

Testing Of Silicon Carbide Neutron Detector For Detection Of Fast Neutrons

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ABSTRACT

As the supply of ^3He diminishes, a need for neutron detectors based on other technologies than ^3He has arisen. In recent years, semiconductor detectors have become popular, especially silicon carbide (SiC) detectors. Such detectors have been developed in the E-SiCure project and further optimized in E-SiCure2 project. Optimizations have been focused on scaling the detection efficiency to thermal neutrons and expanding the detection capabilities to other radiation types, in particular fast neutrons.

A computational study was carried out in search of new neutron converter materials for the detection of fast neutrons. Among the identified candidates, the converter material of choice was potassium chloride (KCl), with two isotopes (^{39}K and ^{35}Cl) having a sufficiently high (n,p) reaction rate. Testing of the fast neutron converter material with SiC detectors was performed at the Jožef Stefan Institute TRIGA Mark II research reactor. In the experiments, two thermal neutron absorbers, cadmium (Cd) and boron carbide ($^{10}\text{B}_4\text{C}$) on copper (Cu) substrate, were mounted in front of the SiC detector to reduce the thermal neutron component as much as possible. From the measurements performed, a clear response to fast neutrons was observed even without the presence of converter material, attributable to recoil carbon and silicon nuclei.

This paper presents the preparation of fast neutron converters and the experimental testing of SiC detectors for fast neutron detection.

1 INTRODUCTION

Neutron detector devices serve many roles in this day and age. In the past years the use has expanded from fundamental research [1] and inspection of nuclear warheads to the prevention of illicit trafficking of radiological materials, primarily through the screening of cargo at borders [2]. Shortages of ^3He have led to increased research in the field of new neutron detection technologies. In recent years silicon carbide (SiC) semiconductor detectors have proven to

be a promising option for neutron detection devices they have a low operating voltage, good gamma discrimination capabilities and are radiation-hard. Prototype SiC neutron detectors were developed and tested in the E-SiCure project (Engineering Silicon Carbide for Border and Port Security), funded by the NATO Science for Peace and Security Program. The project E-SiCure2 is focused on further optimization by scaling the detection efficiency. Emphasis is also given to the expansion of detection capabilities to other types of radiation, especially to fast neutrons.

SiC detectors detect neutrons indirectly by detecting charged particles that result from neutron interactions with a material. This material, called a converter, is placed in front of the SiC detector, configured as a Schottky Barrier Diode (SBD). Converters for thermal neutrons have been extensively studied, especially converters based on ${}^6\text{LiF}$ or ${}^{10}\text{B}$ [3], due to the high reaction rate for production of charged particles.

Potential new converter materials for the detection of fast neutrons were identified in a computational study carried at the Jožef Stefan Institute (JSI) [4]. KCl was chosen as the converter material, with two isotopes (${}^{39}\text{K}$ and ${}^{35}\text{Cl}$) having a sufficiently high (n,p) reaction rate. In this work a KCl neutron converter was realized and tested in the JSI TRIGA Mark II research reactor.

In section 2 the identification and preparation of the converter material are presented. The optimized SiC detector prototype is presented in section 3. Measurement results are shown and analyzed in section 4.

2 CONVERTER FOR DETECTION OF FAST NEUTRONS

Neutrons can only be detected indirectly in a semiconductor detector and there are two approaches to it. The first is to detect particles that are created after neutron interaction with the detector materials (Si, C). In a SiC crystal, the most probable interactions with fast neutrons ($E_n > 10$ MeV) are elastic and inelastic neutron scattering on ${}^{12}\text{C}(n,n'){}^{12}\text{C}$ or ${}^{28}\text{Si}(n,n'){}^{28}\text{Si}$ [5]. In these reactions, part of the energy and momentum of the incident neutron is transferred to Si and C (recoil nuclei), which is detected through the induced charge carriers in the semiconductor upon slowing down. The second is to add a converter layer, where neutrons interact with the converter material and the resulting energetic charged particles are detected by the SiC diode.

