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Characterisation of radiation-induced defects in tin oxide semiconductor using positron annihilation techniques

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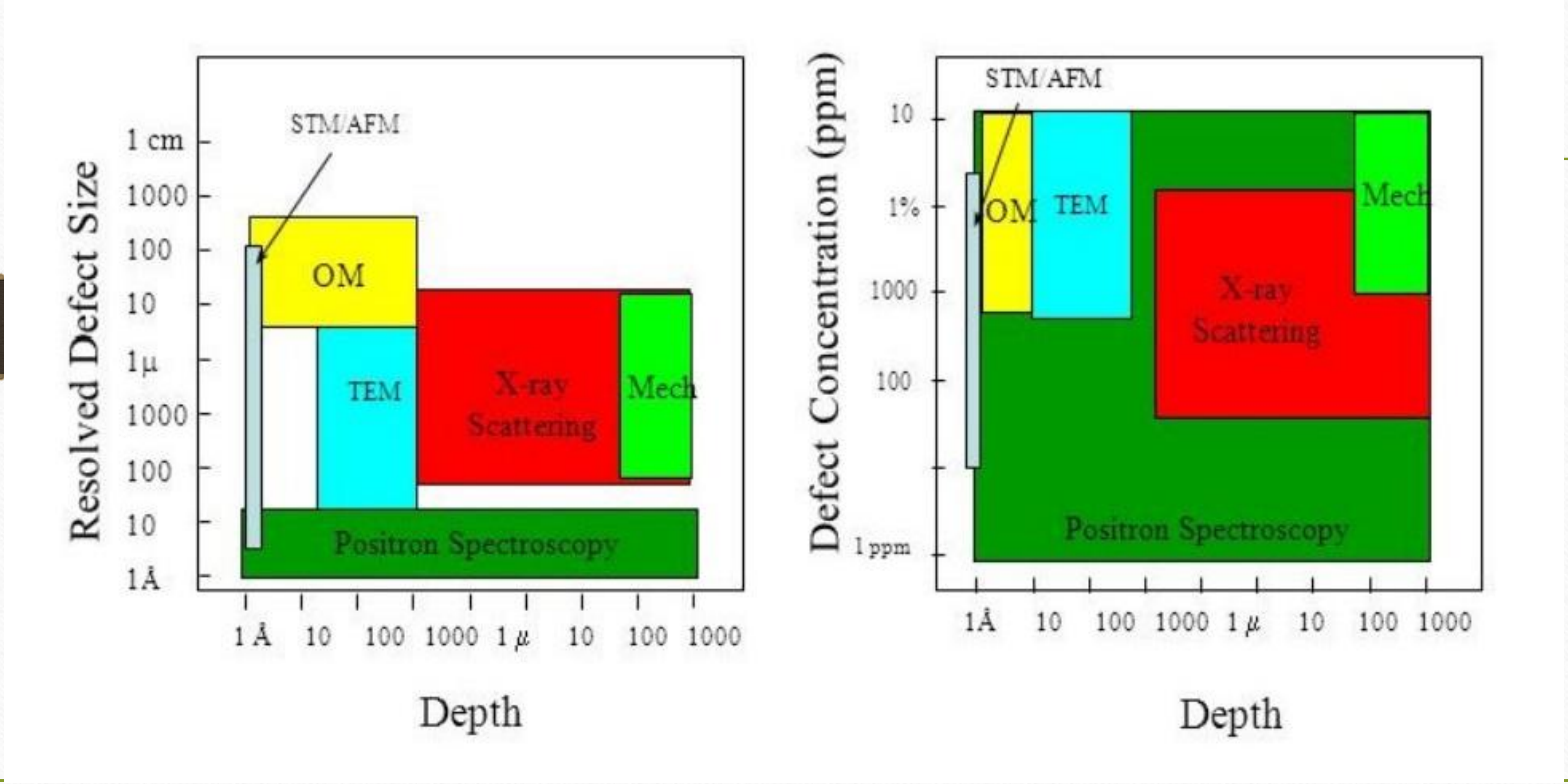
Introduction

Irradiation of materials with neutrons or ions introduce several defects such as vacancies, voids, dislocations, etc.. There are various techniques of identifying these defects such as scanning electron microscopy, transmission electron microscopy which offers high-resolution imaging of the internal structure and thus enabling the detection of defects in the bulk[1].

