

A simple Method for Determining the Effective Removal Cross Section for Fast Neutrons

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Abstract: A method was proposed for determining effective removal cross section (Σ_R) for fast neutrons. The source beam is obtained from a Pu-Be combined source. The transmitted fast neutrons through standards samples, of known effective removal cross sections, are recorded by fast neutron counter (FNC). This FNC consists of a He-3 neutron detector and a paraffin wax moderator was designed to minimize recording of scattered neutrons from the investigated samples. The results obtained are supported by calculation carried out using the MCNP. It was shown that the response of the FNC versus Σ_R (calibration curves) can be used for determining Σ_R for liquid, powder and solid samples. The advantages of the proposed method were discussed and compared with existing methods in literature.

Keywords: fast neutrons; cross section, neutron detector, calibration.

1 Introduction

Effective removal cross-section, Σ_R (cm²/g) is a measure of the probability that fast or fission energy neutron undergoes a first collision, which removes it from the group of the penetrative uncollided neutrons (Kaplan, 1989). Σ_R is the equivalent absorption cross section which fits the attenuation of a slab of material located near a neutron source as measured by a detector separated from the source by a large thickness (more than 50 cm) of water. It is approximately constant for neutron energies between 2 and 12 MeV. The concept of Σ_R is used as long as the shielding material under investigation contains some scattering nuclides. However, when there are no scattering nuclides around the beam another quantity, the total mass neutron cross-section Σ_T (cm²/g), is used. Observed values of the Σ_R are roughly 2/3 of Σ_T for neutrons of energies in the range from 6 to 8 MeV (Glasstone and Sesonske, 1986). Additionally, it was proven that fission neutrons penetrate like 8 MeV neutrons (Perlini et al., 1970).

First measurements of Σ_R were based on a large water tank adjacent to an exit of a neutron beam that was obtained from a reactor. Shielding samples were inserted in the tank adjacent to the neutron exit. Measurements of transmitted neutrons (fast and the thermal neutron dose

rates) were made in the water behind the samples along the source axis (Wood, 1982). All available results for Σ_R were obtained from such measurements (Wood, 1982). A neutron-gamma spectrometer based on organic scintillator detector (stilbene) and pulse shape discrimination technique is widely used for measuring Σ_R (Osman et al., 2015; El-Sarraf and El-Sayed Abdo, 2013; Miller, 1968; McBeth et al., 1971; Knoll, 1989). This spectrometer works in a mixed field of neutrons and gamma rays. It is based on using a fine and fast neutron beam that is obtained from a neutron source. Transmitted fast neutrons from the samples investigated having different thicknesses are analyzed in terms of the Beer-Lambert law of exponential decay for determining Σ_R .

The effective removal cross-sections can be calculated for mixtures, alloys and compounds, with the knowledge of the weight percentages w_i , and the values of $(\Sigma_R)_i$ for each of the consisting elements. This is achieved by following simple addition rules (Kaplan, 1989; Wood, 1982):

$$\Sigma_R = \sum_i w_i (\Sigma_R)_i \quad (1)$$

Programs based on the additional rule (Eq. 1) were developed for calculating Σ_R combined material (El-Khayatt and Abdo, 2009; Elmahroug et al., 2015). Some

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empirical formulas (Wood, 1982) are being used for determining Σ_R for elements in terms of the atomic weight, A or the atomic number, Z . These are given by:

$$\Sigma_R = 0.21A^{-0.56} \text{ cm}^2 \text{ g}^{-1}$$

$$\Sigma_R = 0.00662A^{-1/3} + 0.33 A^{2/3} - 0.211 A^{-1} \text{ cm}^2 \text{ g}^{-1} \quad (A > 12), \quad (3)$$

$$\Sigma_R = 0.190 Z^{-0.743} \text{ cm}^2 \text{ g}^{-1}, \quad (Z \leq 8), \quad (4)$$

$$\Sigma_R = 0.125 Z^{-0.565} \text{ cm}^2 \text{ g}^{-1} \quad (Z > 8) \quad (5)$$

Additionally, other empirical formulas were reported for mono-energy neutrons (Perlini et al., 1970). Semi-empirical models were developed for calculating fast neutron attenuation through hydrogenous shields. Albert and Welton (1950) assumed that the entire total cross section for hydrogen is effective for removal (it is taken to be energy-dependent). However, a fraction of the total cross section (averaged over the fission spectrum) is effective for heavier elements.

