Monte Carlo Simulations of Cosmic-ray Interactions in order to Model Shielding Options for Neutron Multiplicity Counters

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Abstract

The interaction of high-energy cosmic-rays incident on a neutron multiplicity waste assay counter was modeled within the GEANT4 Monte-Carlo simulation code framework. The primary purpose of this study was to aid in the design and construction of shields to reduce the cosmic-ray induced background in neutron multiplicity counters. We demonstrate the feasibility for using the GEANT4 code for this application by comparing the results of this study to experimentally observed background rates.

KEYWORDS: Cosmic-ray shielding, Neutron Multiplicity Counters, Monte-Carlo simulations, GEANT4

1. Introduction

Neutron multiplicity counting is a well established technique to non-destructively assay plutonium and uranium-bearing bulk materials [1] such as nuclear waste. By detecting the multiplicity of neutrons generated by the spontaneous fission of isotopes such as ^{240}Pu and ^{252}Cf, the amount of fissioning material can be quite accurately and relatively quickly (compared to destructive analysis (DA) techniques) determined, if the relative isotopic composition can also be assessed, without opening the containment vessel. When combined with an isotopics assessment, the inventory of special nuclear material can be obtained non-invasively for the entire item and therefore without the sampling errors of DA.

The features of a generic modern neutron multiplicity and coincidence counters (NMC) include a matrix of high-density polyethylene (HDPE) surrounding a cavity of sufficient size to contain the item of interest. The fission neutrons from the item are thermalized in the HDPE and subsequently detected in ^{3}He filled proportional counters that are embedded within the polyethylene. Often a thin layer of cadmium is placed between the cavity and the polyethylene moderator to prevent thermalized neutrons from reflecting back into the cavity and possibly inducing fission. The cadmium also shortens the lifetime of the neutrons in the counter which allows the reduction of random coincidences. For wastes with a high external gamma-ray dose, the cavity may also be lined with an attenuator such as Pb. Pb and Cd along with the any steel work (such as loading systems and turn tables) are important in determining the background rates and should be used sparingly.

The neutron multiplicity counting technique can be used to detect very small amounts of fission materials. Ultimately, the limiting factor that determines the minimum detectable activity (MDA) is the background on the system. The primary source of background is caused by cosmic-ray interactions in the atmosphere that produce high-energy (0.1-10 GeV) neutrons, protons, and muons which “rain” down to the Earth’s surface [2]. These particles can themselves in-turn produce neutrons via e.g. spallation reactions (in both the body of the assay system and in the item being assayed) which ultimately can be detected in NMC’s to

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Table 1. List of the materials placed within the cavity and their molar mass values.

Target Material	Molar Mass ($g.mol^{-1}$)
Carbon	12.011
Water	18.015
Aluminum	26.982
Titanium	47.880
Iron	55.847
Copper	63.546
Tin	118.710
Lead	207.200

even high multiplicities (relative to the number of neutrons emitted per spontaneous fission). These can contribute a high and sporadic score to the correlated neutron signal used to assay special nuclear materials.

To reduce the cosmic-ray induced background, NMC's are often placed within a concrete bunker. Because of the large expense for the construction of a heavily shielded bunker, it is desired to optimize the shield thickness to meet the counters MDA requirements without significant over-engineering. Much of the present data in this regard is based on limited measurements and is often difficult to generalize to meet specific requirements. It is desired to have a model in which to quantitatively predict cosmic-ray induced background on a NMC in order to aid in optimized design of shielding options for these devices.

In order to simulate the cosmic-ray interactions within an NMC (and shielding), the Monte Carlo simulation program GEANT4 [3] was chosen. GEANT4 is a C++ framework for the simulation of particles through matter with particular attention paid to high-energy and nuclear physics applications. While not its original purpose, GEANT4 does include physics models to simulate and track very-low energy radiation (e.g. x-rays) and particles (e.g. thermal neutrons).

It is the purpose of this article to present the first results from a GEANT4 model simulation compared to experimental measurements, to demonstrate the suitability of GEANT4 to aid in the development of quantitative modeling of cosmic-ray backgrounds in non-destructive neutron assay systems.

