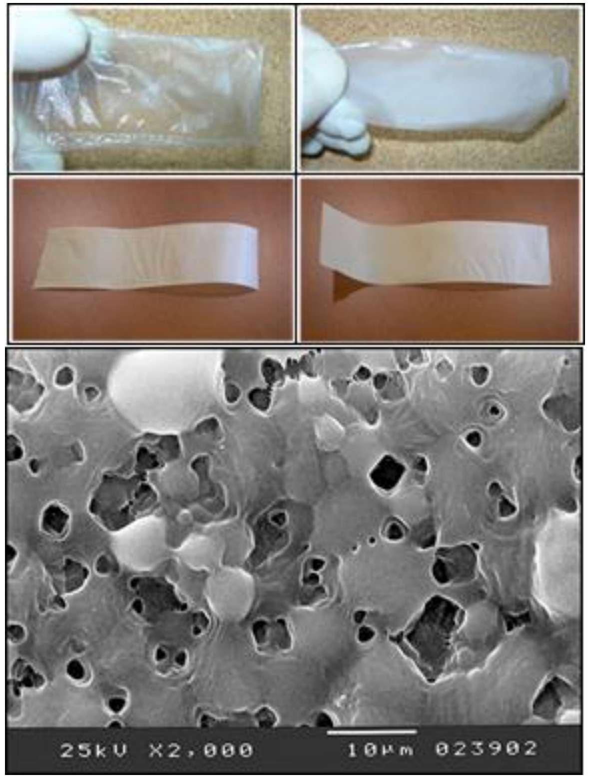
- Being a function of the material's electron density, the determination of the lifetime of the positrons injected into the material allows us to estimate the free volumes size, concentration and distribution inside it.

- PALS has four great advantages:
  - It is a non-destructive technique;
  - It provides a complete study of the free volume structure;
  - It can probe both near-surface and in-depth regions;
  - A PALS spectrometer can operate unattended and for long periods of time.

Objectives

- A PALS spectrometer will be optimised and then applied to the study of the morphology and structure of radiation processed polymer-based and hybrid materials.

- These materials have been developed by the research group, mainly for biomedical applications, such as human tissue repair and conservation of stone-based cultural heritage artefacts.

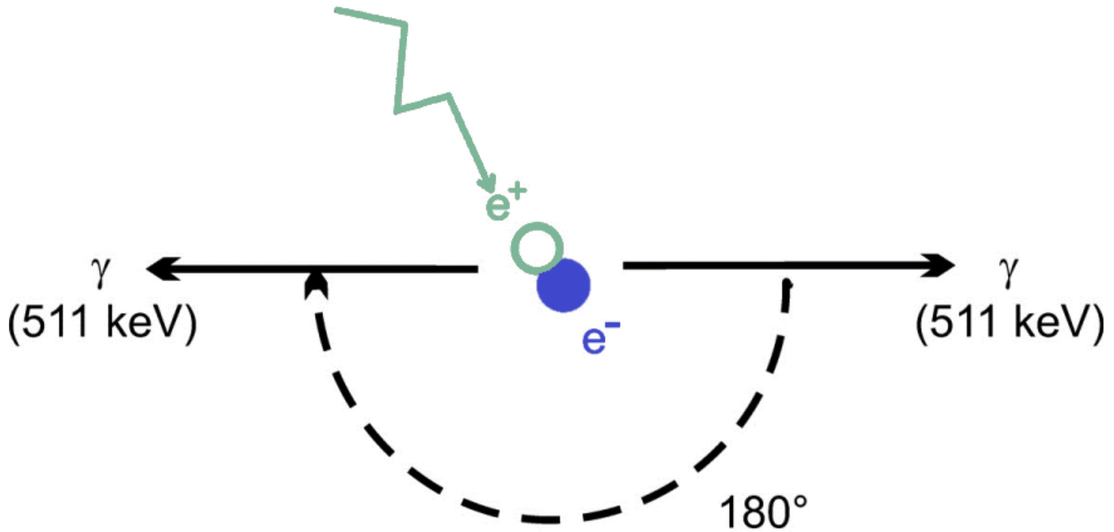


Source: [1]

Source: [1]

Physics background - Positrons in solids

- When injected into a solid, positrons will:
  - Thermalise, by electron ionisation, electron-hole excitations and phonon scattering;
  - Diffuse, with a three-dimensional random walk through the material;
  - Annihilate with an electron in the material.