The total cross section for heavier nuclei is taken to be an empirical energy-independent removal cross section (Σ_R). Removal-diffusion (RD) based methods were developed (see Engineering Compendium on Radiation Shielding, Vol.1, and references cited in). These were based on coupling between the removal concept of fast neutrons and diffusion of the slowing down and thermal neutrons. Some codes based on the RD methods were developed and has already proved to provide good predictions of the neutron penetration in different shields. Additionally, methods based on the solution of the neutron Boltzmann equation were reported for determining Σ_R (see for example El-Sarraf, and Abdo, 2013).

The chemical composition of the sample under investigation should be known to calculate Σ_R using addition rule (Eq. 1) and/or any calculation method based on it. For measuring Σ_R , using the neutron-gamma spectrometer, the following requirements are needed: samples of different thickness should be prepared, a fine beam of neutrons should be used, scattered neutrons resulting from interaction with the sample should be avoided, long acquisition time for collecting neutron spectra is required specially when using low activities of neutron sources. Therefore, a method that avoids the above limitations for determining Σ_R for any kind of sample is required.

The aim of the present work is the development of a method for determining Σ_R for any kind of sample. It is based establishing a calibration curve between transmitted fast neutrons through different standard samples and their Σ_R . Response of transmitted fast neutrons obtained from a Pu-Be source through investigated standard samples is recorded using an assembly of a cylinder made of wax containing a He-3 neutron detector.

2 Experimental Setup

The experimental set-up used in this work is shown schematically in Fig.1. It consists of an empty cylinder (made of polyethylene) of 70 cm diameter and 100 cm length. A tube of 10 cm diameter is fixed along axis of the cylinder. The cylinder is filled with water. The water cylinder was fixed on a wooden table (1 meter height above the ground). A Pu-Be neutron source (1 Curie) is stored below the wooden table in a special chamber, made of borated paraffin wax and surrounded by lead.

The source is raised - through a tube - by a rope into the central tube such that distance between source and one face of the cylinder is 15 cm (exit face of neutrons). The other face of the tube is blocked by paraffin wax. The outlet face of neutrons is covered by Cd except an opening of ~10 cm diameter. Water surrounding the neutrons source moderates emitted neutrons from the Pu-Be source. The water cylinder is surrounded by blocks of borated paraffin to minimize leakage of fast neutrons.

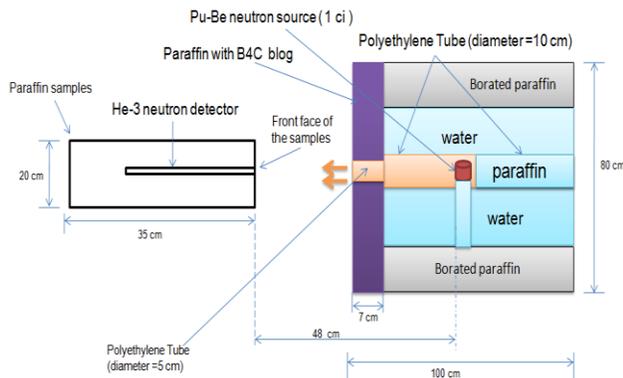


Fig.1 Experimental set-up

Fast neutron counter (FNC) consists of Helium-3 neutron detector (LND-252172) inserted through a moderator made of paraffin wax (20 cm x 20 cm x 35 cm) was used to register fast neutrons transmitted from the investigated samples. The FNC is covered by Cadmium sheets from all sides. A power supply (Canberra model 31060) providing high voltage to the He-3 detector via a preamplifier (Ortec-142PC) was used.

The output signal from the preamplifier was fed to amplifier (Canberra Amp/TSCA 2015A) and subsequently to a counter (Ortec 772) for neutron counting. To check performance of the He-3 detectors, the output signal from the amplifier was fed to multi-channel analyzer cart (MCA) cart with 8000 channels (NDS). The MCA cart was installed on a PC and was controlled by the Genie 2000 software package. The MCA cart was used for acquiring and analyzing spectra resulting from the neutron interactions with the He-3 detector.