Introduction

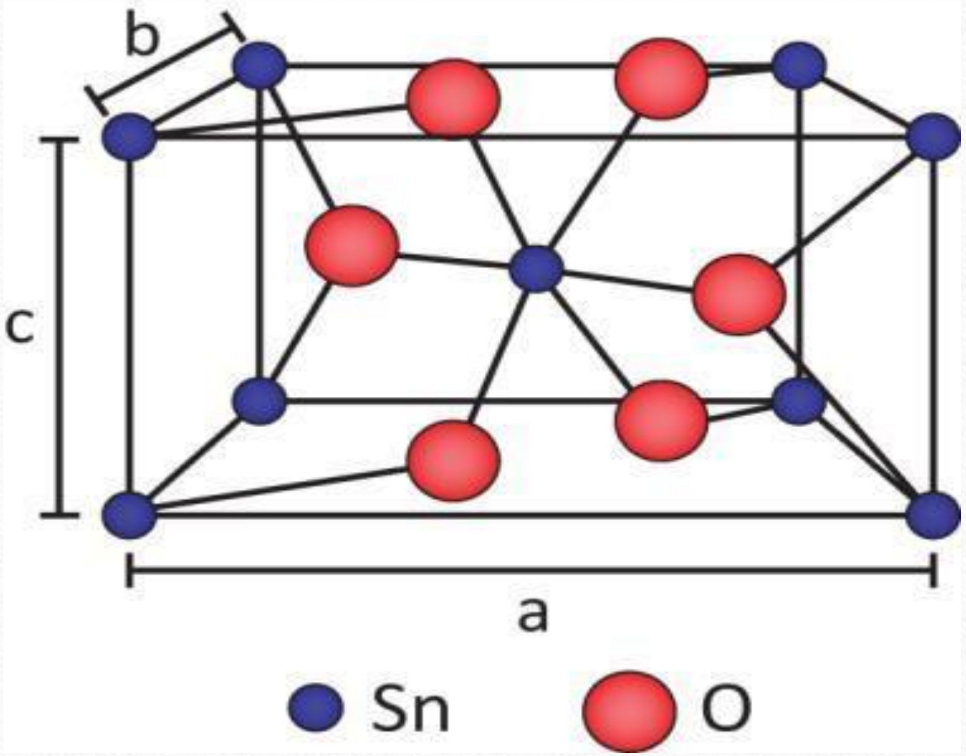
- In this work, we use positron annihilation technique (PAS) to probe defects. PAS is a non-destructive technique and very sensitive small defects such as point defects to large defects.