2. Experimental Measurements

The experimental measurements used for comparison were first reported in Ref. [4]. A summary of these measurements is presented here. The measurements were performed with a high-level neutron coincidence counter of the HLNCC-II design [5]. The device was operated with shift-register electronics [6]. The counter contains eighteen ^{3}He tubes embedded in a cylindrically symmetric HDPE matrix with an 8.5 cm radius and 25.8 cm long internal cavity. The entire device stands about 60 cm high and is about 34 cm in diameter. This counter was measured to have an absolute efficiency of 20.66±0.15% to detect fission neutrons from a ^{252}Cf source placed at the center of the cavity.

Measurements of the total (Totals), and coincident (Reals in the parlance of the shift-register technique) event rates were performed with the counter inside an effectively unshielded aircraft hanger with the detector cavity either empty or containing one of a set of target materials. These materials and their molar masses (A) are presented in Table 1.

For each of these measurements, the total and coincident neutron production rates were determined as a function of the material within the cavity. The mean number of neutrons per cosmic-ray interaction is estimated by

$$\langle n \rangle = 2(R/T)(1/\varepsilon f_2), \tag{1}$$

where $\langle n \rangle$ is the mean number of neutrons, R the number of Reals, T the number of Totals, ε is the counter efficiency, and f_2 is the gate utilization factor. The gate utilization factor (GUF) is a correction to account for lost coincidences due to the limited length of the shift-register coincidence window. All the results were normalized to mass of material within the cavity chamber. The results of this study are presented with the simulated data in Section 4.

3. The Simulation Model

In order to effectively simulate the effects of cosmic radiation on the background of NMC's, it is necessary to have satisfactory descriptions of physical processes covering several GeV down to thermal energies. The GEANT4 Monte-Carlo code framework was chosen for this particular study because it not only has models that describe nucleon and other particle (e.g. muons and pions) interactions at GeV energies, but it also includes thermal neutron and low-energy electromagnetic models to simulate low-energy processes, as well as, intermediate energy models to link the two regimes. Consequently, GEANT4 can simultaneously describe reactions of high-energy cosmic rays in and around the NMC and track any resulting neutrons to thermal energies where they may be detected in the counter's ^{3}He detectors.

For this particular study, GEANT4 version 7.1 was used. Version 3.8 of the G4NDL neutron thermal cross sections library [3] was utilized in the modeling of the low-energy neutron transport. The developed model includes tracking of the created particles listed in Table 2. Also listed in the table are the physics models used for each of the particles. The detailed descriptions of each of the models are found in Ref. [7].

For tracking of the neutrons, the "HP" models listed in Table 2 are used for energies below 20 MeV. These models utilize point-wise cross sections extracted from the Evaluated Nuclear Data File ENDF/B-VI [8]. The modeling of neutron inelastic reactions above 20 MeV utilizes the GEANT4 implementation of the INUCL code [9] for intermediate-energy particle transport. This model includes the Bertini intra-nuclear cascade model with excitons, a pre-equilibrium model, a simple nucleus explosion model, a fission model, and an evaporation model. While these models were developed for hadrons with energies from 100 MeV to 5 GeV, it is also applied to the intermediate energy regime from 20 to 100 MeV.

The HLNCC-II counter was modeled within GEANT using dimensions based on the engineering drawings. Minor features such as the wheels and electronic components were not included in the model, but a floor of concrete was placed beneath the counter to account for the possibility of scattering from the floor back into the counter. Above the counter was another slab of concrete which could be varied in thickness in order to study the effects of attenuation cosmic radiation. This is a crude representation of the actual geometries which varied according to shielding room and setup, but the objective of the objective in these first studies was to test whether GEANT4 was a viable tool.

To model the detector response, the total number of ^{3}He(n,p)T reactions are logged on an event-by-event basis to provide a neutron multiplicity distribution for a given simulation run. The total number of neutrons (Totals) is determined by

$$T = \sum_{i=1}^{256} i N_i \,, \tag{2}$$

Table 2. Particles tracked in the GEANT4 simulation and the respective physics models utilized. The named models are using the GEANT4 nomenclature in Ref. [7].