The most common isotopes used for detection of thermal neutrons are ${}^6\text{Li}$ and ${}^{10}\text{B}$, due to their high thermal neutron cross-sections (energy below 1 eV) and the emission of detectable charged particles (tritons and alpha particles, respectively). The relevant reactions for ${}^6\text{Li}$ and ${}^{10}\text{B}$ are ${}^6\text{Li}(n,t){}^4\text{He}$ and ${}^{10}\text{B}(n,\alpha){}^7\text{Li}$. To expand the detection capabilities of a SiC detector in terms of the neutron energy range, an analysis was performed in search of isotopes with reactions sensitive to fast neutrons that produce charged particles, such as (n,p), (n,d), (n,t), (n, ${}^3\text{He}$), (n, α) (n,t2 α) and elastic scattering on hydrogen [4]. For the analysis, a neutron spectrum calculated using the Monte Carlo particle transport code MCNP [6], using a validated computational model of the JSI TRIGA Mark II reactor [7], in conjunction with the RR_UNC code [8] for calculations of reaction rates, were used.

From the nuclear data library ENDF/B-VIII.0 [9], containing 557 isotopes, isotopes with the highest calculated reactions rates are presented in Table 1 (adapted from [4]). Table 1 lists the reaction types for different isotopes, reaction rates values and the energies at which the integral of the reaction rate is 50 % of the total reaction rate (E 50 %). Reactions produced predominantly by fast neutrons can be identified through this parameter. Reactions with a small value of E 50 % are predominantly sensitive to thermal neutrons, reactions produced predominantly by fast neutrons have a larger value of E 50 %.

All of the isotopes presented are good candidates as neutron converters for use with SiC

Table 1: Calculated reaction rates for candidate fast neutron sensitive reactions, producing charged particles. E 50% represents the energy at which the integral of the reaction rate reaches 50 % of the total reaction rate (adapted from [4]). Reactions with a small value of E 50 % are predominantly sensitive to thermal neutrons, reactions produced predominantly by fast neutrons have a larger value of E 50 %.

Isotope	Reaction	Reaction rate per src. neu. par.	E 50% [MeV]	Natural abundance [%]	Radioactive
H	Elastic scattering	8.98E-05	1.067E-07	99.98	stable
22-Na	(n,p)	4.34E-02	4.567E-08	traces	2.602 y
40-Ca	(n,p)	3.47E-07	4.48332	96.941	stable
58-Ni	(n,p)	3.45E-07	4.53248	68.077	stable
39-K	(n,p)	3.39E-07	3.46221	93.258	stable
35-Cl	(n,p)	3.24E-07	2.99003	76	stable
6-Li	(n,t)	1.41E-03	4.543E-08	7.59	stable
10-B	(n, α)	5.78E-03	4.540E-08	20	stable
33-S	(n, α)	1.13E-06	0.15735	0.75	stable
22-Na	(n, α)	4.41E-05	4.642E-08	traces	2.602 y

detectors, but the most promising ones are ^{40}Ca , ^{58}Ni , ^{39}K and ^{35}Cl , ^{33}S and ^{22}Na with E 50 % energies in the range of several MeV. The (n, α) reaction rate for ^{33}S is higher by an order of magnitude compared to other fast neutron reactions, but the E 50 % energy is significantly lower (0.15 MeV), due to the fact that the reaction is partly sensitive to thermal neutrons. The (n,p) reaction rate of ^{22}Na is seen to be extremely high, making ^{22}Na an interesting candidate as a thermal neutron detector (low E 50 %), however ^{22}Na is radioactive, with a half-life of 2.602 years. The isotopes ^{40}Ca , ^{58}Ni , ^{39}K and ^{35}Cl are solid at room temperature with a high reaction rate. This paper is focused on the converter material KCl as it has two isotopes of interest, ^{39}K and ^{35}Cl .

2.1 Preparation of KCl converter

Solid KCl was dissolved in distilled water and deposited drop by drop onto a substrate. The substrate of choice was cadmium (Cd) as it is a strong thermal neutron absorber. In the measurements two samples were used. The first one was a Cd plate with dimensions of 7.8 cm \times 3.8 cm \times 0.1 cm. On top of the plate a thin layer (53 μm) of KCl was deposited in a shape of a circle with a diameter of approximately 1 cm. The second sample was made of a concave container with inner diameter of 0.9 cm. The KCl layer thickness was 485 μm . The first KCl converter sample can be seen in Figure 2 (third component from the left).

