Simple Experimental Design for Calculation of Neutron Removal Cross Sections

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This article proposes an experimental technique that could be used to determine the fast neutron removal cross section of a given shielding material. Removal cross sections can provide a rough estimate of the fast neutron shielding capabilities of the material in question. Under the right conditions, neutron removal cross sections indicate the probability, per unit path length, that a neutron will be both scattered out of the fast neutron group flux and then subsequently moderated to thermal energies. Removal cross section data are not available for a number of promising shielding materials. The experimental technique proposed in this article will allow for a quick comparison of several candidate shielding materials prior to selecting one for a rigorous shielding analysis. The benefits and limitations of this experimental technique are discussed. The major limitation of this technique is found to be the lack of consideration for neutron absorption reactions, which, in certain cases, can lead one to believe that a certain material is a good shielding candidate, when it is not. Further work on this technique would involve quantitatively comparing the removal cross sections of various materials to their actual fast neutron shielding capabilities, as determined by a shielding analysis code, to see if the predictions made solely from the removal cross sections are valid in practise.

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I. INTRODUCTION

Shielding analyses are one of the most common and important problems within nuclear engineering. There are two primary types of shielding analyses, both of which require one to determine the dimensions and composition of a shield ie. the shielding configuration. The first type is referred to as a biological shielding analysis. Here, the problem is to find a shielding configuration that will reduce dose rates to persons in the vicinity of the radiation source to some predetermined level (generally in the most cost effective way possible). The second type of analysis is that of thermal shielding, where the goal is to attenuate the radiation flux as much as possible, in an attempt to reduce the damage to reactor components caused by the radiation. In both cases, neutrons and gamma rays are the primary concern, as charged particle radiation, in the form of alpha and beta particles, are sufficiently attenuated by air alone.

A. Fast Neutron Shielding

Shielding of neutrons, and in particular, fast neutrons (neutrons with energies above 1 MeV), is one of the more difficult radiation shielding problems one could encounter. This is because:

1 Neutron cross sections for almost all elements vary significantly with both the energy of the neutrons, and the temperature of the materials.
2 Neutron absorption reactions can lead to the production of gamma rays (often of significant energy), charged particles or more neutrons.

Furthermore, as the vast majority of neutrons produced in fission and fusion reactions are fast, the shielding of fast neutrons is particularly relevant to the nuclear industry today. For example, one of the most important considerations in the design of a magnetic confinement based fusion reactor, such as the tokamak reactor, is a thermal shielding configuration for the magnetic coils, which is needed to reduce radiation induced heating to acceptable levels[1].

Unfortunately, absorption cross sections for fast neutrons are, in general, very small and so the direct absorption of fast neutrons is not feasible. As such, the general approach to fast neutron shielding is to slow the neutrons down to thermal energies (between 0.0625 eV and 1 MeV) before trying to absorb them (similar to moderation in a reactor). It can be shown[2], that neutrons lose about half their energy in an elastic collision with hydrogen. As such, hydrogen rich materials are almost always incorporated into a shielding design. Another approach is to slow down neutrons through inelastic scattering with heavy nuclei. It can be shown[2], that the approximate energy lost by a fast neutron in an inelastic collision is given by: (1)

$$\Delta E = E - 6.4\sqrt{E/A}$$

Where E is the energy of the incident neutron in electron volts and A is the atomic mass number of the scattering atom also measured in electron volts. Heavy nuclei tend to work better than hydrogen for very fast neutrons with energies greater than 10^7 MeV (like those produced in fusion reactions) because at these energies, the total hydrogen cross section decreases rapidly[3], while the inelastic scattering cross section of most high Z materials, such as Zirconium or Lead, stay relatively constant[4,5].

B. Problem Statement

In general, shielding analyses are performed using specially designed computer programs that use either a deterministic or Monte Carlo based method. In either case, the codes are both expensive and time consuming to use. As such, there is a need for an easy way to characterize the fast neutron shielding capabilities of many materials quickly and easily. One way this can be done is using the removal cross section method originally developed by Oak Ridge National Laboratories[6] (ORNL). The macroscopic removal cross section of a material, denoted Σ_R, represents the probability, per unit path length, that a single interaction between a fast neutron and a target atom will cause the neutron to eventually slow down to thermal energies and be "removed" from the fast neutron flux. Since the general approach to fast neutron shielding is to slow the neutrons down before absorbing them, neutron removal cross sections could provide an estimate of a given materials ability to do the first half of this approach, the moderation, and could therefore be used as an input into the design of a shielding configuration.

