DEVELOPMENT OF A NEW POSITRON LIFETIME SPECTROSCOPY TECHNIQUE FOR DEFECT CHARACTERIZATION IN THICK MATERIALS

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ABSTRACT

The recent development of Accelerator-based Gamma-induced Positron Annihilation Spectroscopy (AGPAS) has shown the possibility of probing residual stress in thick materials by Doppler broadening measurements [1]. In those measurements, the residual stresses were reflected in the line shape parameters of the 511 keV annihilation peak. However, since positron lifetime spectroscopy is a powerful tool to distinguish between different types of defects, such as dislocations and vacancy clusters [2], it is crucial to enable positron lifetime measurements in AGPAS. In this work, a new method is developed to conduct positron lifetime measurements on thick engineering materials using accelerators. By focusing 1.5 MeV protons from a Van de Graaff accelerator on a thin Al window, coincident γ-rays of 2.8 MeV and 1.78 MeV are induced through (p,γ) reactions. The 1.78 MeV quantum provides a start signal for the positron lifetime spectrometer, whereas the 2.8 MeV quantum bombards the material under investigation. This, in turn, creates a positron, which annihilates with one of the material electrons emitting two 511 keV photons. The stop signal for the positron lifetime is provided by the detection of one of the two 511 keV photons. The measured positron lifetime spectrum depends on the electron densities and hence provides information about the size of open volume defects. The method enables positron lifetime measurements in thick engineering materials up to tens of gm/cm², a thickness not accessible by conventional positron lifetime spectroscopy.

INTRODUCTION

Positron annihilation spectroscopy (PAS) has been known as a valuable tool to characterize materials and study defects [2]. Positron spectroscopy of defects can be performed by means of Doppler broadening spectroscopy of annihilation radiation, angular correlation of annihilation radiation (ACAR) measurements or positron lifetime spectroscopy (PLS). Positron annihilation measurements have been often performed using keV positrons from radioactive sources or positron beams. The range of keV positrons in materials are in the order of few hundred micrometers and hence the depths in materials that can be investigated by positrons are limited to this range. Consequently standard PAS is not able to study defects in thick materials.

Accelerator based γ-ray induced positron annihilation spectroscopy (AG-PAS) [1, 3] is a recently developed technique that conducts positron annihilation spectroscopy by using MeV γ-rays from accelerators instead of using positrons. This technique combines the
This document was presented at the Denver X-ray Conference (DXC) on Applications of X-ray Analysis.

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high penetrability of MeV $\gamma$-rays with the high sensitivity of positrons to defects to provide a highly penetrating sensitive probe. Hence it extends positron spectroscopy of defects to thick materials and a wide variety of new applications. The first measurements of AG-PAS were conducted using bremsstrahlung radiation from a 6 MeV electron Linac to perform Doppler spectroscopy of positron annihilation. In those measurements residual stresses in engineering materials are reflected in the narrowing of the Doppler broadening of the 511 keV peak and in an increase of the annihilation fraction with valence electrons. Residual stresses due to tensile test were measured in low alloy steel [3] and Zr-4 [1]. Residual stresses due to cold work were also investigated in EP-823 and SS-304 [4].

The objective of this work is to develop a method to perform positron lifetime spectroscopy in AG-PAS. The importance of PLS lies in its unique ability to distinguish between different kinds of open volume defects and identify the defect size and its concentration in a single measurement. In the following, the basis of positron lifetime spectroscopy as a method for defect characterization will be demonstrated. Then the positron lifetime spectrometer in AG-PAS will be described.

**CHARACTERIZATION OF DEFECTS BY PLS**

Positron lifetime is the time that a positron spends in a material once it enters until it annihilates with an electron. The annihilation rate $\lambda$, which is the reciprocal of the positron lifetime, is given by [5]:

$$\lambda = \frac{1}{\tau} = \pi r_o c^2 \int |\psi(r)|^2 n(r) \gamma dr,$$

where $\tau$ is the positron lifetime, $r_o$ is the classical electron radius, $c$ is the speed of light, $r$ is the position vector, $\psi(r)$ is the positron wave function, $n(r)$ is the electron density and $\gamma$ is the enhancement factor representing the increase in electron density due to the coulomb interaction between an electron and a positron.

Since the positron lifetime is a function of the electron density at the annihilation site, it provides information about the electron densities and consequently open-volume defects in materials. Standard PLS in bulk materials is performed by using a Na-22 source, which emits a 1.72 MeV $\gamma$-quantum coincident with each positron. The positron lifetime is measured as the time difference between the detection of the 1.27 MeV photon and one of the two 0.511 MeV photons emitted from the positron annihilation event in the material. The measured positron lifetime spectrum is a convolution of the resolution function of the spectrometer and the positron decay curve in material.

In a defect-free metal or semiconductor, all positrons annihilate as free particles from Bloch states with well-defined annihilation rate. This produces a single exponential decay and only one lifetime component exists in the lifetime spectrum. Open volume defects in materials form trapping centers for positrons. This is because a positron is strongly repelled by the nuclei in the lattice due to its positive charge. The absence of a positive charge at a defect site provides an attractive potential that traps positrons at this site.
Positron trapping at defect sites leads to longer positron lifetime because of the lower electron density at defect sites compared to regions without defects.

