Study of neutron induced damage in ADS related metals using PAS and Monte-Carlo simulation

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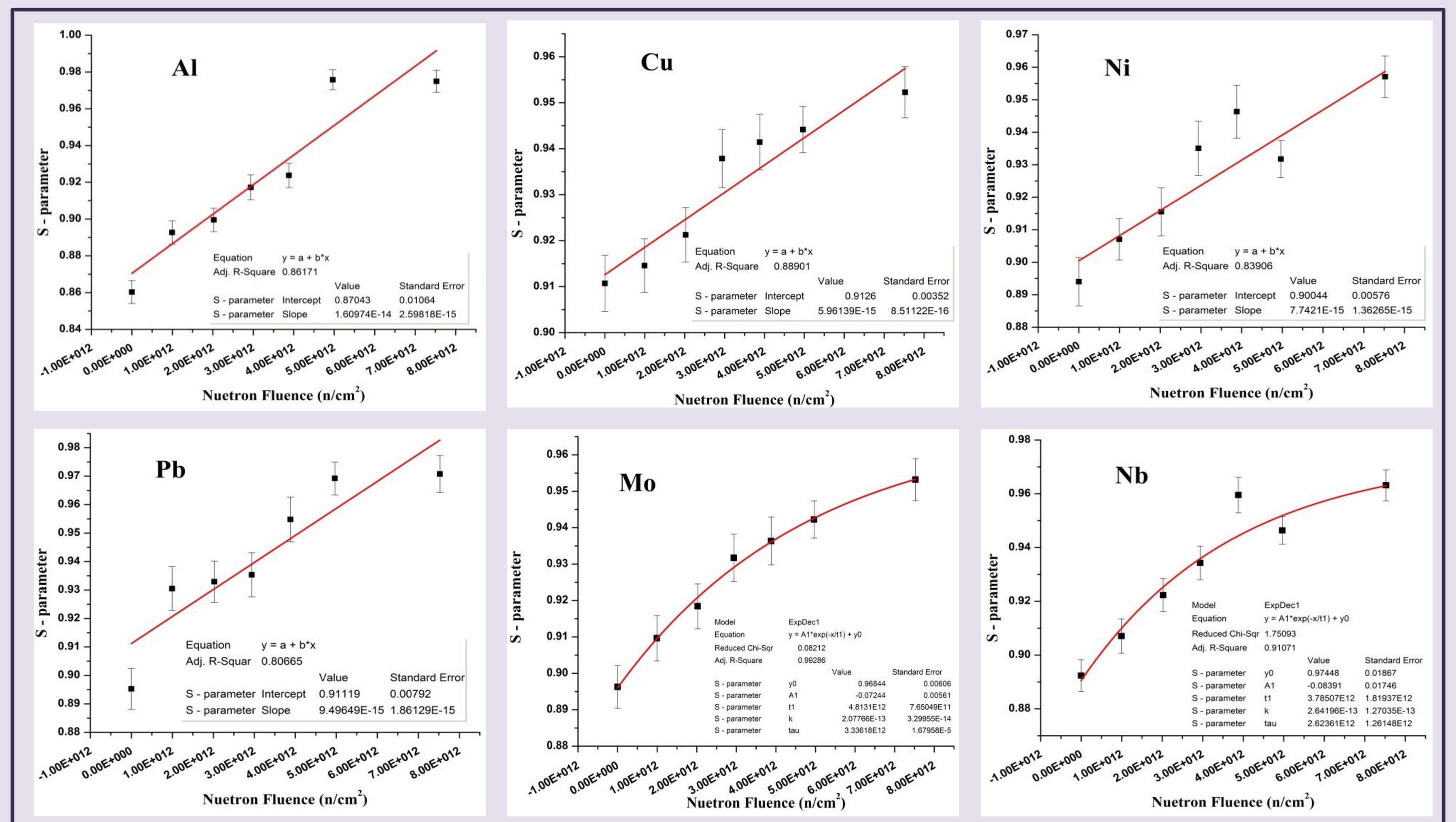
Abstract: In the present work, the effect of neutron irradiation on the samples of accelerator driven subcritical system (ADS) related metals such as aluminum, nickel, copper, niobium, molybdenum, and lead was studied using Positron Annihilation Doppler Broadening Spectroscopy (PADBS). These Samples were irradiated by a 5Ci Am(Be) neutron source. The simulated flux of emitted neutrons at irradiation position, 4 cm from source, is 3.3×10^5 n/sec/cm^2. These measurements of PADBS and electric resistivity were taken for different irradiation period from 35days to 264days. Doppler broadening of annihilation peak for all samples at different irradiation time periods was characterized by the determination of S and W parameter. The S parameter of materials increases with neutron irradiation time or neutron fluence whereas the W parameter decreases. Also, the resistivity increases by irradiation. The Simulation work was also done for the study of radiation damage by neutron irradiation using JA-IPU Monte-Carlo code.

