3-D defect density plots of large scale structural materials with positron annihilation spectroscopy: feasibility and optimization

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1. Introduction

PAS techniques have long been employed to study various defect states and obtain information about electron momentum-density distributions in different materials. PAS measurements based on conventional radioactive sources or positron beams are restricted to probe only up to a few hundred μm in solids due to the low penetrability of positrons in the keV range. The feasibility of utilizing MeV positrons have been also demonstrated [1,2], but it only extends PAS measurements to a few mm depth.

We have been developing accelerator-based in-situ pair-production techniques to produce positrons in thick samples (~ 4-40 g/cm², or ~ 0.5-5 cm in steel). These techniques are called “Accelerator-based Gamma-induced Positron Annihilation Spectroscop” (AG-PAS). AG-PAS employs an accelerator to produce bremsstrahlung, which creates positrons inside the material via pair production. No positron emitters or photon-induced activation are involved in this process. The annihilation radiation is counted by a high resolution HPGe, which allows us to analyze the Doppler broadened 511 keV peak in terms of S and W parameters. This technique combines high penetration of MeV gamma rays with the high sensitivity of positrons to microscopic defects (mono-vacancies, di-vacancies, dislocations, etc). Hence, it creates a

Abstract. We investigated the feasibility of the pair-production based positron annihilation spectroscopy (PAS) technique to generate 2-D and possibly 3-D defect density images (defect density plots) of structural materials widely used in industry. After assessing the possible PAS techniques and performing MCNPX simulations on different positron production mechanisms, we chose to employ Doppler broadening analysis of the annihilation peak coupled with the pair-production method of positron creation using bremsstrahlung photon beams from a linear accelerator. First, to optimize the important parameters of the proposed experimental setup, MCNPX simulations were performed. These parameters are: thickness of the bremsstrahlung beam hardener, detector orientation, thickness of the bremsstrahlung converter, beam energy and current, etc. These parameters were studied to maximize the number of 511 keV counts in the detector and minimize the low energy background photons that Compton scatter into the detector. A Figure of Merit (FOM) was used to quantitatively track effectiveness (or ineffectiveness) of each value for all the parameters. We conducted experiments with the simulated setup and these results were compared to MCNPX calculations.
possibility to add defect density imaging capabilities of large scale structural materials to the conventional PAS methods. Studying these microscopic defects is very important for a number of industries in order to predict structural integrity of materials. Based on MCNPX (Monte Carlo N-Particle eXtended) simulations [3] on different positron creation mechanisms and knowledge from the previous experiments [4] we think that the AG-PAS is the most practical approach for defect studies of thick structural materials in engineering applications.

The biggest limitation of the AG-PAS experiments performed in the past [5–7] was the low repetition rate of the LINAC (200-300 Hz). However, the work presented here is performed by utilizing a 1 kHz High Repetition Rate LINAC (HRRL), which makes defect density imaging with AG-PAS more feasible. In order to demonstrate this feasibility and optimize the experimental setup, FOM analyses have been performed based on the experimental and MCNPX simulation results.

2. Methods and materials

2.1. Experimental setup

A 15 MeV electron beam from the HRRL was bent 270° before striking a 1 mm thick tungsten converter. The accelerator is capable of producing 30-60 ns pulses, up to 100 mA peak current at 1 kHz repetition rate. The generated bremsstrahlung cone was filtered (hardened) by a graphite hardener to reduce the contribution of the low energy photons in the spectrum. The hardened beam went through a 1.2 m thick concrete wall that had two collimators. The primary collimator was a 25 cm long steel cylinder with a 6 mm diameter hole in the center. The secondary (clean up) collimator was a 2.54 cm diameter hole in a 10 cm thick lead block. The highly collimated beam then struck the sample, which in this case was a 0.95 cm (3/8 inch) thick steel punch. The sample was located about 1.5 m away from the wall (figure 1).

![ Experimental setup ](image)

Figure 1. Experimental setup

The 15 MeV endpoint energy bremsstrahlung beam produced photo-electric effect, Compton scattering and pair production processes in the sample. Compton scattering predominantly contributed to the detected low energy background, while pair production created positrons throughout the sample volume, resulting in emission of 511 keV photons. These 511 keV photons were Doppler broadened due to conservation of momentum and contained valuable information about defects in the material. The spectrum emitted from the sample was recorded by a heavily shielded, 20% relative efficiency HPGe detector. The detector orientation was such that there was a 120° angle between the beam line and the detector field of view. The detector shielding was composed of lead bricks and had the following dimensions: 30 cm thick on the wall side,
20 cm thick on the opposite side and 10 cm thick in front, top and bottom. The shielding had 2.54 cm diameter collimator which was covered with a secondary lead hardener to reduce the low energy background even further. The detector side of the concrete wall was also shielded with 20 cm thick and 25 cm tall lead to reduce the scattered photons coming from the beam. The intensity of the bremsstrahlung beam was monitored by a NaI(Tl) detector. The signal from the NaI(Tl) detector was also used to gate the germanium detector so we can record the spectrum coming only from the accelerator pulses. 1 μCi $^{133}$Ba and $^{137}$Cs sources were used for calibrating the HPGe detector.

