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A review on multilayer radiation shielding

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Abstract. Radiation shielding is a body of material that is placed between a radiation source and an object to be protected with the aim of reducing the intensity of radiation at the object's location. It can be made from various materials. These materials can be stacked into a multilayer shield or they can be mixed into a composite shield. The main objective of the present study is to review the list of multilayer shield combinations that have been studied and to highlight the findings on material arrangement and consequent buildup factor. The scope of the study is limited to the results of the performed studies. It was observed that there was no clear method on arranging the layer. Buildup factor was also found to be complicated in multilayer shields. Future studies may focus on new multilayer shielding design with unlisted materials, complementary buildup calculations, and applications of metaheuristics in shielding optimization.

1. Introduction

As nuclear technology advances and is widely used in today's industries, more people are likely to be exposed to ionizing radiation. A shielding is needed to absorb and reduce the intensity of the radiation. It is one of the protection methods recommended by the International Commission of Radiation Protection (ICRP), other than minimizing operation time and maximizing distance. It requires a more careful design as it is usually permanent and not easily replaceable. Besides, it also needs to be able to fulfill other design requirements depending on where the shield is applied such as heat resistance and structural integrity.

Shielding can be divided into two configurations which are composite and multilayer. A composite shield is made up of a base material mixed with additives. This can improve the shielding capability of the material. For example, by varying the amount of additives in a concrete, the density of the material can be increased, resulting in a better performance [1]. However, it can be difficult to obtain a homogenous mixture of the component materials that can result in inconsistent shielding performances [2][3]. Uneven mixing of the composite can also cause pinholes which are pure regions that radiation can penetrate through [4]. This problem can be prevented by using multilayer shielding approach [5].

A multilayer shield consists of two or more layers of different materials. In this arrangement, the incoming radiation will have more chances to be scattered and absorbed by the shield. One of the first instances of using multilayer shield was in 1943 in which concrete and paraffinized wood were used for the graphite-moderated reactor in Chicago. This shield configuration is useful against mixed radiation. For example, a ^{252}Cf source emitting both fission neutrons and prompt gamma rays can be shield with layers of polyboron and concrete [6]. The composite, containing hydrogen and boron atoms, slows down the fast neutrons and absorbing them, while the concrete scatters the resultant gamma-rays. Often, a shield is made from the combinations of composite and multilayer configurations which can be observed later in this review. This is because each of the materials has different shielding properties that they can be mixed and matched to solve a particular problem depending on the application. Shultis and Faw [7] had provided an excellent review on the history of radiation shielding technology.

Currently, there are many types of materials and combinations that have been tested. Ongoing improvements and developments are still being carried out as nuclear technology progresses. The aim of this review is to list some of them and to highlight the trends discovered by the authors. It will focus on the results on the effect of material types and arrangements on the shielding performance and buildup factor. Suggestions for future works will also be provided.

2. Radiation Interactions through Multilayer Shield

As radiation passes through a shielding material, it has certain probability to interact with the atoms of the material. When this happens, it is either absorbed or scattered. In both scenarios, secondary radiation such as beta or gamma is produced. Besides, in the scattering process, the direction of the radiation particles may be changed. Eventually, some of scattered particles are transmitted through the shielding. This phenomenon is known as buildup and it is represented by the buildup factor, *B*. Neglecting this factor may cause huge errors in shielding calculations [8].

For a narrow beam geometry, the intensity of radiation *I* emitted through a single body of material is given by the Beer-Lambert Law. I_0 is the initial intensity, $\mu(E)$ is the attenuation factor of the material at the particular energy *E*, and *x* is the shield thickness. The $\mu(E)$ can be expressed as mass attenuation factor $\mu \ \rho(E)$ for gamma rays and as total macroscopic cross section $\Sigma_t(E)$ for neutrons. When a broad beam geometry is applied, a buildup factor *B* is added to Equation 1 as a correction factor to account for scattered beam.

$$I = I_0 e^{-\mu(E)x} \quad (1)$$

$$I = BI_0 e^{-\mu(E)x} \quad (2)$$

In the case of a multilayer shield as shown in Figure 1, the intensities of radiation emerging from the subsequent layers are dependent on the preceding layers. The resultant intensity I_3 can be derived into Equation 3. Although, based on the commutative property of multiplication in this equation, it would seem that there would be no effect if the order of materials is changed. However, this is observed to be the opposite in various studies as explained later.