Adapted from [2]

We will analyse positron-electron annihilation in materials with

High electron density

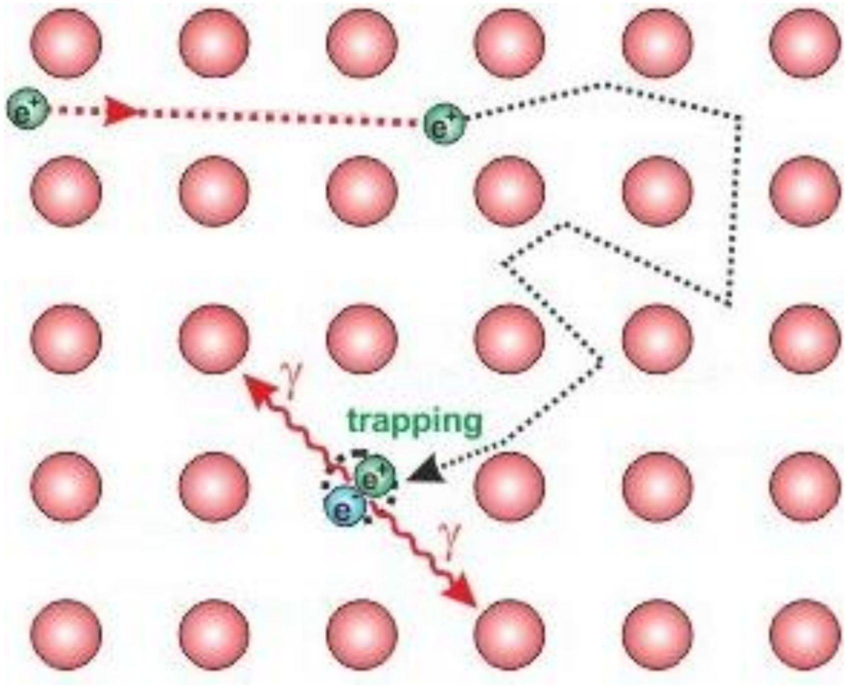
Low electron density

Materials with a high electron density

- Positrons see defects in these materials as strongly attractive sites, due to the locally reduced atomic and electron density.

- The transition of a positron from a free state to a trapped state is called ***positron trapping***.

- In the trapped state, the positrons live longer, as they have more difficulty in finding an electron to annihilate with.



Adapted from [3]

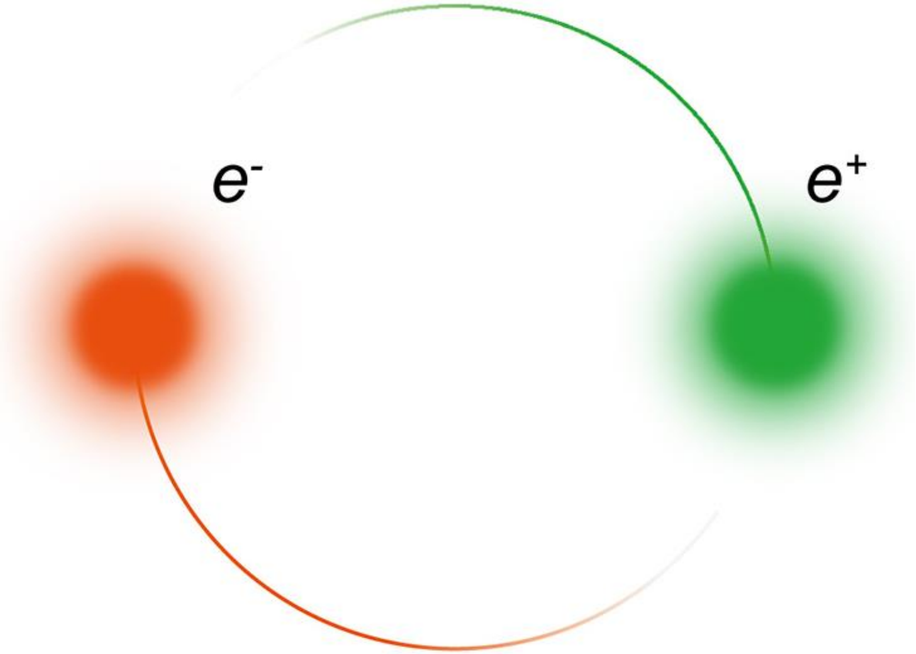
- The existence of positron trapping can reveal:
  - **The concentration of defects in the material;**
  - **The size and internal electronic structure of these defects.**

