

Active Neutron Interrogation Approach to Detect Special Nuclear Material in Containers

R.E. Mayer, A. Tartaglione¹, F. Di Lorenzo¹, C. Sepulveda Soza², M. Schneebeil, P. D'Avanzo, L. Capararo

Neutron Physics Group and Instituto Balseiro, Comisión Nacional de Energía Atómica and Universidad Nacional de Cuyo, Argentina.

¹ also at Consejo Nacional de Investigaciones Científicas y Técnicas, Argentina.

² also at Comisión Chilena de Energía Nuclear, Chile.

Email contact of main author: mayer@cab.cnea.gov.ar

Abstract. Cargo interrogation in search for special nuclear materials like highly enriched uranium or ²³⁹Pu is a first priority issue of international borders security. In this work we present a pulsed thermal neutron based approach to a technique which combines the time-of-flight method and demonstrates a capability to detect small quantities of highly enriched uranium shielded with high or low Z materials providing, in addition, a manner to know the approximate position of the searched material. An experimental test in a 2m thick cargo array is reported.

1. Introduction

The first objective of this work initiative has been the discovery of illicit drugs, explosives and chemical weaponry. Many of the proposed methods reported in the reviewed literature are also neutron based active interrogation [1,2] due to the high penetrability of this type of radiation and its capacity to induce prompt gamma response.

The following step has been the detection of nuclear material for which samples were employed consisting of natural UO₂ in the form of a partial reactor fuel bundle and highly enriched uranium (HEU) in the form of 93% enrichment uranium in aluminium sheets.

Even mass number special nuclear materials (SNM) isotopes have higher spontaneous fission rates, but it is ²⁴⁰Pu the one which can certainly be detected through passive neutron counting in the lowest relevant quantities. Nevertheless, the significant isotopes from a security point of view are odd mass number, namely ²³⁵U and ²³⁹Pu and these show a very low spontaneous fission neutron emission, while at the same time their gamma emissions are easily shielded due to their fairly low energy. These conditions drive efforts to develop applicable active interrogation techniques which induce fission with the ensuing prompt and delayed radiations.

Once more, 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.

The experiments we herein describe are typical initial laboratory tests already reported in a previous paper [8] and new tests employing a 2 meters thick cargo array. As much effort is devoted elsewhere through the use of fast neutron induced reactions, we seek to explore a complementary approach by means of testing the applicability of moderated neutrons. These,

although not penetrating thick objects as a beam, they do diffuse into substances and cause the ensuing reactions.

2. Experimental Set-up

Time-of-flight (TOF) experiments were carried out at the Bariloche (Argentina) electron linear accelerator facility (LINAC). The accelerator operated at 100 Hz frequency, electron pulse width 1 μ s, 25 μ A mean electron current. The 25 MeV electron pulse impinges on a lead target and its bremsstrahlung photons produce fast neutrons by photonuclear effect. Neutrons are moderated in a 40 mm thick polyethylene slab from which a broader pulse of moderated neutrons emerges with an approximate exponential decay of several tens of microseconds time constant. At the end of the extraction tube, through thick concrete shielding with a borated hydrogenous added wall, a 5 cm diameter neutron beam is allowed to irradiate the experimental set-ups.

Fast neutrons emitted from HEU were received by a detector array placed in the “backscattering” position as a one-sided set-up to inspect the samples and the cargo mock-up. In the first experiments five ^3He detectors (10 atm gas filling, 2.5 cm diameter, 15 cm active length), embedded in thick curved paraffin moderator-reflector, completely wrapped in Cd sheet 0.8 mm thick. These detector tubes were inserted in holes drilled 3.8 cm into the 12 cm paraffin block, leaving a minimum of 2.5 cm moderator in front of them and no less than 6 cm behind to reflect neutrons. Such an array is insensitive to thermalized neutrons due to the Cd wrapping and is most adapted to the detection of fission neutrons which slow down in the moderator. Neutron pulses appropriately processed were routed to the TOF encoder and recorded as spectra 2000 channel length and 4 μ s dwell-time each, during a wide time gate after the accelerator pulse. A neutron detector housed in a different

place away from the experiment, acted as accelerator neutron production monitor.