Introduction



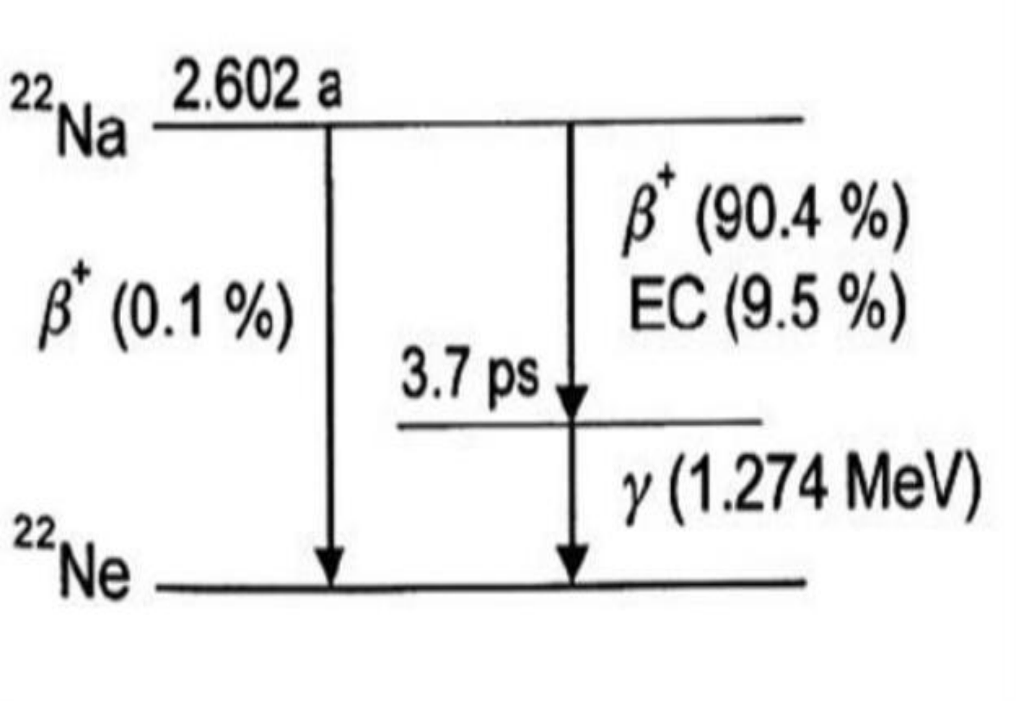
Structure and properties of SnO_2

- Figure 1[2], displays tin oxide structure showing atomic positions of both Sn and Oxygen.
- Tin Oxide (SnO_2) crystallizes crystallizes in a tetragonal rutile structure, which belongs to the P42/mnm space group.
- SnO_2 has lattice constants a = 4.7374 Å and c = 3.1864 Å and the melting point of $1630°C$ with the density of $6.95 g/cm^3$.



Positron emission

- ^{22}Na beta decays into an excited state of $^{22}Ne^*$ as shown in figure 2 [3] .
- 1.27MeV signals the start of positron lifetime[4].
- Positron has energy distribution up to an energy of 0.5MeV and can penetrate deep into a sample.

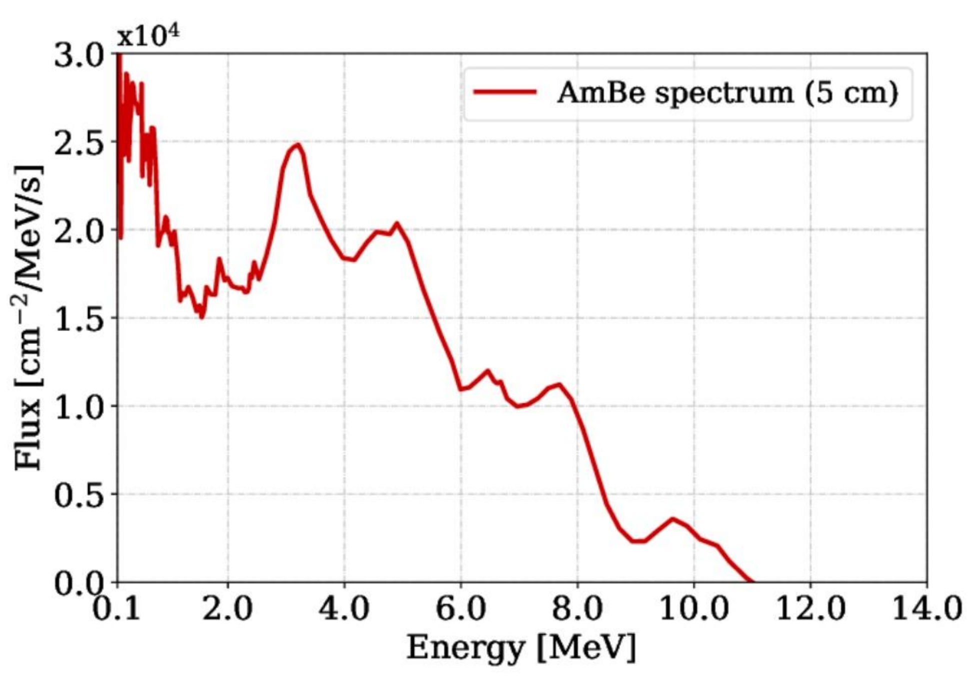


Sample Preparation

- Pellets of diameter 1cm and thickness of 0.6cm were made from powder form of tin oxide as shown in figure 3.
- Mass of powder to be pelletized was calculated from $m = \rho\pi(\frac{d}{2})^2 x\ w$.
- Where w is the width and d is the diameter.

