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Positron annihilation spectroscopy studies of irradiated Fe-based alloys using different radioisotope sources

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ABSTRACT

Positron annihilation spectroscopy as a unique tool for the characterization of radiation-induced defects often faces difficulties with the reproducibility of the experimental data obtained from different irradiation experiments. A particularly challenging problem is the correlation of near-surface and bulk irradiations as well as the correlation of the experiments performed with different types of positron sources. This work addresses some practical issues on the application of radioisotope sources ^{22}Na and ^{44}Ti to the investigation of Fe-based samples. Conventional Kapton-encapsulated ^{22}Na positron sources external to the sample and a ^{44}Ti source internal to the sample are discussed in terms of ion-implanted samples and samples irradiated in spallation neutron source, respectively. While the first type of experiment suffers from the distinct ion and positron stopping profiles, the latter is sensitive to the geometry of the measured sample and the emission of positrons out of the studied volume. The first part of the present work concentrates on theoretical and experimental investigation of the positron stopping in Fe-based materials with particular attention to the near-surface region. In the second part, Doppler broadening data obtained on FeCr samples irradiated in a spallation neutron source are discussed concerning the contribution of out-of-the-sample annihilation due to the use of internal and external positron sources.

1. Introduction

Positron annihilation spectroscopy (PAS) has become a valuable characterization tool for radiation-induced defects in structural materials exposed to harsh radiation environments [1–3]. Two techniques namely positron lifetime spectroscopy (PALS) and Doppler broadening spectroscopy (DBS) enable investigation of vacancy-type defects sized below the resolution of transmission electron microscopy (TEM) [4]. Both techniques, however, often suffer from a poor reproducibility, when used in various irradiation studies. The reason for this can be the difficult reproducibility of the irradiation experiments themselves or an attempt to compare near-surface (ion) implantation [5,6] and bulk (neutron) irradiation experiments [7,8]. Another factor is the contribution of positron annihilation events from outside of the investigated sample volume, particularly when the used positron source is internal to the sample [9]. Nevertheless, a certain fraction of the positrons annihilate outside the investigated sample, regardless of whether the positron source is internal or external to the sample. By conventional measurement using an encapsulated positron source, this is called a “source-component” and it is attributed to the positron annihilation in the encapsulation materials, the surface of the sample or to the gap in the source-sample sandwich respectively. Naturally, such a component does not exist in the measurements utilizing a positron source internal to the sample [9]. On the other hand, due to the emission of positrons outside of the sample, the internal-source method requires an assessment of this type of contribution.

This paper presents theoretical and experimental analysis of the above-mentioned problem. It addresses the feasibility of using an unmoderated ^{22}Na source for the investigation of ion implanted samples and compares this conventional approach with measurements utilizing a ^{44}Ti (sometimes referred to as ^{44}Ti/^{44}Sc or ^{44}Sc) source internal to the sample that was irradiated in a spallation neutron target. The manuscript is structured as follows: Section 2 covers the basics of absorption and emission of positrons by metallic radionuclides as a theoretical background for Section 3, which describes measurements of the absorption of positrons emitted from the ^{22}Na source in thin Fe(Cr) foils and Doppler broadening spectroscopy (DBS) characterization of FeCr alloy irradiated in the spallation neutron source. Results are further

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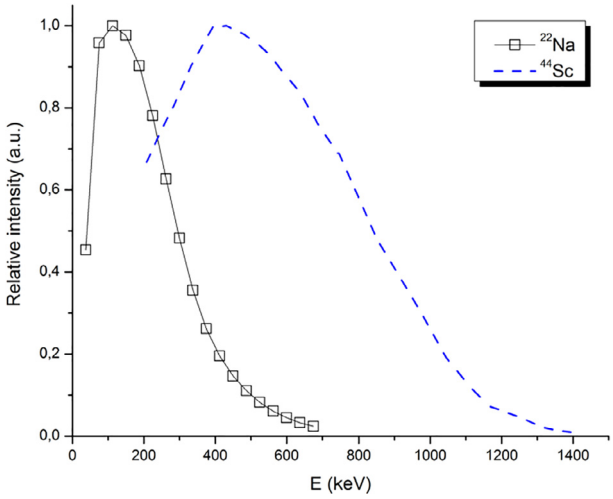

Fig. 1. Experimental beta spectrum of ^{22}Na (measured with 7.5 μm Kapton-encapsulated source using CANBERRA iSolo® alpha/beta counting system) and ^{44}Ti (adopted from [10]).

discussed in Section 4, and Section 5 concludes the present work and points out the directions towards the improvement of the reliability of positron annihilation spectroscopy in the characterization of radiation damage of nuclear structural materials.