Introduction: The neutron irradiation causes some defects in the metals and affects mechanical [1] and electrical properties [2]. Positron annihilation spectroscopy (PAS) is a very power full tool for condensed matter studies and material research. There are several experimental techniques which basically came from nuclear spectroscopy. Hence PAS is closely related with nuclear experimental methods. One important experimental technique is to study Doppler Broadening of positron annihilation peak. The Doppler broadening can be measured using HPGe detector. The shape of annihilation peak is characterized by shape parameters S and W. Here S parameter is the relative area of the central part of the peak and W parameter is the contribution of peak tails to the total peak area. Thus the S parameter shows the contribution of low momentum electrons to positron annihilation and W parameter reflects the contribution of positron annihilation by core electrons with high momentum. In the case of open volume defects the positron is trapped and it is observed by the increase in S parameter [3]. Positron annihilation has widely studied in metals. It makes possible to find out important characteristics of metals so that we can determine mechanical, electrical and magnetic properties of metal [4]. It was observed that S parameter of Doppler broadening measurement for proton irradiated reactor pressure vessel steel increased with proton dose [5]. In our one paper we have irradiated Al, Cu and Brass for a short time period 120 hours and then observed Doppler broadening of annihilation peak and found low changes in S parameter with irradiation time. Substantial changes have shown in the SEM pictures of pristine and irradiated samples which indicate defect production [6]. In another paper Cu, Ni and Pb are irradiated for log time duration up to 264 days then S-parameter and resistivity have been measured for different irradiation time periods. The variation of S-parameter as well as resistivity with the neutron fluence was linearly increasing which shows the continuous increment of defects with radiation [7]. In present paper we have studied the effect of neutron irradiation using PADBS and resistivity measurement. Also the simulation of radiation damage has been done using JA-IPU Monte-Carlo code.

