Monte Carlo study to measure the neutron flux using the prompt gamma-rays in hydrogen

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A Monte Carlo study was realized to use a gamma-ray spectrometer with a 3" $\emptyset \times$ 3" NaI(Tl) detector, with a moderator sphere, to measure the neutron fluence rate. Materials with a large amount of hydrogen are able to slowdown and thermalize neutrons; once thermalized there is a probability of neutron capture in moderator nuclei. When the neutron is captured by hydrogen a 2.22 MeV prompt gamma-ray is emitted. The photons pulse-height spectrum shows a photopeak around 2.22 MeV whose net area is proportional to total neutron fluence rate. The characteristics of this system were determined by a Monte Carlo study using the MCNP 4C code; a model of the NaI(Tl) was utilized, the study includes water and polyethylene as moderators with 3, 5, and 10 inches-diameter spheres that were bombarded with neutrons emitted by a polyenergetic ²³⁹PuBe. With the best sphere the study was extended to determine the response to monoenergetic neutron sources. Calculations were carried out to obtain realistic gamma-rays pulse height spectra, where the 2.22 MeV is observed. In polyethylene spheres the 4.43 MeV photon is also shown due to ¹²C(n, n' γ)¹²C reaction.

Keywords: Monte Carlo; neutrons; moderators; prompt gamma-rays.

Mediante métodos Monte Carlo se ha realizado un estudio con el propósito de utilizar un espectrómetro de rayos gamma, con base en un centellador de NaI(Tl) de 3" $\emptyset \times$ 3" y un moderador esférico, para medir la tasa de fluencia de neutrones. Los materiales que contienen hidrógeno tienen la capacidad de moderar y termalizar neutrones; una vez termalizados los neutrones pueden ser capturados por los núcleos del moderador. Si la captura ocurre en hidrógeno un fotón de 2.22 MeV se produce y el espectro de altura de pulsos muestra un fotopico en torno a los 2.22 MeV, el área neta bajo el fotopico es proporcional a la tasa total de la fluencia de neutrones que incide sobre el moderador. Las características de este sistema han sido determinadas mediante métodos Monte Carlo usando el código MCNP 4C, donde se utilizó un modelo del centellador. Durante el estudio se incluyó como medios moderadores esféricos de agua y de polietileno de 3, 5 y 10 pulgadas de diámetro. Los moderadores se bombardearon neutrones emitidos por una fuente de ²³⁹PuBe. Con el mejor moderador es estudio se extendió para determinar la respuesta del sistema ante neutrones monoenergéticos. Los cálculos se efectuaron tratando de obtener espectros de altura de pulsos de los rayos gamma similares a los observados en el multicanal. Durante el estudio se observaron los fotones de 2.22 MeV. Para el caso del polietileno, también se observó un fotón de 4.43 MeV proveniente de la reacción nuclear ¹²C(n, n' γ)¹²C.

Descriptores: Monte Carlo; neutrones; moderadores; rayos gamma prontos.

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1. Introduction

Several techniques have been investigated for bulk detection of illicit drugs, explosives and other contraband materials. The illicit drugs and explosives can be identified by the determination of the ratios of the elements hydrogen, carbon, nitrogen and oxygen as their major constituents by using different neutron interactions. The characteristics gamma rays 2.22, 4.44, 10.8 and 6.13 MeV respectively emitted by H, C, N and O makes possible the elemental analysis of bulk samples [1-3].

The simplest nuclear reaction is the capture of a low energy neutron by a stable nucleus. Because of the binding energy of the added neutron, after capture the nucleus is left in an excited state, several MeV above ground level. This capture state deexcites within 10^{-14} seconds by emission of gamma-rays [4]. The energies of the gamma-rays are characteristic of the isotopes of the elements and their intensities are proportional to their concentration, thus the prompt-ray spectrum (pulse height spectrum in a multichannel analyzer) car-

ries quantitative and qualitative information of the elements present in the sample. This technique is useful to determine the most low Z elements like H, B, Si, P, S, and Ti which are difficult to measure using the traditional neutron activation analysis. It is also useful to detect heavier elements. However, the poor sensitivity due to low intensity of neutron beams ranging from 10^6 to 10^8 cm²-s⁻¹ is a drawback of this method [5].

HPGe, NaI(Tl) and BGO scintillators are commonly utilized in industrial prompt γ -ray neutron activation analysis (PGNAA). NaI(Tl) and BGO detectors have large γ -ray detection efficiencies; beside they can be used without cooling. in comparison to HPGe detector [6-7]. PGNAA differs from Instrumental neutron activation in that prompt γ -ray emissions from a sample are counted during neutron irradiation rather than counting decay γ 's at some time after irradiation [8].

PGNAA requires the use of a neutron source; this can be a Nuclear Reactor [4,5,9,10], a 252 Cf spontaneous fission source [11,12] and isotopic neutron sources [1,13,14].

Having the fluence-to-dose coefficients, the neutron fluence rate and the average neutron energy of a neutron source, the neutron dose can be easily estimated [15]. A related problem to this procedure is to measure the total neutron fluence rate.

