

# Development of a Triple-Coincidence Positron Lifetime Spectrometer for Nuclear Materials Research

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## ABSTRACT

In this paper, the development of a new positron lifetime spectrometer dedicated to high radioactive materials such as nuclear power plant's mixed oxide fuels and structural steels is discussed. In the case of nuclear fuels, the behaviour of nanoscale defects, which are mainly created by the high radioactivity and high irradiation flux of neutrons and fission products is of primer importance to control the evolution of the fuel properties from its synthesis to its long term depository after its use in reactor. The deterioration of mechanical strength of structural components under high thermal and radiation fluxes is dependent on the microstructural evolution of radiation damage. A new kind of a lifetime spectrometer is required in order to reduce unwanted effects arising from the excess gamma radiation of the high-active nuclear fuel samples.

## 1 INTRODUCTION

A good understanding of nuclear fuel behaviour during normal in-reactor operation, under accident conditions and during long term storage after irradiation, is of great interest for safe and economic nuclear energy production. The fuel properties strongly depend on the chemistry of fission products and actinides in the fuel matrix. In particular, the nature and concentration of point defects are key parameters to establish accurate thermodynamics modelling but yet incompletely understood. Also mechanical strength of structural components under nuclear reactor conditions depends strongly on microstructural defects.

Positron lifetime spectroscopy offers means to study vacancy-type defects that are the primary radiation damage that eventually evolves into larger voids and dislocation loops that are detectable by modern electron microscopy. The method is based on the trapping of positrons in the matter, which is favoured in the presence of empty volume such as point defects or nanovoids [1]. Positrons are produced, for example, via spontaneous  $\beta^+$ -decay of radioactive elements. The radioactive isotope  $^{22}\text{Na}$  is widely used as a positron source due to its practical half-life of 2.6 years. The  $^{22}\text{Na}$  decay produces a neutrino and a positron with an energy between 0 and 540 keV, resulting in less than a hundred

micrometres probing depth for a positron in heavy material such as nuclear fuel. After the decay, the daughter  $^{22}\text{Ne}$  nucleus is left in the excited state that relaxes essentially immediately via 1.27 MeV gamma photon emission (cf. figure 1(a)). In positron lifetime spectroscopy, this photon is used as an indication of positron emission or as a start signal. When a positron annihilates with an electron, the mass of the two particles transforms into two 511 keV gamma photons propagating to opposite directions. These gamma photons can be used as stop pulses. Measurement of the time differences between the detected start and successive stop signals, and collecting sufficient statistics of such events, results in a histogram like those shown in figure 2. This positron lifetime spectrum is analysed as a sum of exponential decay components

$$N(t) = \sum_{i=1}^k I_i \exp(-\lambda_i t), \quad (1)$$

where  $k$  is the number of different annihilation states observed,  $\lambda_i$  is the annihilation rate in each state, and  $I_i$  is the intensity of the observed components. The positron lifetime for each state is defined as the inverse of the annihilation rate  $\tau_i = 1/\lambda_i$ . Typical lifetimes in solids are in the range 100 ... 400 ps, depending on whether the positron annihilates from

