E-SiCure Collaboration Project: Silicon Carbide Material Studies and Detector Prototype Testing at the JSI TRIGA Reactor

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Abstract—In 2016, the "E-SiCure" project (standing for Engineering Silicon Carbide for Border and Port Security), funded by the NATO Science for Peace and Security Programme, was launched. The main objective is to combine theoretical, experimental and applied research towards the development of radiation-hard SiC-based detectors of special nuclear materials (SNM), and by that way, to enhance border and port security barriers. Along the plan, material modification processes are employed firstly to study, and secondly to manipulate the most severe electrically active defects (which trap or annihilate free charge carriers), by specific ion implantation and defect engineering. This paper gives an overview of the experimental activities performed at the JSI TRIGA reactor in the framework of the E-SiCure project. Initial activities were aimed at obtaining information on the radiation hardness of SiC and at the study of the energy levels of the defects induced by neutron irradiation. Several Schottky barrier diodes were fabricated out of nitrogen-doped epitaxial grown 4H-SiC, and irradiated under Cd filters in the PT irradiation channel in the JSI TRIGA reactor with varying neutron fluence levels. Neutron-induced defects in the material were studied using temperature dependent current-voltage (I-V), capacitance-voltage (C-V) and Deep-Level Transient Spectroscopy (DLTS) measurements. Our prototype neutron detectors are configured as 4H-SiC-based Schottky barrier diodes for detection of secondary charged particles (tritons, alphas and lithium atoms) which are result of thermal neutron conversion process in ^{10}B and ^{6}LiF layers above the surface of the 4H-SiC diodes. For field testing of neutron detectors using a broad beam of reactor neutrons we designed a standalone prototype detection system consisting of a preamplifier, shaping amplifier and a multichannel analyser operated by a laptop computer. The reverse bias for the detector diode and the power to electronic system are provided by a standalone battery-powered voltage source. The detector functionality was established through measurements using an ^{241}Am alpha particle source. Two dedicated experimental campaigns were performed at the JSI TRIGA reactor. The registered pulse height spectra from the detectors, using both ^{10}B and ^{6}LiF neutron converting layers, clearly demonstrated the neutron detection abilities of the SiC detector prototypes.

Keywords—Silicon carbide, Neutron detection, Neutron converter, JSI TRIGA reactor

I. INTRODUCTION

Increasingly complex risks like geopolitical instability or decentralized terrorism threats, have led to the urge for deploying nuclear screening systems for detection of illicit trafficking of nuclear materials, and from that, to a growing interest in the field of research and development of new radiation detection technologies suitable for homeland security applications. Recent progress in the manufacturing of high-quality bulk and epitaxial silicon carbide (SiC) and processing technologies for fabrication of SiC-based electronic devices, could enable unprecedented opportunities for future SiC-based detection of neutron and alpha particle emissions. Unlike existing and commonly used gas-based neutron detectors, SiC-based devices have the potential to be simultaneously portable, operable at room temperature and radiation hard. The main objective of the E-SiCure project is to combine theoretical, experimental and applied research towards the development of radiation-hard SiC-based detectors of special nuclear materials (SNM), and therefore to enhance border and port security barriers, which is achieved through studies of material radiation hardness and modification processes in order to manipulate the most severe electrically active defects

which trap or annihilate free charge carriers, by specific ion implantation and defect engineering. This paper presents the experimental activities performed at the JSI TRIGA reactor in the framework of the E-SiCure project. Section 2 presents the study of radiation-induced defects in neutron irradiated SiC diodes with Deep-Level Transient Spectroscopy. Section 3 presents the realization of prototype SiC detectors and initial functional testing with a [241]Am radiation source. Section 4 presents a dedicated experimental campaign performed at the JSI TRIGA reactor aimed at demonstrating the neutron detection capability of SiC-based detector prototypes.