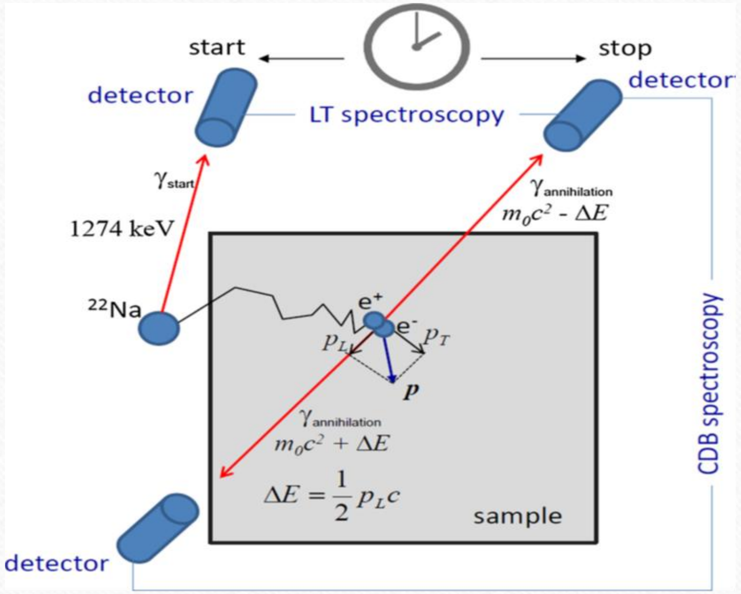
Irradiation of samples using AmBe

- Flux of neutrons from AmBe over a wide range of neutron in figure 4 [7].
- Activity of the source – 140 MBq
- Paraffin wax of thickness 3 cm was used to slow down neutrons and the sample sample was exposed for 24hrs.
- $E=E_0\ e^{-x/\lambda}$ where λ is the mean-free path for neutron scattering in paraffin, x is the thickness of paraffin

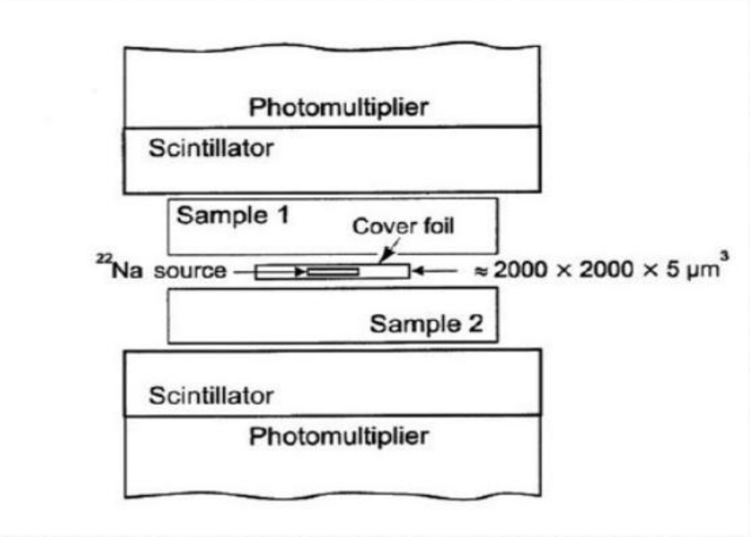


Positron annihilation spectroscopy

Figure 5 [5] below illustrate of how the positron lifetime and Doppler broadening are measured.