Materials with a low electron density

- In these materials, positrons can annihilate in the free state, the trapped state or as a ***positronium (Ps)*** atom. Ps is an unstable bound state of the positron-electron pair.

Ps can exist as

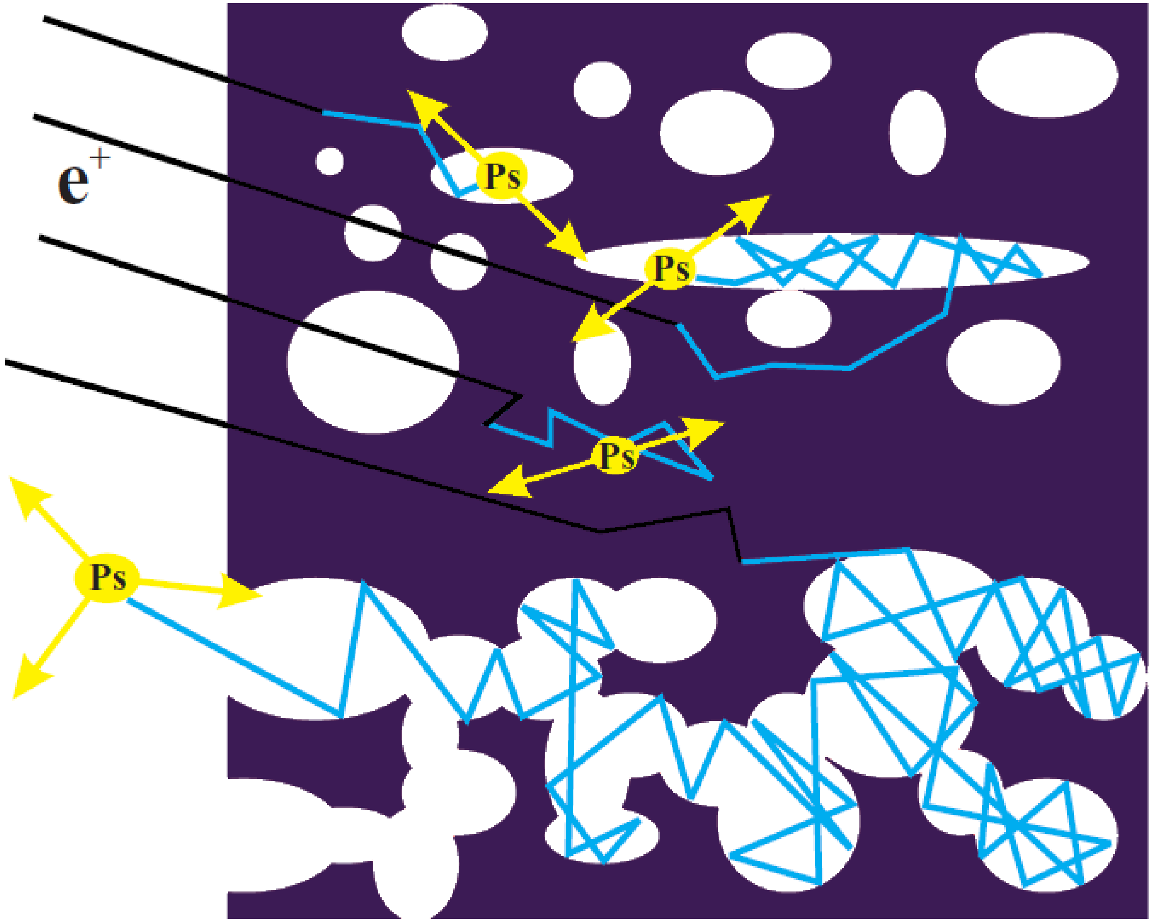
Para-positronium (p-Ps)	Ortho-positronium (o-Ps)
Lifetime of 125 ps in vacuum	Lifetime of 142 ns in vacuum
Decays into 2 γ-rays with energy equal to 511 keV	Decays into 3 γ-rays with energies less than 511 keV