3 Experimental Results and Discussion

3.1 fast Neutron Counter (FNC) Design

The fast neutron counter (FNC) was constructed to record fast neutrons transmitted from the sample investigated avoiding scattered neutrons. This was achieved by carrying out two experiments to choose the best orientation of the He-3 detector that records direct transmitted neutrons, either perpendicular or parallel to the direction of the incident neutron beam. In the first experiment, the He-3 detector was oriented perpendicularly to the direction of the incident neutron beam. Samples made of paraffin wax (20 cm x 20 cm) and having different thicknesses were set behind the detector. The paraffin wax was used as a neutron moderator, since it is mainly composed of hydrogen and carbon. For every thickness added, the thermal neutron count resulting from backscattered and moderated neutrons was registered. With these samples (total thickness of ~ 20 cm) fixed behind the detector, another paraffin wax sheets (20 cm x 20 cm) of different thickness were set in front of the detector and the detector count was registered. The results are shown in Fig 2a. As one can see, the count rate increases as the thickness of the wax samples behind the detector increases, till certain thickness above which the count rate saturates. The neutron count rate increases since thermal neutrons resulting from moderation and reflection (back scattering) of the fast neutrons, increases as the sample thickness increase.

The neutron count rate suddenly increases when samples of thicknesses =1.5 cm was set in front of the detector. This is due to the increase of the intensity of thermal neutrons resulting from fast neutron moderations. With further increase of the sample thicknesses in front of the detector, the neutron count rate decreases because of attenuation of thermal neutrons. Additionally, the detector orientation (perpendicular to the neutron beam) enhances registering scattered neutron from the samples investigated. In the second experiment, the He-3 detector was set parallel to the direction of the incident neutron beam. The wax samples (20 cm x 20 cm) having different thicknesses were inserted through the detector - the first wax sample was touching the window of the detector. For every thickness inserted, the count rate was registered. After reaching 20 cm thickness along the length of the detector, another wax samples were set in front of the detector and the neutron count rate was registered. The results are shown in Fig 2b. The count rate increases as the thickness of the wax sample surrounding the detector increases till certain thickness above which, count rate saturates. This is attributed to the increase of thermal neutrons resulting from moderation of fast neutrons as the thickness of the wax samples increase. It can be observed that the count rate of the thermally moderated neutrons resulting from samples in front of the detector decreases as the thickness increases. Namely, with detector orientation in the same beam direction, scattered neutrons from samples in front of the detector are minimized. So, the

FNC should rely on this geometry. Comparing Fig2a and b, it can be noticed that the count rate of the first experiment is higher than that of the second because the chance of registering scattered neutrons is higher. Thus, the second experiment is preferred for direct fast neutron recording.

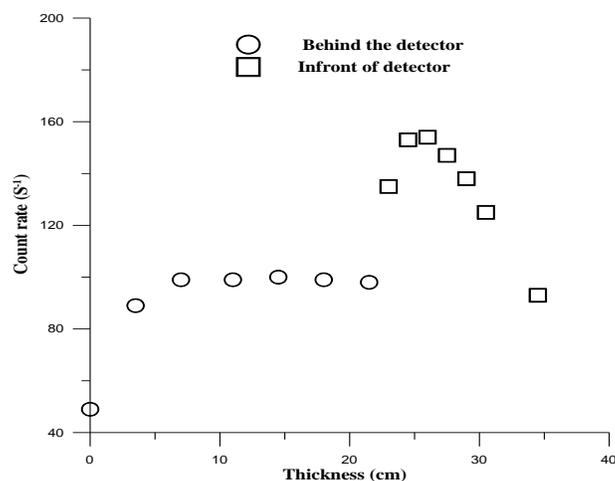


Fig. 2a. Detector count rate versus sample thickness: Detector orientation is perpendicular to the beam direction. Circle symbols are for sample behind the detector (up to thickness=21.5 cm), square symbols are for samples in front of the detector (the thickness of 24.5 cm in the figure is for the first sample of thickness=3 cm).

