Development of neutron interrogation techniques for concealed substances detection in port containers

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This work is aimed at contributing to the effort of nations seeking to control international borders movement of dangerous chemical substances and nuclear material, in accordance with a multitude of agreements signed to that purpose [1].

At this stage, the signature of pure substances: nitrogen (N), uranium (U), chlorine (Cl), chromium (Cr), mercury (Hg), cadmium (Cd), arsenic (As) are trying to be identified and later, to detect their presence in simulated large cargo containers. The technique employed consists in the detection of prompt and early decay gammas induced by incident thermal, epithermal or fast neutrons in the cargo array. Uranium has also been detected through the counting of fast neutrons originated in induced fissions.

Introduction

The use of slow neutrons, whether in the incident beam or thermalized inside the merchandize, for investigating large cargo is being considered as complementary to the use of fast neutrons for this task [2]. Slow neutrons tend to spread in the sample with loss of time of flight (TOF) information but induce absorption reactions due to the high interaction cross sections.

To simulate a lower intensity neutron source, the experiment was placed 6–8 m from the target of the 25 MeV electron linear accelerator (LINAC). The flux on the sample was, approximately, 200 thermal n/cm^2 sec and 90 epithermal n/cm^2 sec (above the cadmium energy cut-off), provided by a 40 mm thickness polyethylene neutron moderator.

Detection from uranium induced fission neutrons was carried out through a 70 cm × 100 cm active area neutron detector incorporating 10 ^{3}He tubes (2.5 cm diameter and 55 cm active length, 4 atm gas pressure), to probe into realistic 2 m cargo arrays with 3 mm steel plates in the incoming and outgoing sides of the simulated container.

To detect gamma emissions, after initial crude experiments, a detection arrangement was set up consisting of four 2″ × 2″ NaI(Tl) detectors with their independent photomultipliers and voltage dividers, coupled to one sole preamplifier designed to reduce the initial dead time after each accelerator burst. Under appropriate shielding, it was possible to sort-out the gamma response to neutron interactions in the presence of a combined intense pulsed high energy X-ray and neutron field.

Apart from the LINAC based neutron pulsed source with its detector arrangement that could be expanded to industrial scale, the use of isotopic neutron sources (^{252}Cf and Am-Be) was also applied in order to visualize the gamma response from substances of interest, experimentally sorting out thermal, epithermal and fast neutron reactions. Detection of outgoing gammas was carried out by a Ge(Li) semiconductor detector and a NaI(Tl) scintillator to ease identification of the signal of interest in the latter. Signal to noise ratio was optimized through a dedicated gamma shielding and different methods were applied to analyze the gamma regions of interest that denote the presence of each tested substance.

For the same measurement, scintillator and semiconductor detectors have the expected important difference in gamma spectra resolution (Figure 1). But at industrial scale level, detectors must be used in great number, which makes scintillators the choice due to their costs and practicality of operation.

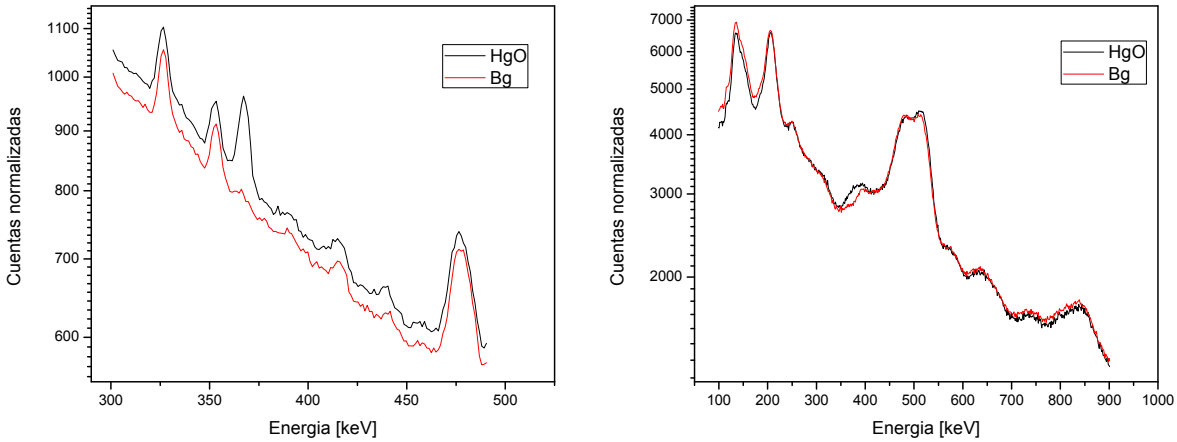

Figure 1. HgO sample compared with background. Left side: Ge(Li) detector, Hg characteristic peak ~360 KeV. Right side: NaI(Tl) detector, Hg characteristic signal.

