Optimisation of Signal-to-Background Ratio for Thermal Neutron Detectors

Ph.D. Thesis booklet

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Introduction

Large-scale material testing instruments are essential tools of modern research, and one of the techniques utilised is the widely-applied neutron scattering. The new flagship of this field is the European Spallation Source (ESS) ERIC, which is currently being built in Lund, Sweden, by the joint effort of 17 European member countries. The ESS has the goal to become the world's leading neutron source for the study of materials by the second quarter of this century [Peggs, 2013]. It is going to be the brightest neutron source in the world, serving instruments beyond the limits of the current state-of-the-art. The ESS is a unique, 'long-pulse' spallation neutron source with 5 MW thermal power, generated by 2 GeV protons hitting a tungsten target, equipped with an innovative 'butterfly moderator'. Due to these properties the ESS will provide an unprecedented neutron yield, which is beneficial to study smaller samples or wider phase space. However, this neutron yield and the other scientific requirements that exceed that of the state-of-the art, – such as time- and position-resolution, large sensitive area, etc. – also challenge the instrumentation, especially the neutron detectors.

One of the traditionally common neutron detectors for scattering experiments has been the ^{3}He-filled proportional counter. It has been widespread due to the excellent neutron absorption and chemical properties (i.e. non-toxic, inert etc.) of ^{3}He, the simplicity of the technique as the neutron converter also serves as the counting gas, and the affordable price and availability of the ^{3}He. ^{3}He is produced as a by-product of the fabrication of nuclear missiles; the tritium used in the warhead decays to ^{3}He with 12.33 year half-life, so it has to be purified regularly. Therefore, the two major suppliers are USA and Russia. Due to its by-product nature, on one hand, the price of ^{3}He was artificially suppressed. However, the events of 9/11 compelled the US Government to increase homeland security, including installation of radiation monitors, especially neutron monitors on state and interstate boundaries all over the US. This led to a sudden increase of demand of ^{3}He, so its litre price increased by more than an order of magnitude. This is the so-called '^{3}He crisis' [Shea, 2010]. This phenomenon highly affected the whole neutronic community as well as the construction of ESS. The decision was made that alternative technologies should be applied wherever it is reasonably achievable, without significant decrease of scientific value.

A potent alternative is the solid ^{10}B-based detector technology, used typically with Ar/CO_2 counting gas [Sauli, 1977]. The performance of these detectors is highly affected by the conversion method: the conversion particles, produced in the $^{10}B(n,\alpha)^{7}Li$ reaction have to escape from the converter layer and reach the sensitive volume of the detector. This dictates the typical coating thickness is 1–2 μm for thermal and cold neutrons, and in order to reach the detection efficiency of the ^{3}He-tubes, multiple converter layers should be applied in a repetitive structure.

These types of detectors are developed with the joint effort of several institutes, including the ESS. This need for a ^{3}He-substitute technology is the major challenge for e.g. the chopper spectrometers as these instruments require large area detectors with a large volume of counting gas. The construction of two direct geometry chopper spectrometers is decided by the ESS, the CSPEC (Cold Chopper Spectrometer) and the T-REX (Bispectral Chopper Spectrometer). The chosen detector for these is the so-called 'Multi-Grid' detector

[Andersen, 2012], invented at the Institut Laue-Langevin (ILL) and now jointly developed by ESS and ILL. This is an Ar/CO_2-filled proportional chamber with solid B_4C-converter, enriched in ^{10}B.

The basic unit of the Multi-Grid detector is the grid, which is an aluminium frame holding aluminium lamellas coated on both their sides with few μm thick converter layers of B_4C. In the detector the grids are structured into columns and modules, and each detector module is placed in a few mm thick aluminium housing. The whole detector arc is constructed from these modules: a ~3 m high cylinder wall with 3–4 m radius, covering typically 180° in angle in the horizontal plane. As the chopper spectrometers are inelastic instruments, the signal is a few orders-of-magnitude smaller than that for elastic scattering, so a driving requirement for these instruments is the excellent Signal-to-Background Ratio (SBR).