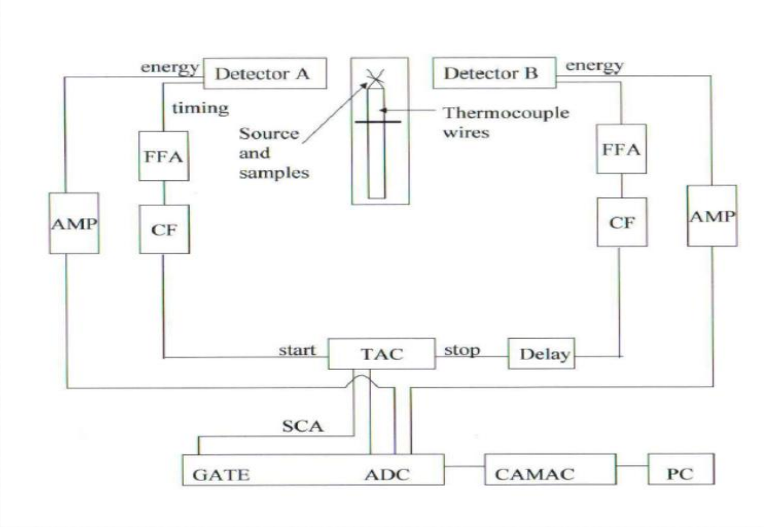


Sample-source sandwich arrangement for the registration of the birth and annihilation photon as start and stop signals is shown in figure 6 [4] .

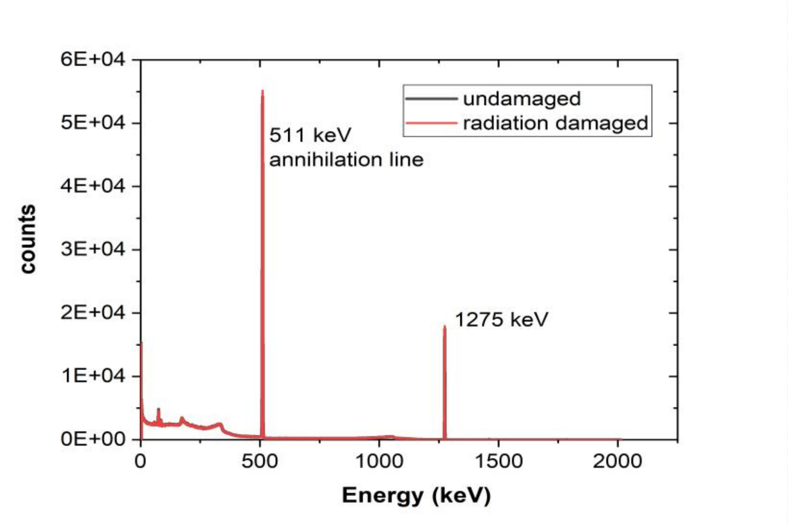


Positron annihilation spectroscopy : Annihilation spectra

Circuit diagram in figure 7 [6] for coincidence measurements for both Doppler broadening and positron lifetime.