Particle	Physics Models
Gamma-ray	Low-energy Rayleigh Scattering Low-energy Compton Scattering Low-energy Photoelectric effect Low-energy e^-e^+ Pair Production
Electron	Multiple Scattering Low-energy Ionization Low-energy Bremsstrahlung
Positron	Multiple Scattering Ionization Bremsstrahlung e^-e^+ Annihilation
Muon (μ^- and μ^+)	Multiple Scattering Ionization Bremsstrahlung Muon Pair Production Muon-Nuclear Interaction μ^- capture at rest
^{4}He, ^{3}He, ^{3}H, ^{2}H, Proton	Multiple Scattering Low-energy Ionization Low-energy Elastic Scattering Low-energy Inelastic Scattering
Neutron	HP Elastic Scattering (< 20 MeV) HP Inelastic Scattering (< 20 MeV) HP Capture (< 20 MeV) Low-energy Elastic Scattering (> 20 MeV) Intra-nuclear Cascade Model (> 20 MeV) LCapture (> 20 MeV)

where T is the number of Totals, i is the multiplicity, and N_i is the number of events that occurred with that multiplicity. The total number of coincidences (Reals) is determined by

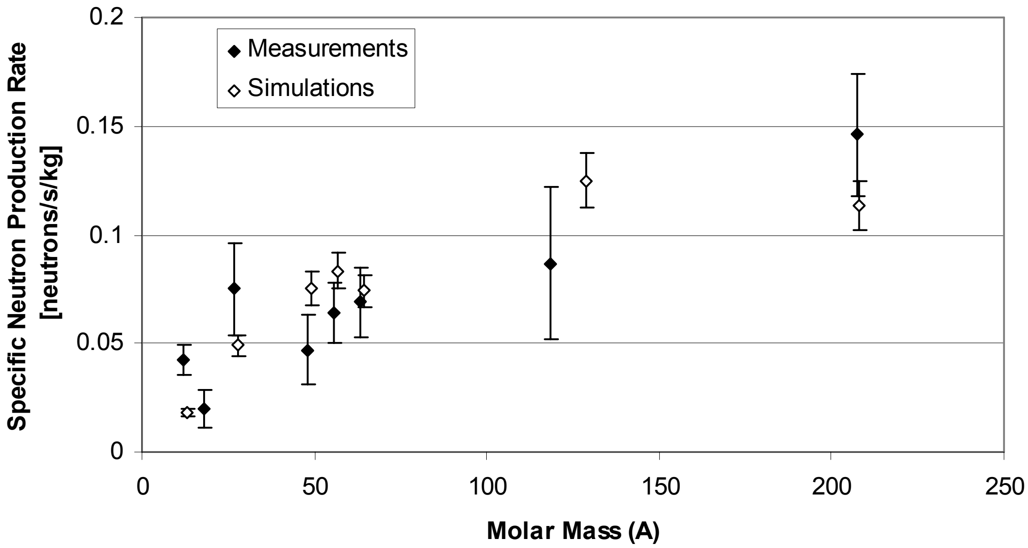
$$R = \sum_{i=2}^{256} \frac{i(i-1)}{2} N_i \, , \tag{3}$$

where R is the number of Reals, and i and N_i are the same as defined in Eq. 2.

It should be noted that there is no specific time requirement between ^{3}He(n,p)T reactions to be recorded. Experimentally, neutron multiplicity counters record only coincidences within a fixed time interval, however, in the experimental analysis a gate utilization parameter is applied to account for the limited gate width. Consequently, the simulated Totals and Reals events are comparable to the experimentally determined values reported in Ref [4].

Because it is not practical to simulate the cosmic-ray flux in its entirety, a series of single tests were performed to gauge the sensitivity of the number of ^{3}He(n,p)T reactions to various parameters. It was found that neutrons and protons show a similar dependence, but because protons also interact via electromagnetic processes they are more readily absorbed by the shielding and are consequently less important than high-energy neutrons. For this study, protons are not considered. Muons also represent a significant portion of the total

Figure 1. Experimentally measured (solid diamonds) and simulated (open diamonds) specific neutron production rate of various target material as a function of molar mass. The simulated results are slightly offset to the right to distinguish them from the experimental results.



cosmic-ray flux at sea level. These particles interact via electromagnetic and weak interactions. In addition, muons have a relatively short half life and are much less massive than protons and neutrons. Consequently, muons are much more penetrating than protons and neutrons. Because of this and their very different reaction behavior (compared to the Strongly interacting nucleons), they are considered separately from neutrons. From previous experimental studies [10], the effect of muons is expected to be at least an order of magnitude less important in creating background in NMC's.