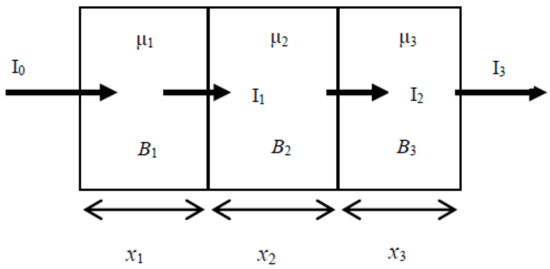

Figure 1. An example of a triple-layer shield [9].

$$I_3 = I_0 \ B_1 B_2 B_3 \ e^{-\ \mu_1 x_1 + \mu_2 x_2 + \mu_3 x_3} \quad (3)$$

On the other hand, Lamarsh and Baratta [10] had outlined four methods to approximate gamma buildup factors of double-layers when exact methods are unavailable. The first, if the materials differ in atomic number Z by only 5-10, is to use the buildup factor of the material with higher B value. It was based on observation that buildup do not vary rapidly with Z, except at low energy.

$$B(\mu x) = B_{max}(\mu_1 x_1 + \mu_2 x_2), \qquad \text{if } Z_1 - Z_2 = 5{,}10 \tag{4}$$

For any Z difference higher than in Method 1, the order of arrangement is considered. If the lower Z material is first, the buildup of the higher material is used. It is assumed that the lower energy photons transmitted from the first layer is absorbed in second layer.

$$B(\mu x) = B_2(\mu_2 x_2), \qquad \text{if } Z_1 - Z_2 > 10, \text{ low Z first} \tag{5}$$

Then, when the higher Z material is first, the buildup is determined by the energy of the gamma-rays E_γ. If E_γ is lower than 3 MeV, the buildup is given by Equation 6. As the flux emitting from the high Z material has a different energy profile from the original source, they are treated as a source in the second layer. When the E_γ is higher than 3 MeV, the buildup is similar to the previous case except that the buildup in the second layer is set at the minimum 3 MeV instead of higher energy. This is based on the assumption that the gamma-rays emerging from Layer 1 have energies about the minimum.

$$B(\mu x) = B_1(\mu_1 x_1) \times B_2(\mu_2 x_2), \qquad \text{if } Z_1 - Z_2 > 10, \text{ high Z first, } E_\gamma < 3 \text{ MeV} \tag{6}$$

$$B(\mu x) = B_1(\mu_1 x_1) \times B_2(\mu_2 x_2)_{min}, \qquad \text{if } Z_1 - Z_2 > 10, \text{ high Z first, } E_\gamma > 3 \text{ MeV} \tag{7}$$

Besides, there is also another formula aside from the ones suggested above. As briefly reviewed by Mann *et al.* [11], there is an empirical formula proposed by Kalos [12][13] for calculating buildup factor in a double-layer shield based on its component materials. The Kalos' formula is shown in Equation 8. It was also validated by Shin and Hirayama using Monte Carlo method [14].