The aim of this study was to apply the prompt gammaray, emitted during the neutron capture in hydrogen, to determine the total neutron fluence rate of a polyenergetic neutron source.

2. Materials and methods

Moderators are materials with low Z nuclei, some of these materials are light water, heavy water, polyethylene, lucite, paraffin, berylium and graphite. Materials with high concentration of hydrogen can reduce the neutron energy in a single head-to-head collision. When neutron reaches thermal energies has a large probability to be captured by hydrogen nucleus producing a characteristic 2.22 MeV photon. The amount of 2.22 MeV photons is directly related to the total neutron fluence rate. In this procedure the size of moderator media is important to guarantee the neutrons moderation and thermalization.

In this investigation the Monte Carlo method, using the MCNP 4C code [16] was utilized to model a spherical moderator located above a 3" \Box 3" NaI scintillator with a cladding made of aluminum and with a base made of lucite. As source term the neutron spectrum produced by a ²³⁹PuBe point-like neutron source was utilized. The model is shown in Fig. 1.

The model is based in the "Grey" neutron detector proposed by Pönitz [17,18] to measure the neutron cross sections. The elemental concentration of detector and moderator was obtained from Seltzer and Berger [19]. Spheres, of light water and polyethylene were used to model the moderator. Three, five and ten inches-diameter spheres were used to determine the best. Neutron source spectrum was obtained from IAEA compilation [20].

During calculations neutrons are emitted by the source, reaching the moderator; neutrons suffer different interactions



FIGURE 1. Model used in the Monte Carlo study



FIGURE 2. Mass attenuation of polyethylene



FIGURE 3. Mass attenuation of NaI.

with moderator nuclei, some of these are absorbed by the moderator or are leaked out from the moderator, other neutrons reach thermal energies and are captured by hydrogen, the neutron capture reaction rate, RR, is given by Eq. (1).

$$RR = \Phi_{TH} \Sigma_{n,\gamma} \exp[-\Sigma_a \rho] \tag{1}$$

Here, Φ_{TH} is the thermal neutron fluence rate, $\Sigma_{n,\gamma}$ is the (n, γ) prompt gamma macroscopic cross section Σ_a is the neutron absorption cross section and ρ is the distance traveled, inside the moderator, by the neutron before it is absorbed.

The RR becomes in a 2.22 MeV volumetric gamma-ray source, S_V , that isotropically emits the photons inside the moderator, then the RR becomes the S_V . The photons must travel from the production site to the NaI(Tl) location. Along its path photons can be attenuated by the moderator sphere before they reach the scintillator. The uncollided gamma-ray flux that reach the scintillator is given in Eq. (2).

$$\Phi_{\gamma} = \frac{B S_V}{2 \mu} \left(1 - \frac{1}{2 \mu R} + \frac{\exp[-2 \mu R]}{2 \mu R} \right) \quad (2)$$

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FIGURE 4. Monte Carlo pulse height spectra of prompt gamma rays produced by the neutrons emitted by a point-like ²³⁹PuBe neutron source impinging on polyethylene and light water moderator spheres.



FIGURE 5. Monte Carlo pulse height spectra of prompt gamma rays produced by the neutrons emitted by monoenergetic, isotoropic point-like neutron sources impinging on 10 inches-diameter polyethylene sphere moderator.

Here, B is the photons build up factor, μ is the linear attenuation coefficient, R is the sphere radius.

In Fig. 2 it is shown the photon attenuation properties of polyethylene. Here, some of them pass through the NaI(Tl) without being detected, while others are totally or partially absorbed; in Fig. 3 the γ -rays attenuation properties of NaI(Tl) is shown. Polyethylene and NaI(Tl) attenuation features were calculated with WinXCOM code [21,22].

From the Monte Carlo calculations the best moderator was the 10 inches-diameter polyethylene sphere. With this moderator the study was extended to determine the system response to isotropic point-like neutron sources. The response was calculated in terms of the expected pulse height spectra observed at a multichannel analyzer, using 256 channels ranging photons from 0 to 5 MeV.

3. Results and discussion

In Fig. 4 is shown the calculated pulse height spectra of 2.22 MeV photons produced by the capture of neutrons in hydrogen. Neutrons are emitted by a point-like ²³⁹PuBe neutron source interacting with 3, 5 and 10 inches-diameter spheres made of light water and polyethylene. The 2.22 MeV photopeak has the maximum for 10 and 5 inches-diameter spheres, however the 5 inches-diameter sphere has a lower base because shows a peak in 4.43 MeV followed by two escape peaks, then the net area under 2.22 MeV photopeak is larger for 10" polyethylene sphere. In all cases the 2.22 MeV maximum is lower for water moderator spheres. Thus the 10 inches-diameter sphere shows the best performance.

Polyethylene spheres show a peak in 4.43 MeV due to ${}^{12}C(n, n' \gamma){}^{12}C$ reaction; this peak is larger for 3" sphere and is lower for 10" spheres also observed, with lower efficiency, in the 10 inches-diameter sphere. This peak is not observed in light water spheres, because here there is not carbon. The other peaks close to 4.43 MeV are its single and double escape peaks. These results are in agreement to the results reported in literature [2,3,7].