3 SiC DETECTOR PROTOTYPE AND SETUP

Two SiC SBD detectors of the same characteristics were used in the measurements. Their active layer thickness was 25 μm and their lateral dimensions were 2 mm \times 2 mm. The SBDs were fabricated by growth of a SiC epitaxial layer onto a SiC substrate [3]. The detector prototype, developed in the E-SiCure project, that was used for the testing of KCl converter can be seen in Figure 1.

Particle event processing and recording was performed with an electronic system consisting of a preamplifier and a shaping amplifier and a multichannel analyzer, operated by a laptop.

In order to avoid the use of mains power, typically a source of electronic noise, a standalone battery powered voltage source provided power to the electronic system. The construction details on the set up can be found in the paper [3].

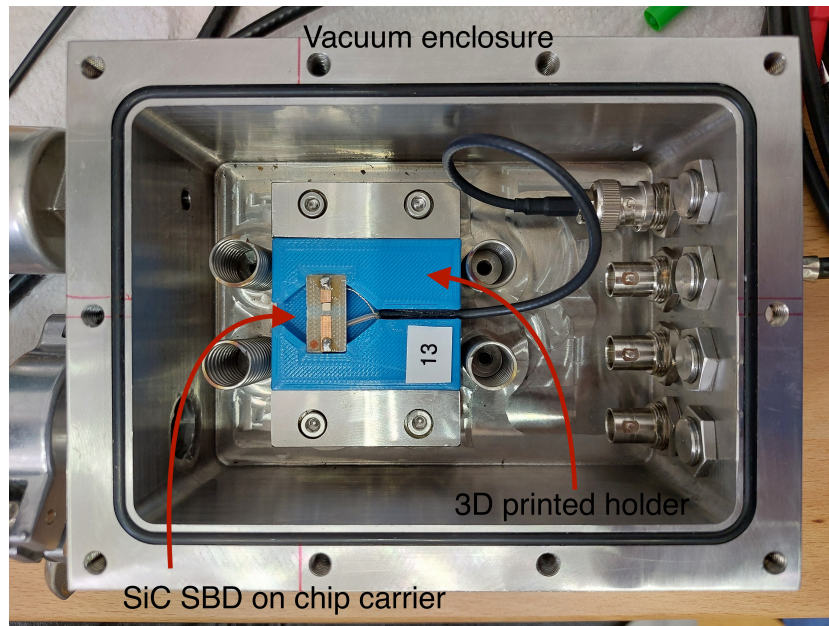


Figure 1: Detector prototype components: SiC SBD mounted onto chip carrier with contacts, installed in 3D printed holder, open stainless steel vacuum enclosure. Not displayed are converter for detection of fast neutrons made from KCl on Cd plate, placed on top of the SiC SBD, and polycarbonate lid.

The converter setup had two versions. For measurements done with a KCl converter only, the set up was the same as in Figure 1 with the addition of the KCl converter on top of the SiC SBD. For measurements done with an additional thermal absorber ($^{10}\text{B}_4\text{C}$ on Cu substrate) 3D-printed plastic spacers were used. The whole set up can be seen in Figure 2.

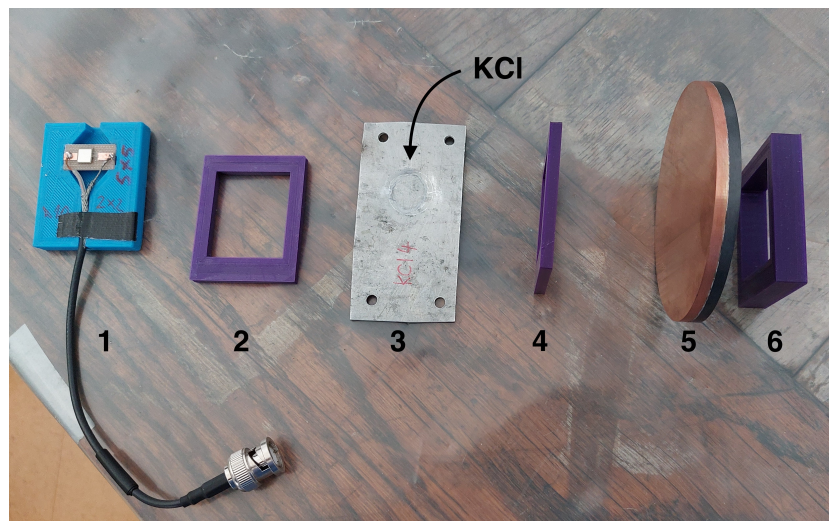


Figure 2: Components located on top of the SiC SBD. From left to right: a 5 mm \times 5 mm SBD (1) (in our case a 2 mm \times 2 mm SBD was used), plastic spacer (2), KCl converter on Cd plate (3), plastic spacer (4), thermal absorber $^{10}\text{B}_4\text{C}$ on Cu substrate (5), plastic spacer (6). The lid of the vacuum enclosure is placed on top of the last spacer.