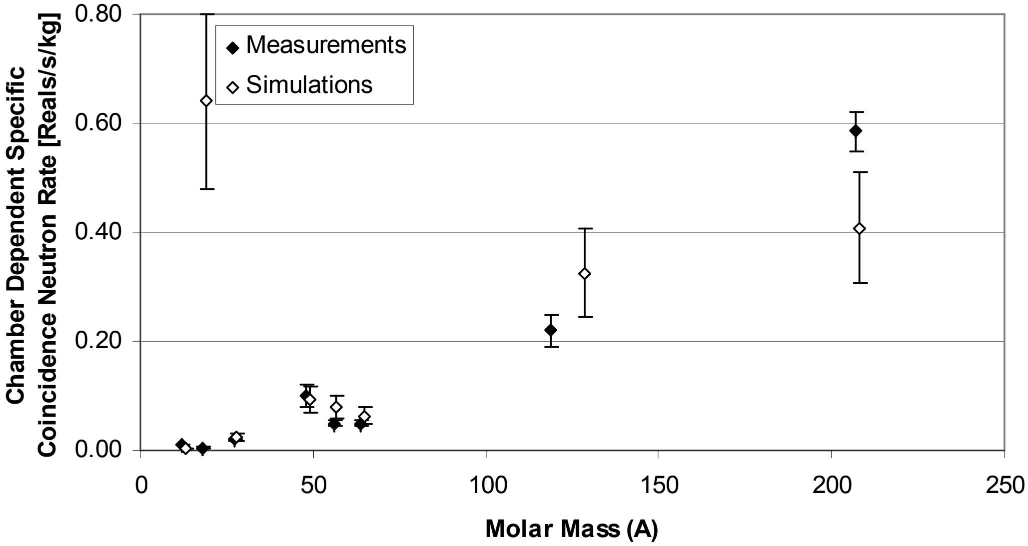
While cosmic rays can be produced anywhere in the atmosphere, the cosmic-ray flux is peaked at vertical angles, so the following simulations assume emission directly from above the counter. The positions of the vertically descending particles are distributed in area equal to the cross section of the counter, such that entire counter is irradiated from above.

To validate the model for fission-regime neutron energies, the neutrons of 2 MeV (the approximate average energy of ^{252}Cf fission-neutrons) were emitted in 4π from the center of the detector cavity. The counter efficiency was determined to be 20.9±0.5%, which is in acceptable agreement to the experimentally measured efficiency of 20.66±0.15%.

4. Results

To simulate the response of the HLNCC-II counter to cosmic-ray interactions, the modeled counter was bombarded from above with neutrons of energies 50, 100, 500, 1000, and 5000 MeV. The ^{3}He(n,p)T multiplicity distributions determined at each energy were additively combined after multiplying the 50, 100, 500, 1000, and 5000 MeV results by 100, 20, 4, 1, and 0.01, respectively. The multiplicative factors are estimated from the calculated energy-dependent cosmic neutron fluxes (abundances) presented in Fig. 8 of Ref. [2]. From these results the simulated Totals and Reals rates were determined using Eqs. 1 and 2.

Figure 2. Experimentally measured (solid diamonds) and simulated (open diamonds) specific coincidence neutron production rate of various target materials as a function of molar mass. The results are normalized to an efficiency and gate utilization factor of unity. The simulated results are slightly offset right to distinguish them from the experimental results.



Successive simulations were run in which the counter cavity was empty, or filled with an equal volume and 3 g.cm^{-3} density of each of the materials listed in Table 1. Due to programmatic reasons, the results for Tin could not be produced at the time of this writing. Alternatively, the results for Tellurium (A = 127.6 g.mol^{-1}) are presented.

Figure 3. Experimentally measured (solid diamonds) and simulated (open diamonds) mean number of neutrons released by cosmic-ray interactions as a function of the molar mass of the target material. The simulated results are slightly offset right to distinguish them from the experimental results.

