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Positron Annihilation – A Novel Tool for Porosimetry in the nm-Range for Closed and Open-Pore Systems

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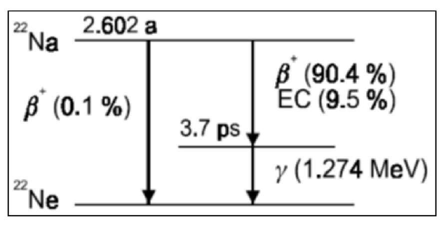
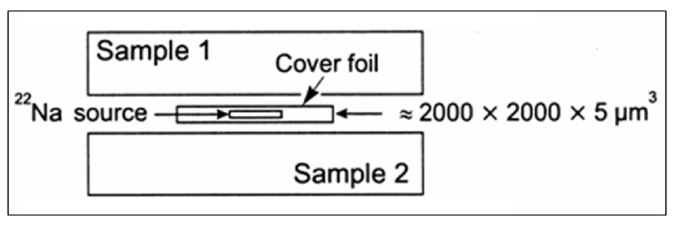
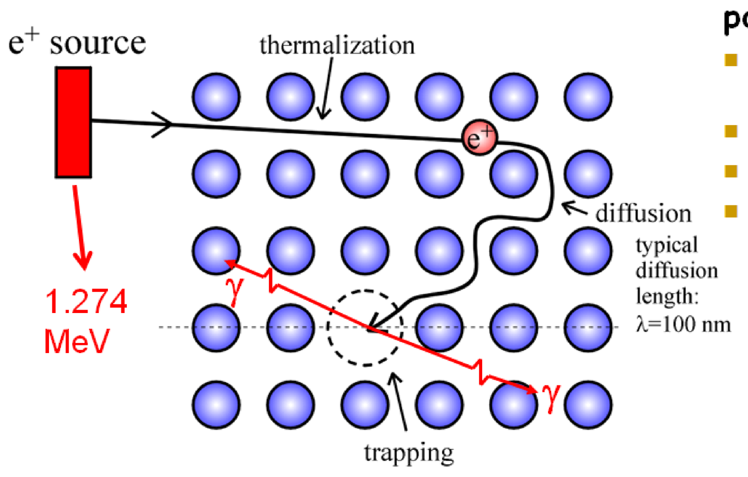
Positron annihilation is used for decades for the study of open-volume lattice defects, such as vacancies, voids, dislocations, and grain boundaries. A positron and an electron can also form **positronium** (Ps). It exist in two states: the singlet state (para-Ps; antiparallel spin) annihilates with a lifetime of 125 ps, independent of the ambient host material. In the triplet state (ortho-Ps; parallel spin) the lifetime in vacuum is 142 ns. When o-Ps is formed in a **porous material**, the lifetime is shortened by the **pick-off annihilation process**. Here, the positron picks up an electron from the pore wall material with antiparallel spin and forms, thus, p-Ps which annihilates almost instantaneously. This process depends on the number of wall contacts, and therefore, the **o-Ps lifetime is a direct measure of the pore size**. The pore size distribution can also be determined. The method is non-destructive and can be used for open and closed pore systems. **The sensitivity limits are 0.2 … 40 nm.**







positrons:

- thermalize (reach thermal energies)
- diffuse
- being trapped
- and annihilate

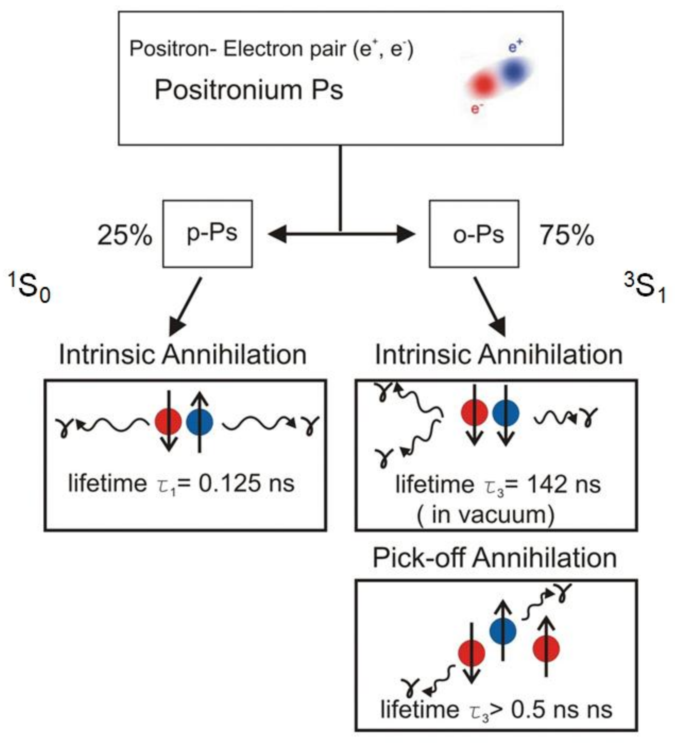
When trapped in vacancies:

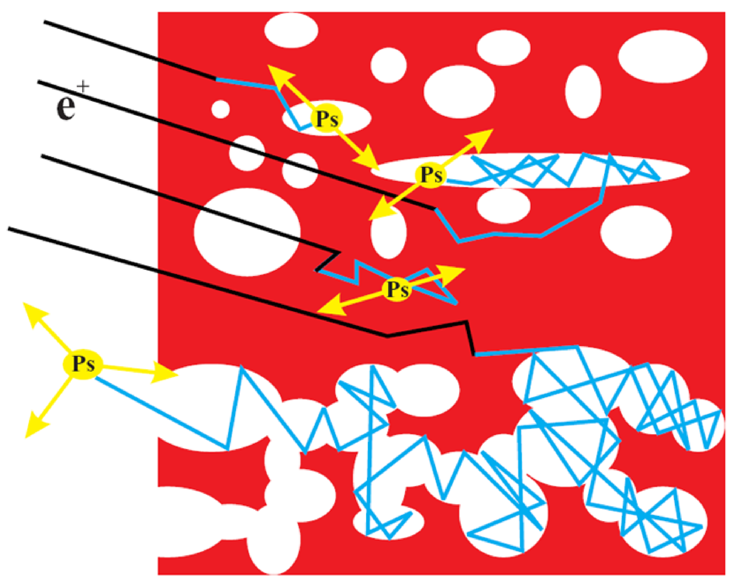
- Lifetime increases due to smaller electron density in open volume

Positrons can be obtained from radioactive ^{22}Na (2.6 a half life). The positrons thermalize and diffuse a short path through the sample material. The lifetime can be measured as time difference between the 1.274 MeV quantum (β^+ decay) and the 0.511 MeV quantum (positron annihilation).

Positronium formation

- in materials without free electrons
- polymers, glass, liquids, gases, …
- Positronium is formed
- Lightest atom: bound state between electron and positron
- Ortho-Ps lifetime is measure for open volume
- Pick-off annihilation

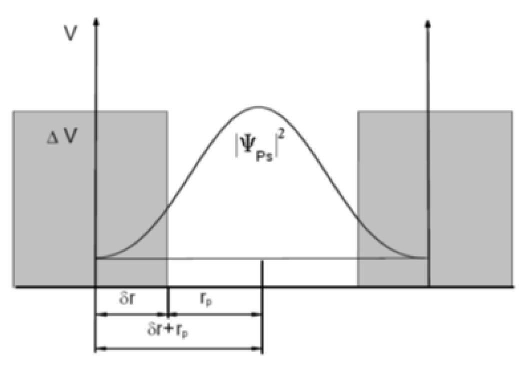




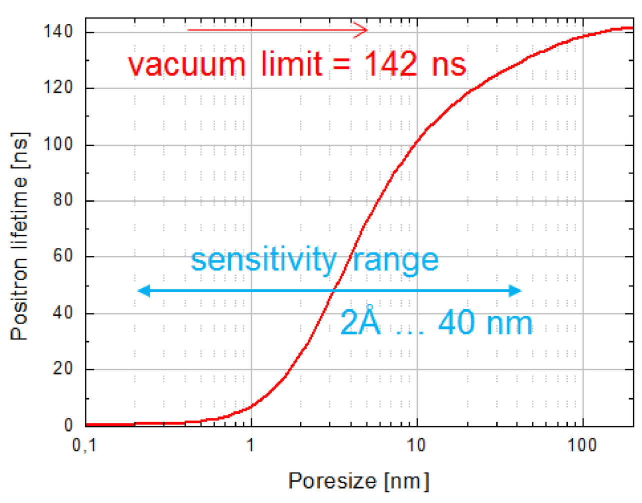
pick-off annihilation:

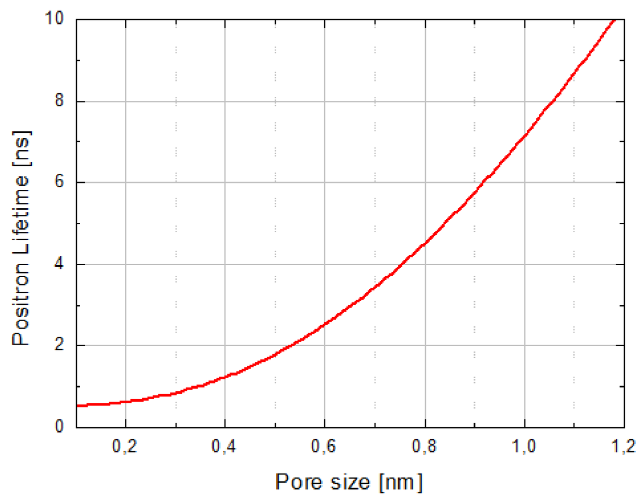
- o-Ps is converted to p-Ps by capturing an electron with anti-parallel spin
- happens during collisions at walls of pore
- lifetime decreases rapidly
- lifetime is function of pore size 0.5 ns … 142 ns
- lifetime can be extracted from spectra
- Also in closed pore systems

o-Ps lifetime = f(pore size) ⇨ quantum-mechanical models

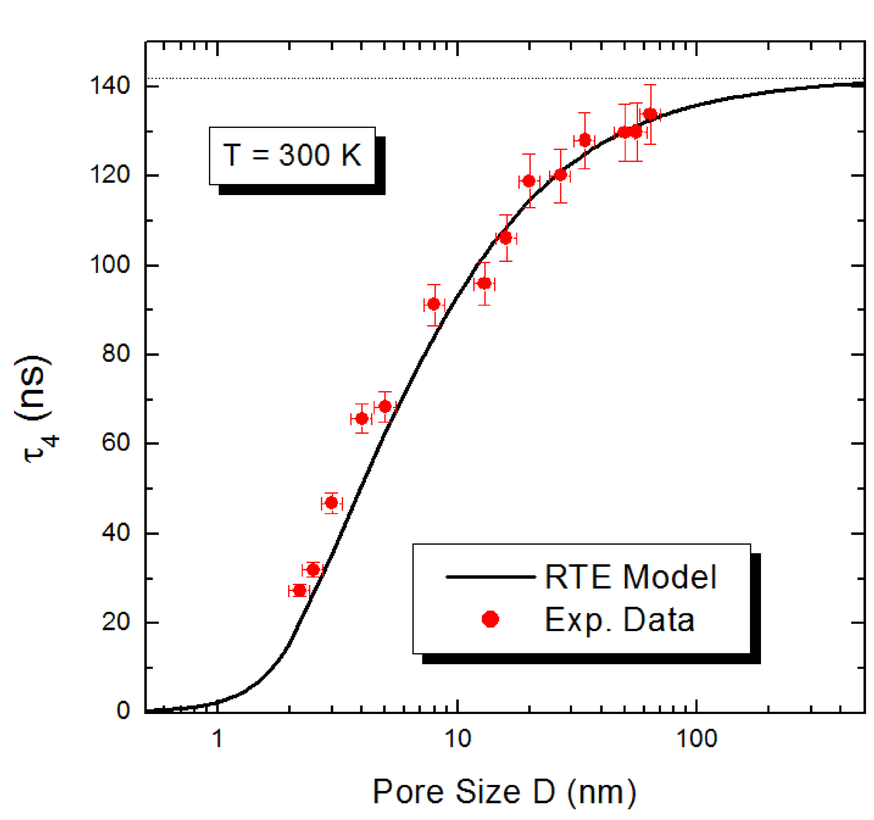


- Tao-Eldrup-Model
  - acceptable for pore size up to 2 nm in diameter
- extended Tao-Eldrup-Model
  - acceptable for pore size up to 100 nm in diameter