II. NEUTRON-INDUCED DEFECTS IN SiC

To study neutron induced defects in SiC, n-type silicon carbide Schottky barrier diodes (SBDs) were fabricated onto nitrogen-doped epitaxial grown 4H-SiC single crystal layers approximately 25 μm thick, their lateral dimensions being 1 mm by 1 mm [1]. The Schottky diodes were irradiated either upon bare exposure or inside Cd thermal neutron filters with a wall thickness of 1 mm in the 250 kW JSI TRIGA reactor in Ljubljana, Slovenia. The irradiations were performed in the Pneumatic Tube (PT) irradiation facility, located in the F24 position in the outermost F ring of the reactor; the neutron fluences ranged from 10^8 n cm^{-2} to 10^{15} n cm^{-2}. The neutron spectrum in the PT irradiation channel was obtained from Monte Carlo neutron transport calculations with the MCNP6 code [2] in conjunction with the ENDF/B-VII.1 nuclear data library [3]. A verified and validated computational model of the JSI TRIGA reactor was employed, based on the JSI TRIGA criticality benchmark model [4], featured in the International Handbook of Evaluated Critical Safety Benchmark Experiments [5]. The computational model is in permanent use at the Reactor Physics Department of the JSI for research purposes (computer code and nuclear data validation) and in support of experimental campaigns performed at the reactor [6][7][8]. Over time, the model has been considerably updated and refined. It has been validated against experimental measurements for calculations of the multiplication factor k_{eff} [4], reactor kinetic parameters [9], neutron flux and reaction rate distributions [10][11][12]. The neutron spectrum obtained from Monte Carlo calculations underwent a subsequent process of neutron spectrum adjustment to measured reaction rate ratios. The adjustment process was performed using the JSI-developed GRUPINT code package [13]. Fig. 1 displays the adjusted bare and Cd-filtered neutron spectrum in the PT facility.

A total of 8 samples were irradiated at different reactor power levels (2.5 W, 25 W, and 250 kW), specifying different irradiation times, in order to span a range of 7 orders of magnitude in the neutron fluence delivered to the samples (10^8 n cm^{-2} to 10^{15} n cm^{-2} nominal fluence range). Activation measurements for the ^{197}Au(n, γ) reaction were performed for each power level used for the irradiations in order to deduce the total neutron flux. The subcadmium flux was obtained from the characterized neutron spectrum and the Cd cutoff energy for the thickness of 1 mm, i.e. 0.55 eV. Table 1 reports
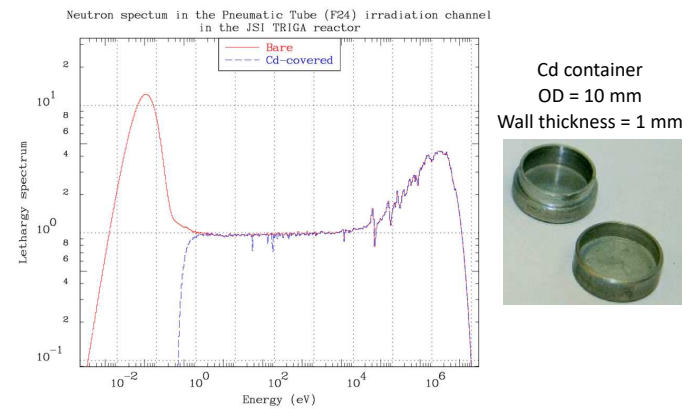

Fig. 1. Neutron spectra in the Pneumatic Tube (PT) irradiation channel, located in the F24 position in the core of the JSI TRIGA reactor. Red: bare spectrum. Blue: spectrum under Cd cover.

details of the neutron irradiation conditions for the selected samples. The calculated uncertainties in the neutron fluences were obtained from uncertainties in the neutron flux and the irradiation time.

TABLE I
IRRADIATION DETAILS, SUBCADMIUM NEUTRON FLUX LEVELS AND 1-σ EXPERIMENTAL UNCERTAINTIES, SAMPLE NEUTRON FLUENCE VALUES AND 1-σ EXPERIMENTAL UNCERTAINTIES.

ID	Irr. time [s]	P [W]	Sub-Cd flux [n cm^{-2}s^{-1}]	Fluence [n cm^{-2}]
R6	3.0	2.5	$3.66 \times 10^7 \pm 2.7\%$	$1.10 \times 10^8 \pm 9.8\%$
R10	30	2.5	$3.66 \times 10^7 \pm 2.7\%$	$1.10 \times 10^9 \pm 2.9\%$
R11	300	2.5	$3.66 \times 10^7 \pm 2.7\%$	$1.10 \times 10^{10} \pm 2.7\%$
R9	273	25	$3.44 \times 10^8 \pm 2.9\%$	$9.39 \times 10^{10} \pm 2.9\%$
R12	2730	25	$3.44 \times 10^8 \pm 2.9\%$	$9.39 \times 10^{11} \pm 2.9\%$
1E13	2.73	250k	$3.66 \times 10^{12} \pm 2.7\%$	$1.00 \times 10^{13} \pm 10.7\%$
IE14	27.3	250k	$3.66 \times 10^{12} \pm 2.7\%$	$1.00 \times 10^{14} \pm 2.9\%$
1E15	273	250k	$3.66 \times 10^{12} \pm 2.7\%$	$1.00 \times 10^{15} \pm 2.7\%$

