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Flux measurements for a DD neutron generator using neutron activation analysis

Walid A. Metwally[1] · Samar El-Sayed[1] · Ahmad Ababneh[1] · David L. Williams[2] · Allan X. Chen[2]

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Abstract The fast and maximum thermal neutron fluxes from the DD-109 neutron generator at the University of Sharjah were experimentally measured by the activation technique using different neutron reactions. The thermal and fast neutron fluxes were found to be 2.960×10^6 and 6.186×10^7 n/cm^2 s, respectively. This was done to verify the modeling results for the optimum moderator thickness needed to maximize the thermal neutron flux. The optimum moderator thickness was found to be between 3.5 and 4 cm. The present data were compared with the detailed MCNP model-based calculation performed in earlier work to simulate the generator.

Keywords Neutron activation analysis · Neutron generator · Neutron moderation · Thermal neutron flux measurement

1 Introduction

Neutron generators are becoming increasingly available to educational institutions for different applications and research purposes [1–3]. The most common neutron generators use deuterium (^{2}H) and tritium (^{3}H) in fusion reactions to produce neutrons. The resulting neutron energy is 2.5 MeV for a deuterium–deuterium (DD) reaction and 14.1 MeV for a deuterium–tritium (DT) reaction. Both types of generators produce a high neutron flux in the range of 10^7–10^{10} n/s for DT sources and 10^5–10^{10} n/s for DD sources [4].

Important applications where neutron generators can be effectively used are Neutron Activation Analysis (NAA) using delayed gamma rays, Prompt Gamma Neutron Activation Analysis (PGNAA), Inelastic Neutron Activation Analysis (INAA), and neutron radiography. These applications require the utilization of fast or thermal neutrons [4, 5]. Activation analysis (NAA or PGNAA) is important to determine the amounts of isotopes in a sample at different concentrations. For many isotopes, the ability to precisely determine their concentrations is enhanced by utilizing either thermal neutrons in the activation process, where the capture cross section is high.

The University of Sharjah acquired a model DD-109.4 M neutron generator from Adelphi Technology [6]. The generator is designed with three neutron ports that can be plugged with polyethylene moderator layers of different thickness to control the neutron flux at different regions in the port. As the thickness of the polyethylene increases in the port, the fast neutron flux decreases and the thermal neutron flux increases until a certain thickness where the thermal neutron flux starts to decrease due to neutron absorption. One of the main intended uses of this generator is to perform neutron activation analysis. As such, it is important to have an accurate measure of both the fast and thermal neutron fluxes in the ports as well as subject the sample to the maximum thermal neutron flux. Maximizing the thermal neutron flux also leads to minimizing the time required to activate the sample and efficient utilization of the neutron generator.

✉ Walid A. Metwally
WMetwally@sharjah.ac.ae

[1] Department of Nuclear Engineering, University of Sharjah, Sharjah 27272, UAE

[2] Adelphi Technology, 2003 E Bayshore Rd., Redwood City, CA 94063, USA

An MCNP [7] model of the neutron generator and the ports has been previously developed [8] in order to analyze the neutron flux in the ports and maximize the thermal neutron flux. The model took into account the neutron flux from both the forward and backscattered neutrons reaching the sample as a result of the polyethylene moderators. In addition, a detailed neutron flux mapping at different moderator thicknesses was presented.

While neutron activation is a common technique for characterizing the flux in neutron generators [9, 10], it is important to perform this characterization for specific combinations of generator types and geometrical configurations. The work herein describes the different neutron activation approaches used to measure the fast and maximum thermal neutron flux that can be obtained from the neutron generator at the University of Sharjah. In addition, this work presents an effective method of maximizing the thermal neutron flux using backscattered neutrons.

2 Neutron generator

The DD-109.4M generator uses the D–D fusion reaction and is driven by an ion beam supplied by a high current ($\leq$ 35 mA) microwave ion source [6]. The deuterium ions (D^+) are accelerated from the ion source's extraction iris to a titanium (Ti) *V*-target where the fusion reaction occurs.

