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Neutron interrogation of shielded/unshielded uranium by a 4 MeV linac

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ABSTRACT

A non-destructive active assay method was developed for revealing illicit trafficking of uranium. Photoneutrons produced in beryllium or heavy water by bremsstrahlung from a linac induced fission in the samples. Delayed fission neutrons were detected by a neutron collar built up of ^{3}He counters embedded in polyethylene moderator. High-enriched uranium samples shielded and unshielded by lead up to 14 mm thickness were detected, with a performance practically unaltered. 25 mg ^{235}U can be revealed in a 1 min interrogation time.

1. Introduction

As is well-known, active methods ("interrogation") are widely used for revealing illicit nuclear materials (NM) by irradiation and detection of the induced response. Neutrons can penetrate high-Z shielding material, induce fission in the NM, and fission neutrons can more effectively be detected than passive γ-rays. The active method does not suffer from difficulties due to shielding encountered in passive γ-spectroscopy. This involves an external source that irradiates the material whether or not it is shielded, and the subsequently emitted radiation is detected and used to characterize the material being present. In order to accomplish such an analysis, active radiation portal monitors are to be deployed at customs and border checkpoints. If the induced radiation comprises neutrons, they may penetrate the shielding as well. In such cases it may be advisable not to take the material out of its holder during the irradiation and counting. Fission neutrons can be distinguished from irradiating ones via utilizing the time correlation among the former (coincidence counting), or via separating delayed neutrons from the primary and prompt fission ones by their time sequence.

In the fields of safeguards and nuclear forensics, pulsed D-T neutron generators represent a sensitive and versatile variant of active interrogation systems, by counting delayed fission neutrons or gammas (see references in Lakosi et al., 2008; Lakosi and Nguyen, 2008). Larger systems designed for inspecting sea cargo relying on D-D (Hall et al., 2007; Slaughter et al., 2007; Church et al., 2007) or photoneutron interrogation by linacs are also common as pulsed neutron sources (references in Lakosi et al., 2008; Lakosi and Nguyen, 2008). Assay of hidden or shielded NM via delayed neutrons and gammas from photo- and neutron-induced fissions, by the use of pulsed 10–20 MeV linacs, has also been reported (Jones et al., 2005; Norman et al., 2005; Kinlaw and Hunt, 2006; Jones et al., 2006,2007; Norman et al., 2007; Sterbentz et al., 2007). Uranium mass of samples embedded in large concrete packages was assessed by photon interrogation and photofission, using 15–30 MeV linac (Gmar et al., 2005). Shielded HEU was interrogated with a 60 keV neutron beam produced by a 2 MeV proton linac (Kerr et al., 2007).

A photoneutron interrogation project has been carried out by applying 4 MeV electron linac of the Institute of Isotopes as a neutron source, to induce fission in low- (LEU) (Lakosi et al., 2008) and high-enriched uranium (HEU) samples (Lakosi and Nguyen, 2008). The electron energy has been converted into bremsstrahlung by a platinum foil, whereas neutrons for interrogation have been produced either in heavy water or beryllium. Delayed neutrons produced in the fissile material have been detected, distinguished from interrogating neutrons by using time discrimination. Having performed the assay of bare uranium previously, results of the interrogation of HEU samples behind lead shielding are presently reported.

Typically about 1% of fission neutrons only are delayed, emitted by fission products. It is necessary therefore to count delayed neutrons for long enough time, in order to achieve good statistics. It means that pulsing neutron sources are necessary, and delayed neutrons can be counted in the interval between pulses. As a result of the continuously pulsing irradiation, the intensity of delayed neutrons goes into saturation with an amplitude depending on the pulse repetition rate. By halving the rate, saturation intensity halves as well, in parallel to the mean intensity of the electron current. It has been shown (Lakosi and Nguyen, 2008) that 20 s irradiation allows a saturation degree

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of about 80%, therefore initial irradiations of 20 s at least were applied before starting irradiation-measurement cycles.

2. Experimental

Electron pulses, duration of 2.6 μs, have been fired with a repetition rate of 25 Hz. Selected from 50, 25, 12.5, or 6.25 Hz, this was established to be the optimum setting in previous experiments (Lakosi et al., 2008; Lakosi and Nguyen, 2008).

Bremsstrahlung was generated on a 20mm × 30 mm size, 0.9 mm thick platinum converter positioned at 3 cm distance from the exit window of the linac. The diameter of the electron beam was about 2 cm at converter distance.

Neutron production was due to (e,gamma) and (gamma,n) double conversion. Beryllium and heavy water was applied alternatively as photoneutron converter. Their (gamma,n) reaction thresholds are 1.67 and 2.23 MeV, respectively. The neutron energy available from the ^{9}Be(gamma,n)^{8}Be and D(gamma,n)H reaction is up to 2.33 and 1.77 MeV, respectively, at 4 MeV endpoint energy bremsstrahlung. However, the yields abruptly vanish above around 0.9 MeV neutron energy, whereas maximum intensity of the spectrum of evaporated neutrons is at about 0.5 MeV, even at much higher (15–50 MeV) linac energies (Sáfár and Lakosi, 1994, Facure et al., 2005).