In Fig. 5 it is shown the pulse height spectra of 2.22 MeV photons produced during the neutron interaction of isotropic and monoenergetic neutron sources impinging on the polyethyelene 10 inches-diameter sphere. Here, the lower response is observed for 0.1 MeV neutrons; for larger energies the 2.22 MeV photopeak is approximately independent of neutron energy. From neutrons with energies larger than 5 MeV the 4.43 MeV photons due to ${}^{12}C(n, n' \gamma){}^{12}C$ is observed.

The area under the 2.22 MeV is approximately constant for neutrons whose energy range from 0.5 up to 12 MeV, this feature indicates that this system can be used to measure the total neutron flux for mononoenergetic and polyenergetic neutron sources.

4. Conclusions

During transport in moderator media neutrons loose energy until they become thermal; with this energy neutrons are captured by hydrogen producing a characteristic 2.22 MeV photon.

The prompt gamma photon is measured by a NaI scintillator, these response can be obtained by a gamma-ray spectrometer with a multichannel analyzer that shows the pulse height spectrum, where the area under the 2.22 MeV is proportional to total neutron flux.

Three diameter spheres, made of water and polyethylene, were utilized to determine the response of this system. The best performance was obtained with the 10 inches-diameter sphere, where the area under the 2.22 MeV photopeak is the largest. For polyethylene spheres a 4.43 MeV is observed due to ¹²C(n, n' γ)¹²C reaction.

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The 10 inches-diameter polyethylene sphere with a 3" $\emptyset \times 3$ " NaI has approximately the same area under the photopeak for neutron from 0.5 to 12 MeV, this allows to measure the neutron flux for monoeneretic and polyenergetic neutron sources. For neutrons ranging from 0.5 to 12 MeV the 4.43 MeV is also observed.

This Monte Carlo study will be extended for different

- A. Pazirandeh, A. Maryam, and S. Farhad Masoudi, *Appl. Radiat. Isot.* 64 (2006) 1.
- P.A. Dokhale, J. Csikai and L. Oláh, *Appl. Radiat. Isot.* 54 (2001) 967.
- T. Cywicka-Jakiel, J. Loskiewicz, M. Nezamzadeh and G. Tracz, *Appl. Radiat. Isot.* 51 (1999) 419.
- R.L. Paul and R.M. Lindstrom, J. Radioanal. Nucl. Chem. 243 (2000) 181.
- 5. A.G.C. Nair et al., Nucl. Instrum. Meth. Phys. Res. A 516 (2004) 143.
- R.P. Gardner and Ch.W. Mayo, *Appl. Radiat. Isot.* 51 (1999) 189.
- R. Proctor, S. Yusuf, J. Miller and C. Scott, *Nucl. Instrum. Meth. Phys. Res. A* 422 (1999) 933.
- D.L. Anderson, Y. Sun, M.P. Failey and W.H. Zoller, *Geostandards Newsletter* 9 (1985) 219.
- D.L. Anderson, W.C. Cunningham and E.A. Mackey, "Neutron capture prompt-γ activation analysis of foods". Published in Schruzer, G.N. (editor). Biological Trace Element Research. The Humana Press Inc. (1990).
- K. Sudarshan et al., Nucl. Instrum. Meth. Phys. Res. A 457 (2001) 180.
- 11. S.J.S. Ryde, W.D. Morgan, C.J. Evans, A. Sivyer and J. Dutton, *Phys. Med. Biol.* **34** (1989) 1429.

neutron sources, as well as, the experimental verification of this performance.

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- 12. M. Borsaru, M. Biggs, W. Nichols and F. Bos, *Appl. Radiat. Isot.* **54** (2001) 335.
- R.J. Shypailo and K.J. Ellis, J. Radioanal. Nucl. Chem. 249 (2001) 407.
- D.L. Collico, M.A.J. Mariscotti and S.R. Guevara, Nucl. Instrum. Meth. Phys. Res. B 95 (1995) 379.
- H.R. Vega-Carrillo, E. Manzanares, V.M. Hernández, E. Gallego and A. Lorente, *Rev. Mex. Fis.* 51 (2005) 494.
- J.F. Breismeister (editor), "MCNPTM-A general Monte Carlo N-particle transport code", Los Alamos National Laboratory Report LA-13709. (2000).
- 17. W.P. Pönitz, Nucl. Instrum. Meth. 58 (1968) 39.
- 18. W.P. Pönitz, Nucl. Instrum. Meth. 72 (1969) 120.
- S.M. Seltzer and M.J. Berger, Int. J. Appl. Radiat. Isot. 33 (1982) 1189.
- R.V. Griffith, J. Palfalvi, and U. Madhvanath, U. (editors). "Compendium of neutron spectra and detector responses for radiation protection purposes". International Atomic Energy Agency, *Technical Report Series No. 318*. Vienna (1990).
- L. Gerward, N. Guilbert, K.B. Jensen and H. Levring, *Radiat. Phys. Chem.* 60 (2001) 23.
- 22. L. Gerward, N. Guilbert, K.B. Jensen and H. Levring, *Radiat. Phys. Chem.* **71** (2004) 653.