$$B(\mu x) = B_2(\mu_2 x_2) + [B_2(\mu_1 x_1 + \mu_2 x_2) - B_2(\mu_2 x_2)] \times K(\mu_1 x_1) . C(\mu_2 x_2) \tag{8}$$

$$K(\mu_1 x_1) = \frac{B_1(\mu_1 x_1) - 1}{B_2(\mu_1 x_1) - 1} \tag{9}$$

$$C(\mu_2 x_2) = \begin{cases} 1 & \text{for } high\text{-}Z/low\text{-}Z \text{ order} \\ exp(-1.7\mu_2 x_2) + (\alpha/K)[1 - exp(-\mu_2 x_2)] & \text{for } low\text{-}Z/high\text{-}Z \text{ order} \end{cases} \tag{10}$$

$$\alpha = \frac{(\mu_c/\mu_t)_1}{(\mu_c/\mu_t)_2} \tag{11}$$

Using transport calculations, some modifications to the correction factor were made by Burke and Beck [15] as shown in Equation 12. In this case, γ is the ratio of Compton mass attenuation coefficient of the first layer to the second layer.

$$C(\mu_2 x_2) = \begin{cases} exp(-\mu_2 x_2/\gamma) + 1.5[1 - exp(-\mu_2 x_2)] & \text{for } high\text{-}Z/low\text{-}Z \text{ order} \\ exp(-\mu_2 x_2 \times \gamma) + (\alpha/K)[1 - exp(-\mu_2 x_2)] & \text{for } low\text{-}Z/high\text{-}Z \text{ order} \end{cases} \tag{12}$$

$$\gamma = \frac{(\mu_c/\rho)_1}{(\mu_c/\rho)_2} \tag{13}$$

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The formula was further adjusted by Lin and Jiang [16] whom had approved the use of the empirical method for point isotropic source. This is shown in Equation 14-16. β is the ratio of total mass attenuation coefficient of the second material to the first material.

$$C(\mu_2 x_2) = \begin{cases} exp(-1.08\beta\mu_2 x_2) + 1.13\beta\ell(\mu_2 x_2) & for\ high\text{-}Z/low\text{-}Z\ order \\ 0.8\ell(\mu_2 x_2) + (\gamma/K)exp(-\mu_2 x_2) & for\ low\text{-}Z/high\text{-}Z\ order \end{cases} \quad (14)$$

$$\beta = \frac{(\mu_t/\rho)_2}{(\mu_t/\rho)_1} \quad (15)$$

$$\ell(\mu_2 x_2) = \frac{B_2(\mu_2 x_2) + 1}{B_1(\mu_2 x_2) + 1} \times [1 - exp(-\mu_2 x_2)] \quad (16)$$

2.1. *Observations on Buildup Factor*

Unlike in the case of a single layer shielding, the buildup in a multilayer shielding is more complicated [17][18]. This is because a monoenergetic radiation flux that passes through the first layer may result in a flux with a spectrum of energy due to absorption and scattering. This makes it difficult to determine the attenuation factor (cross sections, linear attenuation, mass attenuation coefficient) for the shielding calculation. Therefore, many studies had been done to calculate the buildup factor for various multilayer shielding designs using simulations and experiments.

Buildup factor is dependent on the type of radiation, its energy, the type of shielding material, and the geometry involved [19]. It is also significantly affected by the material arrangement and the thickness of each layer [17]. While the buildup was observed to increase with thickness, the effects of changing the material order were varied. Abbas [20] deduced that a lead-water gamma shielding had a greater buildup than the opposite order at 1 and 2 MeV, but the case was reversed at 6 MeV. Al-Arif and Kakil [9] saw no considerable difference in measured buildup factor when the order of material was modified. They also noticed that the buildup factor had a strange dependency on the atomic number and the photon energy. At low energy, the buildup increased as atomic number decreased. At high energy, it increased as atomic number increased. Meanwhile, Mann *et al.* [11] stated that at fixed energy, a double layered shield in a low-Z/high-Z orientation always results in a lower buildup as compared to a single material with the same optical thickness. This difference would keep increasing as the optical thickness increases.

3. List of Material Combinations

Table 1 (a) and (b) below show the multilayer shielding designs found in literature. Most of them employed combinations of light and heavy materials to shield against neutrons and photons of gamma-rays or X-rays. Some of them focused on determining the buildup factor which is important in shielding calculations. However, not all of them involved simulations that were coupled with experiments.