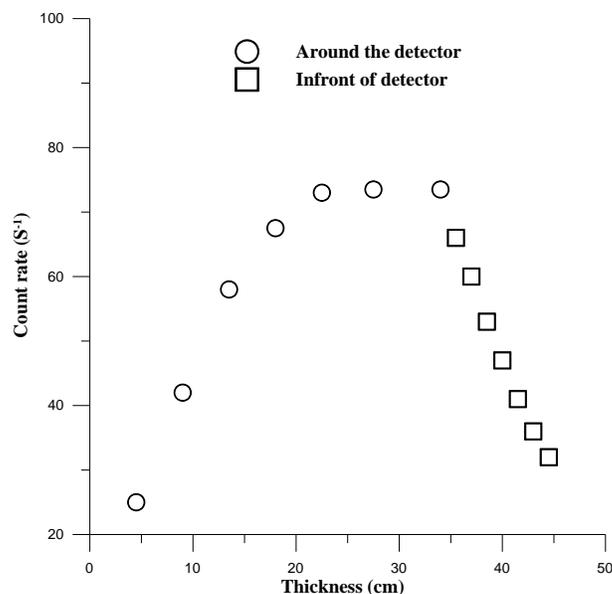


Fig.2b. Detector count rate versus sample thickness: Detector orientation is parallel to the beam direction. Circle symbols are for sample surrounding the detector (up to thickness=34 cm), square symbols are for samples in front of the detector (the thickness of 35.5 cm in the figure is for the first sample of thickness =1.5 cm).

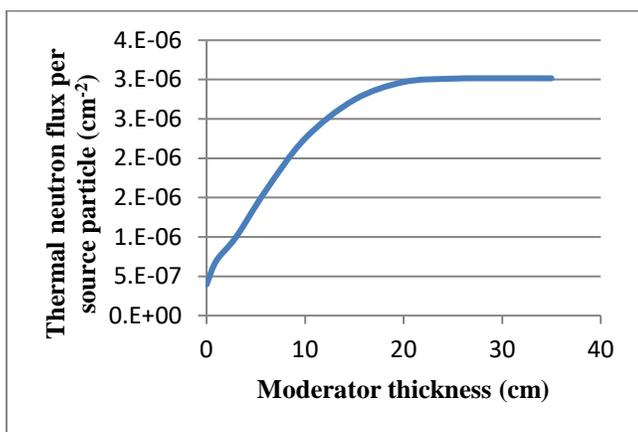


Fig. 3a. Thermal neutron flux inside moderator versus moderator (wax) thickness calculated by MCNP.

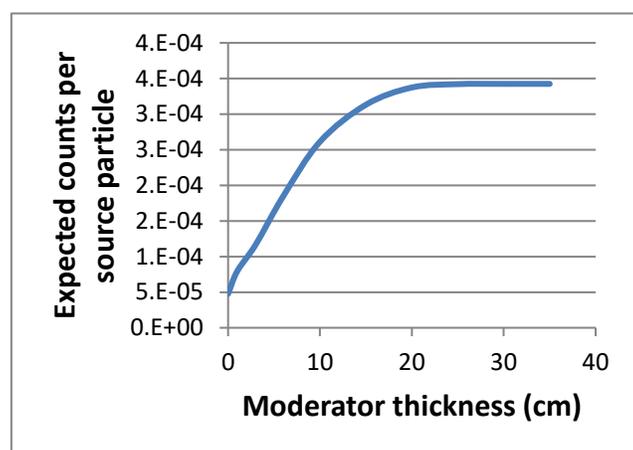


Fig.3b. The expected counts for the He-3 detector inside the moderator versus moderator thickness calculated by MCNP.

The detector response for second experiment was calculated by the MCNP5 computer code using a three dimensional model (Fig. 1) for both paraffin wax and polyethylene samples. The samples in front of the detector were not included in these calculations. Results for both paraffin wax and polyethylene samples are similar. The results obtained for the wax samples are shown in Figs.3a and b, for the thermal neutrons inside the moderator and expected count in the He-3 detector, respectively. As one can see, the expected counts saturates at moderator thickness from 15 to 20 cm in consistent with the experimental results. The response of the FNC for incident neutron beams having different energies (neutron spectrum) was calculated using the MNCP code. The results are shown in Fig.4. The FNC converts all incident energy to thermal neutrons , however with different efficiencies.