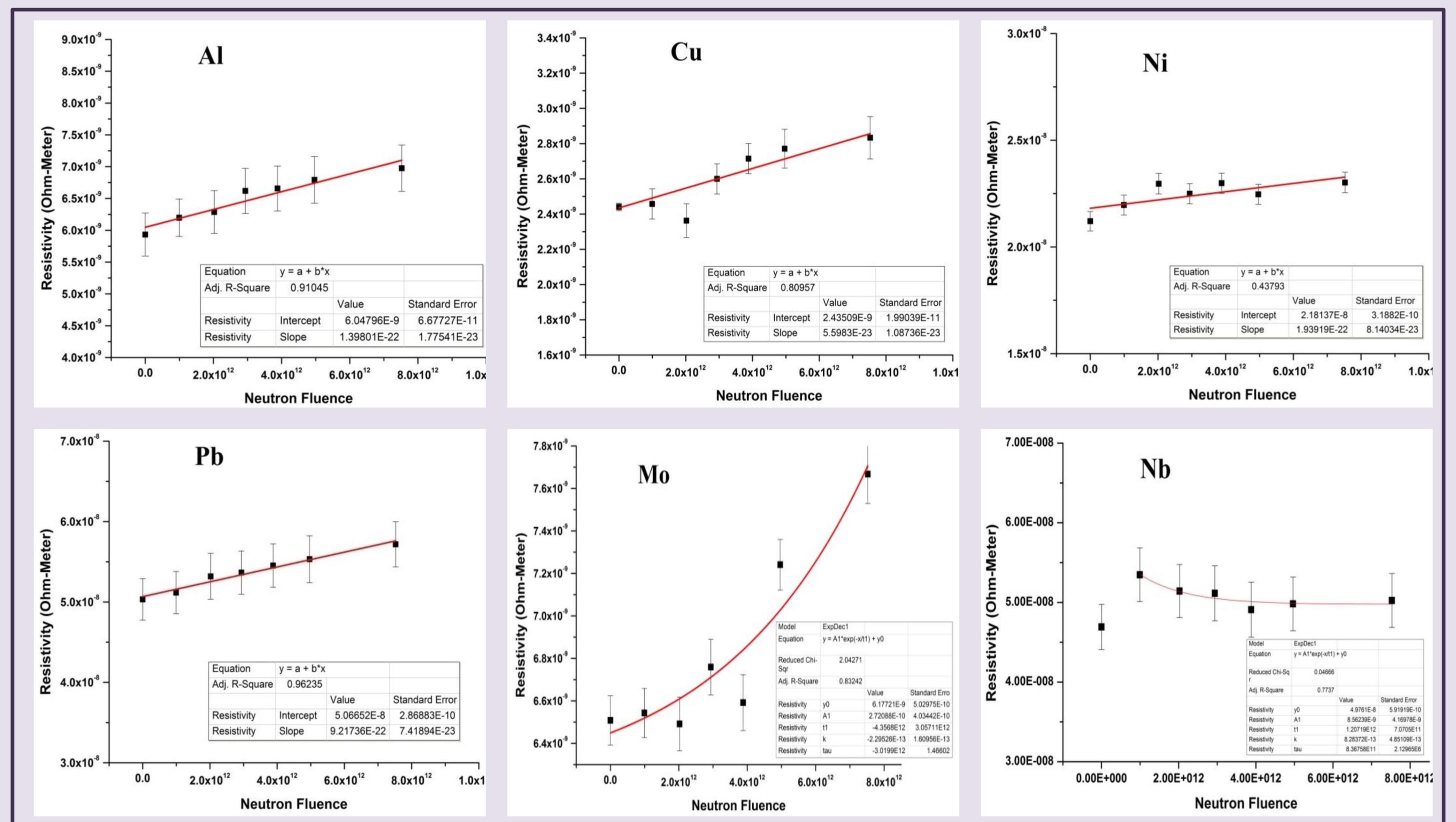
Experimental Set-up: The Am(Be) neutron source was used for the irradiation of metal samples. A 5Ci Am(Be) source placed at the centre of a cylindrical tank filled with paraffin. The samples are kept at a 4 cm distance from the neutron source. The simulated neutron flux at irradiation position is 3.3×10^5 n/cm^2/sec [8]. Experimental set up of positron annihilation spectroscopy is shown in figure-1. In the set up, radioactive sodium (Na^{22}) was used as a positron source [9]. The activity of the positron source is 35mCi. The end-point energy of ^{22}Na is at 545 keV. **A** high purity germanium (HPGe) detector was used to detect positron annihilation gamma. The relative efficiency and FWHM of detector at 1332.5 keV are 16.4% and 1.63 KeV. The distance of the detector window from the sample was 19.5 cm. A multichannel analyzer (MCA) working with Genie 2000 data acquisition system is used to store and further analysis of the gamma spectrum. Keithley Current Source and Nano-voltmeter have been used to measure the resistance of samples at different irradiation states.

Experimental Setup of PAS in the Laboratory.

Experimental Data: The Samples have been irradiated by placing them near the neutron source at a distance of 4 cm. These samples were taken out from the irradiation position for different time periods from 35 days to 264 days to investigate the effect of neutron irradiation. These samples are examined by positron annihilation Doppler broadening spectroscopy (PADBS) and resistivity measurements at different irradiation states.



Variation of S-parameter of positron annihilation peak with neutron fluence.



Variation of Resistivity of materials with neutron fluence.

Simulation Data: The Monte Carlo JA-IPU code has been developed in the High Energy Nuclear Physics laboratory, University of Rajasthan, Jaipur [8] for the simulation of the radiation damage in materials induced by intermediate and high energy neutrons [10]. Simulation of radiation damage in SiC and Nb has also been done by this code [11]. The code has been written in Microsoft Visual Basic 6.0 language [12] and it can be run in many operating systems like Windows XP, 7, 8 as well as Linux. Initially, some essential information is required for the simulation work by JAI-PU code.