Table 1(a). Summary of multilayer shielding combinations.

Author	Shielding Materials	Remarks/Application
Fuse *et al.* (1970) [21]	Iron | Water	• Slabs of iron movable in water for 2.5 MW reactor • MAC-RAD code was used as comparison to experiments
Kuspa (1972) [19]	Aluminum | Lead	• Double-layer slabs against 1, 4, 6, and 8 MeV gamma rays • Monte Carlo (MC) method was employed to calculate buildup
Mandour & Hassan (1987) [17]	Iron | Carbon	• Buildup calculation using MC method for 14.1 MeV neutrons
Shin & Hariyama (1998) [18]	Concrete | Iron | Water	• Buildup calculation using EGS4 MC code for 1 and 10 MeV gamma-rays
Hu *et al.* (2008) [22]	Iron | Polyamide composite | Lead	• Optimization using genetic algorithm metaheuristic coupled with MCNP • Combination of composite and multilayer shielding for ^{252}Cf neutron source and ^{60}Co gamma source
McCaffrey *et al.* (2009) [23]	Metal elastomers bilayers	• Air kerma attenuation measured using commercial metal/elastomer test layers • EGSnrc MC code for 30−150 keV X-rays
Hossain *et al.* (2010) [6]	Poly boron | Borax mixed concrete	• Shielding against ^{252}Cf neutron source with detection using BF_3 long counter detector • MCNP calculations for 14 MeV neutrons
Kim and Moon (2010) [24]	(2-layer) LiH | W (8-layer) LiH | W | depleted U | Fe | TiH_2 | ZrH_2 | Pb | B_4C	• Optimization using genetic algorithm for shielding in spaceship conditions • Dose calculation using ANISN code
Abbas (2012) [20]	Water | Lead	• Buildup calculation using EGS4 MC code for 1, 2, and 6 MeV gamma rays
Gaber *et al.* (2013) [25]	Boron oxide glass | Epoxy ilmenite	• Double layer shields for neutrons and gamma rays resulting from ^{252}Cf fission source • Simulation using MCNP-4C2 MC code
Kim *et al.* (2015) [4]	Sendust alloy polymer | Tungsten	• Lamination of tungsten and composite films to shield against X-rays of 150 keV
Al-Arif & Kakil (2015) [9]	Aluminium | Iron | Lead	• Various combinations of shielding layers for gamma energies 0.662 and 1.25 MeV • Measurements of attenuation coefficient experimentally using ^{60}Co, ^{137}Cs sources and NaI (TI) scintillation detector
Mann *et al.* (2016) [11]	Aluminium | Limestone	• Buildup calculation using Geometric Progression method for gamma energies 0.5, 1.0, 2.0, and 3.0 MeV

Table 1(b). Summary of multilayer shielding combinations (cont.).

Author	Shielding Materials	Remarks/Application
Whetstone and Kearfott (2016) [26]	Steel | Polyethylene	• Alternating layers of shielding materials for an active neutron interrogation system • MCNP5 simulations for 1.0, 2.5, 4.0, 6.0, 8.0, 10.0, 12.0, and 14.1 MeV neutrons
Hadad *et al.* (2016) [27]	HDPE | Lead	• Optimization of shielding for prompt gamma neutron activation analysis with ^{252}Cf neutron source by Monte Carlo analysis
Sariyer & Küçer (2018) [28]	Concrete | Iron-contained materials (FeB, Fe_2B, stainless steel)	• Double layer of 1 m of concrete and varying thickness of iron layer against neutrons generated from proton interactions • FLUKA code was used to calculate dose distribution.
Park *et al.* (2018) [5]	Polymer composites containing tungsten or bismuth-tin alloy | BiSn layered sheet	• Lamination of W composite film or BiSn composite film onto a BiSn-coated layered sheet for 150 keV X-ray shielding
Cai *et al.* (2018) [29]	Six combinations of polyethylene, iron, boron carbide, and lead	• Optimization using genetic algorithm to shield against fission spectrum of ^{235}U