Neutron induced defects in the samples were studied using temperature dependent current-voltage (I-V), capacitance-voltage (C-V) and Deep-Level Transient Spectroscopy (DLTS) measurements. I-V and C-V measurements were carried out using a Keithley 6487 Picoammeter/Voltage Source and a Keithley 4200 Semiconductor characterization system. DLTS and Laplace-DLTS measurements were carried out using a Boonton 7200 capacitance meter, a NI PCI-6251 DAQ and Laplace DLTS software. DLTS and Laplace-DLTS measurements were performed in the temperature range from 100 K up to 380 K. Temperature dependent I-V measurements revealed that the fast neutron irradiation did not affect the transport properties of SBDs for neutron fluences lower than 10^{12} n cm^{-2}. An increase in the series resistance and decrease of the capacitance of neutron irradiated samples was observed first after the neutron fluence reached the value of 10^{12} n cm^{-2}, followed by even more pronounced changes in the sample irradiated with the fluence of 10^{13} n cm^{-2}. The quality of SBDs was not satisfactory for DLTS measurements above

the neutron fluence of 10^{14} n cm^{-2}. Temperature dependent C-V measurements revealed uniform changes in the free carrier concentration, showing a small decrease for the highest fluences.
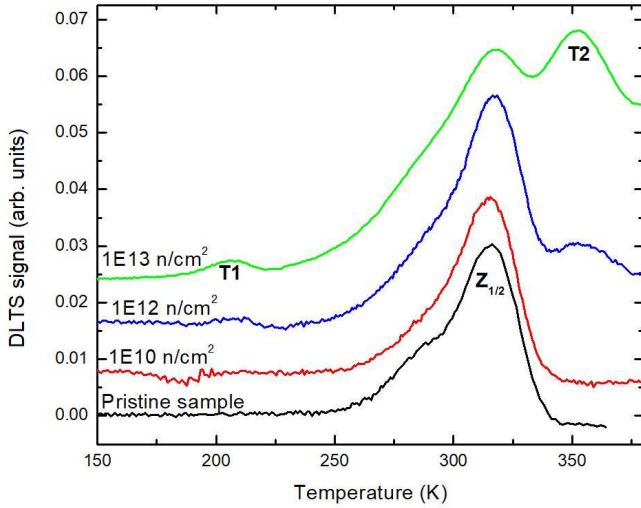

Fig. 2. DLTS spectra of pristine and selected neutron irradiated SBDs. Measurement setting are: emission rate 50 s^1, reverse bias V_R = 10 V, pulse bias V_P = 0.1 V, and pulse width τ_p = 10 ms. The spectra are shifted vertically for clarity.

Fig. 2 shows DLTS spectra for the pristine SBD and selected neutron irradiated SBDs. In the pristine sample, only one broad and asymmetric DLTS peak is observed. The estimated activation energy for electron emission was 0.64 eV. This defect, known as $Z_{1/2}$ has already been reported and was ascribed to the carbon vacancy V_C(=/0) transition [14]. V_C is known as one of the most stable defects in SiC, acting as a strong minority carrier recombination center in n-type material. The intensity of $Z_{1/2}$ slightly increases with the neutron fluence. Additional DLTS peaks, which we have labelled as T1 and T2, have been detected for neutron fluences of 10^{12} n cm^{-2} and 10^{13} n cm^{-2}, respectively. The activation energies for electron emission are estimated at 0.36 eV and 0.70 eV. The T1 and T2 traps have been reported in the literature in numerous instances, however, information regarding these defects, which regularly appear in implanted or irradiated 4H-SiC samples are very limited and controversial. We suggest that T1 and T2 are intrinsic, simple radiation induced point-like defects.