The neutron cross-section for the JA-IPU code was taken from the evaluated cross-section data given in the ENDF-BVII.0 library [13] and the algorithm of IOTA code [14] is used in JA-IPU code for the calculation of the interaction cross-section.

In the present work, the Frenkel pair density, displacement per atom, defect production efficiency, and damage energy cross-section are determined for neutron irradiation on the samples [15,16]. The Values of parameters calculated by Monte-Carlo simulation for radiation damage using JA-IPU code are given in the table.

Element	En (eV)	N_d	DPA/yr	T_{dam} (keV)	σ (Barn)	σT_{dam} (Barn-keV)	$\eta = N_D/N_{NRT}$ (T_{dam})
Al	5.13E+05	1.83E+03	5.27E-11	1.36E+01	2.68E+11	8.45E+09	9.05E+00
	2.14E+06	5.87E+03	1.69E-10	6.09E+01	8.55E+11	6.48E+10	6.45E+00
	6.12E+06	2.58E+04	7.45E-10	2.97E+02	3.75E+12	1.58E+12	5.79E+00
Ni	5.65E+05	1.66E+03	2.99E-10	4.04E+00	4.68E+11	3.87E+09	2.76E+01
	2.14E+06	6.63E+03	1.19E-09	1.57E+01	1.87E+12	3.13E+10	2.86E+01
	5.56E+06	2.30E+04	4.14E-09	5.78E+01	6.52E+12	3.90E+11	2.71E+01
Cu	5.13E+05	3.27E+03	9.91E-11	2.33E+00	9.30E+11	4.80E+08	9.85E+02
	2.14E+06	8.51E+03	2.58E-10	5.85E+00	2.30E+12	3.01E+09	5.23E+02
	7.40E+06	4.36E+04	1.32E-09	3.02E+01	1.20E+13	1.93E+11	3.31E+02
Mo	5.13E+05	1.01E+03	9.20E-11	2.95E+00	4.10E+11	2.90E+09	2.29E+01
	2.14E+06	2.00E+03	1.83E-10	7.00E+00	8.27E+11	1.27E+10	1.96E+01
	6.73E+06	8.04E+03	7.34E-10	2.85E+01	3.29E+12	1.51E+11	1.90E+01
Nb	5.13E+05	2.00E+03	2.23E-11	1.41E+01	5.26E+11	1.46E+10	9.68E+00
	2.14E+06	1.88E+04	2.10E-10	1.29E+02	4.86E+12	6.55E+11	9.90E+00
	6.12E+06	2.29E+04	2.55E-10	1.64E+02	5.91E+12	1.16E+12	9.42E+00
Pb	5.13E+05	8.00E+02	5.14E-12	3.26E+00	5.85E+11	3.40E+09	1.61E+01
	2.14E+06	9.51E+03	6.11E-11	4.20E+01	7.03E+12	2.81E+11	1.52E+01
	6.12E+06	1.85E+04	1.19E-10	8.50E+01	1.38E+13	1.27E+12	1.46E+01

Result and Discussion:

Experiment : The increased value of the S parameter of irradiated samples indicates the generation of open volume defects by neutron irradiation [3,7]. It can be seen that the S parameter is linearly increasing with neutron fluence for the samples of the materials having fcc lattice structure (Al, Ni, Cu, and Pb). It shows that neutron irradiation continuously increasing the defects in these materials. However, for the samples having bcc lattice structure (Mo and Nb), the S parameter increases exponentially which indicates that the defect production due to neutron irradiation tends to increase with neutron fluence and saturate at higher neutron fluence.

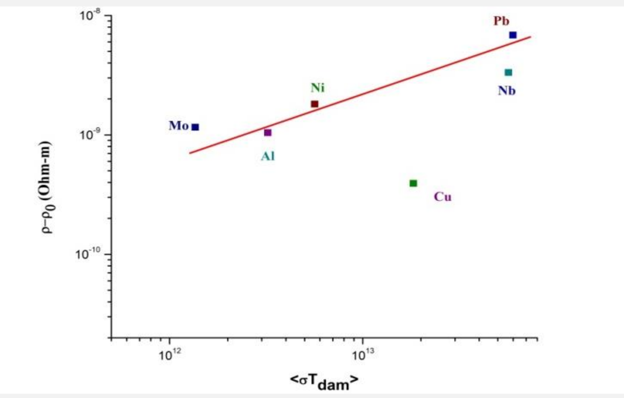
Similarly, the lattice defects created due to displacement of atoms by neutron irradiation also alters the electrical resistivity of material [2]. The resistivity of Al, Ni, Cu, and Pb samples increases linearly with neutron fluence whereas resistivity for Mo increases exponentially with neutron fluence. But, the resistivity of Nb increases for the first irradiation state and then decreases exponentially and saturates for higher neutron fluence. The saturation value of resistivity is higher than the pristine value.