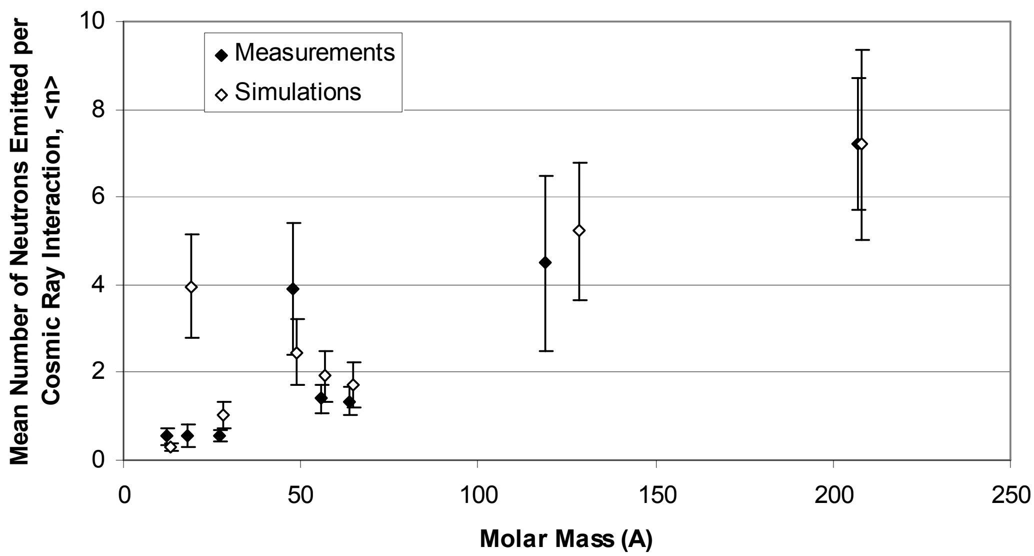
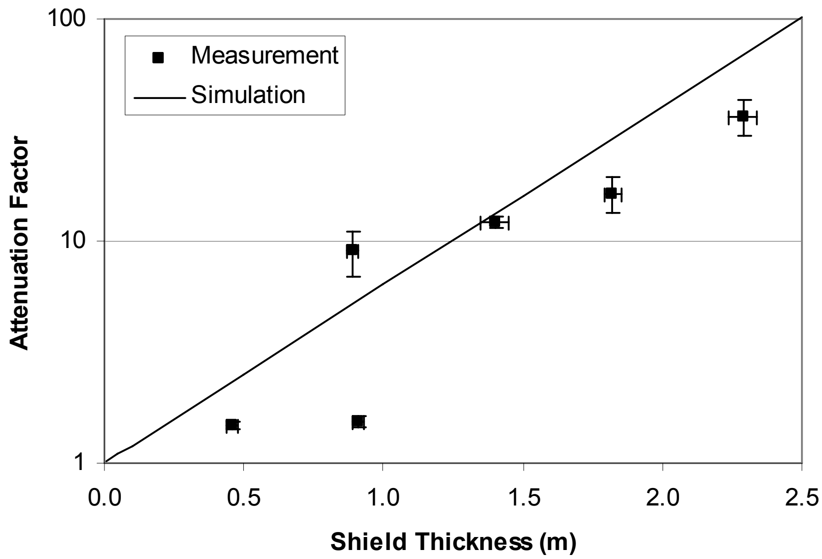


Figure 4. Attenuation factors for experimentally measured and simulated Totals rates as a function of thickness of concrete shielding above the neutron coincidence counter.



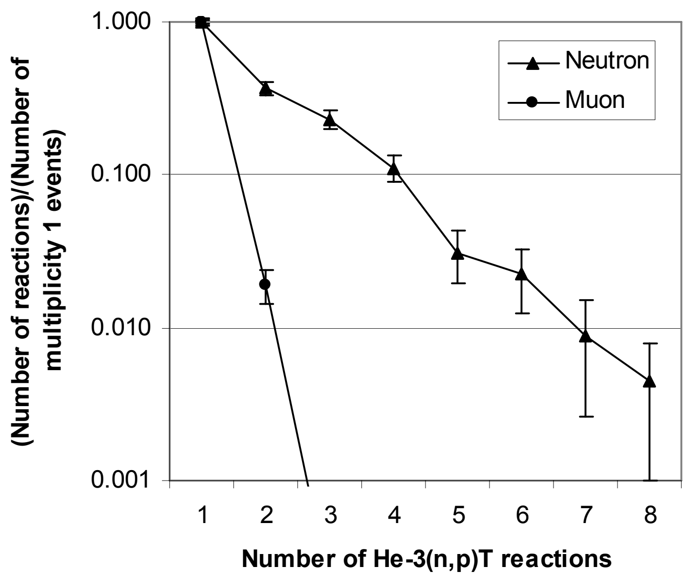
The results of the simulated specific neutron production rate are compared with the experimental results in Fig. 1. The experimental results are reproduced from Fig. 1 of Ref. [4]. The simulation values are normalized to the measurements by fitting the results (excluding water, see comments below) with a single scaling parameter. In general, the simulated trends are very comparable to the measured values. The neutron production rate calculated for water is about 15 times greater than measured value. The reason for this discrepancy is not yet understood, but considering the good agreement for the other materials, it is likely that this significant disagree is specific to water and not indicative of the model as a whole. It is speculated that this discrepancy may be related to inappropriate modeling of the (n,p) reaction which would be highly enhanced in water compared to other materials because of water's hydrogen content. It should be noted that water at 3 $g.cm^{-3}$ is also a severe non-realistic case, which may result in additional inconsistencies.