The sample distance to the target-moderator ranged from 6 to 8 m away, outside the accelerator bunker. The low intensity falling on the sample was ~ 200 thermal n/cm²/sec and ~ 90 near epithermal n/cm²/sec (above cadmium cut-off energy). This situation mimics a smaller neutron source than the one actually used.

The uranium test samples were two, HEU diluted in aluminium matrix in the form of flat rolled sheets and a partial reactor natural UO₂ fuel bundle (FB) 50 cm long. Their characteristics were as follows :

HEU - 235U mass= 27.5 g
Total sample mass=152.8g
18% U-235 in Al
Max.U-235 irradiated by direct beam=
10.7g

FB - 235U mass= 38.38 g
Number of fuel pins= 13
Total UO₂ mass= 6.05 kg
Probable irradiated fraction= $\sim 1/10$

3. The experiments

The first test has been to check if sample response is due to interaction with thermal neutrons. Fig.1 is the result of irradiating HEU with the whole incident spectrum and through a Cd filter which possesses very high capture cross section for slow neutrons.

This thermal vs. epithermal interrogation through the Cd difference method allowed to confirm that detected response was being produced by the thermal component in the incident neutron beam.

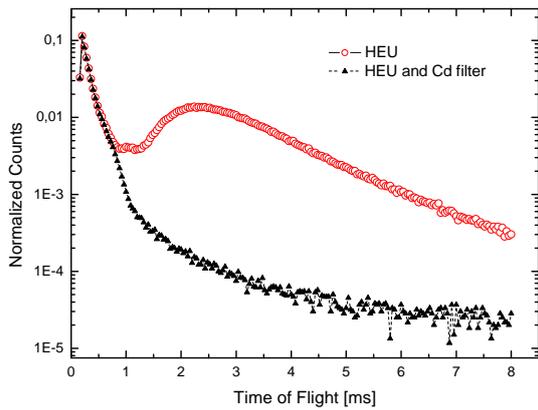


Fig.1. Fast neutron detection from HEU as function of slow neutron incident TOF, under full spectrum and under epithermal (Cd filtered) irradiation.

The next step has been to test if the observed fast neutron response is attributable to HEU. For that purpose, two other samples were irradiated, one of similar Al content and shape as the aluminium containing the enriched uranium and the other made of lead. Results are shown in Fig.2 which leaves no doubt as to the fission origin of observed fast neutrons.

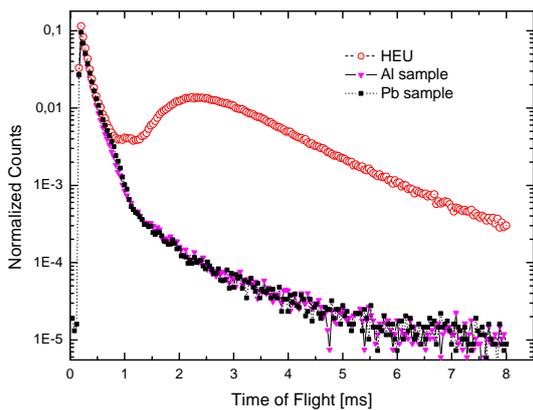


Fig.2. TOF fast neutron signal for different samples: HEU, Al and Pb.

Position sensitivity was tested moving the sample away from the detector. Two of these positions, 90 cm apart from each other, produce the graphs shown in Fig.3.