However, the application of new materials and structures in high neutron flux raise new questions and may result in new issues to encounter. The neutron activation of Ar is a well-known issue for nuclear facilities, so the exposure of the large volume of Ar/CO_2 (with 70–90% Ar) contained in various neutron detectors should be considered, as well as the activation of other detector components, such as the aluminium housing. Most instruments at the ESS will operate with thermal or cold neutrons. This leads to a high average reaction rate, which means the activation of Ar and other detector components could be of concern in terms of activity release as well as occupational exposure in the measurement hall. Moreover, the accompanying prompt- and decay-gamma radiation may also affect the measurement, contributing to the radiation background.

Another background source of concern is the scattered neutron background. The novel, large area detectors have a complex structure, typically made of aluminium, carrying the converter layers, and this massive aluminium content should be considered as a source of intrinsic scattered neutron background, i.e. induced within the detector itself. This phenomenon is especially relevant for the Multi-Grid detector, having ~3 tonnes of aluminium in a detector arc. Due to the complexity of this aluminium structure, multiple scattering volumes and surfaces have to be considered as a source of background, but this complex structure also leaves room for effective background reduction via the application of a targeted, multi-element shielding. Thanks to the great effort that has been invested in the development of neutron scattering simulation tools in the past few years, this otherwise hard-to-study intrinsic detector background can now be investigated in detail via realistic Monte Carlo simulations.

Objectives

This thesis takes part in the development of the Multi-Grid solid boron-carbide-based detector, with the aim to optimise the SBR via the development of an advanced internal detector shielding, in order to suppress the contribution of the neutron-induced background radiation produced in the detector itself. For this purpose, neutron induced gamma and scattered neutron background are determined in the detector as well as the neutron induced activity, distinguishing the sources of background via detailed Monte Carlo simulations.

Contrary to the highly geometry-dependent neutron scattering phenomenon, the neutron activation and the neutron-induced gamma background are generic issues among all Ar/CO_2-filled neutron detectors. Therefore, this study aims to provide an easy-to-scale, conservative estimation of prompt- and decay-gamma yield produced in the aluminium housing and the counting gas, for a typical incident neutron energy region of the ESS (0.8–227 meV, 0.6–10.0 Å). These data serve as input for further radiation safety planning in the ESS, e.g. in terms of potential occupational exposure. The impact of the neutron-induced intrinsic gamma background on the measurement is also shown through the Neutron-to-Gamma Response Ratio (NGR) comparing the gamma and neutron count rates. Similarly, the neutron-induced activity is determined for the whole incident neutron energy range, originating from the housing and the counting gas, providing input for further considerations of activity emission (i.e. ^{41}Ar from continuous-flush operation of the detectors) and nuclear waste management. These studies were performed parallel with MCNP simulations and analytical calculations.

The other major area of this thesis is the Monte Carlo simulation study of the intrinsic scattered neutron background and its impact on the measured signal, and the potential of background suppression with the application of complex internal shielding. The afore-described Multi-Grid detector is chosen as an example for this. The simulations have been performed with Geant4. Different sources of the intrinsic scattered background are considered, e.g. neutron scattering on the aluminium grid structure and the counting gas, scattering on the detector vessel, and especially on the entry window. The latter is a well-known challenge of neutron detector development, as it is an important mechanical structure item, being part of the vacuum interface. In order to put the impact of these sources into perspective, they are also compared with some instrument-related background sources, such as the scattering on the sample environment and the tank gas of the measurement chamber. In the study elastic and inelastic scattering are simulated as well as interaction with crystalline materials (i.e. aluminium in this case), including both Bragg diffraction and inelastic/incoherent processes. Therefore, the second aim of the study is to *i)* develop and validate a detailed, parameterised and flexible, realistic Geant4 model of the Multi-Grid detector, *ii)* use this model to distinguish and quantify the components of the intrinsic scattered neutron background from different sources and *iii)* optimise the SBR in the Multi-Grid detector via background suppression with advanced shielding design, and with fulfilling all these steps, to provide a novel, holistic approach for detector development.