III. REMOVAL CROSS SECTIONS

To understand the concept of removal cross sections, consider the arrangement shown in Figure 1. Here, a thin cylindrical fission source of radius R, emits S neutrons per second towards a detector that is aligned with the centre of the source. The entire arrangement is surrounded by water. This set up is a simplified version of the experimental apparatus used in the original ORNL experiments. Letting G(r) represent the flux from a point source of neutrons that only emits one fast neutron per second, it can be shown[2] using the results of the ORNL experiments[6] that the flux at some distance, r, away from the fission source is given by:

$$\emptyset(r) = 2\pi S \int_0^R G(r) z dz \qquad (2)$$

Furthermore, beyond a distance of approximately 40 cm, if $4\pi r^2 G(r)$ is plotted on a semi log scale with distance from the source, the resulting curve is linear, so that G(r) can be written as[2]:

$$G(r) = \frac{A}{4\pi r^2} \exp\left(-\Sigma_{RW} r\right) \qquad (3)$$

Where A is a constant equal to 0.12 and Σ_{RW} = - 0.103 cm^{-1} is the removal cross section of water. Equation 3 states that, beyond 40 cm of distance travelled in water, fast neutrons are attenuated at a rate of Σ_{RW} neutrons per unit path length travelled.

Since G(r) is the flux from a point source of neutrons emitting 1 neutron per second, it follows that the flux at some distance, r, away from a point source emitting S neutrons per second, that is shielding by a given material of thickness t, and surrounded by water, is given by[2]:

$$\emptyset(r) = SG(r) \exp(-\Sigma_R t) \qquad (4)$$

Where Σ_R is the removal cross section of the material. This can be generalized for an arrangement surrounded by any hydrogenous material, rather than just water, by replacing the removal cross section of water in equation 3, with the macroscopic removal cross section of hydrogen for the given material (the microscopic cross section for hydrogen doesn't change but the macroscopic cross section does change with the hydrogen density of the material).

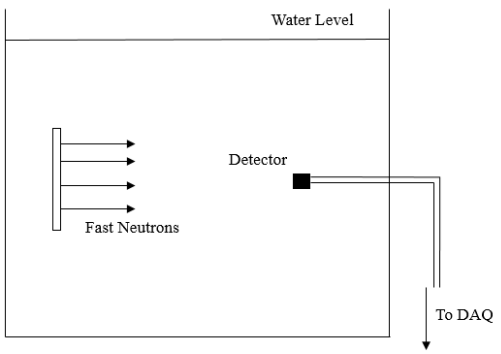

Fig. 1 Simple depiction of the original Oak Ridge National Laboratories study

IV. EXPERIMENTAL DESIGN

Although removal cross sections for various materials were found experimentally in the original Oak Ridge experiments[6], and have been calculated theoretically by various researchers since then,[7,8] there are many promising materials which do not have removal cross sections tabulated. For example, both ^{113}Cd and ^{73}Ge do not have removal cross section data available, while both could be promising materials for fast neutron shielding when combined with hydrogen, given their relatively large atomic number and absorption cross sections for thermal neutrons compared to other elements. The remainder of this article will focus on a simple experiment that can be used to determine the removal cross section of any given material.

A. Experimental Setup

The experimental technique proposed below is similar to

the original experiments performed by ORNL, however, it is much simpler to perform. The experimental apparatus is shown in Figure 2. Here a small sphere of a radioactive element, such as ^{252}Cf, is used as a point source of neutrons. Of course, a point source cannot actually exist in real life, however, for all practical purposes, if the distance between the source and the detector is much larger than the radius of the sphere, then it is well approximated as a point source.