Source: [4]

Materials with a low electron density

- In materials with free volumes, Ps atoms are attracted to these areas, due to a lower nuclear and electron charge density.
- In there, ***"pick-off" annihilation*** can occur: the positron does not annihilate with its bound electron, but instead with an electron from the walls, in a 2 γ-rays emission process.
- The annihilation probability depends on the void size. In smaller voids, more Ps-wall collisions will take place, which results in a faster "pick-off" annihilation.



Adapted from [5]

Annihilation processes

Positron state	Process	Characteristic time scale
Free e^+	2γ self-annihilation	0,1 – 0,4 ns
Trapped e^+	2γ self-annihilation	0,2 – 0,5 ns
p-Ps	2γ self-annihilation	125 ps
	2γ pick-off annihilation	> 1 ns
o-Ps	3γ self-annihilation	142 ns
	2γ pick-off annihilation	> 1 ns

High electron density materials (Free e^+, Trapped e^+)

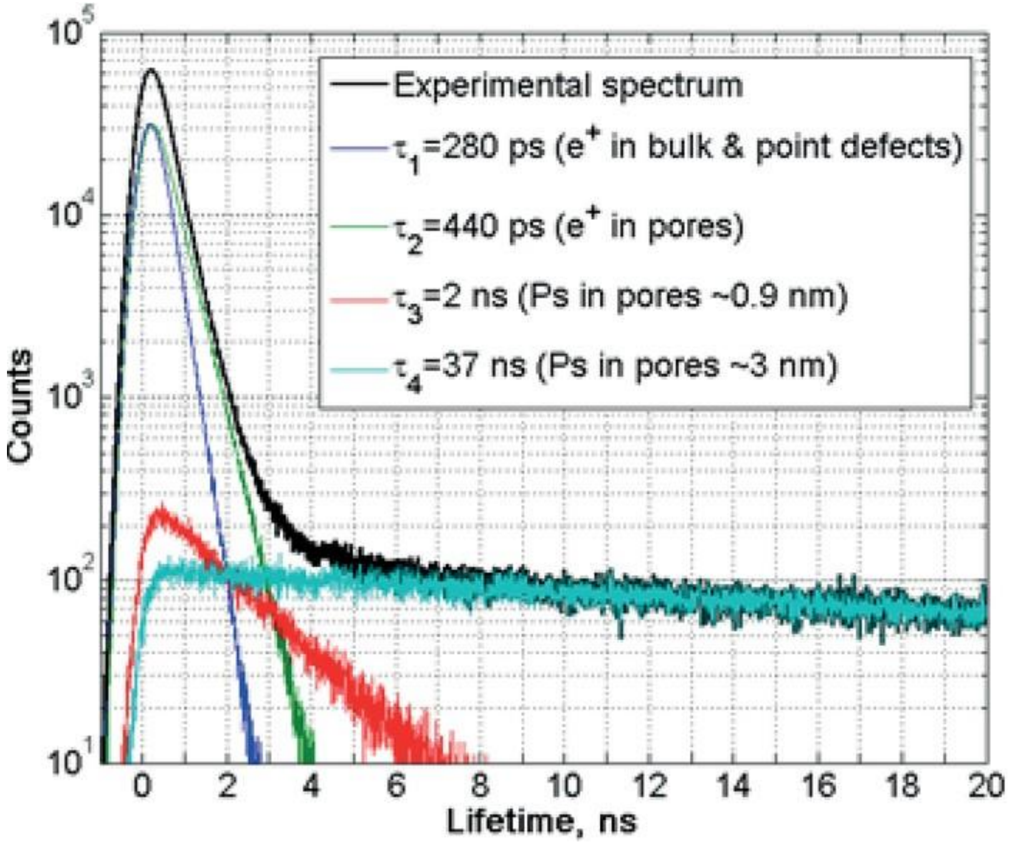
Low electron density materials (all states)

Dependent on the free volume structure (Trapped e^+ 2γ self-annihilation; p-Ps and o-Ps 2γ pick-off annihilation)

PALS technique

- In the PALS technique, ***the lifetime of positrons is measured*** as the elapsed time between their implantation into the sample being studied and the detection of any γ-ray resultant from their annihilation.