The DD109.4M has two major levels of shielding: (1) the inner shielding and moderator and (2) the outer shielding. The outer shielding is made of a 7.62 cm-thick high-density polyethylene (HDPE) that can be opened by sliding the two sides apart, exposing the inner shielding and moderator. The inner shielding is made of an outer layer of 1.27 cm lead, followed by layers of 2.54 cm borated polyethylene (BPE) forming 15.24-cm-thick horizontal walls and 10.2-cm-thick floor and ceiling. The next layer is the HDPE moderator which is cut to contour around the generator head. Three cylindrical ports (in red) are cut directly through walls of outer and inner shells and the moderator reaching into the generator head (source). Disks of HDPE can be inserted in the ports to form various thicknesses of moderation, thereby varying the distribution of thermal and fast neutrons. The minimum distance at which the moderators can be placed is 7.62 cm. HDPE and borated polyethylene plugs are also utilized in the ports, if needed, to block the neutrons from being emitted in the surrounding area. The shielding arrangement and two of the ports are shown in Fig. 1.

3 Theory

The activation process of a sample and its corresponding activity as a function of time are shown diagrammatically in Fig. 2.

When a sample is irradiated with a flux, Φ, neutrons/cm^2/s it becomes activated; the sample activity at the end of the irradiation is given by [11, 12],

$$A = N \cdot \Phi \cdot \sigma \left[1 - e^{(-\lambda \cdot t_i)}\right], \tag{1}$$

where N is the number of atoms in the sample, σ is the neutron absorption cross section, λ is the decay constant, and t_i is the irradiation time.

The number of counts under the peak, C, measured by a detector of intrinsic peak efficiency η, between the start of the measurement, t_1, and the end of the measurement, t_2, (with respect to the end of the irradiation time which is defined to be $t = 0$) is given by:

$$C = Y\eta \int_{t_1}^{t_2} A_{(t=0)} e^{(-\lambda t)} dt = A_{(t=0)} Y \frac{\eta}{\lambda} \left[e^{(-\lambda t_1)} - e^{(-\lambda t_2)}\right], \tag{2}$$

where Y is the yield of the gamma ray being measured. Consequently, the activity at the end of the irradiation time, based on the measured number of counts, is:

$$A_{(t=0)} = \frac{C\lambda}{Y\eta[e^{(-\lambda \cdot t_1)} - e^{(-\lambda \cdot t_2)}]}, \tag{3}$$

where $A_{(t=0)}$ is the activity at the end of the irradiation period. Combining Eqs. 1 and 3 results in the following equation to calculate the neutron flux of the generator

$$\Phi = \frac{C\lambda}{NY\eta\sigma\{e^{(-\lambda \cdot t_1)} - e^{(-\lambda \cdot t_2)}\}\{1 - e^{(-\lambda \cdot t_i)}\}}. \tag{4}$$

Equation 4 requires that the number of atoms in a sample is known. For compounds, two or more elements are simultaneously irradiated, and the elemental abundance needs to be taken into account. The activity of each element at $t = 0$ can be independently determined because of the differing gamma ray signatures.

Gold foils are commonly used for thermal neutron flux measurement because it is readily activated, emits a single gamma ray, and has a relatively long half-life. Another single gamma ray emitter is aluminum, but it has a much shorter half-life. Indium foils are commonly used for the fast neutron flux measurements by utilizing the inelastic scattering of neutrons.

