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Abstract

Neutron generators (NG) being devices where neutron outputs are accomplished electrically, suffer from fluctuations in their outputs. Of particular importance are the short term variations that may affect individual data acquisition runs. Thus when using NGs for quantitative neutron-induced gamma-ray spectroscopy, the neutron output must be continuously monitored in real time, and normalization procedures subsequently applied to properly evaluate the gamma-ray spectra. Using a plastic scintillator, we developed a scheme for detecting fast neutrons that relies firstly, on recording a spectrum and, secondly, on establishing a region-of-interest (ROI) that may effectively discriminate against gamma rays that are always present in a neutron field. We discuss the optimization of these procedures for a field system to measure carbon in soil.

Introduction

A 14 MeV neutron generator is used to measure C in soil (1) as part of the terrestrial C sequestration studies. Fast neutrons impinge on the soil matrix and undergo inelastic scattering reactions (INS) with C and other elements. In the process characteristic prompt gamma rays of 4.43 MeV are emitted by C. These gamma rays are detected and analysed for quantitative measurements of C.

NGs produce 14 MeV neutrons by using the deuterium-tritium fusion reaction. This is accomplished by ionizing deuterium and accelerating the deuterons under high voltages of 40-100 kV on to a tritium target. Since the operations are electrical in nature the neutron outputs suffer from fluctuations. Thus when using NGs for quantitative neutron-induced gamma-ray spectroscopy, the neutron output must be continuously monitored, and normalization procedures subsequently applied to properly evaluate the gamma-ray spectra. These procedures must consider systematic drops in neutron output due to continued usage, along with short-term variations that may affect individual runs. The first case is best addressed by using copper-foil activation. In this procedure, a copper disk is placed at a pre-determined position on the NG, exposed to neutrons for a fixed amount of time, and the induced radioactivity measured with a gamma detector. Any statistically meaningful decrease in the foil's activity over this period suggests the need to adjust the instrument's operating parameters to increase the neutron output. In the second case, the neutron output needs to be monitored in real time for each individual run. This can be best accomplished by using neutron detectors. Many organic crystals, liquids and plastics scintillate when bombarded with nuclear radiations (2). However, since neutron fields are always contaminated with neutron-induced gamma rays, this

dictates the choice of the detector. In organic materials, gamma- rays interact primarily with atomic electrons by the Compton effect whereas neutrons scatter elastically from the nuclei of the atoms in the scintillator. Organic scintillators are good neutron detectors since they contain light elements, particularly hydrogen. The scattering interaction transfers some portion of the kinetic energy of the neutron to the hydrogen, resulting in a recoil nucleus whose recoil can be detected easily by the scintillation it produces in the organic material. However, distinguishing the gamma-ray response of the scintillator from the composite response to gamma-rays and fast neutrons is critical for an accurate determination of the fast neutron output of the NG.

Using a plastic scintillator, we developed a scheme for detecting fast neutrons that relies, firstly, on recording a spectrum and, secondly, on establishing a region of interest (ROI) in the neutron spectrum that may effectively discriminate against gamma-ray contamination. We discuss the optimization of these procedures for a field system to measure carbon in soil.

Methods

A plastic scintillator (0.75 in diameter x 0.5 in thickness) has been used for monitoring the neutron output of the NG. The pulse output from the PMT was input directly to a digital multi-channel analyzer. For all neutron spectra, the NG was operated at a HV= 46.9 kV and beam current, I_b= 41.1 µA. The pulsing frequency was 10 kHz with duty cycle as 25%.

Fast neutron detection

(a) Gamma-ray response and determination of neutron region-of-interest (nROI).