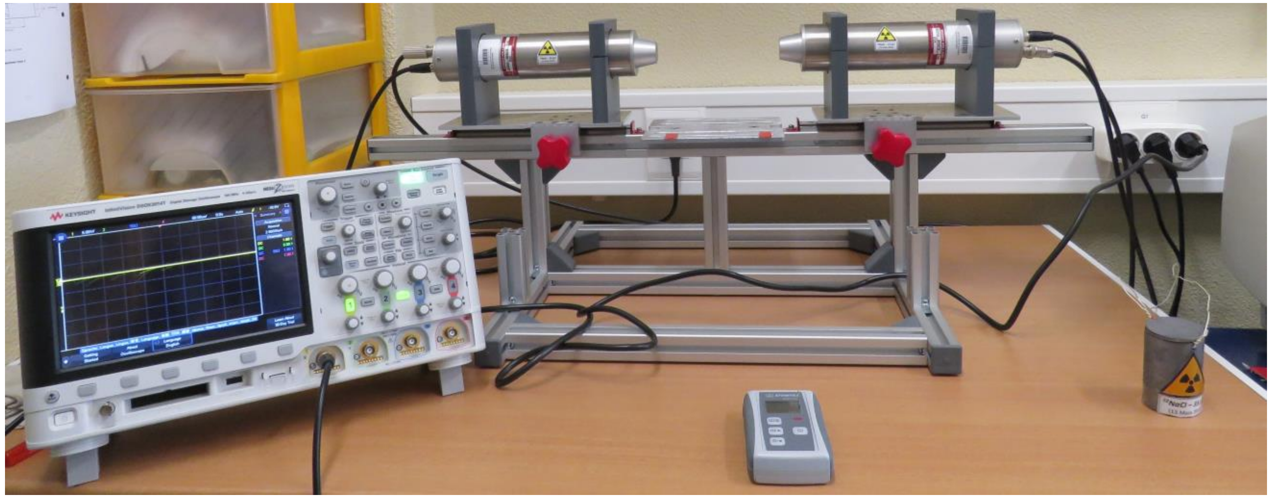
- After some detections, a ***positron lifetime spectrum*** starts to be drawn that contains several ***exponentially decaying lifetime components***, corresponding to different annihilation processes, together with a background, both convoluted with a time resolution function of the detectors and electronics.

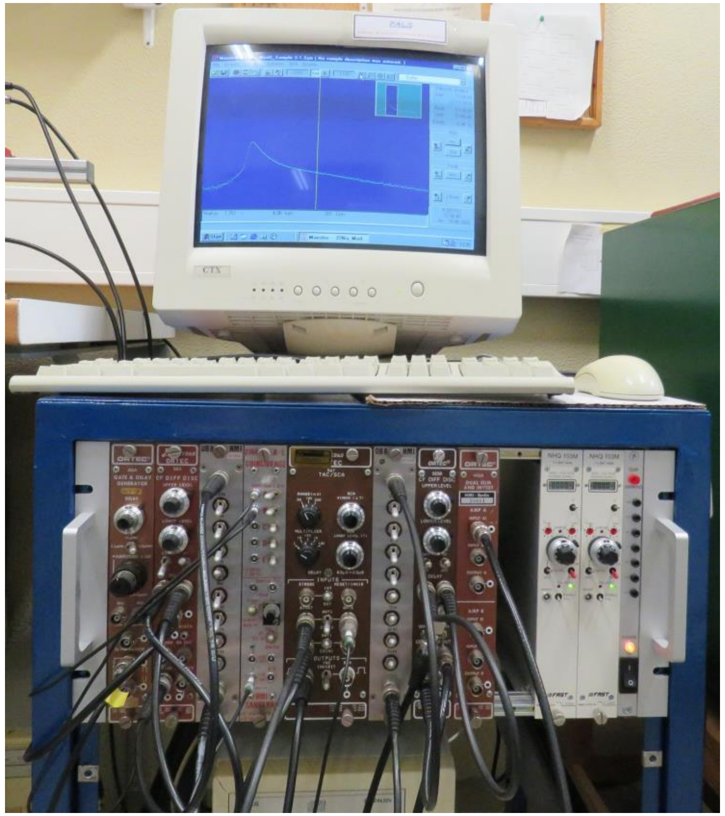


Source: [6]