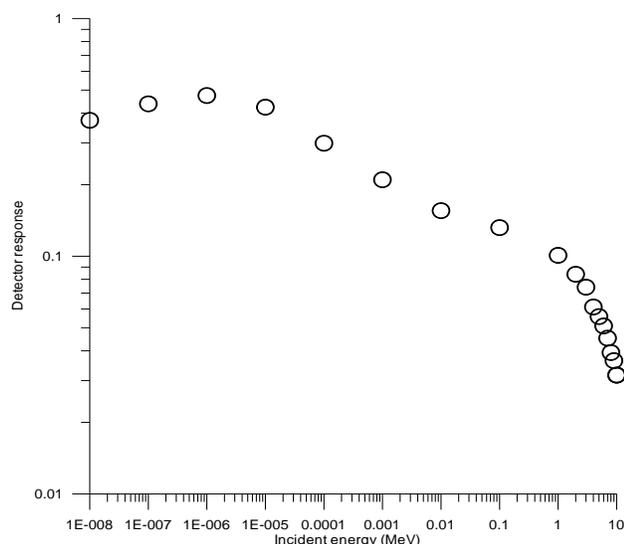


Fig. 4. The expected probability per one particle of the source for the He-3 detector inside the moderator versus incident neutron energy calculated by MNCP.

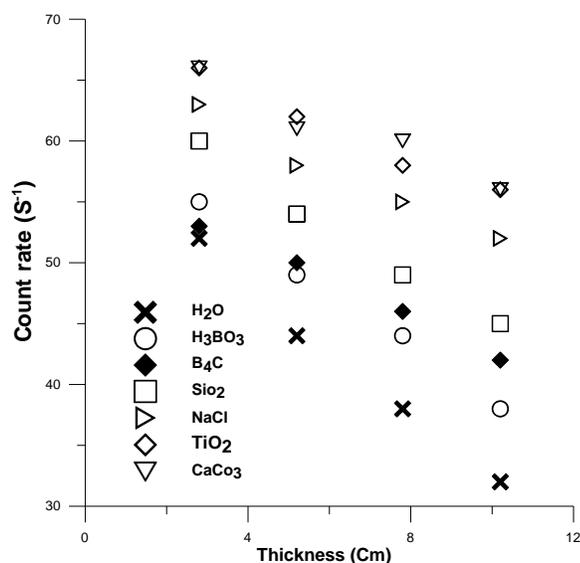


Fig.5. The response of the FNC versus sample thickness for some standard samples.

3.2 Effective Removal Cross Section

The following standard samples (binary mixtures and solutions) consisting of two chemical compounds with different weight percentages were prepared: NaCl and SiO₂, NaCl and H₃BO₃, H₂O and NaCl, and H₃BO₃ and SiO₂. Additionally, binary mixtures consisting of chemical compounds inserted between and around iron slabs were prepared as follow. The calculated effective removal cross section for these samples altogether with their weight fractions are listed in Table 1.

A plastic container (20 cm length x 20 cm width) consists of four cells (cell width = 3.5 cm) was set between the FNC face and the outlet of the neutron beam. The response of the FNC as a function of sample thickness was determined for the following standard samples: H₂O, H₃BO₃, B₄C, SiO₂, NaCl, TiO₂, and CaCO₃. The results are shown in Fig. 5. As one can see, the response mainly decreases as both the thickness of the sample and its Σ_R increase. This means discrimination between samples characterized with different values of Σ_R can be obtained. Three cells was removed from the beam direction leaving only one cell between the face of the FNC and the outlet beam of the incident neutrons – distance between the counter face and outlet beam ~4 cm. The cell was filled with the first 25 samples listed in Table 1. The response of the FNC versus Σ_R for these samples is shown in Fig.6 (lower results). As one can see, response of the FNC decreases as Σ_R increases for all samples. The results were least squared fitted and found to follow a power law function (R-squared = 0.95) given by:

$$R = 269 * (\Sigma_R)^{-0.15} \quad (6)$$

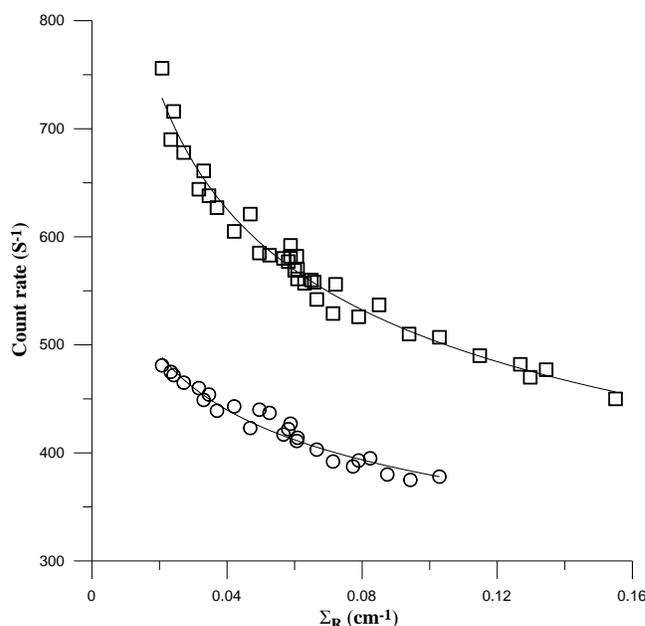


Fig. 6. The response of the FNC versus Σ_R . The circle symbols represent samples of thickness 3.5 cm. The square symbols represent samples of the dimension: length = 9 cm and diameter = 8 cm.

The value of the exponent of the power law function determined from least square fitting is an indication of the sensitivity of the results. If both the thickness of the sample investigated and the incident neutron flux increase, the sensitivity increases. This was achieved by modifying the experimental set-up shown in Fig. 1. The B₄C blog was removed and the face of the FNC was shifted to touch the outlet of the neutron beam. A cylindrical container made of plastic (length= 9 and diameter 8 cm) was used to accommodate the standard samples. The container was

inserted along the center tube facing the neutron source. Based on these modifications, an experiment was carried out using most of the standard samples listed in table 1. The FNC response versus Σ_R is shown in Fig. 6 (upper results). As one can see, R decreases as Σ_R increases. Additionally, the results were found to follow a power law function (R-squared = 0.97) given by:

$$R = 294 * (\Sigma_R)^{-0.23} \quad (7)$$

Table 1. The calculated fast neutron effective removal cross section of the standard samples.

Sample	Σ_R (cm ⁻¹)	Sample	Σ_R (cm ⁻¹)
H ₂ O	0.1023	NaCl/ SiO ₂	0.0526
H ₃ BO ₃	0.0666		0.0496
B ₄ C	0.0714		0.0421
SiO ₂	0.0588		0.0346
NaCl	0.0272		0.0317
TiO ₂	0.0242	NaCl/H ₃ BO ₃	0.0606
Graphite (C)	0.0773		0.0567
Acetone	0.0824		0.0468
Sulfur	0.0201		0.0370
Polyethylene grains.	0.0789		0.0331
CaCO ₃	0.0234	NaCl/H ₂ O	0.0943
Citric Acid	0.0581		0.0874
AlCl ₃ .6H ₂ O	0.0609	H ₃ BO ₃ / SiO ₂	0.0600
Fe	0.1689		0.0609
Fe/ Citric	0.0721		0.0629
Fe/CH ₂	0.085		0.0650
Fe/H ₂ O	0.155		0.0658
Fe/H ₃ BO ₃	0.1230	Fe/SiO ₂	0.1270
Fe/NaCl	0.1148	C ₂ H ₆ O	0.0939

The sensitivity (indicated by the value of the exponent of the power law function (0.23)) is better than the corresponding one of the previous experiment. It was improved by a factor of 1.53.

Based on these calibration results, Σ_R can be determined for any sample by measuring its FNC response.

The neutron-gamma spectrometer based on organic scintillator detector (stilbene) and pulse shape discrimination technique is the most popular method for determining Σ_R (Osman et al., 2015; El-Sarraf and El-Sayed Abdo, 2013; Miller, 1968; McBeth et al., 1971; Knoll, 1989). This method requires preparation of samples having different thicknesses and the use of fine beam of fast neutrons. Preparation of samples having different thicknesses is tedious and not possible all the time. Additionally, it is time consuming especially when using neutron sources of having small activities. In comparison with this method, the proposed method is not only simpler and quicker but also it is less expensive. The method is self-validated since 39 standard samples of different types were used.

The FNC response versus energy was extensively studied in literature – see for example Lacoste, 2010; Hyeonseo Park et al., 2013; Kenichi Watanabe et al., 2011 and references cited in). In their work flat response of the neutron counter versus neutron energy was achieved. The effect of such behavior on the obtained result of the present work is not investigated due to the limitation of our set up.

4. Conclusion

A method was proposed for determining fast neutron effective removal cross section. The method was verified using 39 standard samples of different materials. The method can be applied for liquid, solid and mixture of samples. Cylindrical samples of the dimension length = 9 cm and diameter = 8 cm at best used. The proposed method is not only simpler and quicker than existing methods in literature but also it is less expensive.

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