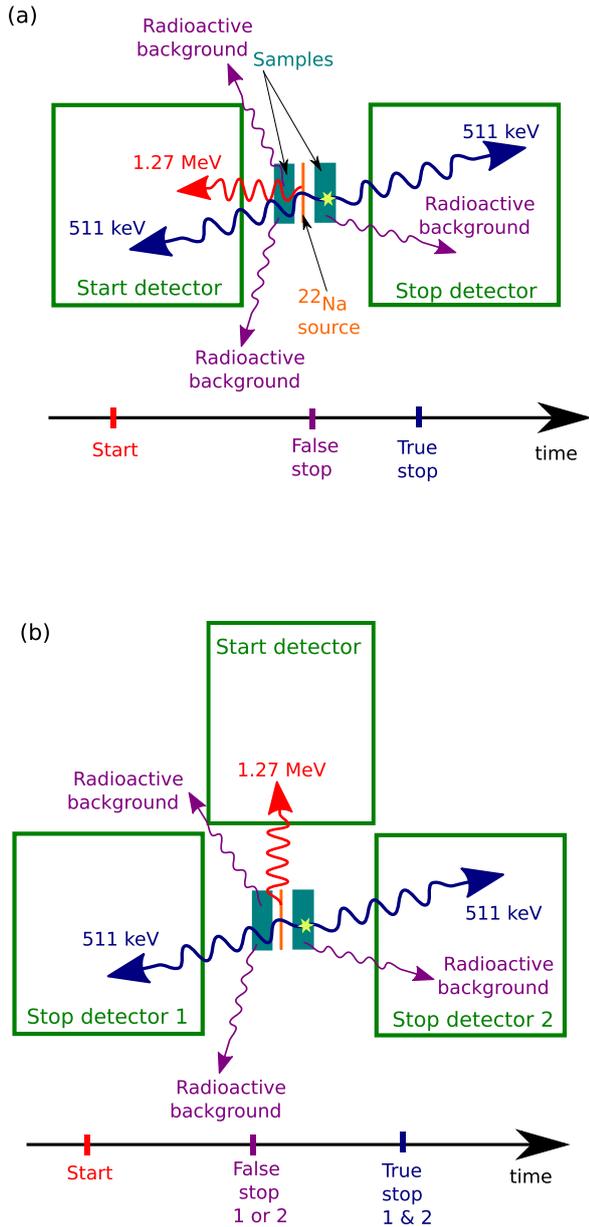


Figure 1.(a) Principle of the standard two-detector coincidence set-up in case of highly radioactive samples. (b) Principle of the proposed three-detector coincidence setup for radioactive materials research.

the delocalized state in the crystal lattice or as trapped at an open volume defect. Different vacancy defects produce different lifetime components, as a rule the larger the open volume the longer the lifetime. An important parameter when applying this method is the average lifetime  $\tau_{ave} = \sum_i I_i \tau_i$  that coincides with the center of mass of the spectrum and is very stable and accurate with experimental uncertainty well below 1 ps. References [1, 2] are suggested further reading for details about the technique in studies of solids. Positron lifetime spectroscopy has been successfully applied as a non-destructive technique to studying defect

behaviour in non-irradiated nuclear fuel  $UO_2$  [3,4] and structural steels [5,6,7].

Even though nuclear fuel materials' decay chains do not contain  $\beta^+$ -active elements [8], the successive coupled gamma photon emissions will be recorded in the lifetime spectrum as false lifetime events [9]. To overcome the above challenge, one can add a third detector to discriminate the wrong stop events from the true events (cf. figure 1(b)). The third detector should be placed on the opposite side of the other stop detector as illustrated in figure 1(b), as annihilation gamma photons propagate in opposite directions. On the contrary, no angular correlation has been observed in the radioactive decays of elements present in the materials to be studied. Such approach is currently under development to allow highly radioactive nuclear material research.

In this paper, we introduce the problematics related to positron lifetime measurements of medium-to-high-active nuclear materials and suggest means to construct a feasible three-detector measurement setup.

## 2 EXPERIMENTAL METHODS

### 2.1 Research objective

Currently, positron lifetime experiments with two detectors can be performed only for low-activity materials (activity below 100 kBq), which is very low considering the activity of more than 10 MBq observed in typical mixed oxide samples or steel activated in a nuclear reactor. The main objective is to design and test a new type of an experimental positron lifetime setup that would suit better for medium-to-high-activity materials.

Another objective related to the development of the new setup is related to the mobility and reliability of the device. Because of the high cost and safety issues related to transportation of highly radioactive materials, it would be much more convenient to develop a compact positron lifetime setup easily transferable to the dedicated facilities such as VTT Centre for Nuclear Safety in Finland.