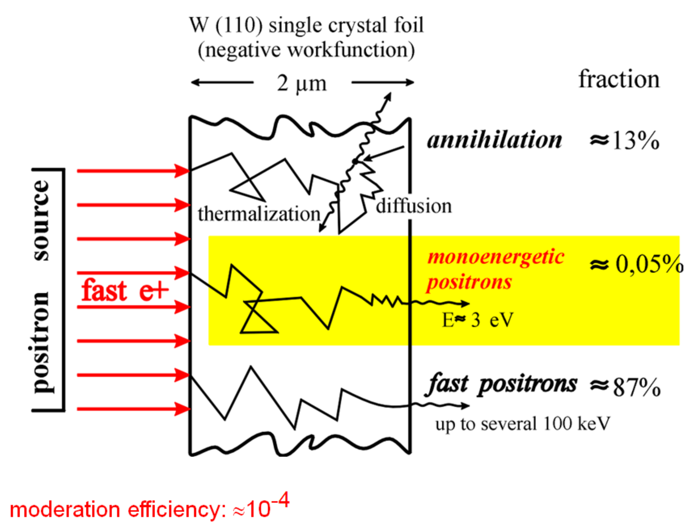


Controlled Pore Glass



- we measured porous glass in a broad pore size range
- pore size obtained by LN_2-adsorption method
- for T=300 K general agreement to the RTE model
- calibration curve for the correlation of o-Ps lifetime and pore size

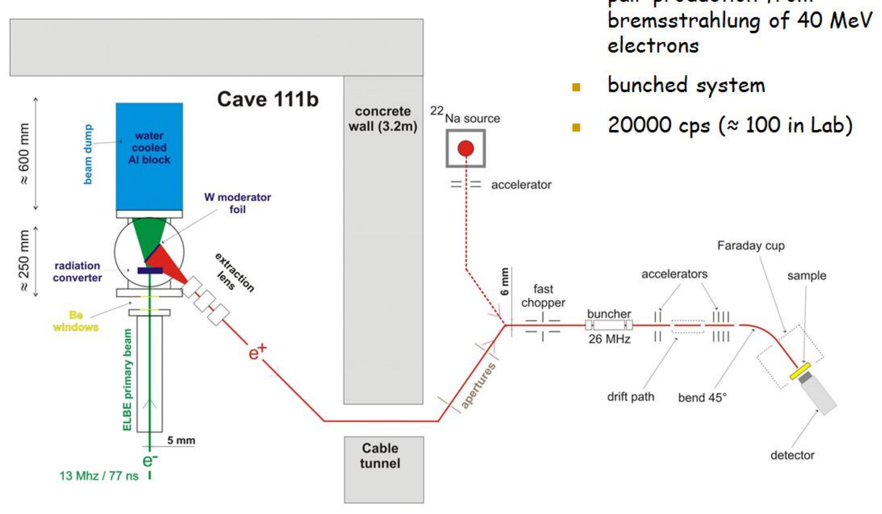
Moderation of positrons for study of thin layers (< 1 µm)

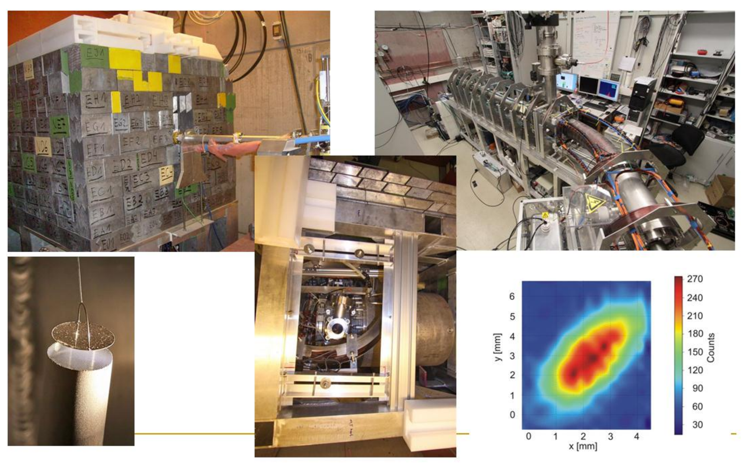


Positrons of ^{22}Na sources have a high energy (up to 540 keV) and penetrate as deep as 1mm into the sample. Thus, they are not useful for the study of thin layers of < 1 µm. However, they can be moderated to form a monoenergetic positron beam of a few keV which can be accelerated to obtain depth profiles of the layer.