Applied methods

The MCNP6.1 code is used for the gamma background and activation simulations, and analytical calculations are also performed to confirm the simulated results. The Ar/CO_2 counting gas volume is approximated as a generic, 10 cm × 10 cm × 10 cm cube, surrounded by a 5 mm thick aluminium box made of Al5754 alloy, representing the detector frame. The detector geometry has been irradiated with a mono-energetic neutron beam in the range of 0.82–227.23 meV (10–0.6 Å), from a mono-directional disk source of 8.5 cm radius at 50 cm distance from the surface of the target volume. Both the determined activity concentration and gamma intensity are scaled to a 10^4 n/cm^2/s irradiating neutron flux, conservatively estimated for standard ESS operational conditions.

The scattered neutron background study is performed with Geant4 simulations, where the aluminium-generated intrinsic background now can be simulated with the usage of the recently developed NXSG4 and NCrystal tools, which allow to model neutron interaction with crystalline materials including both Bragg diffraction and inelastic/incoherent processes. The simulations are performed within the ESS Coding Framework [Kanaki, 2018], developed in the ESS Detector Group. A detailed, realistic and flexible model of the Multi-Grid detector is implemented and validated by reproduction of measured data (ToF, flight distance, energy transfer) received from demonstrator tests at the IN6 instrument at ILL, and the CNCS instrument at SNS. For this purpose, three specific detector models are derived from the general one: the two prototypes from the validation experiments, and the planned detector module for the CSPEC instrument at ESS. The sources of scattered neutron background are identified and distinguished in the CNCS model, while the impact of intrinsic scattering on the SBR is studied in the CSPEC model, e.g. the neutron scattering on the aluminium housing, especially on the vessel window is studied and discussed. For the optimisation of SBR via advanced shielding, an ideal total absorber and common shielding materials, such as B_4C, Cd, LiF and Gd_2O_3 are tested in typical shielding geometries. In this way, the background-reducing capability of shielding applied at different locations is determined, and then the respective impacts of commonly used shielding materials are compared for each geometry.

New scientific results

1. I developed a general MCNP model for the neutron activation in Ar/CO_2-filled detectors with aluminium housing. I determined the neutron induced activity, decay and prompt-gamma yield of 80/20% by volume Ar/CO_2 and Al5457 materials of this typical detector, irradiated with mono-energetic beams in the range of 0.6–10 Å incident neutron energy. I optimised the selection of data from appropriate cross-section libraries to be used for further similar studies, giving sufficient agreement with the analytical calculations in terms of spectra, yield (max. 5–10% difference for main components) and activity (agreement within the margin of estimated uncertainty). I studied the activation of ^{40}Ar as source of gamma background for neutron signal and as source of activity release. I found that the typical daily activity production in Ar/CO_2-filled detectors, e.g. a large area detector of a chopper spectrometer, is $1.3\cdot10^6$ Bq/day (with 1.8 Å neutron irradiation of ~10^7 cm^3 volume), which can be reduced to a negligible level (considering the targeted few GBq/day total release of the facility) with 1 day retention. I also determined a particular Signal-to-Background Ratio (SBR), with the background limited to the prompt- and decay-gamma yield of the counting gas, and I found that the SBR is between 10^9–10^{10} for the whole energy range, meaning that the neutron induced gamma background of the Ar/CO_2 counting gas is negligible and no suppression is required. **[P1]**

2. I developed a detailed, realistic and scalable Geant4 model of the solid ^{10}B-enriched boron-carbide converter based, Ar/CO_2-filled Multi-Grid detector. The model was developed within the ESS Coding Framework, where I used the NXSG4 and NCrystal tools for handling the crystal structure of specific materials, therefore the effects of neutron absorption, coherent and incoherent scattering were simulated. This is the first implementation of scattering model in cold neutron scattering, which allows quantifying continuous scattered neutron background effects, they are narrower that the apparent resolution. The modelled background levels are 5–7 orders-of-magnitude lower that the always present elastic peak. The models given in **Thesis 1** and **Thesis 2** allow a detailed understanding of very low level gamma and scattered neutron background effects, giving a new approach for neutron detector development and optimisation of design for best available SBR. **[P2]**