### 2.2 Technical development

The detector materials and geometry of the three-detector setup need to be optimized to achieve reasonable count rate for experiments. Because of geometrical restrictions, the count rate optimization is much more critical than in the conventional two-detector setup. In traditional face-to-face two-detector lifetime experiment the scintillator material

of the detector is often plastic, because it scatters less gamma photons than for example BaF<sub>2</sub> or NaI detectors. However, the efficiency of the plastic scintillator is rather low. In the three-detector setup the efficiency must be improved by choosing more efficient scintillator material or larger plastic scintillator for one of the detectors. Preferentially the modified detector should be the other STOP-detector in order to maintain good time resolution in the measurements. Both experimental and Monte-Carlo simulations are currently under progress in order to determine the best setup configuration to adopt.

Another field of technical development is related to digital pulse processing and digital triggering of digitized pulses. Nowadays digitizers up to 5 GS/s with 8 bit accuracy can be found with reasonable price [10]. The low speed limit for digitizing positron lifetime signals is considered 1 GS/s [11]. 5 GS/s can be considered sufficient for three-detector coincidence setup even though the digitizer would be shared for four inputs [10]. Also the triggering condition for successive pulses can be programmed in some digitizers or digital oscilloscopes. Digital pulse processing and triggering are currently in routine use in two-detector setups in our laboratory at Aalto University.

### 3 RESULTS

In order to justify the pros and cons of the three-detector setup, one needs to first evaluate the effective impact of additional radiation originating from the samples in conventional two-detector setup. As a first step, the additional radiation was introduced by a 500 kBq <sup>60</sup>Co source in between the START and the STOP detector. Well-known silicon samples, sandwiched with regular <sup>22</sup>Na positron sources, were used for producing real positron lifetime data.

The <sup>60</sup>Co source cause random false lifetime events due to its two successive fast gamma photon decays of 1.173 MeV and 1.333 MeV. The measured spectra are presented in figure 2 and the average spectral backgrounds are given in table 1. Regardless of the energy windows dedicated to the START and the STOP detector, addition of the <sup>60</sup>Co source to a 950 kBq <sup>22</sup>Na positron source caused 3% increment to the total count rate (table 2). This unavoidable detection of high-energy gamma photons is caused by the Compton continuum in the scintillator detectors.

Peak to background ratio or true coincidence to random coincidence count rate of the coincidence measurement can be approximated with

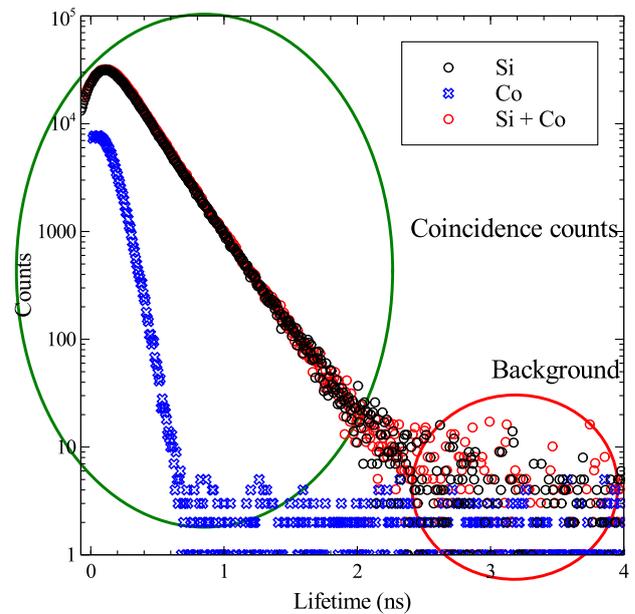


Figure 2. The two-detector lifetime spectra of silicon reference sample with 950 kBq <sup>22</sup>Na source, 500 kBq <sup>60</sup>Co source and the combination of the two.