4 FAST NEUTRON DETECTION WITH KCl CONVERTER TESTING

4.1 Energy calibration

With the aim of testing converters for fast neutron detection, an experimental campaign was performed at the JSI TRIGA reactor at the end of May 2022. The first step of any measurements with SiC detectors is functionality verification, the second is energy calibration (relation between energy and channel number). Both were achieved with the use of an ^{241}Am alpha source. The first measurements were without neutron or radiation sources, with an applied HV bias of -50 V and with no converter material. The detector was under vacuum. A large peak was observed at low channel numbers, attributed to electronic noise, and it can be seen in Figure 3. At higher channels no counts appeared. The energy calibration was made using an ^{241}Am alpha source with a nominal activity of 416 kBq. A clear peak at high channel numbers was observed in the spectra, due to alpha particles emitted from ^{241}Am .

The measurements addressed in this paper were performed in three different instances, hence three different calibrations were performed. In every measurement the alpha peak of ^{241}Am ($E = 5485.9$ keV) appeared in some specific channel range. In the measurements, we observed a slight difference in ^{241}Am alpha particle peak position (less than 3%) which can be attributed to several possible causes, e.g. different overall capacitance values of the detectors, etc. A linear relationship between the channels and energies was assumed with intercept at zero and an approximate energy calibration was obtained.

4.2 Neutron irradiation testing

After the energy calibration a series of measurements with neutron irradiations were performed in a large irradiation room in the concrete biological shield of the JSI TRIGA reactor, called the Dry Chamber. The Dry Chamber is connected with the reactor core by a graphite thermalizing column [10]. In the Dry Chamber a fission plate containing ^{235}U can be lifted to increase the fast neutron flux by the addition of fission neutrons. All the measurements were performed under vacuum at full reactor power (250 kW).

Firstly, the measurements were performed only with the bare diode, with the fission plate up and down (up meaning in use, present in the neutron beam, down meaning not in use, in its storage position). A schematic representation of the measurements can be seen in Figure 3 on the right. The two measurements were then repeated with a Cd plate with no deposited KCl converter, and lastly with a Cd plate with a KCl converter deposit, with a thickness of $53\ \mu\text{m}$. A schematic representation of the measurements can be seen in Figure 4 on the right.

In the second part of the measurements the set up included a second thermal neutron absorber $^{10}\text{B}_4\text{C}$ on Cu substrate, displayed in Figure 2. The aim was to measure only fast neutrons and minimize counts due to thermal and epithermal neutrons. Measurements were performed with the fission plate up and down.

The third part of the measurements was performed with a different SBD from the same series and with the second sample of KCl converter, with the fission plate up. The second sample KCl converter has a much higher thickness of $485\ \mu\text{m}$. A schematic representation of the measurements can be seen in Figure 5 on the right.

4.3 Measured spectra analysis

The pulse height spectra measured only with the bare diode with the fission plate up and down are displayed in Figure 3. A neutron response is clearly visible in the spectra (ranging

from approximately 600 keV to 6500 keV). The peak at low energies is attributed to electronic noise, with the neutron signal appearing as pulses of higher amplitudes. To determine the neutron count rates, a threshold was set at 600 keV. The integral count rates per second over energies are of the order of 10 cps. We observed a higher count rate when the fission plate was up. Additionally, counts were measured at higher energies, up to 8500 keV. The measured counts are attributable to recoil carbon and silicon nuclei.