Standard gamma -ray calibration sources such as ^{137}Cs, ^{60}Co and [207]Bi were taped to the front face of the plastic scintillator and the gamma spectra were recorded separately. For the response to high energy gamma-rays the natural cosmic ray background was acquired over a 14h counting period. Fig.1 shows the scintillator response to pure gamma rays together with a neutron spectrum recorded by placing the detector over the NG. It can be seen that the response to gamma rays ends at channel number 380 whereas the 14 MeV neutron spectrum extends up to channel 500. Accordingly, the region between channel numbers 380-500 has been assigned to the fast neutron response of the detector that is free from the neutron induced gamma ray contamination and is denoted as nROI.
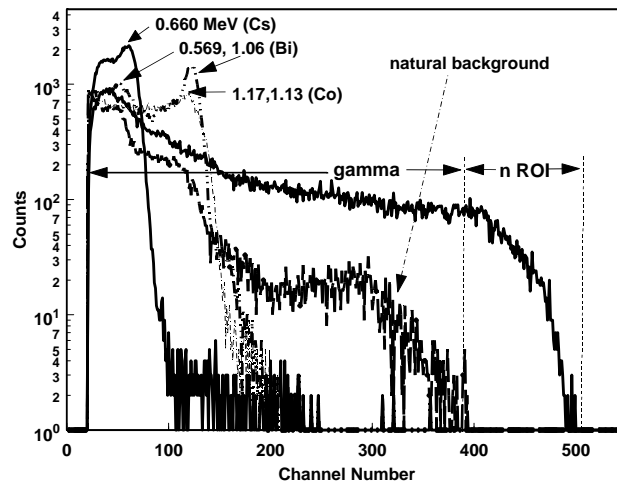
Fig.1 Plastic scintillator response to pure gamma-rays and fast neutrons.

(b) Stability of the system for different incoming count-rates and pile-up studies.

Having established the nROI it was necessary to determine if the gamma-ray pile-up at different incoming count rates (ICR) underwent random summing to contaminate the nROI. For this purpose, the plastic scintillator was positioned inside a lead housing above a 10.3 mCi ^{137}Cs source and the incoming count rate (ICR) to the detector was varied by changing the height of the scintillator above the source. For the lowest ICR of 0.4 kcps, the scintillator was removed from the lead housing and the gamma spectrum was recorded with a ^{137}Cs calibration gamma source taped to the front face of the scintillator. For each ICR, the spectrum was recorded for a live time (LT) of 0.5 h over a period of 8h.

(c) Influence of variation in the neutron induced background on the neutron spectrum

The neutron induced gamma-ray component that can be varied to study the effects on the composite neutron-gamma spectrum and the nROI are the neutron induced gamma rays that are emitted from the soil. This was accomplished by mounting the plastic scintillator in the holder on the NG and progressively raising the NG to different heights from the soil surface. At each height the neutron spectrum was recorded for a LT of 300s.

Validation of nROI

To validate the response of the nROI to changes in the fast neutron flux, the neutron fluxes were measured independently by Cu foil activation and by the plastic scintillator at two locations each on the NG, (1) over the target line and (2) at a distance 10" from the target line.

The reaction used for Cu foil activation is ^{63}Cu (n, 2n) ^{62}Cu, which has a threshold energy of 11.5 MeV and is counted for 0.51 MeV gamma-rays from positron

annihilation. The foils were irradiated for 5 min, followed by a transfer time of 0.5 min, and then counted for 5 min. Neutron spectra were recorded for a LT of 5 min.

Results and Discussion

Pile-up effects and stability of detection system

Fig.2 shows the response of the plastic scintillator to different ICRs of 0.660 keV gamma-rays from ^{137}Cs together with a 14 MeV neutron spectrum. It can be seen that pile-up of gamma rays begin from the lowest ICR of 0.4 kcps and is significant for the highest ICR of 46.0 kcps employed in the present study. The random summing of gamma rays spread over 100 channels beyond the Compton edge which is at channel number 71 determined as half height point of edge maximum. However there is no contribution of pile-up effects to the nROI. At the highest ICR of 46.0 kcps, the total counts in the nROI was 6 counts.

