Detection of Concealed Fissionable Material by Delayed Neutron Counting

W. Rosenstock¹, T. Köble¹, M. Risse¹ and W. Berky¹

¹ Fraunhofer-INT, Euskirchen, Germany

Email contact of main author: wolfgang.rosenstock@int.fraunhofer.de

Abstract. One of the greatest challenges in detecting concealed nuclear material by non-contact means is the measuring of uranium, especially highly enriched uranium, in particular if it is surrounded by additional shielding. Hence, gamma detection is not a promising option. However, neutrons provide a high probability to penetrate typical shielding materials well. Whereas plutonium emits enough neutrons from spontaneous fission to be measured by passive methods in most cases, highly enriched uranium, particularly in casings, does not emit sufficient neutrons to be detected passively. Therefore, we particularly examined the active interrogation with a neutron generator. A clear proof of the existence of special nuclear material is the emission of delayed neutrons after induced fission by neutrons.

We performed measurements with a small, light-weight neutron generator, which can be carried by one person. The time structure of delayed neutrons was measured in order to detect and identify hidden or shielded nuclear material in geometrical configurations whereof only the outer shape is known but little or no information on the inner structure is available. A block of depleted uranium was irradiated repeatedly by a sealed neutron tube from a 14 MeV neutron generator for different time intervals. The delayed neutrons were measured by a neutron "slab" counter consisting of 6 He-3 tubes moderated by high density polyethylene. After the end of each interrogating neutron pulse we analyzed the delayed neutrons in different time intervals, ranging from 3 s to 300 s and thus recorded the "decay curves" of the delayed neutrons. We optimized neutron irradiation and measuring time to get information on the existence of fissionable material in a short time.

These experiments show that fissionable material can be detected clearly and easily in a suspicious object without any information on the inner geometry and the surrounding moderating material within a very short time (several minutes).

1. Introduction

In the current worldwide security situation with many possible terroristic threats there is an increased interest in technical capabilities for the localization and verification of illicit nuclear material in-situ with non-contact methods. For example the US White House Homeland Security Council expressed their concerns about these nuclear and radiological threats. One of the most severe scenarios is a nuclear detonation of a 10 kiloton nuclear device [1]. In this scenario stolen highly enriched uranium is used by terrorists in an improvised nuclear device. The given scenario concerns the whole incident including the detonation. Obviously for such a kind of scenario it is absolutely essential to have technical means at disposal for finding and identifying the improvised nuclear device before the detonation occurs. The localization of such a nuclear device requires high sensitive mobile detection systems both for gamma and neutron measurements. If a suspicious object is found it is essential to analyze the object in order to determine whether nuclear material is involved or not. Here active interrogation with neutrons produced by a light-weight neutron generator may play an important role. A clear proof for the existence of special nuclear material is the detection of delayed neutrons which is possibly by means of such a neutron interrogation technique. In this paper we describe experiments using this technique and present options for optimizing it.

2. The origin of delayed neutrons

In some cases, passive detection methods are insufficient for the detection of special nuclear material and its discrimination from industrial radioactive material. Whereas Plutonium emits a significant amount of neutrons produced by spontaneous fission which can be detected in a direct way this is not the case for Uranium. As for Uranium, active neutron interrogation methods need to be performed. When irradiating Uranium with neutrons, fission processes take place, emitting prompt neutrons. The fission products (typically with atomic masses in the range between 95 and 135, e.g. Ba, Kr, Xe, Sr, among others) in turn create new nuclides by β decay. During the kinetics of these β decay reactions, nuclides with a considerable surplus of neutrons are produced which leads to alternate reactions to the β decay mechanism. These reactions take place when nuclides are created for which the emission of neutrons is energetically preferable to a further β decay. In the course of these alternate reactions, delayed neutrons are produced which then serve as a means of identification of Uranium. After the end of the irradiation, the number of these delayed neutrons decreases exponentially with time. Neutron irradiation of other heavy materials such as Pb does not lead to the emission of delayed neutrons. Hence a discrimination of special nuclear material is possible by means of active neutron interrogation methods.

From the measured "decay curve" of delayed neutrons one may derive a clue for the calculation of the nuclide vector of the special nuclear material in question.

A problem may occur when additional nuclear material is present, emitting neutrons by spontaneous fission or (α,n) reactions. The number of these neutrons remains constant with time, overlaying the decay curve of delayed neutrons. The stronger the source of spontaneous fission neutrons, the more complicated the evaluation of the decay curve. In some of those cases coincidence measurements may help to obtain reasonable results.