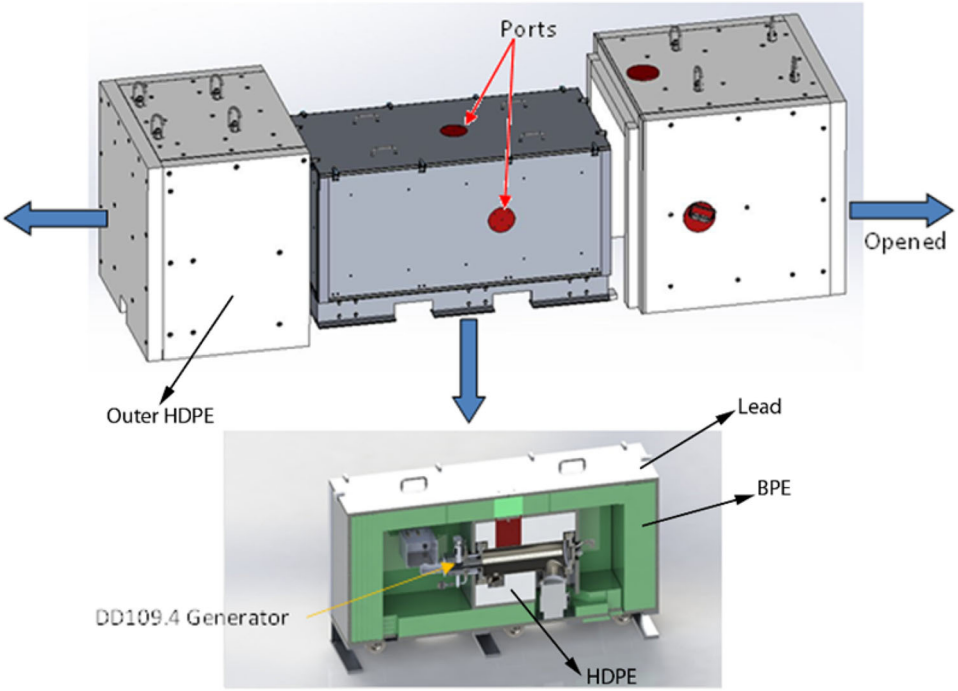

Fig. 1 (Color online) Shielding and moderator assembly of the DD109.4M neutron generator

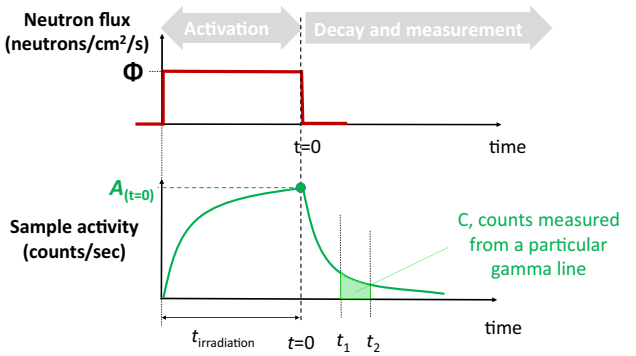

Fig. 2 (Color online) Activation of a sample, decay, and measurement durations

4 Experimental approach

In order to determine the moderator thickness needed to maximize the thermal neutron flux, aluminum samples were activated in the top port at different polyethylene thicknesses. Equation 5 shows the radioisotopes produced as a result of the neutron absorption by aluminum.

$$\begin{aligned} &^{27}\mathrm{Al} + \mathrm{n} \rightarrow {}^{28}\mathrm{Al} \rightarrow {}^{28*}\mathrm{Si} + \beta^{-}, \\ &^{28*}\mathrm{Si} \rightarrow {}^{28}\mathrm{Si} + \gamma, \\ &E_{\gamma} = 1.7789 \text{ MeV } (100\%). \end{aligned} \tag{5}$$

The method is based on the thermal neutron reaction ^{27}Al (n, γ)^{28}Al which has cross section of 0.231 barn. The radioisotope produced (^{28}Al) has a half-life of 2.245 min and decays by emitting a beta particle to the excited of ^{28}Si, which subsequently decays to the ground state by emitting a γ-ray of energy 1.7789 MeV.

Six aluminum samples (mass ranging from 2.6 to 3.23 g) were irradiated for 20 min in the upper port of the neutron generator. Each sample was irradiated with different thickness of moderator; ranging from 1.9 to 5.71 cm (0.75–2.25 inches). After the samples were irradiated, they were placed on the surface of the Canberra BEGe detector (FWHM 1.55 keV at E_{γ} = 1332.5 keV of ^{60}Co, with an efficiency of 19.2% and a peak-to-Compton ratio of 55.9:1). The detectors were surrounded by graded shielding mainly made of lead to minimize the background. A gamma ray spectrum from each sample was collected for 7 min using the Canberra ProSpect software. The waiting time between the end of the irradiation and the spectrum collection was 2 min.