The detection system was found to be stable over the 7h that it was monitored at ICRs of 1.8 and 3.8 kcps. The mean total counts in the spectrum for 0.5h counting periods were 2707841 ± 1758 and 6817896 ± 1581 for ICRs of 1.8 and 3.8 kcps respectively. The errors are in accordance with statistical fluctuations. It must be mentioned here that under normal operating conditions of the NG (HV, 46.9 kV and beam current, Ib, 41.1 μA), the ICR is ~ 1.4 kcps.
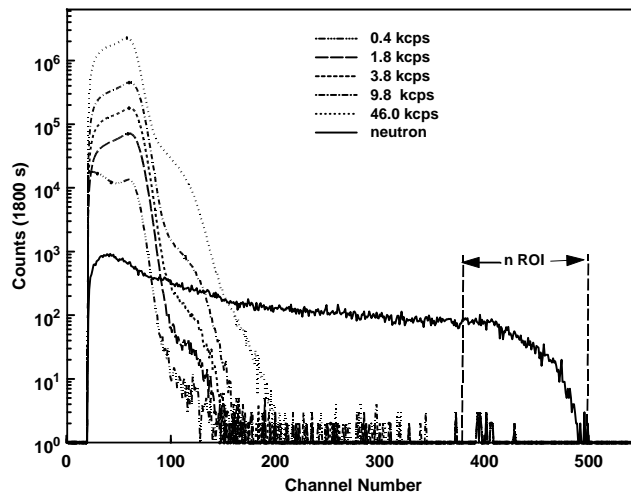
Fig.2 Pile-up of gamma rays at different ICRs.

Effects on the neutron spectrum for variable neutron induced gamma-rays.

Having earlier determined the nROI of a spectrum as the difference between a composite neutron-gamma spectrum and that of the response of the plastic scintillator to pure gamma rays it was essential to test the nROI under variable neutron induced gamma-ray conditions but keeping the operating conditions of the NG constant. Fig. 3 shows the effects on the total counts in the composite neutron-gamma spectra and also on the counts in the nROI as the NG was progressively raised from the soil surface. The nROI counts at each height was normalized to the total composite counts for the position when the NG was on the soil (lowest position). It can be seen that the composite counts decreased rapidly at first as the NG was moved away (with increasing height) from the neutron induced gamma-rays that are produced in the soil reaching a steady state at a height of about 15 inches.
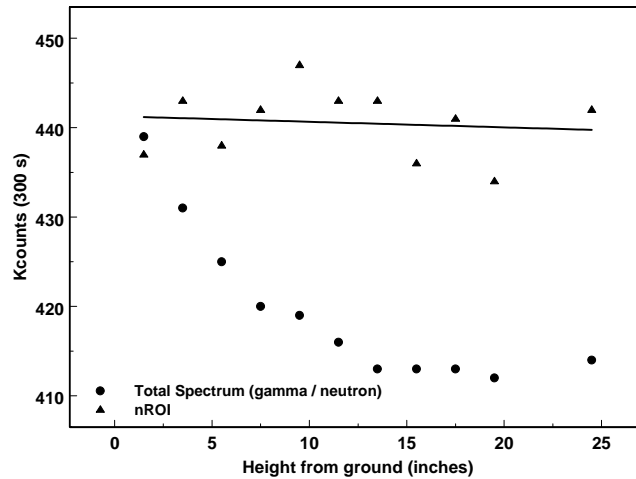
Fig.3 The total and nROI counts as a function of height above soil

At this and higher positions, it appears that the albeido effects from the soil have disappeared and the only neutron induced gamma-ray component would be due to the surrounding material(shielding and construction material) around the detector. On the other hand, it can be seen that the nROI did not suffer from the albeido effects indicating that the nROI selected is free from the gamma - ray contamination. The mean nROI fluctuation for the height range studied was 440684 ± 3778 (0.85%) counts and reflects the neutron output fluctuation over the 1h experimental run required for this part of the study.