3. Neutron generator and pulsing options

The neutron generator Genie 16c (manufactured by SODERN) consists of a neutron emission module (neutron tube), a very-high-voltage (VHV) power supply unit, and a PC unit for running the generator's control software [2]. These components are shown in figure 1.

The tube is vacuum-sealed with a pressure in the range from 10^{-5} to 10^{-3} mbar when in operation. It contains a Penning type ion source, a VHV acceleration capacity, a target, and a gas reservoir. The system features two tubes. One whose target consists of a film of Deuterium (D) and Tritium (T) hydride and whose reservoir of a mixture of D and T produces (primarily) 14 MeV neutrons (DT tube), the other which only contains D produces 2.5 MeV neutrons (DD tube). The maximum neutron output of the tubes is: $2 \cdot 10^8$ n/s (DT), $2 \cdot 10^6$ n/s (DD). The tubes have a length of 71.5 cm, a maximum diameter of 11.5 cm, and a weight of only 8 kg and are therefore feasible for in-situ experiments. The neutron generator is powered by 12 V which may be taken from mains power supply or from a car battery. The typical operation time with battery is some hours.

The control case contains the electronic components necessary for operating the tube. It is also portable with a weight of just 7 kg, a length of 49.2 cm, a height of 41.2 cm, and a thickness of 24 cm and looks like a small suitcase (when closed). The power supply allows for an operation with either a continuous or a pulsed neutron output.

The operation parameters are set and controlled with special software which is run on a laptop computer connected to the power supply case via serial link. One of three operation modes can be selected: continuous, pulsed, or burst. Figure 2 illustrates these different operation modes. The maximum acceleration voltage of the tubes is 110 kV; the maximum beam current is 60 μ A. In the control software which was supplied with the generator, the cycle time in the burst mode may be selected within the range of 1 ms to 18 s, the repetition time in the pulsed or burst mode between 10 μ s and 18 s. The time range may be extended by customized software.



Figure 1: Essential components of neutron generator Genie 16c, on the left: laptop computer with control software, top right: control case, bottom right: neutron emission module.



Figure 2: Illustration of operation modes of Genie 16c.

4. Experimental setup

A 14 MeV neutron generator was used to produce the neutrons. The generator was pulsed with different neutron emission times ranging from 3 s to 300 s. The repetition time of the pulses varied between 3 s and 300 s, too. During the time interval after the neutron pulse of the neutron generator, when no primary neutrons are present, the delayed neutrons were measured. The neutrons were measured by a slab counter which uses 6 ³He-tubes of 2.54 cm diameter and 33 cm length embedded in a high density polyethylene moderator as detector. A discriminator and pulse forming unit shapes the signal to a 50 ns wide rectangular pulse which are then counted.

The investigated object was a cube of size $5 \times 5 \times 4 \text{ cm}^3$ made of depleted Uranium metal encapsulated in a thin steel casing. For comparison to non-fissionable material, a block of lead with the same size was used. Some experiments were carried out with additional PE moderator around the Uranium (see figure 3). In earlier experiments, which were carried out with the previously utilized neutron generator, the dependence of the number of delayed neutrons from the irradiation time had been determined [3].

This set-up is especially suitable for the inspection of suspicious objects in-situ because it can be arranged easily and quickly. A typical situation is shown in figure 4.



Figure 3: Experimental setup with neutron generator, measured object and neutron slab counter. The little picture shows the object in detail. The block of 1.8 kg depleted uranium was inserted in a PE block.



Figure 4: Experimental setup with a suitcase as inspected object.

5. Data acquisition

The neutrons were measured by the slab counter and converted to pulses which were then recorded by a multi channel scaling (MCS) system¹. The MCS-system records the time when the neutron arrives at the detector and finally generates a time distribution, the "decay curve" of the delayed neutrons. We resume the principle of multi channel scaling here. A triggering signal of the neutron generator correlated to the neutron emission starts the counting procedure. The neutron pulses then are counted for a specified time interval and the result is put into the first channel of the spectrum. Then the channel in the spectrum is advanced and the neutron pulses of the next counting interval are put into the next channel. This counting interval is called dwell time. When the last channel of the spectrum is reached, the system again waits for a triggering signal which restarts the counting procedure with channel one.

6. Basics of theoretical calculations

The time structure of the delayed neutrons in the experiment was simulated with the Monte-Carlo program MCNPX [4]. Therefore the experimental set-up was rebuilt in the simulation in detail. MCNPX simulates the micro-physical processes selecting the individual physical process according to its probability.