Results

Uranium detection

The screening against special nuclear material (SNM) is a whole chapter in itself. The reviewed literature includes irradiation of a cargo container with high-energy gamma rays to induce photo-fissions (and neutron-induced fission as well) in shielded nuclear material [3,4]. Other papers propose to irradiate the container with intermediate [5] and high-energy pulsed neutrons [6,7] and exploit neutron moderation caused by the cargo container contents.

In our experiments [8,9], the uranium test samples were two, highly enriched uranium (HEU) diluted in aluminium matrix in the form of flat rolled sheets (^{235}U mass = 27.5g, max. U-235 irradiated by direct beam = 10.7 g) and a partial reactor natural UO_2 fuel bundle (FB) 50 cm long (^{235}U mass = 38.38 g, probable irradiated fraction = ~1/10).

Fast fission neutrons emitted from the samples, irradiated by the low intensity 50 mm neutron beam described in the Introduction, were received by a detector array placed in the "backscattering" position as a one-sided set-up to inspect the cargo mock-up. First experiments [9] employed five ^{3}He cadmium filtered moderated detectors, highly insensitive to thermalized neutrons due to the Cd wrapping and most adapted to the detection of fission neutrons, which slowed down in the moderator. Neutron signals were routed to the TOF encoder and recorded as 2000 channel spectra, 4 μs dwell-time each, during a wide time gate after the accelerator pulse. A

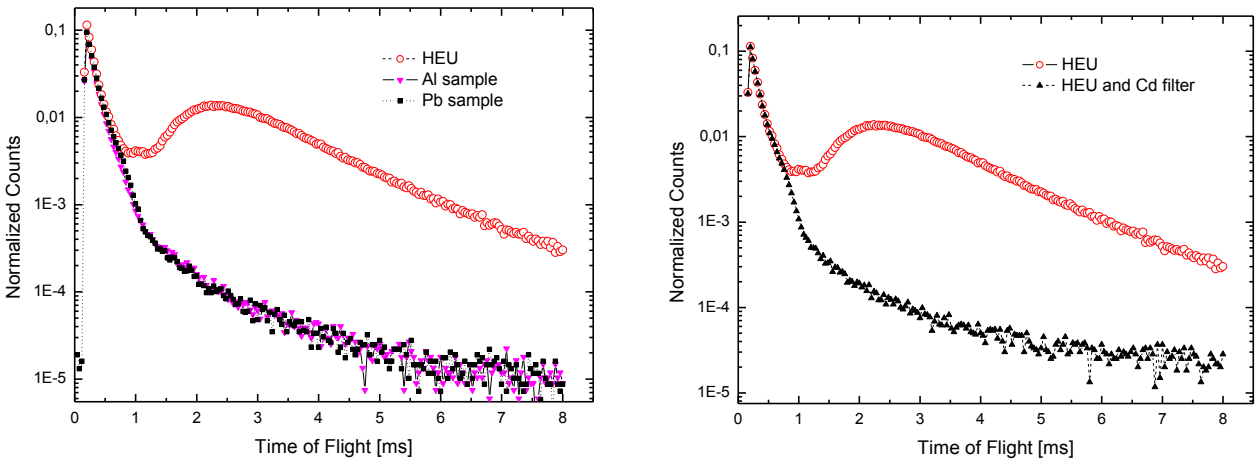

Figure 2. Fast neutron detection from HEU as function of neutron incident TOF [9,11]. Left side: signal for different samples; (upper) HEU, (lower) Al and Pb. Right side: (upper) under full incident spectrum and (lower) under epithermal (Cd filtered) irradiation.

Figure 3. LINAC experimental setup for fuel bundle (at left) inside simulated container. Neutrons impinge from the right. Large area neutron detectors are seen at both sides of image.

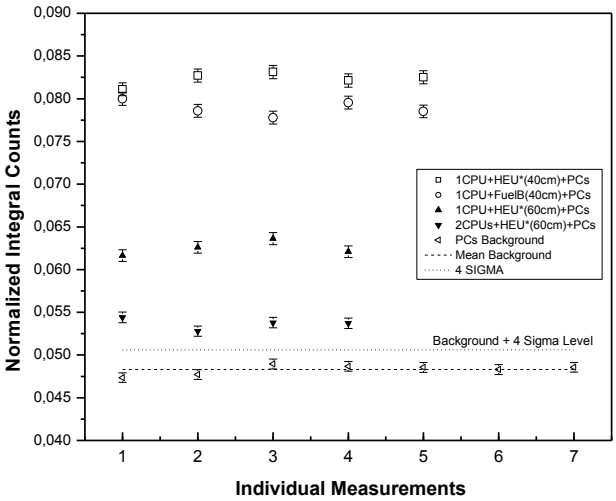

Figure 4. Integral counts for each 5 min recording time individual TOF spectra for HEU and FB hidden behind different number of PCs. The mean background 4-sigma level is indicated for comparison [9,10].

neutron detector housed in a different place away from the experiment, acted as accelerator neutron production monitor.

