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Use of prompt gamma emissions from polyethylene to estimate neutron ambient dose equivalent

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ABSTRACT

The possibility of using measured prompt gamma emissions from polyethylene to estimate neutron ambient dose equivalent is explored theoretically. Monte Carlo simulations have been carried out using the FLUKA code to calculate the response of a high density polyethylene cylinder to emit prompt gammas from interaction of neutrons with the nuclei of hydrogen and carbon present in polyethylene. The neutron energy dependent responses of hydrogen and carbon nuclei are combined appropriately to match the energy dependent neutron fluence to ambient dose equivalent conversion coefficients. The proposed method is tested initially with simulated spectra and then validated using experimental measurements with an Am–Be neutron source. Experimental measurements and theoretical simulations have established the feasibility of estimating neutron ambient dose equivalent using measured neutron induced prompt gammas emitted from polyethylene with an overestimation of neutron dose at very low energies.

1. Introduction

The measurement and estimation of dose equivalent quantities are important for ascertaining radiological safety in neutron generating nuclear facilities. The dose equivalent outside the shield around these facilities such as particle accelerators is dominated by the neutron component [1]. The type of monitor employed in practice for many years is the so-called rem counter that works on the principle of thermalization of fast neutrons and subsequently measuring the fluence of these thermal neutrons. The simulated response matrix of the rem counter to monoenergetic neutrons is used to estimate the neutron ambient dose equivalent, $H^*(10)$. The response of such a rem counter is considered acceptable for neutron energies between thermal and about 10 MeV, although the monitor practically underestimates $H^*(10)$ in the energy range from thermal to about 1 eV and overestimates it in the energy interval 1 eV–100 keV. Above 10 MeV the response reduces significantly, leading to an underestimation of the ambient dose equivalent [2,3]. Moreover, the response of the thermal neutron detectors used in rem-meters changes with time requiring calibration of the instrument at regular intervals (almost every year). It is always desirable to develop new techniques of measurements and theoretical simulations for dose assessments, though many such techniques exist including both passive and active methods of neutron dose estimation. One such technique is the use of activation foils of different elements, where delayed gamma rays emitted from those elements due to the interaction of neutrons are used to estimate neutron dose. Prompt gamma rays are also emitted almost instantaneously from such neutron induced reactions. In this paper, a novel technique for estimating the neutron dose from the prompt gamma intensities is proposed in detail.

Prompt gamma measurements are generally utilized as a field-deployable technique for estimating the composition of large samples, concentration of toxic elements in human organs, oil logging etc. [4–7]. In the above said applications, the elements of interest are present in trace quantities and hence a high intensity neutron source is required. However, if any bulk moderator material is used as the target specimen, then the amount of prompt gammas emitted will be larger and will serve to mitigate conveniently the requirement of a strong neutron source. Measurement of prompt gammas emitted due to thermal and higher energy neutrons absorbed in materials embedded inside a thermalizing medium can be possibly employed to estimate the ambient neutron dose equivalent. This can be done using a combination of some judiciously selected materials such that the combined energy-differential probability of prompt gamma emissions matches well in shape with the distribution of energy dependent neutron fluence to ambient dose equivalent conversion coefficients (DCC). Hereafter the abbreviation DCC will stand for neutron fluence to ambient dose equivalent conversion coefficients unless explicitly mentioned otherwise. In the present work possibility of using High Density Poly Ethylene (HDPE) material for such application is studied using the Monte Carlo simulation code FLUKA (2011.2c) [8,9].

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However, the problem of determining the sample composition or the neutron dose from the measured peak areas is difficult because it is nonlinear. The spatial and energy distributions of the differential neutron fluence are modified by the large sample in a non-trivial, non-separable and composition dependent way. This problem, however, can be addressed by a proper Monte Carlo simulation using an accurate geometry and composition of the sample. The response of the system thus estimated will give the probability of a monoenergetic neutron incident on the sample will generate a prompt gamma which after escaping the sample (with or without having interactions inside the material) will deposit its energy to a gamma detector (spectrometer).