The results of the simulated specific neutron coincidence rate are compared with the experimental results in Fig. 2. The experimental results are reproduced from Fig. 2 of Ref. [4]. The simulation values are normalized using that same parameter determined for Fig. 1. Again, the simulated results are very comparable to the measured values. Also comparable to Fig. 1, is that the calculated result for water is significantly greater than measured result.

The mean number of neutrons emitted per cosmic-ray interaction was calculated from the simulated results using the approximated experimental prescription defined by Eq. 1. For the simulated results the GUF was set to one since all time bias corrections are included in the experimental values. The mean neutron rates from the simulation are compared with the experimental results in Fig. 3. The experimental results are reproduced from Fig. 3 of Ref. [4]. Excellent agreement is obtained for all materials, with the exception of water.

Also reported in Ref. [4] are the results of measured attenuation factors of the Totals and Reals background rates as a function of concrete thickness above the HLNCC-II counter. The experiment results showed that the attenuation factors are essentially equivalent for both the Totals and Reals values. Additional simulations were performed with the counter cavity filled with iron and with varying amounts of concrete shielding above the counter. For these

Figure 5. Simulated multiplicities of $^{3}He(n,p)T$ reactions per incident 1 GeV neutron (triangles) or muon (circles) normalized to the number of single $^{3}He(n,p)T$ reactions



simulations the neutrons were emitted in an area ten times the radius of the counter. In this way scattering from positions that would normally miss the unshielded counter could be accounted in the case were the shielding may actually result in an increased rate.

The comparison of the simulated and measured Totals attenuation factors are presented in Fig. 4. The simulated results agree reasonably well with the experimental results, however, there is a systematic bias in the simulation compared to the data is suggestive that further study into the model, as well as, a more quantitative understanding of the cosmic-neutron energy distribution are necessary.

The total flux of cosmic-muons is about twice as much as compared to cosmic-neutrons [2], however, unlike neutrons, in which the flux is strongly biased to low energies, the muon flux is relatively constant as a function of energy. Initial studies indicate that the muon interactions only produce events of very low multiplicity. This is illustrated in Fig. 5 in which the multiplicity distribution resulting from the impact of a 1 GeV neutron is compared to that of a muon of similar energy. One can see from this figure that high-energy neutrons produce secondary neutrons up to very high multiplicities. In contrast, the 98% of 1 GeV muons producing events in the coincidence counter have only multiplicity one. The result of this is that the muons would primarily contribute to Totals rates of the background, while the Reals rates are strongly influenced by cosmic-neutron events. A detailed study of the effects of muons on the coincidence counter backgrounds is still in progress.

5. Conclusions

The GEANT4 Monte-Carlo simulation framework has been studied to determine its suitability for reproducing cosmic-ray background rates in neutron multiplicity and coincidence counters. The model has been validated by comparing the simulated results to measured background rates of an HLNCC-II counter when the device's cavity is filled with different materials. The model was also compared to measured background attenuation factors due to concrete shielding above neutron counter.

The overall agreement between the measurements and the simulated results is considered to be excellent in this context. The most significant deviations are observed in the neutron production rates when the counter cavity is filled with (high-density) water. These deviations indicate a possible deficiency in the present model description, in particular for neutron-proton reactions. The good agreement obtained between simulation and experiment shielding attenuation factors provides additional confidence that the utilized models are appropriate. In order to provide quantitative predictions of shielding effects, further investigation is required as to the details of the energy distributions of the cosmic-ray particles and the application of a more realistic incident angular distribution, as well as, further studies of the effects of muons on neutron multiplicity and coincidence counters. The effect of shields other than monolithic concrete would be a primary area of focus in new shield design studies.

Overall these results provide a promising outlook that the GEANT4 can be useful a design and optioneering tool for demanding applied non-destructive assay applications of this kind. It represents a new level of sophistication not previously available to this field.

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