Thermal vs. epithermal interrogation through the Cd difference method allowed confirming that detected response, shown in Figure 2, was being induced by the thermal component in the incident neutron beam. Two other samples were irradiated, one of similar Al content and shape as the aluminium containing enriched uranium and the other made of lead. Results shown in Figure 2 leave no doubt as to the fission origin of observed fast neutrons. Later tests, carried out with the wider 70 cm × 100 cm active area neutron detector with 10 ^{3}He tubes described in the introduction, included the fission sample inside 2 m thick cargo mock-ups with 3 mm steel walls to mimic a real container (Figure 3).

A figure-of-merit approach [9,10] was employed to decide initial and final TOF spectra channels for the integration of fast neutron counts, maximizing contrast. Figure 4 depicts these results compared with mean background (container without uranium) plus its 4-sigma level proposed in Reference [11].

Nitrogen detection

To test the capacity to detect nitrogen which may signal the presence of explosives [1], experiments were devised to measure nitrogen proportion respect to hydrogen content and with respect to iron, through the N/H and N/Fe count ratios over selected regions (ROI) of the gamma spectra [2]. Through 5 minute recording times, the sensitivity to N_2 presence appeared clear, but extrapolation of these results to more than 2 m thick cargo arrays is far from straightforward, especially when many cargo components are nitrogen bearing. An example was a LN2 sample "hidden" among several 1 kg cooking flour bags [2]. The presence of the N_2 sample was not completely obscured by the surrounding flour, where even its own N was "seen" by the experiment when the LN2 was removed from the neutron beam. In these determinations the volume of LN2 irradiated by the direct beam (neutrons) was 125 cm^3.

Other chemical substances detection

The Cl detection with the accelerator was the only case of interest where the simulated container carried a significant amount of chlorinated contaminants (Figure 5). The contrast obtained made it clear that control of such non-declared chemical substances is highly feasible.

Other substances whose characteristic gamma emissions were tested, where Cr, Cd, As, liquid Hg, HgO (Figure 1). This characteristic Hg line evinced by the peak around 360 keV was seen both with the accelerator and with the Am-Be isotopic source. Arsenic instead has escaped detection so far.

Final comments

The above described neutron flux allowed the experiments on the samples to be performed during 5 minute counting times, at realistic distances, and with a modest gamma detector array and a wide area neutron detec-

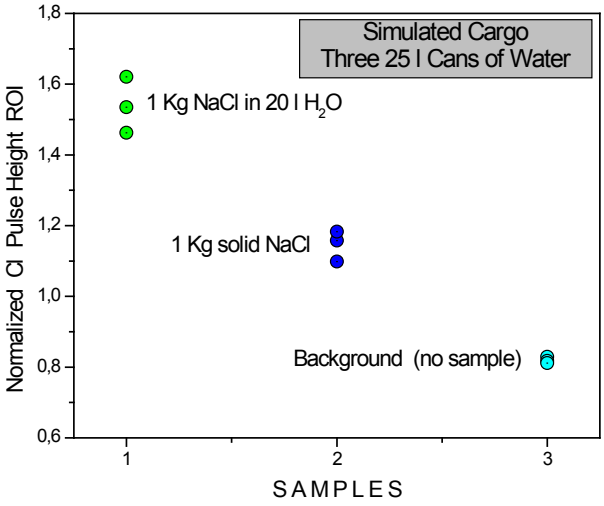

Figure 5. Simulated container full of 25-liter water cans. Later loaded with NaCl solution (1 kg in 20 l) and compared to a 1kg solid NaCl box [9,11].

tor. A neutron flux only five times greater (1000 thermal n/cm^2 sec) would produce the same results in one minute.

The use of fast, thermal and epithermal neutron beams for the identification of substances raises some expectation as to the possibility of a complementary tool to other current scanning methods. The identification of complex spectra from real merchandize arrangements requires the construction of a database of usual merchandise spectra to detect significant deviations in cargo composition.

For the particular case of the screening against illegal traffic of special nuclear material, the very small (27.5 g U-235) sample was marginally detected to a depth of 80 cm, with the reduced incident thermal flux and the wide area neutron detector almost in contact with the container wall, when the interfering slice of merchandise gave a transmission less than 0.01. It seems clear that scanning from both sides of the container will be a necessity. The addition of a fast neutron flux that undergoes moderation inside the cargo should shift this detection limit farther.

The analysis to identify mixed chemical substances "in the field" turns very complex in practice. The addition of a technique that employs neutrons of different energy ranges bears some increased promise and prompts us to continue developing this tool.

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