Validation of nROI

The nROI response to changes in fast neutron flux using the plastic scintillator are compared with Cu foil activation responses to the same changes. Table 1 shows the nROI counts and count ratios of responses at two different spots on the NG along with the Cu

foil results obtained from the same spots. It can be seen that the ratios of counts of the two positions are similar for the two independent methods.

Table 1: Comparison of nROI of neutron spectrum and Cu foil activation counts ratios.

Position on NG	nROI counts (300s) (Plastic scintillator)	Net ^{62}Cu counts (Cu foil activation)
A (Target line)	31926	576
B (10" away from A)	7203	140
Counts at A/ Counts at B	4.4	4.1

Normalization of net peak areas of soil elements of INS gamma spectra.

INS spectra of sand were recorded over a sand pit on four different days through a month. The net peak area counts of the major soil elements are shown in Table 2 along with their counts normalized to the mean neutron output as obtained from the neutron detector nROI counts. The nROI counts were obtained in real time during each experimental run. It can be seen that the fluctuations of the elemental counts are in accordance with the NG output variation viz the mean elemental counts have ~ 10.3 % error which is close to the mean nROI fluctuation of ~ 11%. When normalized to the mean nROI, the fluctuations of the net peak area counts are close to statistical fluctuations. The neutron output fluctuations as determined by the mean nROI over a period of 1.5 years has been observed to be ~ 8 %. The n detection system stability has been monitored regularly over the same period by attaching a ^{207}Bi source to the plastic

scintillator front face and acquiring a gamma spectrum for a LT of 1000s. The mean counts in the spectrum has been found to be 90300 ± 643 (0.7%).

Table 2: Net peak area counts (LT, 1800s) of soil elements obtained from INS spectra.

Si (1.78 MeV)	C(4.43 MeV)	O (6.13 MeV)	H (2.22 MeV)	nROI counts
572402 ± 2627 (525583)	33627 ± 1745 (30876)	178820 ± 1847 (164193)	47689 ± 1194 (43788)	185884
575084 ± 2632 (525068)	32472 ± 1751 (29647)	182472 ± 1847 (166602)	48205 ± 1194 (44012)	186938
461064 ± 2719 (532575)	28248 ± 1803 (32629)	139592 ± 1914 (161242)	38944 ± 1237 (44984)	147762
493739 ± 2751 (519750)	31282 ± 1835 (32930)	161024 ± 1925 (169507)	39951 ± 1244 (42055)	162138
525572[*] ±57210,10.9%	31407[*] ±2314,7.4%	165477[*] ±19636,11.8%	43697[*] ±4928,11.2%	Mean (nROI) 170680 ±19094,11.2%
525744[**] ±5262,1.0%	31521[**] ±1543,4.9%	165386[**] ±3514,2.1%	43709[**] ±1219,2.8%	

Values in parentheses are counts normalized to the mean nROI.
*   Mean counts
** Mean counts (Normalized)
The values expressed as % are CVs.

Conclusion

A plastic scintillator has been used to monitor the neutron output of a pulsed NG in real time during acquisition of INS gamma-ray spectra. The nROI determined within a neutron spectrum that is recorded has been shown to be free of the neutron induced gamma-ray contamination and the counts therein reflects the changes in the neutron output of the NG as validated by independent Cu foil activation. When INS gamma-ray spectra are normalized to a mean neutron output, the errors due to the output fluctuations are eliminated. The neutron output fluctuations over a period of ~ 1.5 years has been found to be about 8 %.

References

(1) L. Wielopolski, I. Orion, G. Hendrey, H. Roger, "Soil carbon measurement using inelastic neutron scattering," IEEE Trans.Nucl.Sci., 47,914 (2000).

(2) G.F. Knoll, Radiation Detection and Measurement, 2nd ed. New York Wiley, (1979).