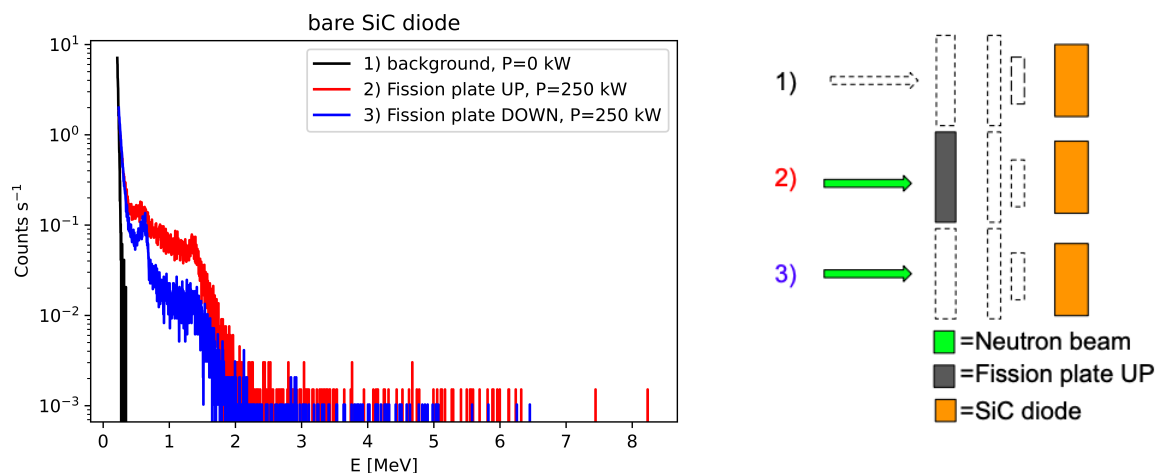


Figure 3: Left - pulse height spectra from bare SiC diode without neutron source (P=0 kW), bare SiC diode with fission plate up and down (P=250 kW). Right - schematic of the measurements set up.

Pulse height spectra measured with a SiC diode, Cd plate with and without the addition of KCl film can be seen in Figure 4. The spectra were measured with the fission plate up to increase the fast neutron component. The two spectra appear one over the other, meaning that the thinner KCl film has no appreciable impact.

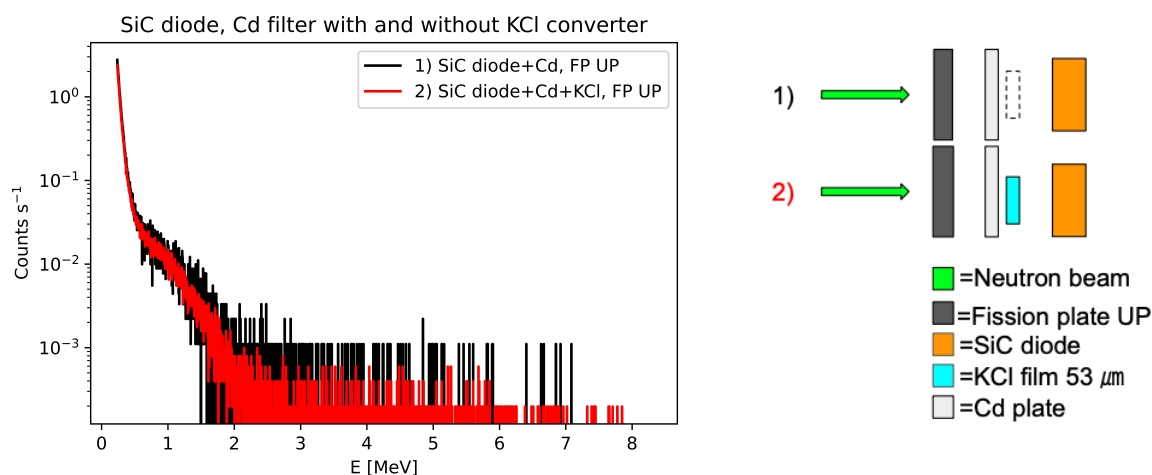


Figure 4: Left - pulse height spectra measured with the fission plate up with a SiC diode and a Cd plate with no KCl film and a SiC diode with a Cd plate with a KCl film. Right - schematic of the measurements set up.

In Figure 5, we can see a comparison between three different spectra measured by a SiC diode, with a Cd plate (no KCl converter deposit), a Cd plate with KCl deposit (53 μm) and a Cd plate with a thicker KCl deposit (485 μm). A ¹⁰B₄C thermal and epithermal neutron filter

(thickness 4 mm, on Cu substrate) was introduced above the Cd plate in the measurement with the thinner KCl deposit to further reduce the epithermal component in the incident neutron spectrum.