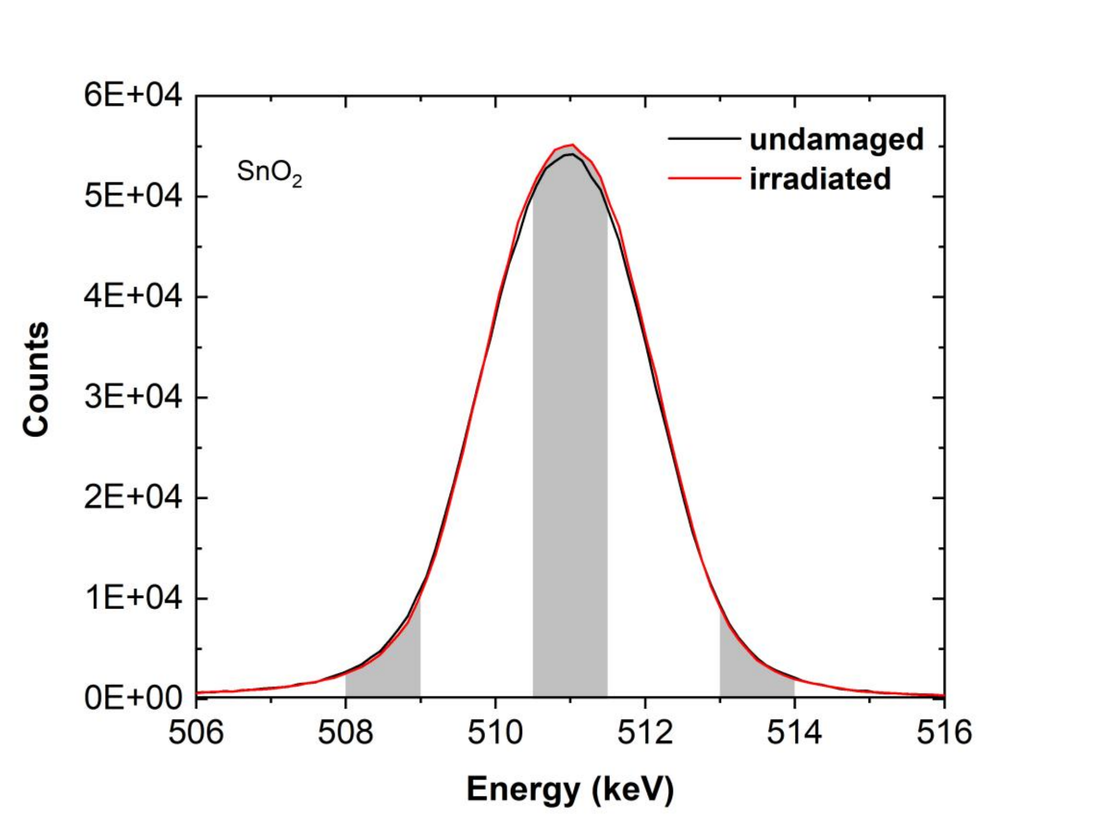


The annihilation spectrum obtained as shown in figure 8 below with energy resolution of HPGe (High Purity Germanium 2keV full width at half maximum (FWHM) at 511keV.



Positron annihilation spectroscopy: Annihilation parameters

- Figure 9 is the annihilation curve expanded around the centroid.
- S-parameter was calculated using this formular: s-parameter $= \frac{A_s}{A_{total}}$
- For A_s we used energy interval of $511 \pm 0.5\ keV$.
- And A_{total} we used energy range 511keV.



Positron annihilation spectroscopy: Annihilation parameters

Table 1 shows the calculated annihilation values and parameters. Also, table 1 clearly shows that the quantity of defects (irrespective of the type) is large in irradiated sample than in unradiated sample. The type and nature of defects are obtained through the positron lifetimes and their respective intensities.