2. The absorption and emission of positrons by metallic radionuclides

The distribution of positrons produced by a radioactive source cannot be described analytically in a straightforward and accurate way. Experimentally obtained β^+ spectrum of ^{22}Na and ^{44}Sc are shown in Fig. 1. The spectrum of ^{22}Na was measured with a 7.5 μm Kapton-encapsulated source using the CANBERRA iSolo® alpha/beta counting system. Data for ^{44}Sc are adopted from literature [10]. Due to a short half-life (3.97 h) of the ^{44}Sc, it is hereinafter referred to as ^{44}Ti, its mother isotope (half-life 60 years). The maximum energy of positrons from these sources is 545 keV, or 1450 keV for the ^{22}Na or ^{44}Ti isotope respectively. Since the implantation depth of such positrons is few hundreds of micrometres in most solids, any experimental characterization of miniaturized samples must consider the likelihood of positrons escaping the studied system. These contribute to the experimental spectra with the annihilation either on the sample surface or in the sample holder material. On the other hand, near-surface studies, such as ion-implantation studies need to be interpreted with respect to the positron absorption profile, due to the distinction with the displacement damage profile.

2.1. Absorption profile of ^{22}Na positrons in Fe

The implantation profiles of ^{22}Na positrons in various materials were studied by several authors in the past [11–13]. In these works, the fraction of positrons stopped at certain depth was described either by a single exponential law [12,13] or by a sum of two exponential functions [11]. The implantation profile for a given material can be reasonably estimated also by discretization of the continuous beta spectrum of ^{22}Na and calculation of the mean implantation depth for individual energy intervals. Using an approximation to monoenergetic positrons ($z\,[\mathrm{nm}] = 40 \times E[\mathrm{keV}]\hat{}1.6/\rho\ [\mathrm{g.cm}^{-3}]$ [14]) one can obtain an implantation profile, which is in good agreement with theoretical calculations quoted above (Fig. 2).

From the theoretical calculations, one can obtain the maximum implantation depth of ^{22}Na positrons in Fe to be close to 100 μm, with 95% stopping for 60 μm and a mean implantation depth of 25 μm.

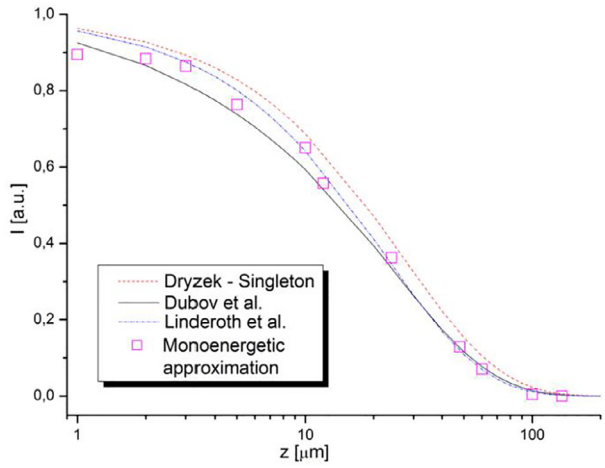

Fig. 2. The implantation profile of ^{22}Na positrons in Fe, calculated either using different exponential decay laws published in literature [11–13] or by an approximation to the stopping of monoenergetic positrons.

2.2. Emission of positrons from Fe-based alloys containing ^{44}Ti/^{44}Sc

One of the main advantages of utilizing positron sources internal to the sample for positron annihilation spectroscopy is the uniform distribution of positrons across the sample volume. Although there is no external source needed and thus no annihilation takes place in the encapsulation envelope, a certain fraction of positrons escapes out of the sample and annihilate for instance in the sample holder or in the detection system respectively.

On the other hand, the use of an external positron source in a sandwich geometry with two identical samples can minimize the fraction of escaping positrons by increasing the number of those absorbed in the sample. The sacrifice here is the contribution from the encapsulation foil. While positron lifetime spectroscopy has an effective procedure for subtracting the “source component”, it is practically impossible to do this in Doppler broadening spectroscopy. One can argue that the contribution of, for instance, Kapton encapsulation is constant and as such it can be neglected in the DBS measurements. This is, however, not the case in the investigation of spallation samples, containing fluence-depending activities of β^+ radioisotopes. A significant uncertainty can be introduced by measuring a series of samples of one material irradiated to different fluencies, without considering the variable contribution of the external source encapsulation (Kapton).

The unavailability of a quantitative assessment of the “source component” for the Doppler broadening spectroscopy favours the use of positron sources internal to the sample, such as a ^{44}Ti source. However, the tendency of minimizing the volume of the samples irradiated in neutron environments increases the probability of positron emission outside the sample.

To calculate the emission of positrons out of the sample and thus to estimate the impact on the measured spectra, we have used MCNPX (Monte Carlo N-Particle eXtended) code. MCNPX is a widely spread calculation code based on Monte Carlo algorithms used to simulate interaction of radiation with matter. It contains high-quality physics and has access to the most up-to-date cross-section data [15]. The code enables the tracking of nearly all particles at nearly all energies, whereby the nuclear data tables beside model physics are employed. The source of positrons was modelled as a volume source spread uniformly through an infinite iron plate of different thicknesses. The energy spectrum of the source positrons was taken from the reference [10] and is depicted in Fig. 1. The flux of escaping positrons was calculated using F1 tallies summed for both outer planes of the iron plates. The number of histories used in the calculations was 10^6. Fig. 3 shows the results of the simulation for an infinite plate (x, y > > z) of thickness z,