After determining the optimum moderator thickness, gold foils were activated in the top port at this optimum thickness to determine the maximum thermal neutron flux. Equation 6 shows the radioisotopes produced as a result of the neutron absorption by gold.

$$\begin{aligned} &^{197}\mathrm{Au} + \mathrm{n} \rightarrow {}^{198g}\mathrm{Au} \rightarrow {}^{198*}\mathrm{Hg} + \beta^{-}, \\ &^{198*}\mathrm{Hg} \rightarrow {}^{198}\mathrm{Hg} + \gamma, \\ &E_{\gamma} = 411.8 \text{ KeV } (95.6\%). \end{aligned} \tag{6}$$

The method is based upon the thermal neutron reaction ^{197}Au (n, γ)^{198g}Au which has a cross section of 98.65 barn. The radioisotope produced (^{198g}Au) has a half-life of 2.6947 days and decays by emitting a beta particle to the

excited status of ^{198}Hg, which subsequently decays to the ground state by emitting a γ-ray of energy 0.4118 MeV. Four gold samples (mass ranging from 0.08 to 0.09 g) were irradiated for 30 min.

For the fast neutron flux measurements, indium samples were irradiated in the port without the polyethylene moderator. The ^{115}In (n, n′)^{115m}In reaction was utilized in these measurements. The radioisotope produced (^{115m}In) has a half-life of 4.486 h and decays to the ground state of indium by emitting a 336.22 keV gamma ray. Two indium samples of mass 0.490 ± 0.001 and 0.430 ± 0.001 g were irradiated for 20 min.

The irradiation times used were more than the minimum time required for constant sample activity during the activation process. The measurement times were the minimum to collect at least 10 thousand net counts in the peak of interest.

The efficiency curve of the BEGe detector was determined using gamma rays of known energy from a set of point radioisotope sources. The efficiency at the 411.8 and 336.22 keV energies was determined using a natural logarithmic interpolation in the efficiency curve (Fig. 5).

5 Results and discussion

A typical experimental spectrum obtained from one of the aluminum samples is shown in Fig. 3. The accumulated spectra were corrected for the background, dead time, and sample mass. No correction was made for the sample decay because the irradiation, waiting, and measuring times were the same for all samples analyzed.

Figure 4 shows a plot of the net counts per gram under the 1.7789 MeV peak as a function of moderator thickness. The counts are mainly proportional to the thermal neutron flux incident on the sample. Figure 4 shows that the maximum counts are obtained at a moderator thickness of approximately 3.8 cm. Also shown in Fig. 4 are the uncertainties in the counts. It is noted that only a discrete set of moderator thicknesses, as shown in Fig. 4, were available for testing. Some uncertainties were not accounted for in these results such as those resulting from the sample dimensions, moderator thickness, and sample positioning with respect to the neutron beam. Given the uncertainties in this experiment, the maximum counts are better defined by a range of moderator thickness from 3.5 to 4 cm, which aligns well with the previously obtained MCNP results [8] which show that an optimum moderator thickness can be found in this range.

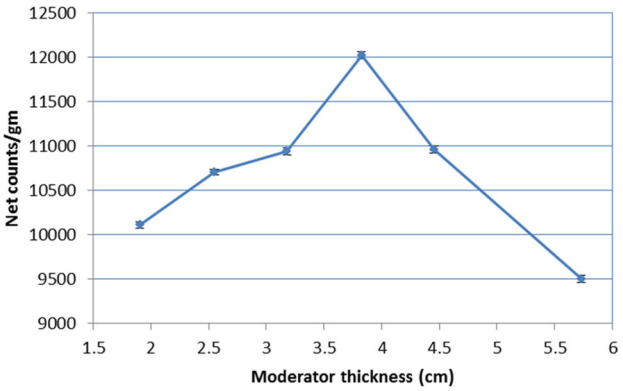

Fig. 4 Total counts under the 1.7789 MeV peak at different moderator thicknesses

The resulting efficiency curve for the BEGe detector used in these experiments is shown in Fig. 5. Also shown in Fig. 5 are the calculated efficiency values using the Canberra LabSOCS software. A good agreement is shown between the measured and calculated efficiencies.