3. I derived the models of two demonstrators from the general scalable model of **Thesis 2**: the 6-column Multi-Grid demonstrator detector that had been tested on the IN6 at ILL and the 2-column Multi-Grid demonstrator detector that had been tested on the CNCS at SNS. With these I validated the built model against the measured data both qualitatively (matching scattering profiles) and quantitatively within a sufficiently low margin of error. For this purpose, I reproduced the measured Time-of-Flight spectra and the studied scattering phenomena from the IN6 experiment with 3.15, 3.87 and 44.87 meV incident neutrons. I also reproduced Time-of-Flight, energy transfer and fight distance results from the CNCS experiment, within a sufficiently low margin of error in terms of the elastic peak, and the profile of the scattered neutron background. In addition, in the CNCS model I simulated several detector and instrument effects (e.g. scattering on the detector grid, the detector vessel, the Ar/CO_2 tank gas and a simplified sample environment) that had appeared in the measured data sets at different energies in 1.0–8.0 meV incident neutron energy range. **[P2]**

4. I performed a simulation study on the 2-column Multi-Grid demonstrator model of **Thesis 3** and determined the impact of various effects apparent in the scattering neutron background on the measured data, e.g. Time-of-Flight, energy transfer, flight distance. In the simulation I highlighted the background contribution of the different neutron scattered components, e.g. scattering on the detector grid, the detector vessel, the Ar/CO_2 tank gas and a simplified sample environment. I found that the contribution of scattered neutron background from the detector is 10% of the total simulated background below the aluminium Bragg edge, and 60% above the Bragg edge. The detector's scattered neutron background contribution is 0.1-2% of the elastic peak for the whole energy range, evenly given by the grid structure and the housing. This background contribution shows high energy-dependence and scales with the volume. Therefore, these sources of background have to be considered and optimised for full-scale detectors. **[P2]**

5. Due to **Thesis 4**, I performed a simulation study on the aluminium detector window as a source of scattering neutron background with different incident neutron energies in the range of 1.0–8.0 meV. I determined the scattering neutron rate coming from the sidewalls of the aluminium vessel and the entrance window at different thicknesses and incident neutron energies. I showed that below the aluminium Bragg edge, the background contribution of the neutron scattering on the detector frame and aluminium vessel is 0.2–0.4% of the elastic peak that is negligible. Above the Bragg edge the total background contribution is 1.7–4% of the elastic peak. I found that with a realistic 2–5 mm window thickness 80-96% of the total scattered neutron background contribution of the detector and the vessel is given by the detector frame and the sidewalls of the vessel. Therefore, the aluminium window has only a minor contribution to the scattered neutron background, and its thickness can be optimised for engineering requirements, without any relevant influence on detector operation. **[P2 and P3]**

6. On the basis of **Thesis 4 and 5,** I used the developed and validated Geant4 detector model to reduce the intrinsic neutron scattering in the Multi-Grid module of the CSPEC Cold Chopper Spectrometer of ESS. I performed this study with two different approaches. On one hand, as the conversion layer can also be interpreted as shielding, I determined the effect of the long blade coating on the efficiency and SBR. I concluded that the efficiency can be increased by 8–19%, and the SBR can be increased by 8–14% in the 5.1–511 meV energy region with the application of 1 μm $^{10}B_4C$ coating on the long blades. In terms of cost over neutron or SBR, the moderate increase in cost that can be expected by coating the long blades can be justified by the accompanying increase in SBR. On the other hand, I determined the background-reducing capacity of common shielding geometries, end-shielding, interstack-shielding and side-shielding, by applying black material. I showed that the most effective shielding geometries are the end-shielding, absorbing 10–60% of neutrons above 5.1 meV, and the side-shielding, absorbing 5–10% of neutrons through the whole energy range, while the interstack-shielding, that is the most difficult to apply, has only minor importance. Common shielding materials, B_4C, Cd, Gd_2O_3 and LiF are tested for each shielding type, and 1 mm of B_4C or Cd is proven to be equally efficient shielding as the total absorber. With these materials as a combination of end-, side- and interstack-shielding, the SBR can be raised by 50–106% for the 0.81–511 meV region. **[P3]**