The spectrum measured with $^{10}\text{B}_4\text{C}$ and the thinner KCl film is lower almost by a factor of 2 above approximately 0.5 MeV compared to the spectrum measured by a SiC diode with a Cd plate, due to the neutron absorption properties of $^{10}\text{B}_4\text{C}$ in the resonance region and consequently the lower number of recoil nuclei. There is an observable difference in the pulse height spectrum measured using the thicker KCl film: up to 1 MeV it is almost identical to the spectrum measured with the Cd plate only (no KCl film), between 1 and 2 MeV it drops off more steeply. However, the experiments are as yet inconclusive as to whether the KCl film has an appreciable impact on the fast neutron detection capabilities, possibly due to different measurement setups.

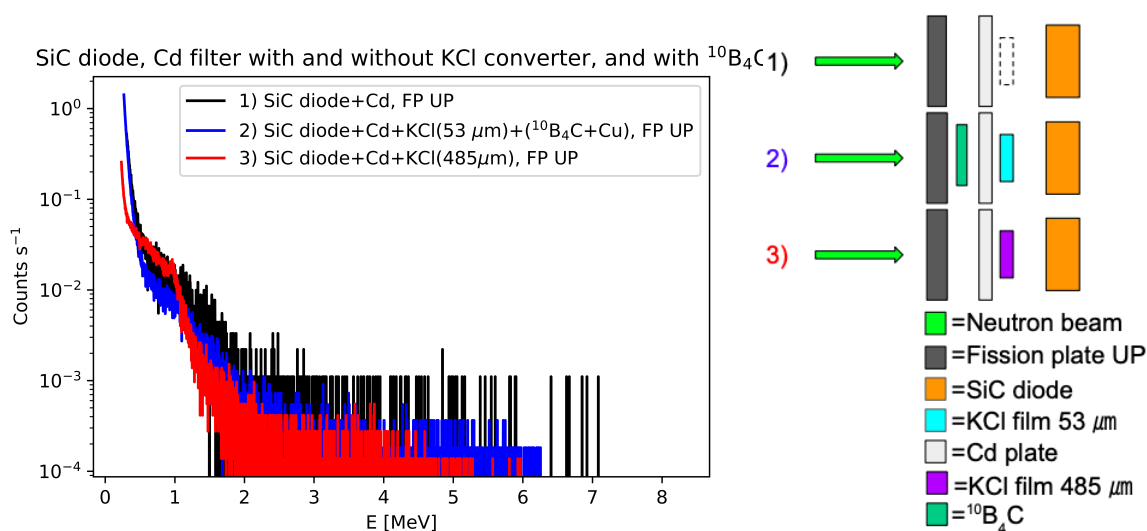


Figure 5: Left - pulse height spectra measured with the fission plate up with SiC diode with Cd plate without KCl film, SiC diode with a Cd plate with a KCl film (485 μm) and a SiC diode with a KCl film (53 μm) with thermal absorber $^{10}\text{B}_4\text{C}$. Right - schematic of the measurements set up.

5 CONCLUSION AND FUTURE WORK

This paper presents the search for fast neutron converting materials suitable for the detection of fast neutrons with semiconductor devices, the preparation and testing of an example converter with KCl as the material of choice.

It is demonstrated that the SiC detector has a fast neutron response without any converter. The measurements using KCl converter films of different thicknesses are inconclusive in terms of the fast neutron detection capabilities. The major issue in distinguishing the converter effect is the fast neutron response of bare SiC diodes, due to scattering reactions.

To be able to clearly distinguish the converter effect, in future work systematic measurements with the same set up will be performed. Firstly, the $^{10}\text{B}_4\text{C}$ will be used for measurements with all different KCl converter samples. Secondly, a shield for the SiC diode, machined or 3D-printed from borated polyethylene, is envisaged to reduce recoil nuclei in the SiC diode. A schematic of the shielded detector is displayed in Figure 6.

The presented results have increased our motivation to further study fast neutron converters and have enabled new ideas towards this goal.

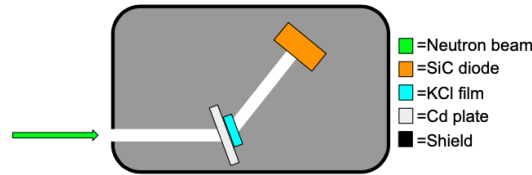


Figure 6: Schematic of the measurement setup with additional shield for SiC diode.

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