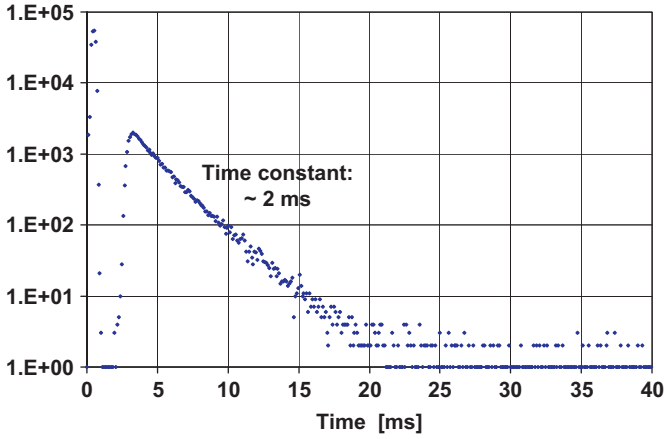

Fig. 1. Time spectrum of the D_2O converter alone, without U sample. 25 Hz, 1.9 μA, 1000 cycles.

A home-made neutron collar, as reported earlier (Lakosi et al., 2006; Lakosi and Nguyen, 2008), consisted of concentric polyethylene cylinders of an outer size of 300 mm diameter and 470 mm total length. The innermost ring (of 20 mm wall thickness) forms a measurement cavity for the material to be assayed. In between the outermost and inner cylinders, 12 proportional counters (type SNM-28) of diameter 32mm × 308 mm length each, filled with ^{3}He gas to a pressure of 4.0E5 Pa (4 atm) served as neutron detectors. Based on previous experiences, Cd foil was not used. Heavy water or beryllium photoneutron converters of a mass of 105 and 170 g, respectively, were applied at the top of the collar.

The signal processing electronics, consisting of a 400 channel analyzer in multiscaler mode of operation as a time analyzer, was almost the same as reported previously (Lakosi and Nguyen, 2008). The multiscaler receives commands from a PC through a micro-controller. Triggering the analyzer was synchronized with the linac control command pulse. A channel width (dwell time) of 100 μs was selected for the present measurements.

Irradiation-measurement cycles of up to 1000 were carried out at 25 Hz. As experienced previously (Lakosi et al., 2008; Lakosi and Nguyen, 2008), time spectra acquired at 25 Hz show that the pulse length of prompt (interrogating and fission) neutrons cover a half of the 40 ms interval between two pulses. Exponential decay with a time constant of about 2 ms was observed, with a total pulse length of 20 ms. Thus, the effective time remaining for delayed neutron measurement is about 20 ms, no matter if using heavy water or beryllium converter, as observed previously.

An irradiation-measurement cycle lasts for 40 ms at 25 Hz. Thus, 1000 cycles last for 40 s. In Fig. 1 a time spectrum of the D_2O converter alone is displayed, taken during 1000 cycles at a mean electron current of 1.9 μA, while no U sample was in the measurement cavity. It was practically the same using the beryllium converter as well.

Time spectra of HEU oxide powder samples of 10.5, 5.5, and 2.3 g mass of 36% enrichment, as well as of a 0.53 g sample of 90% enrichment are seen in Fig. 2. All the spectra were taken by the D_2O converter, during 1000 irradiation-measurement cycles. The mean electron current was uniformly 1.9 μA at 25 Hz. In order to reach a sufficient degree of saturation, 20 s irradiations were

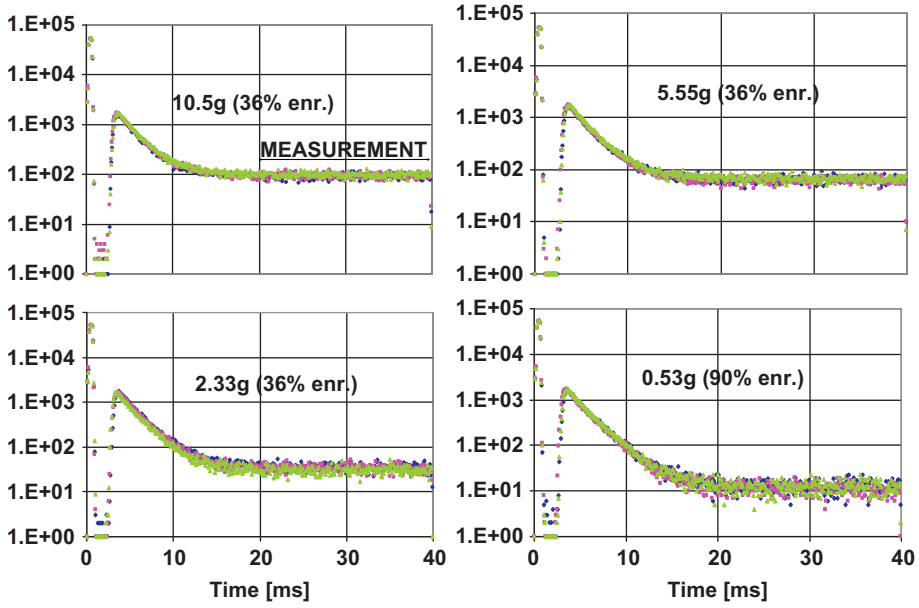

Fig. 2. Time spectra of 10.5, 5.5, 2.3 g (36% enr.), and 0.53 g UO_2 (90% enr.) samples without and with Pb holders.