The parameters that we optimized in this setup were: thickness of the tungsten converter, thickness of the graphite hardener, orientation of the detector and the thickness of the secondary lead hardener. Experiments were conducted to optimize the graphite and lead hardeners and also to measure background rates in this experimental environment with different shielding configurations.

We introduced a $FOM$ definition to quantitatively track effectiveness of different values of different parameters. The idea was to maximize the 511 keV count rate per unit total rate in the detector. So, the obvious definition was

$$FOM = \frac{R_{511}}{R_T}$$

where $R_{511}$ and $R_T$ are the 511 keV and total count rates respectively.

In a pulsed environment like ours, one has to take careful measures to address detector dead time and pulse pile-up issues. We know that under the conditions $T < \tau < (1/f - T)$, where $T$, $\tau$ and $f$ are pulse width, detector dead time and repetition rate respectively, which is certainly satisfied in our case ($T = 30 \text{ ns}$, $\tau = 10 \mu\text{s}$, $f = 1 \text{ kHz}$), the true event rate $n$ and the recorded count rate $m$ are related by the following expression: [8]

$$n = f \ln \left( \frac{f}{f - m} \right)$$

When the dead time losses are small (desired condition for our experiment) under the condition $m \ll f$, equation (2) can be simplified by expanding the logarithmic term

$$n = \frac{m}{1 - m/2f}$$

We decided to stay at 10% pile-up level which sets the upper limits for the accelerator peak current and consequently recorded total rate. In the first approximation, when only two pulse pile-up is significant, the pile-up can be expressed in terms of $m$ and $f$ the following way:

$$PU = \frac{2(n - m)}{n} = \frac{m}{f}$$

where we used equation (3) for $n$. This tells us that 10% pile-up means observed 100 counts/s at 1 kHz repetition rate. Hence, we kept the total rate at about 100 counts/s and looked at $R_{511}$ as a function of thickness for both graphite and lead hardeners. The reason we used graphite as a primary hardener is that low Z materials are effective bremsstrahlung filters, which has been demonstrated experimentally in the past [9]. This is simply due to the fact that the Compton scattering cross-section is proportional to $Z$, while pair production goes approximately as $Z^2$. Thus, the ratio of Compton interactions to pair production events in the hardener goes as $1/Z$. On the other hand, a high Z material is desired as a secondary hardener since there are no pair production events caused by the photons coming from the sample.

The beam background coming from the accelerator side of the wall was also measured for two shielding configurations of the lead next to the wall. One was the shielding described above and the other with added shielding: 30 cm thick and 50 cm tall.
2.2. MCNPX simulations

MCNPX version 2.5f was employed to perform simulation for optimizing the tungsten converter, both, graphite and lead hardeners and the detector angle. The experimental setup described above was replicated for the MCNPX geometry. The bremsstrahlung converter was optimized by simply looking at the pair production events per electron in the sample as a function of the tungsten thickness. The biggest obstacle encountered during the other simulations was the “thick penetration problem”. Since one can only run $2 \times 10^9$ source particles in version 2.5f, it was extremely difficult to simulate the whole process in a single run and generate reliable statistics even with the weight window technique. So, we simulated the process in two steps: step one was finding the bremsstrahlung energy distribution in the sample while step two was assigning this distribution to the source photons and sending them mono-directionally (ignoring the angular divergence justified by the collimation) towards the sample in order to determine the resulting spectrum in the detector. All the other parameters were optimized by using the same FOM definition as in the experimental case.

All the MCNPX simulations were done on a cluster called BREMS (Beowulf REsource for Monte-carlo Simulations) at the physics department of Idaho State University. BREMS is made of 21 nodes: 12 nodes with two 2.0 GHz Opteron CPUs and 9 nodes with two dual-core 2.0 GHz Opteron CPUs - a total of 60 processing cores.

3. Results and discussions

3.1. Background

Let’s clarify the term background first. In our optimization problem the only useful information is the 511 keV counts. We call everything else - “background”. The total background can be broken down the following way

$$ R_T = B_1 + B_2 + B_3 + R_{511} $$

where:

- $B_1$ - is the natural background plus counts from the calibration sources
- $B_2$ - is the beam background coming from the accelerator side of the wall
- $B_3$ - is the Compton scattered photons from the sample

So, another way to state our problem is to minimize $B_1$, $B_2$ and $B_3$ for a fixed $R_T$. Based on the not-gated spectrum with the beam-off and the width of the gate, we estimated that in the gated spectrum $B_1 \approx 8-9$ counts/s when $R_T = 100$ counts/s and there isn’t much we can do about optimizing $B_1$.

$B_2$ was measured by removing the sample with the beam-on for two different shielding configurations. The results are presented in figure 2. With the added shielding, $B_2$ ranges about 8-13% of the $R_T$. We could have reduced this background if we had more room for shielding. $B_2$ becomes the major limitation with thick hardeners, because we need to increase the current to keep the $R_T$ fixed.