III. PROTOTYPE DETECTION SYSTEM AND INITIAL TESTING

Two main components of any detector system are 1) the detector of particle events and 2) the electronic system for processing and recording of events. The prototype detectors were based on hexagonal 4H-SiC-based Schottky diodes with several active layer thicknesses, ranging from 25 μm to 170 μm, their lateral dimensions being 1 mm by 1 mm. The detectors consisted of SBDs surface-mounted onto chip carriers with electrical contacts. The chip carriers were enclosed

in 3D-printed plastic holders in order to completely enclose the electrical contacts under high voltage (Fig. 3a). The holders had an opening above the SBD to enable charged particles to reach the SBD surface. The electronic system for particle event processing and recoding consisted of a preamplifier and a shaping amplifier PCB modules (CR-150-R5 and CR-160-R7) manufactured by CREMAT, and a digital signal processing multichannel analyser (DSP-MCA) manufactured by AMPTEK (model no. 8000D), connected to a laptop computer (Fig. 3b). Power to the electronic system (preamplifier and shaping amplifier) was provided by a standalone battery-powered voltage source, in order to avoid the use of mains power. The latter often carries electronic noise which can negatively affect the measurement performance. The voltage source also provided power to a separate high voltage bias module. Reverse negative bias was connected to the front Schottky contact, and the back Ohmic contact of the prepared prototype detectors was grounded. All initial tests have been performed in low-AC-noise environment of the ANSTO detector lab.
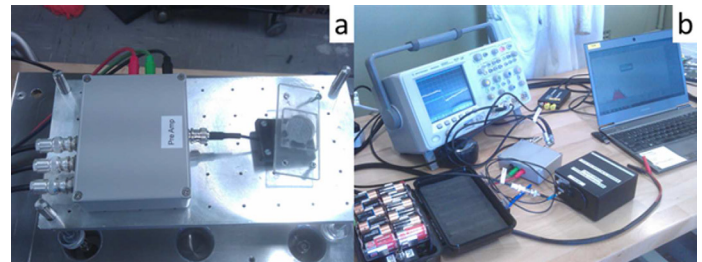

Fig. 3. Detector system under development: a) A SBD detector being tested using a ^{241}Am source. b) Electronic system for operation of a detector and data acquisition during bench testing at ANSTO.

The detection set-up was tested using a precision pulse generator and a ^{241}Am source of energetic alpha particles. For a low capacitance input ($\sim$10 pF) to the preamplifier stage we used a CREMAT CR-110-R2 preamplifier chip and tested the detection energy resolution with the shaping PCB module equipped with the CR-200-R2 chip with shaping times of 0.5 and 1.0 μs. The total gain for event signal amplification was kept constant. Pole/Zero (P/Z) was adjusted for different shaping constants. Recorded events were sorted by the MCA in 2k-channel energy spectra. The pulser peak and the ^{241}Am peak were fitted with Gaussians, given by the following expression:

$$y = y_0 + \frac{A}{w\sqrt{\frac{\pi}{4\ln 2}}} \exp\left(-\frac{4\ln 2(x - x_c)^2}{w^2}\right) \qquad (1)$$

where y is the number of detected counts in the channel x, x_c is the centroid, A determines the height of the peak and w the peak width. Fig. 4 shows recorded alpha particle spectra. Although the energy resolution for line-pulse detection is very similar for reverse bias voltages of 50 V and 100 V, the calculated width, FWHM and corresponding energy resolution for reverse bias of 50 V have lower values compared to reverse

bias of 100 V. Therefore we performed neutron detection tests using a reverse bias of 50 V and a shaping constant of 1 μs. The initial testing results demonstrated well the functionality of the detector prototypes and the electronic measurement system.
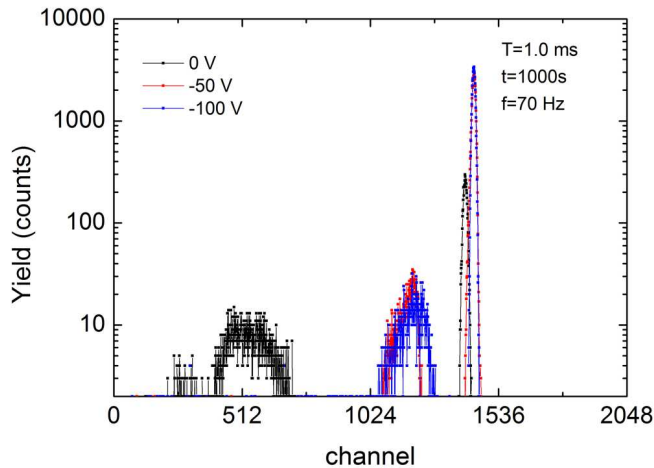

Fig. 4. Recorded ^{241}Am alpha particle spectra during initial testing with the prototype detector system and the following settings: precision pulse generator frequency: 70 Hz, reverse bias voltages: 0 V (black), -50 V (red) and -100 V (blue), total acquisition time: 1000s.