PALS technique

- To perform a PALS analysis, it's necessary to have:
  - A source of positrons;
  - An experimental setup, for the detection of the positrons' lifetimes;

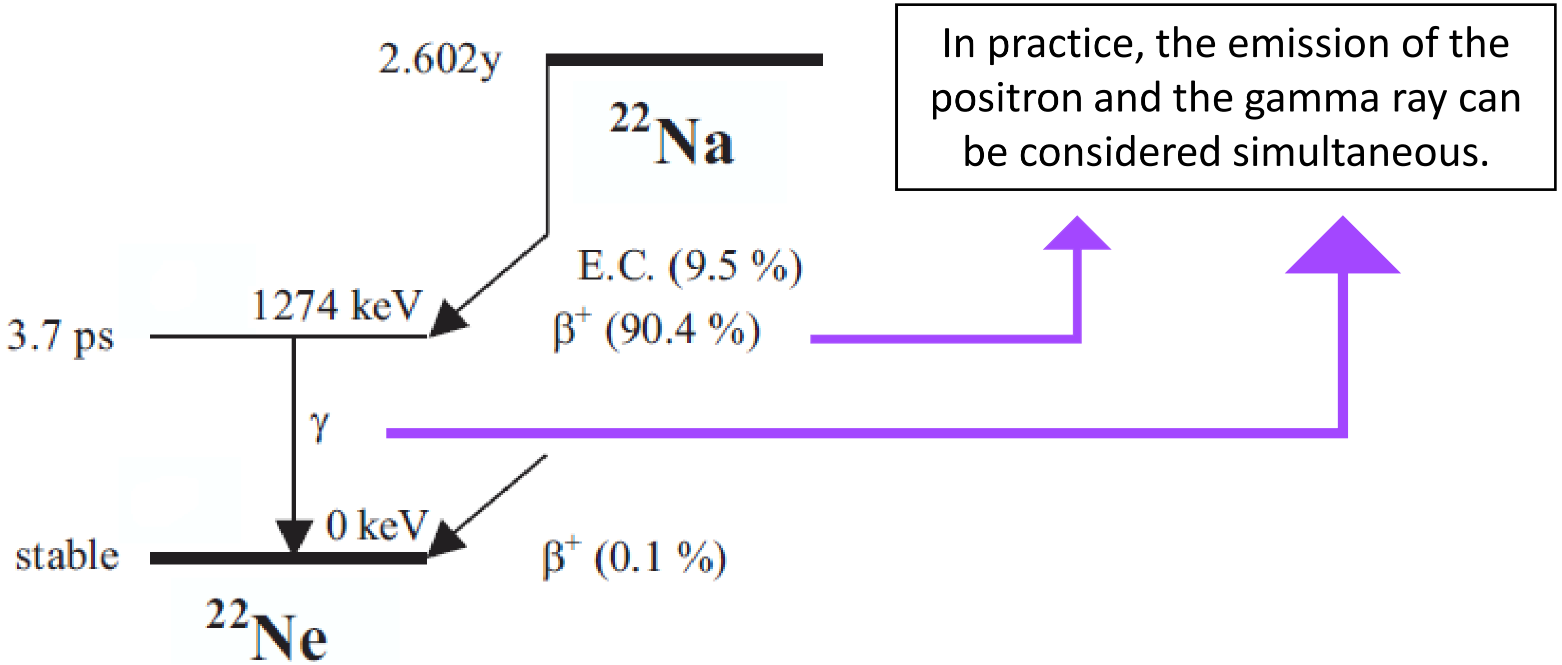




Positrons source

- In this work, a $^{22}\boldsymbol{Na}$ radioactive source will be used as the positrons source. It is easy to handle and substitute and has a suitable half-life.