Delayed neutrons with similar decay time are usually sorted into 6 groups and a mean decay time is assigned to each group. Newer publications try to sort the delayed neutrons into 8 groups [5], but the benefit is not obvious and MCNPX libraries still refer to the 6 groups. Generally, cross sections, decay times and energy spectra were taken from ENDF/B-VI, release 6 [6], for the fission isotopes from the T16 2003 evaluation of LANL [7].

The time dependent neutron flux in the experiment was calculated with MCNPX to study the influence of different shielding material. Therefore, the experimental set-up described above (see figure 5) was simulated. It turned out that shielding material of considerable thickness does not influence the ability to detect fission material by delayed neutrons.

¹ accuspec system manufactured by canberra



Figure 5: Theoretically calculated number of delayed neutrons from induced fission in depleted Uranium with different kinds of shielding (neutron irradiation time 3 s).

7. Experimental results and comparison

Experiments were carried out to determine the most suitable neutron irradiation time and the delayed neutron measurement time. The experiments showed that after 60 s irradiation time a maximum in the counts of delayed neutrons had been reached. This can be seen from figure 6. In the present experiment we only looked at shorter times thus reducing the total measuring time for one measurement.



Figure 6: Number of the delayed neutrons plotted against different neutron irradiation times. The integration time for the measured neutrons is 200 s. The solid line serves to guide the eye.

The delayed neutrons produced by irradiation of depleted Uranium (with and without PE moderation) and lead (with PE moderation) were measured. In addition, the influence of shielding material (Pb, Fe) was calculated by MCNPX. Results are shown in figure 7.



Figure 7: Number of the delayed neutrons plotted against different object composition. The results for Pb in PE, U and U in PE are obtained from measurements with an integration time of 3 s starting after the neutron pulse. The neutron irradiation time was 60 s. The results for U in PE shielded with Pb and U in PE shielded with Fe are calculations with MCNPX see fig. 8. The PE had a thickness of 22 mm, Pb and Fe shielding material of 5 cm.

We also measured the decay curve for various irradiation and delayed neutron measurement times. The data were fitted to a constant and the strength of up to six exponential decay functions whereas the decay times were kept fix at the values found in ENDF/B-VI. An example of a fit to experimental data with 3 s irradiation and 3 s delayed neutron measuring time is given in figure 8.



Figure 8: Measurement of the decay curve of the delayed neutrons of depleted uranium moderated by *PE*. Irradiation time: 3 s, measuring time for the delayed neutrons: 3 s. Fit curve: constant plus four exponentials.

In principle it is possible to gain information on the isotopic composition of the fissionable material by looking at the "decay curve" of the delayed neutrons.

8. Conclusion and further investigations

The neutron interrogation experiments described above clearly show that modern small, lightweight and powerful neutron generators are suitable for the detection of SNM in the field. A discrimination of SNM from industrial radiological material is perfectly possible within a reasonable period of time. This method is therefore especially suitable for the examination of suspicious objects in-situ.

Further experiments may include investigations to evaluate the total measuring time which is optimal for the in-field situation. An optimization of the experimental process with regard to gaining information on the type of SNM present would also be valuable. Moreover, information concerning the quantity of the material in question could be derived from the results. Concerning the countermeasures against terrorist actions, it will be helpful to gain information on the influence of moderating and shielding material. This could then be taken into account when inspecting sites where SNM is suspected.

References

- [1] http://www.globalsecurity.org/security/ops/hsc-scen-1.htm.
- [2] Genie 16 Instructions for use, SODERN, ref. no. 4011 143 6934 0F (manual), 2001.
- [3] P. Hilger, T. Köble, W. Rosenstock: Measurement of the time structure of delayed neutrons after induced fission in special nuclear material. In: C. Foggi (editor); E. Petraglia (editor); European Commission, Directorate-General for Enterprise and Industry: Proceedings of the 21st ESARDA annual meeting, Luxembourg: Office for Official Publications of the European Communities, 1999, S. 321-325.
- [4] Denise B. Pelowitz, ed.: MCNPX User's Manual, Version 2.6.0, Los Alamos National Laboratory report, LA-CP-07-1473 (April 2008).
- [5] J.L. Rowlands: Delayed neutron data in 8 time groups, Jefdoc-976, NEA, Nov 2003.
- [6] J. M. Campbell, S. C. Frankle, and R. C. Little, ENDF66: A Continuous-Energy Neutron Data Library Based on ENDF/B-VI Release 6, LANL memorandum LA-UR-03-954, Los Alamos National Laboratory, Los Alamos, NM, 2003.
- [7] Robert C. Little: MCNP Neutron Library T16_2003, LANL memorandum LA-UR-04-4520, Los Alamos National Laboratory, Los Alamos, NM, 2004.