Table 1 summarizes the results obtained from the irradiation of the four gold foils. The net counts were calculated using the baseline stripping method, and the flux was calculated using Eq. 4

The variation in the measured thermal neutron flux can be attributed to their sample position relative to the neutron source [8]. While the results reported in Table 1 are listed

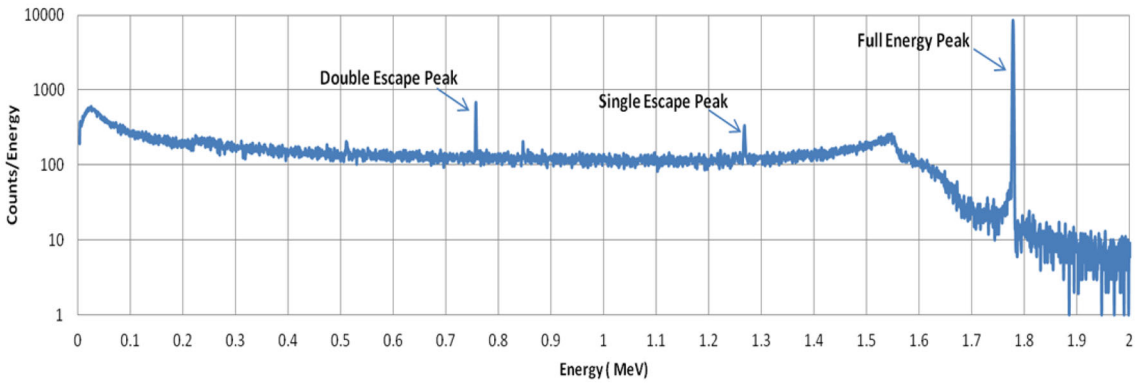

Fig. 3 Typical experimental spectrum from Al sample irradiation

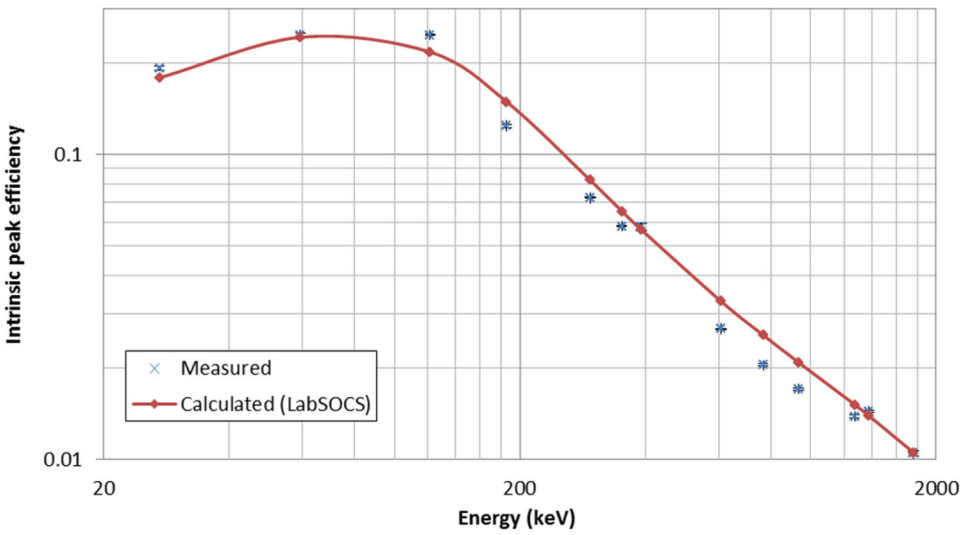

Fig. 5 (Color online) Gamma ray efficiency as a function of energy for the BEGe detector