The biggest potential for optimization is in $B_3$, because it constitutes about 70% of the total count rate. The both, graphite and lead hardeners and the detector angle optimizations were targeted to minimize $B_3$.

3.2. Tungsten converter

The simulation results for the tungsten converter are demonstrated in figure 3. The pair production events in the sample per electron as a function of tungsten thickness is shown for a 15 cm thick graphite hardener. If the tungsten is too thin there is simply not enough high Z material to produce energetic bremsstrahlung efficiently. The pair production events in the sample slowly drops with thickness, because we start to loose high energy photons via pair
Figure 2. $B_2$ background for two shielding configurations. Before - lead shielding, 20 cm thick and 25 cm tall. After - lead shielding 30 cm thick and 50 cm tall.

Figure 3. MCNPX simulation results for the tungsten converter. Pair production events in the sample as a function of the converter thickness.

Production in the converter. This is only a part of the full picture. Increased tungsten thickness means increased bremsstrahlung and therefore increased $B_2$. For example, even though the graph shows that the pair production events at 0.5 mm is slightly better than at 2 mm, 2 mm tungsten produces about 3 times more bremsstrahlung than 0.5 mm. So, we conclude that around 0.5 mm tungsten is the most effective thickness for our purposes.

3.3. Graphite hardener

The effect of the graphite hardener to $B_3$ is demonstrated with the experimental spectra in figure 4 and figure 5 for 4.2 cm and 48.1 cm graphite respectively. In figure 5 the low background allows us to see the Compton edge of the 511 keV peak. 356 keV and 662 keV are $^{133}$Ba and $^{137}$Cs lines. The observed 511 keV count rate ($R_{511}$) was about 10 counts/s.

Figure 4. Experimental spectrum with a 4.2 cm graphite. Note the $^{137}$Cs and $^{133}$Ba lines that were measured synchronously.

Figure 5. Experimental spectrum with a 48.1 cm graphite. Note the $^{137}$Cs and $^{133}$Ba lines that were measured synchronously.
Figure 6 shows the simulated bremsstrahlung spectra in the sample for different graphite thicknesses. We can clearly see the effect of the hardening. Figure 7 compares three data sets against each other. Two experimental data sets are $FOM$ as a function of graphite thickness: upper one including only $B_3$, lower one including all the background. The third one is the simulation results of $FOM$ including only $B_3$. The simulation and the experimental results are close to each other for thin hardeners. The reason they deviate from each other as the thickness increases is that the simulation ignores angular divergence of the bremsstrahlung beam, which becomes more and more significant for thicker hardeners. As the lower experimental data set suggests, $B_2$ is the major limitation for thick hardeners.

![Figure 6. Simulated bremsstrahlung with different thickness of graphite hardener.](image1)

**Figure 6.** Simulated bremsstrahlung with different thickness of graphite hardener.

**Figure 7.** Experimental and simulation results for graphite hardener

### 3.4. Detector angle

Simulated spectra for different detector angles with a 15 cm graphite hardener are presented in figure 8. We can identify three features of each spectrum - first, they all have the Compton edge of the 511 keV peak itself at about 340 keV; second, the Compton background, $B_3$, decreases as the angle increases which is what one should expect based on the angular dependence of the Compton scattering cross-section; and also, the Compton edge of the 15 MeV end-point bremsstrahlung shifts left with increasing angle which is consistent with the Compton scattering kinematics.

![Figure 8. Compton edge and background for different detector angles.](image2)

Figure 9 is the 3D plot of $FOM$ as a function of the detector angle and the graphite thickness. According to the results, the optimal angle appears to be at around 140°.

### 3.5. Lead hardener

Finally, experimental and simulation $FOM$ analysis for the lead hardener are shown in figure 10. The reason that they strongly disagree as the thickness increases, is again the beam background $B_2$, which is ignored in simulations but becomes a major problem at high accelerator currents due to a concrete wall that is not sufficiently thick ($\sim 1.2$ m) to eliminate this background (figure 2).

![Figure 9. 3D plot of FOM for different detector angles and graphite thicknesses.](image3)

### 4. Future work

The knowledge obtained from this work will be exploited to prepare the setup for defect density imaging experiments using AG-PAS. A well-collimated bremsstrahlung beam will strike a bundle
Figure 8. Simulated detector spectra for different angles

Figure 9. FOM as a function of hardener and detector orientation

Figure 10. Experimental and MCNPX FOM results for lead hardener

Figure 11. Simplified scheme of future experiments

of steel samples with different degrees of mechanical damage. Also a well-collimated HPGe detector will look at one irradiated voxel at a time. This way we can map $S$ (or $W$) parameter as a function of the position $(x, y)$. The simplified scheme is shown in figure 11.

5. Conclusions
We conducted experiments and MCNPX simulations to determine optimal values of important experimental parameters for defect density imaging with AG-PAS. We also uncovered some of the major limitations and difficulties we will face to produce such an image. Regardless of these limitations, based on the experimental 511 keV rates ($R_{511}$) obtained, we conclude that defect density mapping of large scale engineering samples with AG-PAS seems feasible.

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