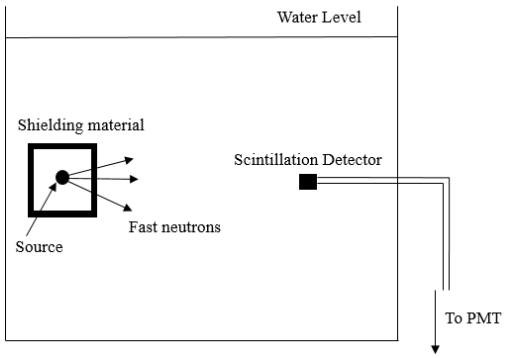

Fig. 2 Proposed Experimental Set-up

Californium-252 sources are common in radiation science laboratories, with fresh sources having activities between 10^7 and 10^9 neutrons per second[9]. However, the half life of ^{252}Cf is only about 2.5 years and on average, relatively few fast neutrons are emitted. In order to account for these factors, the unshielded fast neutron flux must be determined prior to performing the experiment. This can be done using a simple set up similar to that shown in Figure 3. Here, the neutron detector is only sensitive to neutrons with energies greater than 1 MeV (the threshold energy for fast neutrons). To obtain the best results, this calibration should be performed in a vacuum. Fiber optic scintillation detectors such as those developed by Krishnan et al.[10] are perfect for this application. If a detector of radius, r, is placed at a distance, d, away from the source, and the detector counts N_f fast neutrons per second, then the total number of neutrons emitted by the source of radius r_s per cm^2 per second, S, is given approximately by:

$$S = 4\pi N_f \left(\frac{d}{r}\right)^2 r_s^2 \tag{5}$$

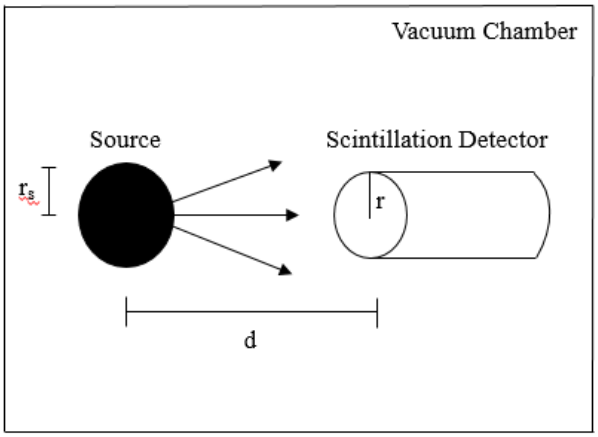

Fig. 3 Calibration Technique

Once the calibration has been performed, the remainder of the experiment is quite simple. Referring to Figure 2, placed immediately next to the source (or as close as is reasonable) is the shielding material to be tested. The material dimensions must only be as large as the radius of the sphere, so that any neutrons that are emitted towards the detector must also pass through the shielding material, but in practise, it will often be easiest to encapsulate the source with the shielding material. Finally, the detector must be placed at least 40 cm away from the shielding material, so that the simple relationships shown in equation 4 remain valid. With the source strength known from equation 5, and the value of G(r) computed for the particular arrangement, equation 4 can be re-arranged for the removal cross section of the shielding material:

$$\Sigma_R = -\frac{1}{t}\ln\left(\frac{\emptyset(r)}{SG(r)}\right) \tag{6}$$

V. RESULTS AND DISCUSSION

Shown in Figure 4 are the results one might expect to get from the fast flux detector with a source strength of 10^9 neutrons per cm^2 per second, shielding thicknesses of 10 cm and the removal cross sections[2] shown in Table I below. The unshielded flux (ie. The flux with only water between the source and detector) is shown as well.

Table I Removal Cross Sections for Various Materials[2]

Material	Macroscopic Removal Cross Section (cm^{-1})
Sodium	0.032
Iron	0.168
Zirconium Oxide	0101
Concrete [a]	0.089

[a] Containing 6% water by weight

The curves in Figure 5 were generated in MATLAB using equation 4. Inspecting the results, one can see that adding any shielding material shows improvements over

just water, with heavy elements providing the best attenuation. However, the results are not exactly as predicted. Iron, with an atomic number of 26 shows better fast neutron attenuation than Zirconium with atomic number 40. This is due to the difference in the densities of the two materials, with Iron having a density of 7.874 g/cm^3 and Zirconium Oxide having a density of 5.68 g/cm^3. Since Oxygen has a removal cross section of zero[2] this has no impact on the results. This indicates that it is not just the energy lost per interaction that matters, but also the total number of interactions that the neutron undergoes before exiting the shield.