IV. TEST AND THE JSI TRIGA REACTOR

Following the results obtained from the initial testing, an experimental campaign was performed at the JSI TRIGA reactor in May 2018 aiming at demonstrating the neutron detection capabilities of the prototype detectors. For the test, four new detectors were realized, enclosed in 3D-printed plastic holders, and housed in turn in custom-made aluminium enclosures. The detector prototypes were equipped with thermal neutron converting materials for neutron detection. Due to the very large thermal neutron cross sections of ^{10}B and ^{6}Li isotopes (around 3843 b and 938 b respectively, at an incident neutron energy of 0.0253 eV [15]), the chosen converting materials were enriched ^{10}B, ^{6}LiF and ^{10}B$_4$C powders. The ^{10}B and ^{6}LiF powders were applied onto plastic film and mounted above the openings of the 3D-printed SBD holders. One of the SBDs was realized with a capping layer of ^{10}B$_4$C, in physical contact with the SBD. For the preparation of the ^{10}B$_4$C capping layer, firstly, the whole surface of a SiC SBD mounted onto a chip carrier was covered with an adhesive material in order to prevent leakage current. After drying, ^{10}B$_4$C powder was dropped onto the adhesive surface on the SiC SBD and encased in a second adhesive layer. Fig. 5 shows two realized prototype detectors used in the experimental tests (fully enclosed and open).

Firstly, the detectors were tested with no neutron converters (except for the detector with the ^{10}B$_4$C capping layer) and no radiation sources. All the obtained spectra showed a large peak at low channel numbers, which was attributed to electronic noise, and no counts appeared at higher channels. It was also observed that counts started to appear at higher channels if the laptop computer was connected to mains power or if the mousepad of the laptop computer was touched. Therefore, all the subsequent measurements were performed with the laptop computer running only on battery power and using a wireless mouse for control. The detectors were then tested using a ^{241}Am source, its nominal activity being 416 kBq (reference date 29.5.2018). For the three detectors without the ^{10}B$_4$C capping layer a clear peak due to the alpha particles emitted from ^{241}Am at high channels was observed, confirming the functioning of the SBDs as detectors of charged particles. The peak was not observed for the detector with the ^{10}B$_4$C capping layer, indicating that the detector was not functioning as expected. Following the functioning tests, the neutron converter films were applied to the detectors and neutron irradiations were performed in the Dry Chamber of the JSI TRIGA reactor. The Dry Chamber is a large irradiation room in the concrete body of the reactor, connected with the reactor core by a graphite thermalizing column [16]. It is mostly used for radiation hardness testing of detectors, electronic components and systems [17][18]. Measurements were taken firstly with a 25 μm thick prototype detector equipped with ^{10}B converter film at the following power levels: 10 kW, 50 kW, 100 kW, 180 kW and 250 kW. In all the recorded spectra we observed a significant number of counts at higher energy channels, attributed to alpha particles and recoil ^{7}Li particles originating from ^{10}B(n, α) reactions. We also observed a distinctive structure in the spectra, attributed to the different energies of the secondary particles. Fig. 6 shows the recorded spectra using the ^{10}B converter film.