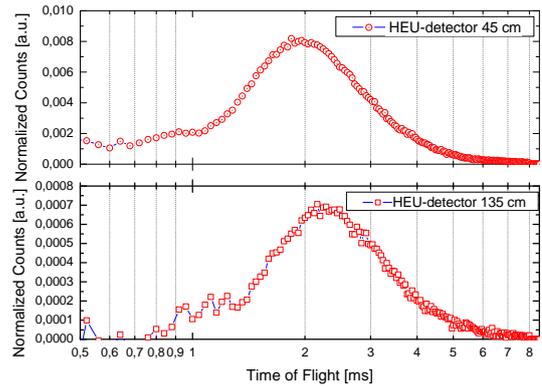


Fig.3. TOF sensitivity to HEU position with respect to fast neutron detector, for two distances differing 90cm. In both cases, the Al sample background spectrum subtracted.

The time difference is 300 μ s. It is appreciated even with such a broad distribution due to the times of arrival at the HEU sample of the incident thermal neutrons. With an extended sample this information would not be useful, but if SNM is hidden as a small object, this sensitivity allows to identify its position as central or lateral inside the container.

First tests of concealing shielding were performed using lead, polyethylene and common cooking flour. Those results appear in Figs.4 and 5 for five minute irradiations.

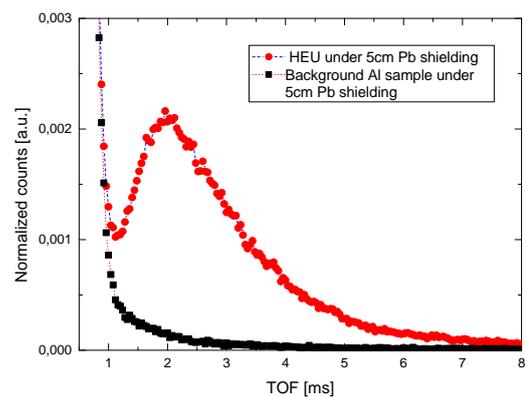


Fig.4. 5 minute TOF neutron spectra from HEU and from Al sheet (background) hidden under 5 cm Pb in both cases.

As expected, lead is no important neutron stopper, but it is possible to appear in such

thicknesses as a means to conceal the SNM gamma emission. The other substances with greater scattering and absorbing power are more effective in perturbing the neutron response. The polyethylene shielded the HEU towards the detector with no neutron reflection from behind thus constituting the most unfavourable condition.

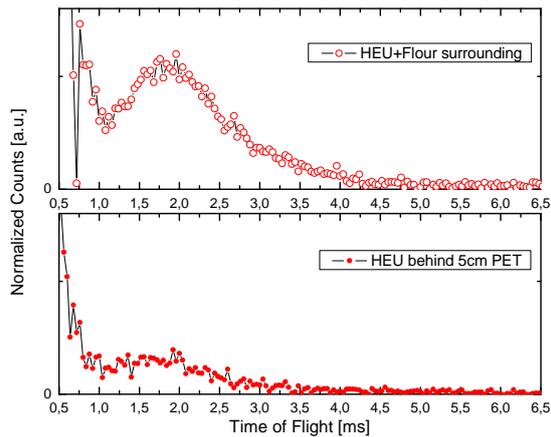


Fig.5. TOF neutron spectra from HEU surrounded by 9 cm wheat flour and hidden behind 5 cm polyethylene (PET) slab with no neutron reflector behind.

After these promising initial experiments, a more realistic set-up was tested. A 2 m thick cargo mock-up was prepared piling up computer monitor CRT screens and complete PCs in the most dense arrangement possible, behind 3 mm thick steel wall. Also, a new smaller detector was constructed incorporating six ^3He tubes with moderator and reflector (polyethylene) and complete Cd wrapping to shield away slow neutrons. Once more, the detector placed in front of the mock-up and at one side of the incoming moderated neutron beam, represented the laboratory approximation to a minimal real scanner.

Five minute TOF spectra recordings were repeatedly done for the two active samples described (HEU and natural UO_2 FB). Several irradiations with no active sample were also recorded at different times to assess background counting in the

presence of such cargo mock-up. These are displayed in Fig.6 as the summation of individual 5 min spectra corresponding to similar cases.