Some of these multilayer designs were found to yield better results than that of when using them separately. This might be because the subsequent layer is able to absorb the scattered radiation of lower energy emanating from the preceding layer. Fuse *et al.* [21] observed that a heterogenous arrangement of thick iron-water-thin iron was superior than a homogenous mixture of iron slabs and water. Shin and Hirayama [18], when calculating buildup factors using EGS4 code, discovered that some multilayer configurations produced lower buildup factors than their single component materials. Moreover, using two complementary metal-elastomer bilayers could decrease the overall shielding weight while providing attenuation equivalent to that of pure lead [23]. This was found to be useful in shielding garments. A polymer-tungsten bilayer had been determined to achieve better attenuation than a single layer of 0.2 mm tungsten [4]. Mann *et al.* [11] concluded that in gamma-ray protection, dual layered shields were more effective than single layer shields. A more recent study showed that bilayers of iron-containing materials were better than single layer of concrete in shielding neutrons [28].

Another interesting note to be highlighted here is the work by Hu *et al.* [22], Kim and Moon [24], and Cai *et al.* [29]. Their studies implemented an optimization technique called the genetic algorithm (GA) metaheuristic to design the most optimal multilayer and composite shield from a given set of materials. They were able to determine the best thickness ratios of individual layers and come up with new composite materials that showed better attenuation than lead oxide. Promoted by John Holland in 1970s, the GA method was inspired by natural evolution. This algorithm randomly generates a solution from a set of components, selects the most fit solutions, and then, uses them to build new generations which improve upon the previous generation. This technique may be helpful for optimizing multilayer shields that require various goals and constraints.

3.1. *Effects of Layer Arrangement*
Currently, there is no general agreement on the effects of changing the layer arrangement on the shielding performance. Some authors found that there was a significant difference when the layers were interchanged. Fuse *et al.* [21] demonstrated that the arrangements of the iron slabs and water could affect the resultant neutron flux and gamma dose rates. It was proven that the difference between the most optimum and the poorest performing arrangement was noticeable. The thickness of the layers and the order that they were assembled could also modify the value of neutron buildup factors which had to be considered in neutron calculations [17]. For neutron shielding, placing a hydrogenous material first was found to be better than the opposite order [6]. Meanwhile, Whetstone and Kearfott [26] claimed that there was no significant difference in shielding performance when the arrangement of materials was changed. This might due to the thin alternating layers of steel and polyethylene in all designs which resulted in a similar transmitted neutron spectrum.

In the case of X-ray shielding, McCaffrey *et al.* [23] observed that low-Z upstream/high-Z downstream could yield up to five times more attenuation than that of the reverse order at 50 kVp. However, this difference would decline and vanished at 150 kVp. Although, for the 150 kVp X-rays, Kim et al. [4] had results showing that high-Z/low-Z order was better than the opposite arrangement. They argued that as the X-rays passed through the high-Z layer, they would lose their energy where they could easily be shielded by the low-Z layer. Nonetheless, the authors stated that there was no optimal combination that could provide effective shielding for a wide energy range, thus the choice of material would have to be tailored for the particular X-ray energy in any application.

4. Conclusions
The main purpose of this study is to review the multilayer shielding used in literature. The designs that have been evaluated were listed. Focus was given on the effect of changing the material arrangement and also observations on buildup factor. The buildup factor for a multilayer shielding was found to be complicated and there was no clear trend on how to effectively arrange the materials. Research opportunities may lie in testing new multilayer designs. They also need to be accompanied by complementary buildup factor calculations. Future studies may also investigate on the application of metaheuristics in multilayer shielding optimization which may be useful in improving radiation protection. Examples of other metaheuristics are simulated annealing, tabu search, and ant colony optimization.

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