- ^{22}Na decay scheme:



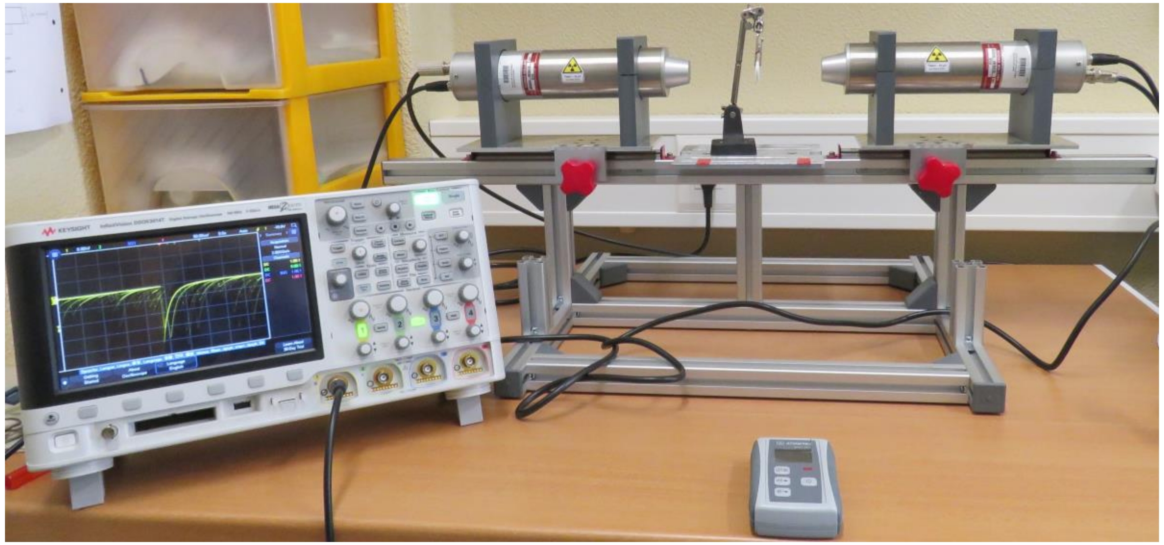
Adapted from [7]