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Utilisation of the results

The results of this thesis have already been applied in various projects related to the ESS. The calculated gamma yields and activity-production data provided input to the radiation protection and safety planning of the ESS. The performed Geant4 Multi-Grid detector simulations have continuously provided input for the improvement of detector design in the past 3 years, e.g. for the decision on the application of long blade coating, detector window thickness, shielding design. Moreover, the validation of the developed Multi-Grid detector model has already been used as part of the validation of the ESS Coding Framework [Kanaki, 2018]. Finally, the presented holistic approach of the utilisation of complex and detailed Monte Carlo detector simulation in shielding optimisation and background reduction has already been applied in the development of other detectors, such as the Multi-Blade detector [Galgóczi, 2018].

Thesis point related publications

[P1]. E. Dian, K. Kanaki, R. J. Hall-Wilton, P. Zagyvai, Sz. Czifrus: Neutron activation and prompt gamma intensity in Ar/CO_2-filled neutron detectors at the European Spallation Source, *Applied Radiation and Isotopes* **128**, (2017) pp. 275286, doi: 10.1016/j.apradiso.2017.06.003

[P2]. E. Dian, K. Kanaki, R. J. Hall-Wilton, A. Khaplanov, T. Kittelmann, P. Zagyvai: Scattered neutron background in thermal neutron detectors, *Nuclear Inst. and Methods in Physics Research, A* **902** (2018) 173–183, doi: 10.1016/j.nima.2018.04.055

[P3]. E. Dian, K. Kanaki, A. Khaplanov, T. Kittelmann, P. Zagyvai, R. Hall-Wilton: Suppression of intrinsic neutron background in the Multi-Grid detector, (2019) *JINST* **14** P01021, doi: 10.1088/1748-0221/14/01/P01021

Additional publications

[P4]. E. Dian & R. Hall-Wilton, Input to ESS SSM (RP safety) application, 2016.

[P5]. BrightnESS Deliverable Report: D4.5 − Simulation and Generic Multi-Grid design, doi: 10.17199/BRIGHTNESS.D4.5

[P6]. E. Dian, K. Kanaki, R. Hall–Wilton, A. Khaplanov and T. Kittelmann: Shielding optimization study for ^{10}B-Based large area neutron detectors with detailed Geant4 model, *2016 IEEE Nuclear Science Symposium, Medical Imaging Conference and Room-Temperature Semiconductor Detector Workshop (NSS/MIC/RTSD)*, Strasbourg, 2016, pp. 1-3.
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[P7]. K. Kanaki, XX. Cai, E. Dian, R. Hall-Wilton, A.Khaplanov, T. Kittelmann:
New simulation tools and reproduction of CNCS results using Geant4 (conference talk)
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[P9]. E. Dian *et al.*, "Validation of Detailed Geant4 Model for Thermal Neutron Scattering using the Results of Multi-Grid Detector Prototype Test at CNCS at SNS," *2017 IEEE Nuclear Science Symposium and Medical Imaging Conference (NSS/MIC)*, Atlanta, GA, 2017, pp. 1-3.
doi: 10.1109/NSSMIC.2017.8532930

[P10]. BrightnESS Deliverable Report: D4.8 - Report on Simulation Tools for Detector and Instrument Design and Simulations for Early Instruments,
doi: 10.17199/brightness.d4.8

[P11]. E. Dian, E. Klinkby, CJ Cooper-Jensen, D. Párkányi, D. Hajdu, J. Osán, G. Patriskov, U. Filges, PM. Bentley:
Preparation for activation measurements of concrete and PE-B4C-concrete to be applied for shielding at the European Spallation Source. Journal of Physics: *Journal of Physics: Conference Series*. 2018; **Vol. 1021**, No. 1.
doi: 10.1088/1742-6596/1021/1/012050

[P12]. E. Dian, K. Kanaki R. Hall-Wilton, A. Khaplanov, P. Zagyvai:
Geant4 Study for Intrinsic Scattered Neutron Background in Multi-Grid Detector (conference talk)
International Workshop on Position Sensitive Neutron Detectors,
Jülich, 2018