Simulation :The order of DPA/yr for these samples is almost the same (~10^{-7}). It can be seen that the element-wise DPA are as follows-

$$DPA_{Pb} < DPA_{Nb} < DPA_{Mo} < DPA_{Al} < DPA_{Cu} < DPA_{Ni}$$

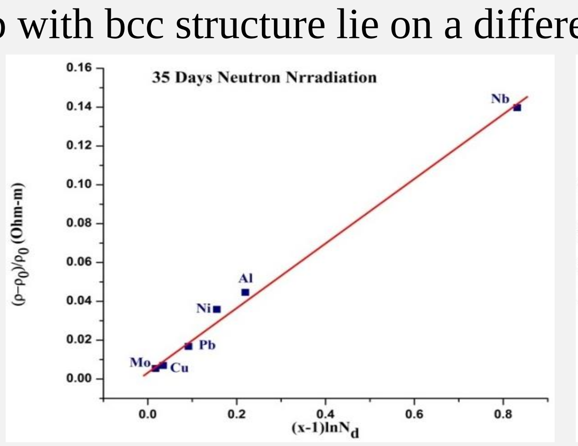
The efficiency (η) of defect production is large for all samples at low neutron energy then rapidly decreases and becomes almost constant at high neutron energy. The fact behind the decrease of efficiency value with the increase of neutron energy is that the neutron makes only a few collisions inside the sample at higher energies and there are more chances of its escape out because of the thin sample size.

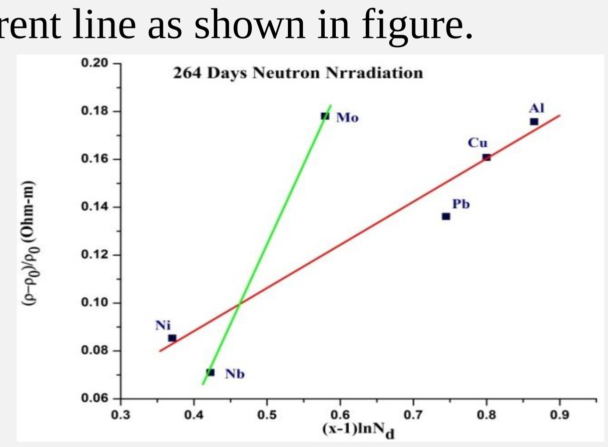
The resistivity as well as damage energy cross-sections (σT_{dam}) increase with neutron energy. The element-wise values of enhanced resistivity, ($\rho - \rho_0$) Ohm-m due to 264 days irradiation by Am(Be) source and simulated average damage energy cross-section, <σTdam> barn-keV have been plotted in figure. The points are almost at the same straight line. This straight-line relation is irrespective of material and establishes the hypothesis envisaged by Broeders and Konobeyev [15].



The rate of change in resistivity can also be related to the number of displaced atoms in the sample. According to J.A. Brinkman [17], the resistivity is related to the disordered volume of the sample and finally it can be correlated with number of displacements [18]. The relation between the rate of change of resistivity and $(x\text{-}1)\ln N_d$ is shown in figures for the 35 days and 264 days irradiation state of samples. For initial neutron irradiation the rate of change of resistivity of all samples has a linear relationship with $(x\text{-}1)\ln N_d$. But at the higher irradiation state of 264 days four elements Al, Ni, Cu, and Pb having fcc structure lie on one line whereas two elements Mo and Nb with bcc structure lie on a different line as shown in figure.

The Mo and Nb were also shown different behavior in the variation of resistivity and S and W parameters. These observations conclude that the defect production by neutron irradiation also depends on the lattice structure of the materials.





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