Experimental setup

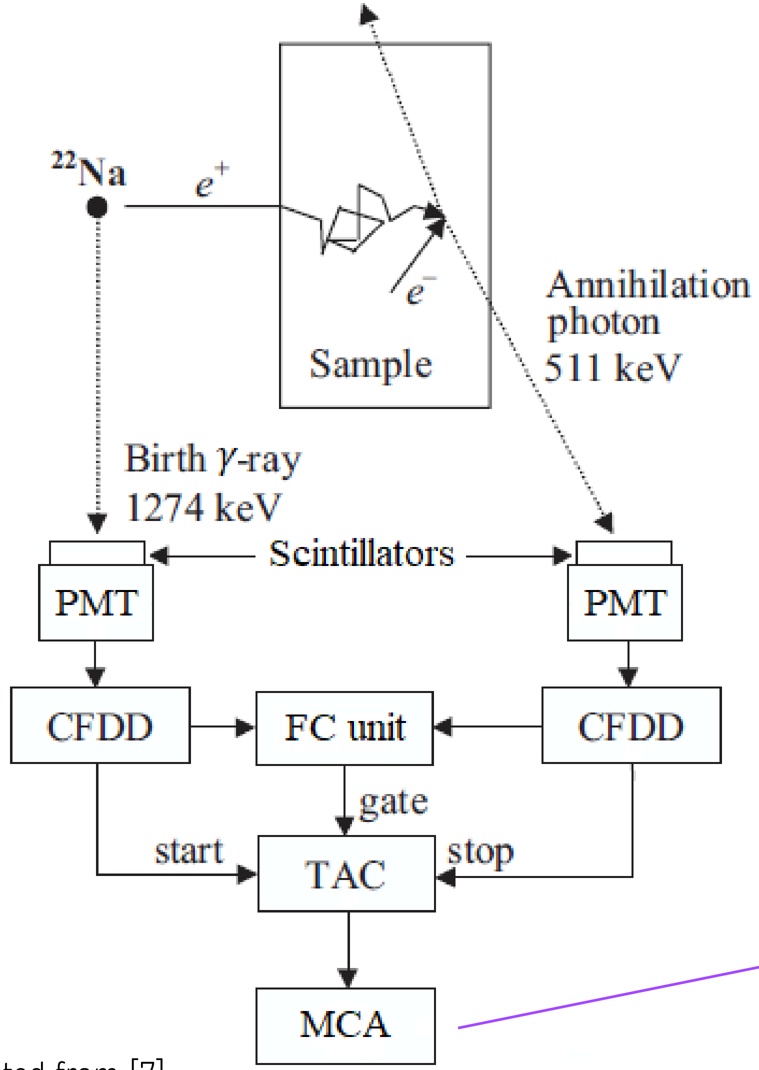
- The two fast scintillators detect

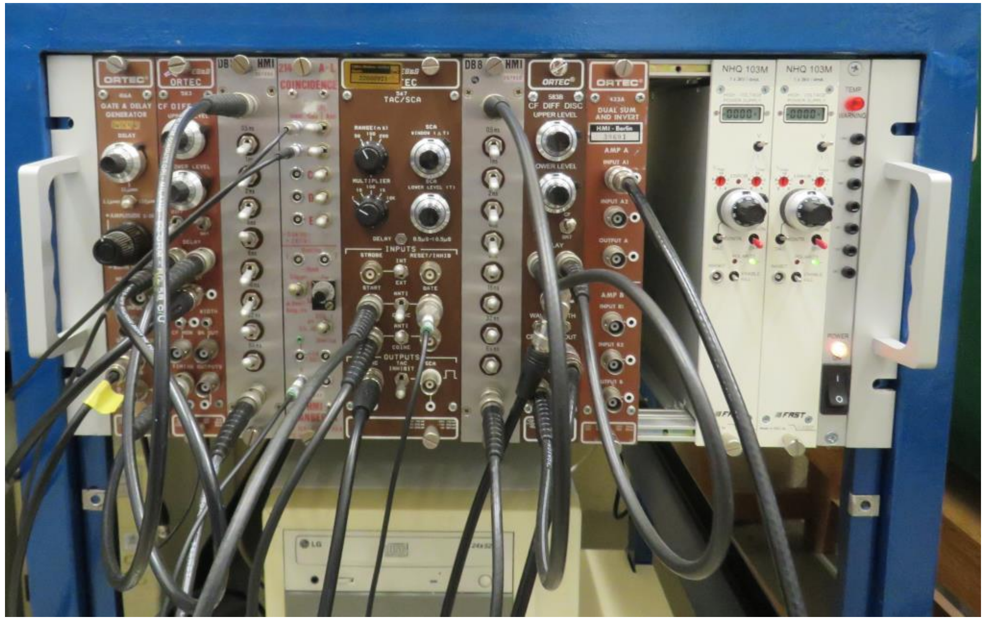
Start signal	Stop signal
↓ corresponds to	↓ corresponds to
Detection of the 1.274 MeV γ-ray from the de-excitation of ^{22}Ne	Detection of a 511 keV γ-ray resultant from an annihilation
↓ associated with	↓ associated with
Implantation of the positron into the material	Positron annihilation in the material

The time difference between the two signals gives the ***positron lifetime***.



Experimental setup





Positron lifetime spectrum: number of annihilation events of the implanted positrons versus their lifetime.

Adapted from [7]

Work plan

Task	Month					
	1	2	3	4	5	6
Familiarise with the principles of operation of PALS system: acquire energy spectra of ^{60}Co and ^{22}Na sources; spectra time calibration	■					
Acquire PALS spectra for reference materials, with careful choice of parameters to include on the software for data reduction	■	■				
Revise and improve the fast coincidence electronic system for PALS. Repeat and compare new obtained spectra with the ones obtained in Task 2			■			
Application on the characterisation of macromolecular materials				■	■	
Thesis writing and scientific work dissemination					■	■

References

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[5] David W. Gidley, Hua-Gen Peng, and Richard S. Vallery. “Positron Annihilation as a Method to Characterize Porous Materials”. Annual Review of Materials Research 36.1 (2006), pp. 49-79. DOI:10.1146/annurev.matsci.36.111904.135144.

[6] Yu. A. Akmalova et al. “Positron Lifetime Spectroscopy of Silicon Nanocontainers for Cancer TheranosticApplications”. KnE Energy 3.2 (Apr. 2018), 1–9. DOI:10.18502/ken.v3i2.1784.

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