Table 1 Results of thermal neutron flux measurements

Sample	Weight (g)	Net counts under the 0.4118 MeV peak (counts)	Activity (Bq)	Flux (n/cm^2 s) $\times$ 10^6	Average flux (n/cm^2 s)
Au#1	0.083 $\pm$ 0.001	36,140	385	2.875	2.960 $\times$ 10^6 $\pm$ 2.7%
Au#2	0.078 $\pm$ 0.001	36207	385	3.064	
Au#3	0.072 $\pm$ 0.001	31,701	340	2.927	
Au#4	0.080 $\pm$ 0.001	35,835	384	2.975	

as those for thermal neutrons, they are in fact corresponding to both thermal and epithermal neutrons. At the optimum moderator thickness, the epithermal neutron flux is four times less than the thermal neutron flux [8]. Taking into account the lower neutron cross section, and based on our previous MCNP calculations, the epithermal neutron contribution to the flux calculations in Table 1 does not exceed 2%.

A similar approach was followed for the indium sample irradiation. The calculated fast neutron flux of 6.186 $\times$ 10^7 $\pm$ 6.1% n/cm^2 s.

6 Conclusion

The neutron activation technique was used to experimentally measure the fast neutron flux, the optimum moderator thickness required to maximize the thermal neutron flux, and the maximum thermal neutron flux from the DD neutron generator at the University of Sharjah. The optimum moderator thickness was found to be between 3.5 and 4 cm. The thermal and fast neutron fluxes were found to be 2.960 $\times$ 10^6 and 6.186 $\times$ 10^7 n/cm^2 s, respectively. The thermal neutron flux measurements revealed some non-uniformity in the flux values. More sample irradiations will need to be performed in the future to generate a detailed neutron flux map and investigate the neutron flux profile.

References

1. M.K. Sharma, A.B. Alajo, X. Liu, MCNP modeling of a neutron generator and its shielding at Missouri University of Science and Technology. Nucl. Instrum. Methods A **767**, 126–134 (2014). https://doi.org/10.1016/j.nima.2014.08.011
2. U. Pujala, L. Thilagam, T.S. Selvakumaran et al., Analysis of neutron streaming through the trenches at linac based neutron generator facility, IGCAR. Radiat. Prot. Environ. **34**, 262–266 (2011). https://doi.org/10.4103/0972-0464.106194
3. J. Fuquan, G. Deshan, C. Daowen et al., An neutron generator-based NIPGA system for on-site analysis. Nucl. Sci. Technol. **21**, 1 (2010). https://doi.org/10.13538/j.1001-8042/nst.21.63-64
4. IAEA, Neutron Generators for Analytical Purposes, IAEA Radiation Technology Reports. Series No. 1, Vienna (2012)
5. V. Zaichick, S. Zaichick, Measurement of trace element content in bone samples using long-lived neutron activation products and high-resolution gamma-ray spectrometry. Nucl. Sci. Technol. **27**, 13 (2016). https://doi.org/10.1007/s41365-016-0002-0
6. H.H. Vainionpaa, C.K. Gary, J.L. Harris et al., Technology and applications of neutron generators developed by adelphi technology. Phys. Procedia **60**, 203–211 (2014). https://doi.org/10.1016/j.phpro.2014.11.029

7. X-5 Monte Carlo Team, MCNP—A General Monte Carlo N-Particle Transport Code. Version 5, University of California, Los Alamos National Laboratory (2003)
8. W.A. Metwally, M. Ballaith, A. Chen, Thermal Flux Maximization from a DD Neutron Generator. Trans. Am. Nucl. Soc. **114**, 338–340 (2016)
9. S. Jakhar, C.V.S. Rao, A. Shyam et al., Measurement of 14 MeV neutron flux from D-T neutron generator using activation analysis. In *IEEE Nuclear Science Symposium Conference Record, Dresden, Germany* (2008)
10. T. Malkiewicz, G. Lhersonneau, G. Simpson et al., Measurement of fast and thermal neutron flux from the d + D reaction using the activation method. [Research Report] LPSC11300 (2011)
11. G.F. Knoll, *Radiation Detection and Measurement* (Wiley, Hoboken, 2010)
12. N. Tsoulfanidis, S. Landsberger, *Measurement and Detection of Radiation* (CRC Press, Boca Raton, 2015)