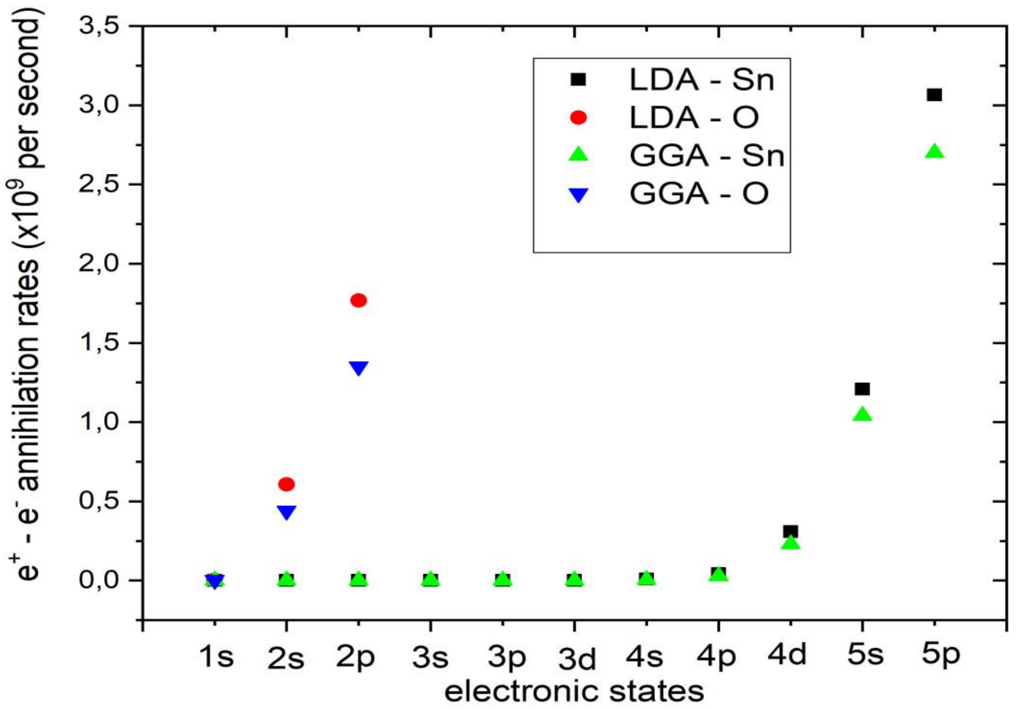
	Unradiated	**Irradiated**
Centroid area	48999	478720
Total area	1267485	1279038
S-parameter	0.370812	0.374281
W-parameter	0.036882	0.033784

Theoretical approach

Two-component density functional theory (TCDFT) was used. Positrons annihilate in the bulk, defect as well as with low and high concentration electrons. Therefore, positrons can annihilate with electrons belonging to the individual atoms.

Theoretical approach: Annihilation rates

Using local density approximation (LDA) the annihilation rates are overestimated especially for low momentum electrons. Figure shows the comparison between LDA and the generalized gradient approximation (GGA) which consider the variational nature of charge density[8].



Theoretical approach: Lifetime components

- Positron lifetimes $(\frac{1}{\lambda})$ are given by:

	LDA	GGA
Bulk	220 ps	240 ps
Defect	245 ps	263 ps

- Annihilation rate, λ, is given by

$$\lambda = \pi r_o^2 c \int n_+(\mathbf{r}) n_-(\mathbf{r}) \gamma(n_-(\mathbf{r})) dr \quad (3)$$

$\gamma(n\_(r))$ is the enhancement factor in the Local Density Approximation (LDA) and in the Generalized Gradient Approximation (GGA).

Conclusion

Doppler broadening clears shows that defects are created by neutron irradiation by investigating the centroids of the unradiated and irradiated spectra. S-parameter for unradiated is smaller than the S-parameter of irradiated SnO_2. The second positron lifetime component of 263 ps suggest the formation of point defects. The next stage is to utilize variable slow positron beam to calculate the stopping profiles

References

1. Kirk, M., X. Yi, and M. Jenkins, *Characterization of irradiation defect structures and densities by transmission electron microscopy.* Journal of Materials Research, 2015. **30**(9): p. 1195-1201.
2. Cui, Z., *Solution processed metal oxide thin films for electronic applications*. 2020: Elsevier.
3. Ter-Pogossian, M.M., M.E. Raichle, and B.E. Sobel, *Positron-emission tomography.* Scientific American, 1980. **243**(4): p. 170-181.
4. Krause-Rehberg, R. and H.S. Leipner, *Positron annihilation in semiconductors: defect studies.* 1999.
5. Iwai, T. and H. Tsuchida, *In situ positron beam Doppler broadening measurement of ion-irradiated metals–Current status and potential.* Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 2012. **285**: p. 18-23.
6. Chantry, P., *Doppler broadening in beam experiments.* The Journal of Chemical Physics, 1971. **55**(6): p. 2746-2759.
7. Ceccheto, M.e.a., IEEE Transactions on Nuclear Science, 2021. **99**.
8. Selim, F., *Positron annihilation spectroscopy of defects in nuclear and irradiated materials-a review.* Materials Characterization, 2021. **174**: p. 110952.