In this paper attempts have been made using theoretical simulations to demonstrate the feasibility of estimating neutron ambient dose equivalent from the measured peak area of the characteristic gamma rays emitted from hydrogen and carbon nuclei present in a HDPE cylinder. In order to achieve this, an estimate of the response matrix for mono energetic neutrons has been done using the FLUKA code. The response matrix consists of two parts; one for the 2.2 MeV prompt gammas from neutron capture by hydrogen and the other for 4.43 MeV prompt gammas from neutron interactions with carbon. These two responses are suitably combined to produce a close match to the energy dependent DCC for neutrons [10]. Several different test neutron spectra are used to calculate area under the gamma peaks (both 2.2 MeV and 4.43 MeV) in a NaI detector and consequent estimation of neutron dose for comparison with the dose computed by folding those test spectra with DCC. Finally, to validate the Monte Carlo simulations experimentally measured spectra are compared with the Monte Carlo simulated results. In the present simulations, gamma rays generated from the neutron interactions in the surrounding non-sample materials and contributing to gamma channel of interest have not been accounted when compared with the experimental measurements. Instead, measurements done in the absence of the sample and then subtracted from the spectrum measured with the sample are considered adequate to eliminate back ground contributions. The present method, utilizing secondary gammas instead of generally used secondary charged particles generated from neutron interactions, is expected to suffer from loss of detection efficiency. However, this problem can be reduced by increasing the mass of the HDPE, the number and size of the gamma detector. Considering the increase in bulk of the system, it can very well be used as a fixed monitor, if not as a portable one.

2. Theory

In order to develop the proposed technique of dose estimation, it can be assumed that when a gamma spectrum is taken, the significant constituents (e.g. hydrogen and carbon) of the HDPE sample can be identified through well resolved peaks in the gamma spectrum and it is possible to determine the net area of these peaks, excluding the continuum part of the spectrum. The net peak areas A_i, with the continuum background subtracted is related with the incident neutron energy distribution φ_j and the response matrix R_{ij} as follows:

$$A_i = \sum_j R_{ij}\varphi_j \tag{1}$$

where the subscript i indicates the constituent element (hydrogen or carbon in the present case) and j denotes the energy bin of the incident neutron energy distribution. Here, R_{ij} is the probability that a neutron emitted from the source in the energy bin j will interact inside the HDPE cylinder, undergo a possible change in energy, generate a gamma photon such that it is detected by the NaI detector and contributes a count in the peak area A_i. Alternatively, after going through all these processes as described above, the neutron from the source makes a contribution R_{ij} to the peak area with probability one. The detail of the simulation is described later in the text.

Now, the elements R_{ij} are made to fit the energy dependent DCC [10] denoted as F_j such that

$$F_j \cong \sum_i C_i R_{ij} \tag{2}$$

The parameters C_i can be obtained using the least square fitting in the following way. In Eq. (2) the inequality can be replaced by an equality by introducing a term ε_j as

$$F_j = \sum_i C_i R_{ij} \pm \varepsilon_j \tag{3}$$

where ε_j represent the error corresponding to the j-th energy bin are unknown and have contributions that may be positive or negative. However, ε_j^2 is always positive such that

$$\sum_j \varepsilon_j^2 = \sum_j \left(F_j - \sum_i C_i R_{ij}\right)^2 \tag{4}$$

The parameters C_i can be estimated by solving the simultaneous equations obtained using the conditions

$$\frac{\partial\left(\sum_j \varepsilon_j^2\right)}{\partial C_i} = 0 \quad \text{for all } i. \tag{5}$$

The neutron ambient dose equivalent $H^*(10)$ is related to the neutron fluence as:

$$H^*(10) = \sum_j F_j \varphi_j \tag{6}$$

Using Eq. (2) one gets

$$H^*(10) \cong \sum_i C_i \sum_j R_{ij}\varphi_j = \sum_i C_i A_i \quad \text{(using Eq. (1))} \tag{7}$$

Eq. (7) gives a method of approximating neutron ambient dose equivalent from measured peak areas of prompt gammas emitted from polyethylene due to neutron interactions. The estimate is as good as the agreement between the two sides of Eq. (2) i.e., smaller the ε_j^2, the better is the agreement.

3. Materials

High Density Polyethylene (HDPE) is a good neutron moderator containing hydrogen and carbon and is used as a moderating material in many neutron rem counters to thermalize fast neutrons. Once the thermal and fast neutrons interact with polyethylene, prompt gammas of energy 2.2 MeV from hydrogen and 4.43 MeV from carbon nuclei are emitted. The cross-section for prompt gamma emissions from these two elements are plotted in Fig. 1. As it is seen from Fig. 1(a) the element hydrogen (^{1}H) has a very high cross-section [11] near thermal and lower energies to produce ^{2}H and gammas in its subsequent decay. The nuclear decay scheme of ^{2}H is nearly complete and well known therein reducing the ambiguity in prompt gamma lines. In addition to the capture gammas from hydrogen, a 4.43 MeV prompt gamma is also emitted because of the ^{12}C (n, n′γ) ^{12}C reaction of fast neutrons with carbon present in HDPE. But the cross-section of this particular reaction has a threshold of about 4 MeV and has resonances at 8 MeV and 11 MeV [12] as it is evident from Fig. 1(b). Thus the prompt gammas emitted due to this reaction are highly energy dependent. The HDPE used in the present work has density of 0.92 g cm^{-3} and is considered to be composed of carbon and hydrogen in the weight fraction of 0.856 and 0.144 respectively.

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