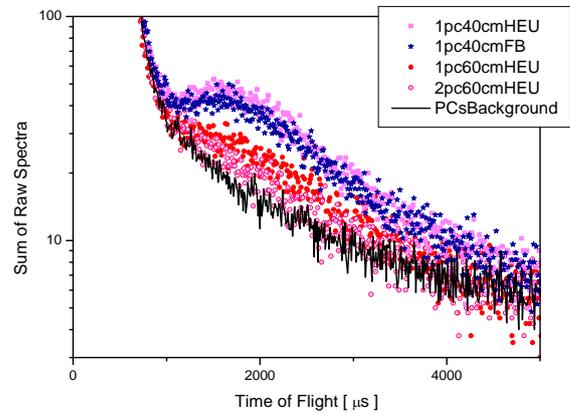


Fig.6. Fast neutron detection as function of incident slow neutron TOF, for the two types of U sample placed at different depths within the 2 m thick cargo array of computers and CRT screens.

In order to integrate neutron counts for each individual spectrum seeking to maximize contrast between different cases, it is necessary to decide the initial and final time channels which maximize resolution.

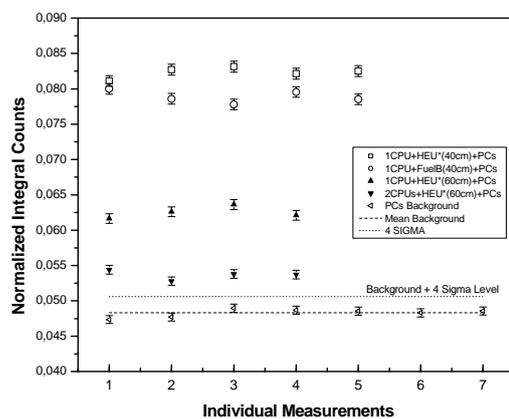


Fig.7. Integral counts for each of the 5 min recording time individual spectra described in Fig.6. The mean background 4 sigma level is indicated for comparison.

A figure of merit approach was used to decide upon this maximal condition. The result is displayed in Fig.7 as TOF

spectrum integrals of individual 5 min runs.

The high correlation among different runs of the same condition as well as the resolution between cases is clear. Fig.7 also conveys an idea of the sensitivity of the method even under such modest conditions of incident thermal neutron flux and very limited detector active area. The contrast with background counting is made clear through comparison with the 4 background standard deviation level, which was calculated propagating each individual Poisson uncertainty into the mean squared dispersion of the measured background points.

It must be added that delayed neutrons were also counted successfully after irradiations, thus providing a useful confirmation tool at no extra cost. For that purpose, a combination of detector systems was tested for the sake of maximizing counts when neutrons from the sample are moderated by hydrogenous goods in the cargo.

Overcoming reduced neutron penetration due to a surrounding thick hydrogenated matrix, will be explored through alternative irradiation with fast neutrons and the ensuing moderation inside the matrix, although with loss of TOF information

4. Conclusions

Having placed the sample 6 to 8 m away from the target mimics a smaller neutron source; this is done to test the detection capability that a small pulsed neutron source would achieve. The low intensity falling on the sample has been in the order of 200 thermal n/cm²/sec and 90 near epithermal n/cm²/sec (above cadmium cut-off energy). This reduced flux and the limited amount of sample irradiated by the 50mm neutron beam, allowed most

experiments to be carried out during 5 min counting times.

High slow neutron fission cross section allows the 4 σ level suggested by IAEA-TECDOC-1312 (2002) to be reached even for samples in the tens of grams range. Delayed neutron emission can be added as a confirmation tool at no extra cost.

In weakly scattering arrays, TOF provides approximate information about SNM position.

Finally, the beam and the neutron detector should be extended to cover a much increased portion of viewed container slice, retaining the high “tens of grams” sensitivity achieved. Screening against SNM contraband through investigation of realistic volumes, like a container slice put in evidence by previous X-ray scanning, seems promising even with a small incident thermal/epithermal neutron flux.

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