Measurements were continued with a 69 μm thick prototype detector equipped with ^{6}LiF converter film, at the following reactor power levels: 10 kW, 50 kW, 100 kW, 170 kW and 250 kW. As for the ^{10}B converter film, with the ^{6}LiF converter film we also observed a significant number of counts at higher energy channels, attributed to tritons and alpha particles originating from ^{6}Li(n, t) reactions. We also observed a distinctive structure in the spectra, which was qualitatively different from
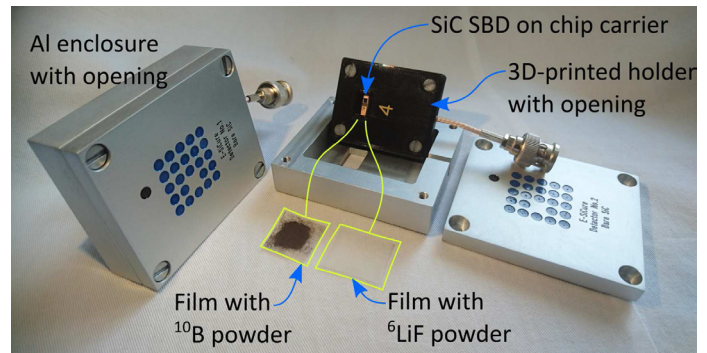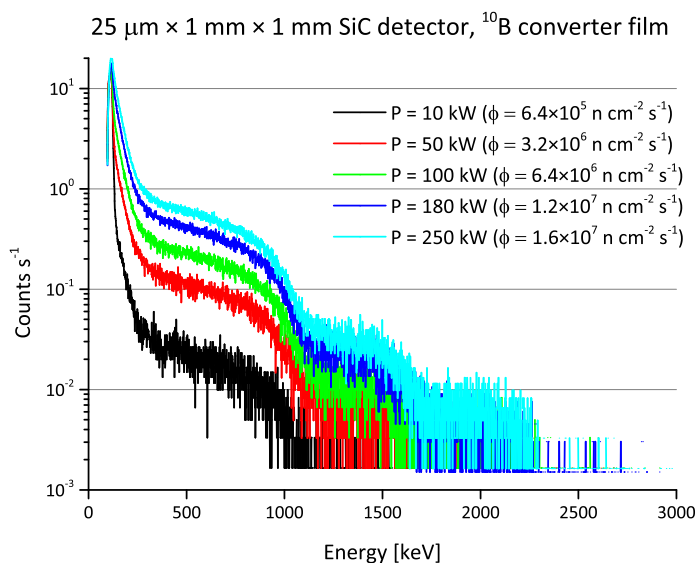

Fig. 5. E-SiCure prototype detectors. Left: assembled detector prototype in aluminium enclosure. Right: prototype detector components: SiC SBD mounted onto chip carrier with contacts, installed in 3D printed holder with opening, converter films (with ^{10}B and ^{6}LiF powder), open aluminium enclosure with opening.

Fig. 6. Measured spectra with a 25 μm SiC detector equipped with ^{10}B converter film in the JSI TRIGA reactor Dry Chamber at different reactor power levels.

the one observed using ^{10}B converter film. Fig. 7 shows the recorded spectra using the ^{6}LiF converter film.
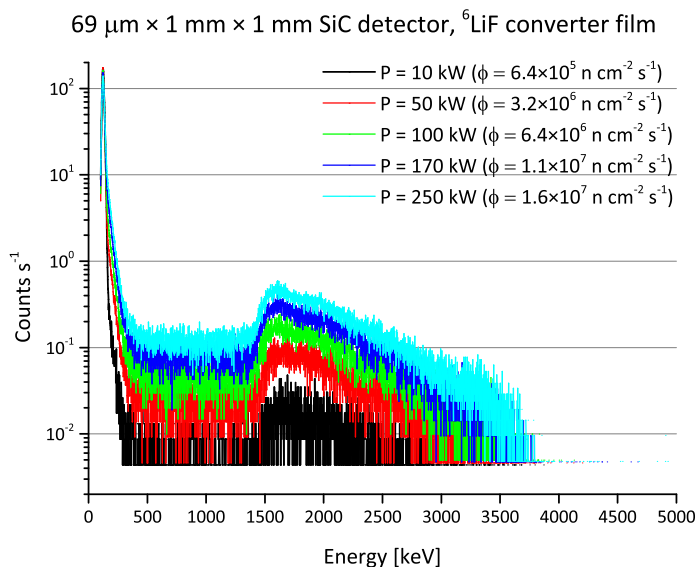

Fig. 7. Measured spectra with a 25 μm SiC detector equipped with ^{6}LiF converter film in the JSI TRIGA reactor Dry Chamber at different reactor power levels.

V. CONCLUSIONS AND FUTURE WORK

This paper gives an overview of the activities performed at the JSI TRIGA reactor in the framework of the E-SiCure project. The project aims at studying SiC material properties for the detection of SNM for border and port security. SiC-based SBDs displayed very good radiation hardness properties. Irradiation tests of detector prototypes, equipped with ^{10}B and ^{6}LiF converter films were performed in the Dry Chamber

of the reactor, clearly demonstrating the ability of small SiC SBDs (side dimensions of 1 mm × 1 mm, thickness below 200 μm) to detect alpha and recoil charged particles originating from neutron interactions. Further studies and irradiation testing will be carried within the E-SiCure project with the objective to optimize the detector and converter properties for maximal neutron detection efficiency.

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