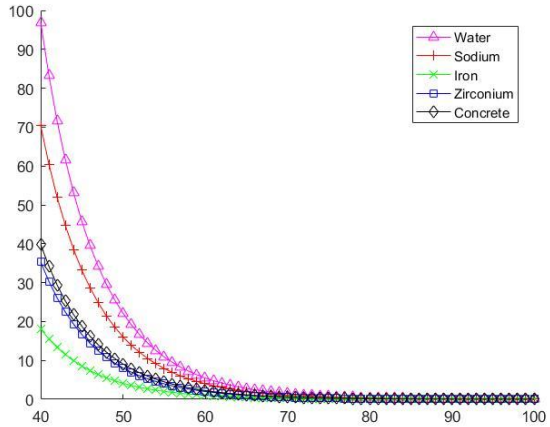

Fig. 4 Fast neutron flux as a function of distance from the source

To understand equation 3, consider that, as mentioned earlier, neutrons lose about half of their energy in an elastic scattering event with hydrogen and that the total hydrogen cross section increases rapidly with decreasing neutron energy. As such, after a fast neutron scatters off a hydrogen atom, the mean free path that it travels before a subsequent collision decreases. This continues until the neutron has lost most of its energy. Because of the continually decreases mean free path, this occurs very close to the original scattering event[2]. As such, a single scattering collision not only removes the neutron from the fast flux, but also ensures that it will eventually be moderated to thermal energies. However, without the presence of a heavy nuclei to provide the initial scattering site, there is a much smaller chance that this chain of scattering will occur. This explains why the materials in Table I all show improvement over water in terms of fast neutron attenuation.

There are two major limitations of this technique. The first is that it can only be used for hydrogen rich materials that have moderation properties similar to water. However, as mentioned, most shielding materials already contain substantial amounts of hydrogen, so this isn't a huge issue. That being said, there are some promising shielding materials, such as Magnesium Boride[11] (MgB_2) that could not be analysed using this technique. The second major limitation is that the removal cross section provides no information about what will happen during moderation or once the neutrons reach thermal energies. This can lead one to believe that a particular candidate material would make a good neutron shield, when in reality it might not. An example of such a material is Iron. From the results shown in Figure 5, it would appear that out of the materials examined, Iron is the best shielding material for fast neutrons. However, when Iron absorbs a thermal neutron, it can emit gamma rays with energies of 7.6 and 9.3 MeV[9]. Although Iron is effective at fast neutron attenuation, if a gamma ray shield was not incorporated into the design, significant amounts of radiation could also be produced by the shield.

At this stage of the research, it is difficult to tell whether the predictions made by removal cross sections would hold in real life. One reason for this is that there are relatively few studies comparing different shielding materials under the exact same conditions. That being said, Hayashi et. al. performed a simulation of various shielding configurations for use in a tokamak reactor[12]. They tested F82H steel, composed mainly of Iron, and Zirconium Hydride (ZrH_2), along with other materials. Their results showed Zirconium Hydride to be a much better shield than F82H steel. This could be due to a difference in the number of Iron and Zirconium atoms per unit volume in the two materials, however, it is difficult draw a firm conclusion. As such, beyond performing the proposed experiment, further work will need to be done to quantitatively compare the results of the proposed experiment to simulated shielding configurations.

VIII. SUMMARY AND CONCLUSIONS

This article presents an experimental technique that could be used to characterize the fast neutron shielding capabilities of candidate shielding materials prior to performing a detailed shielding analysis with the materials in question. It was shown that a simple experimental set up can be used to determine the removal cross section of a candidate material. However, the question of whether a removal cross section provides a good estimate of the fast neutron shielding capabilities of the material remains to be answered. To answer this question, comparisons between the predictions made by removal cross sections, and the results of rigorous shielding analyses would have to be made.

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