Table 1. Spectral properties of the regular lifetime experiment with Si reference samples, <sup>60</sup>Co source and the combination of the two. The activity of sodium and cobalt sources were, at the time of measurements, around 950 kBq and 500 kBq respectively.

	Si	Co	Si+Co
<b>Average background / 8 ps channel</b>	14.5	1.2	21.2

$$\frac{R_C}{R_R} = \frac{1}{A\tau}, \quad (2)$$

where  $A$  is the activity of the sample and  $\tau$  width of the coincidence window.

Table 1 shows that the spectral background increases expectedly (equation (2)) due to the excess activity in the measurement. The decreasing peak-to-background ratio is the reason for limiting positron source activity in the measurements. The lifetime components of the silicon are respectively 221(1) ps and 222(1) ps with and without <sup>60</sup>Co. One can conclude that the results are not significantly changed for 222 ps average lifetime component while adding the fast coincidence source of the activity of  $1/2A_{Na}$  to the measurement.

To study further the effect of excess fast coincidences on the results, we reduced the activity

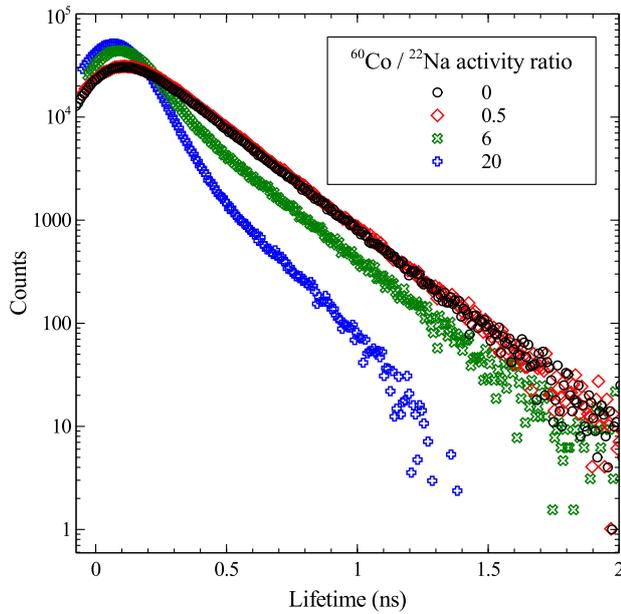


Figure 3. The effect of excess fast coincidences to the background reduced lifetime spectra.

Table 2. Average decay components of the lifetime spectra with additional fast-coincidence events. The spectral resolution was 270 ps (FWHM).

$^{60}\text{Co} / ^{22}\text{Na}$ activity ratio	Fast coinc. count rate (%)	Average decay constant of a spectrum (ps)
0	0.0	222(1)
0.5	2.9	221(1)
6	101.5	147(1)
20	459.1	81(1)

of the positron source while using the same 500 kBq cobalt source. Figure 3 shows that the lifetime spectrum was significantly distorted when the fast-coincidence count rate was of the same order as the annihilation count rate (and the activity of the cobalt source around 6 times higher than the positron source). The average decay components of the spectra or the average positron lifetime components show also a significant change when the fast-coincidence count rate increases. The decay components are calculated in table 2 using the time resolution of 270 ps (FWHM) for the setup.

## 4 CONCLUSIONS

Positron lifetime spectroscopy is suitable and proven method for vacancy-type defect characterization in nuclear materials. However, the excessive activity in irradiated fuel or structural

steels distorts measurements results in a regular two-detector setup.

In this paper the prerequisites for developing a three-detector coincidence setup are discussed. This paper also presents the main challenges for making conventional positron lifetime measurements for radioactive materials. The activity of a positron source cannot be increased a lot above 1 MBq, because the peak-to-background ratio would suffer. On the other hand, the spectrum will be significantly distorted, if the relative fast-coincidence count rate will increase.

With a three-detector setup, one can potentially overcome the limitations demonstrated for a two-detector setup with a cost of reduced count rate.

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