Intense mono-energetic positron beam MePS at ELBE / HZDR in Rossendorf

- positrons are generated by pair production from bremsstrahlung of 40 MeV electrons
- bunched system
- 20000 cps (≈ 100 in Lab)



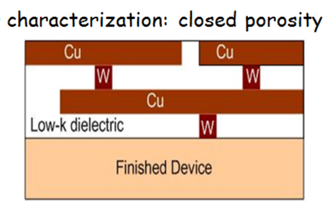


Study of thin low-K layers by the MePS system at ELBE / HZDR

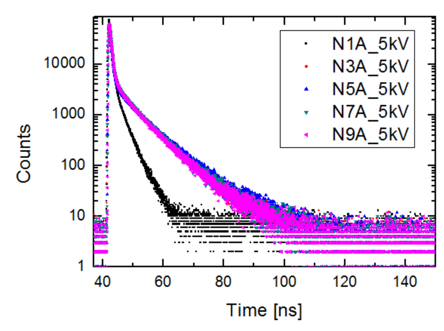
- modern ultra-large scale microprocessors suffers from long relaxation times
- information transport is limited by product R×C
- R has been decreased: Copper technology (instead of Al)
- C is relatively high when SiO_2 is used as isolation layer; ε_r=4

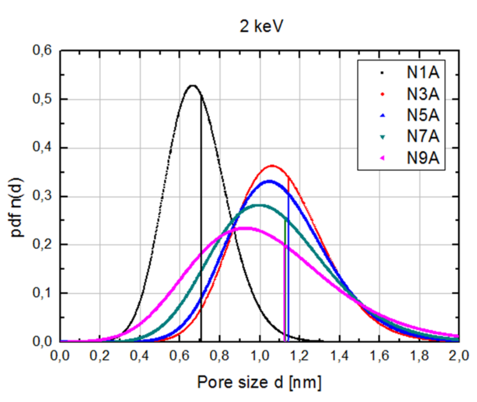
$$C = \frac{\varepsilon_0 \varepsilon_r A}{d}$$

- low-k (small ε_r = 2…2.5) layers may help
- these are layers with micropores with pore size of d≈1 nm with high porosity
- problem for characterization: closed porosity

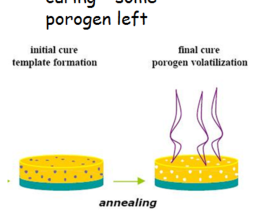


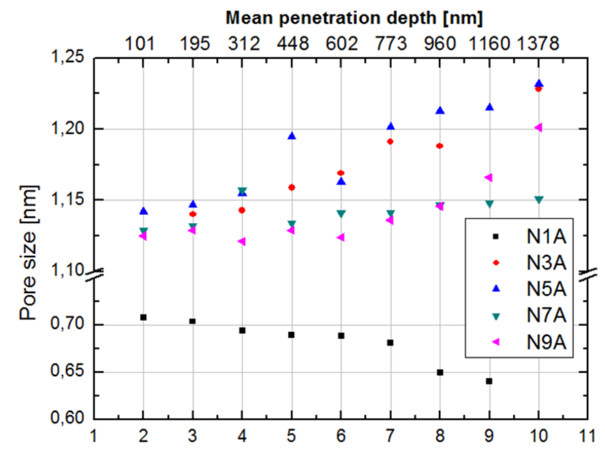
- measurements after different treatments of the layer
- lifetime spectra from 5 samples measured at the same positron energy of 5 keV = 350 nm depth





- dispersion of lifetime gives the size distribution of the pore system
- N1A: incomplete curing – some porogen left





- monoenergetic positrons can be used to depth scar the layer
- monoenergetic positrons are obtained by moderation

Poster is available at http://positron.physik.uni-halle.de
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