With the presence of open volume defects, positrons annihilate from different states, each gives a characteristic lifetime $\tau_i$. The resulting positron lifetime spectrum $N(t)$ consists of multi-time components:

$$N(t) = \sum_{i=1}^{k+1} \frac{I_i}{\tau_i} \exp\left(-\frac{t}{\tau_i}\right),$$

where: $k+1$ the number of time components in the spectrum relating to the $k$ number of defect types in the specimen and one time component from positron annihilation in bulk material, $\tau_i$ is each individual lifetime and $I_i$ its intensity in the spectrum. The measured lifetime spectrum can be fitted by a sum of exponential decay components convoluted with the resolution function of the spectrometer. Each lifetime component and its intensity could be extracted from the fit. The lifetime value is related to the defect size and its characteristics, whereas the relative intensity for each lifetime component determines the defect concentration.

**POSITRON LIFETIME SPECTROSCOPY IN AG-PAS**

AG-PAS utilizes MeV $\gamma$-rays to implant positrons inside the material via pair production. The induced positrons thermalize and then annihilate with the material electrons emitting 511 keV annihilation photons. Lifetime spectroscopy in AG-PAS can be performed by using bremsstrahlung radiation from a short-pulsed electron linac. The pulse width should be in the order of picoseconds since positron lifetimes are in the range of few hundred picoseconds in most of the materials. This type of electron Linac is not widely available. Coincident MeV $\gamma$-rays represents another method to perform positron lifetime measurements in AG-PAS. Proton capture in some low atomic number materials such as Al-27 and C-13 induce coincident MeV $\gamma$-rays. One $\gamma$-quantum can be used to indicate the positron birth where as the other $\gamma$-quantum penetrates the specimen under investigation creating a positron. One of the two 0.511 MeV photons emitted from the positron annihilation event indicates the positron death.

Proton capture in Al-27 forms Si-28 in high-energy excited states [6]. Subsequent transitions to the ground states induce coincident MeV $\gamma$-quanta. Figure 1 shows the energy levels of Si-28 nuclei due to $(p, \gamma)$ reactions in Al-27. Both 2.8 and 3.2 MeV $\gamma$-quanta are emitted due to transitions from high-energy excited states to the first excited state, which decays to the ground state in 475 fs emitting a 1.78 MeV $\gamma$-quantum. This $^{27}$Al $(p,\gamma)^{28}$Si reaction has a strong resonance at 992 keV proton energy [7]. To obtain these coincident $\gamma$’s we focused 1.5 MeV protons from a Van De Graff accelerator on thin Al window mounted at the end of the proton beam line. The 1.78 MeV photon
emitted from the Al window provides a start signal for the positron lifetime spectrometer whereas the other γ-quanta (mostly the 2.8 MeV photon) penetrates the specimen generating a positron inside it. The induced positron annihilates with one of the material electrons emitting two 511 keV photons. The detection of the one of the 511 keV photons provides a stop signal for the spectrometer.

Two BaF$_2$ detectors coupled to fast photo-multiplier tubes are used to record the start and stop signals. Figure 2 shows a fast-slow coincidence system to collect the positron lifetime spectrum, where the time measurement is performed in a fast channel and the energy selection is performed in a slow channel. An energy window was adjusted at 1.78 MeV for the start signal and another energy window was adjusted at 0.511 MeV for the stop signal. The coincidence between the high-energy 1.78 γ-quanta and the 0.511 MeV γ-quanta sharply reduces the random background in the spectrum. The timing resolution of the spectrometer was measured using a Co-60 source, which emits two coincident γ’s of 1.17 and 1.33 MeV, and found to be 210 ps.

Figure 2. Fast slow coincidence system for PLS based on proton capture induced coincident MeV γ-rays
Using this spectrometer, we measured the positron lifetime in pure thick materials of Pb, Al and Cu. Both Al and Cu were annealed before the measurements to get rid of any defects. Pb was not annealed because it is a self-annealing material. The measured positron lifetimes were: $194 \pm 2$ ps for Pb, $162 \pm 3$ ps for Al and $114 \pm 5$ ps for Cu. These lifetime values are in complete agreement with the published positron lifetime values experimentally and theoretically, giving us a complete confidence in the performance of the new positron lifetime spectrometer. More details about the positron lifetime spectra can be found elsewhere [8]. The presented technique extends PLS to thick materials because of the high penetration of $\gamma$-rays. In addition it has additional advantages over standard PLS because it yields lifetime spectra with single exponential positron decay from the pure defect free metals and semiconductors with no other lifetime components. One of the biggest problems of standard PLS is that the measured spectrum always contains unwanted lifetime components from positron annihilation in the source materials and from surface effects.

CONCLUSION:

We upgraded AG-PAS technique by a new method to perform positron lifetime spectroscopy based on using proton capture induced coincident MeV $\gamma$-rays. This approach extends PLS to thick materials and eliminates some of the associated problems with standard PLS technique. The importance of PLS lies in its ability to identify different kinds of defects in material and determine their concentration in a single measurement.

ACKNOWLEDGEMENTS

This work has been supported by Inland Northwest Research Alliance under contract ISU001.

REFERENCES