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Differential Die-Away Analysis for detection of ^{235}U in metallic matrix

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ABSTRACT

Differential Die-Away Analysis is a powerful tool for detecting small quantity of fissile material even if it is shielded or placed in non-fissile matrix. The technique can be used to monitor and characterize fissile content for nuclear waste assay. In this paper, we have discussed the application of differential die away technique for detection of small quantity of fissile material in nuclear waste assay. Feasibility experiments to optimize various parameters have been carried out for detection of ^{235}U in metallic matrix and reported in this paper. A minimum quantity of 1 g of ^{235}U in 150 kg of metallic matrix has been detected in the experimental configuration being reported.

1. Introduction

Detection and/or quantitative estimation of nuclear materials has both safety and security applications – from nuclear material accounting at the back-end of nuclear fuel cycle to detection of special nuclear material. A specific application, which is the focus of this paper, is the detection of residual fissile material in metallic hull matrix. At the back end of nuclear fuel cycle, it is vital to minimize loss of nuclear materials during fuel reprocessing by monitoring residual quantities of fissile material in metallic hull matrix. Feasibility experiments to optimize various parameters have been carried out using ^{235}U and reported in this paper.

Although most fissile material naturally emit neutrons and/or γ-rays, the intensity of the spontaneous radiation is low, and the energies of the γ-rays are fairly low in most cases. The passive gamma techniques are often inaccurate as these measurements get contaminated due to interference from the intense gamma rays from other fission and activation products. Also, gamma radiation from the natural decay of ^{235}U and ^{239}Pu is relatively soft and effectively attenuated by a few centimeters of shielding such as lead or iron. Passive neutron counting (gross counting and/or coincidence counting) may be used for monitoring of wastes but this measurement can only quantify the total spontaneous fission neutron emission rate (due to emissions from even mass nuclides such as ^{238}U, ^{240}Pu and ^{244}Cm). To quantify specific transuranic nuclides or fissile isotopes (such as ^{235}U or ^{239}Pu) one needs to correlate it with fuel burnup and isotopic composition. The assay is complicated by the fact that additional neutron are generated due to (α, n) reaction resulting from the alpha activity of actinides present in the waste and their interaction with the light elements such as fluorine and oxygen also present in the matrix. Passive neutron counting is effective when the quantity of fissile material to be assayed is large and is not typically suited for detection of residual fissile material as in the case of hull monitoring.

Because of the limitations of passive gamma and neutron based methods for such an accurate accounting application, active neutron interrogation technique has been developed to improve sensitivity and minimum detection limit. Active neutron interrogation approach involves bombarding the sample with external neutron source and thereby inducing additional fission in the sample and counting the emitted neutrons due to fission. The advantage of active neutron interrogation technique is that it can detect very small quantities of fissile materials directly even in the presence of high neutron and gamma background and is much more sensitive compared to passive techniques which are indirect measurement of fissile material with large error margins. Active neutron interrogation determines the total fissile content of the waste to be measured (comprising the fissile nuclides ^{235}U, ^{239}Pu and ^{241}Pu).

In this paper, we discuss one particular form of active neutron interrogation namely Differential Die Away (DDA) technique [1–6] which is one of the most sensitive methods for detection of fissile material. We have optimized and calibrated our system for detection of ^{235}U in metallic matrix. This paper presents experimental results of detection of uranium in a simulated hull drum. This is first step in implementation of this technique under actual conditions and this allows us to investigate various aspects of this technique in controlled conditions.

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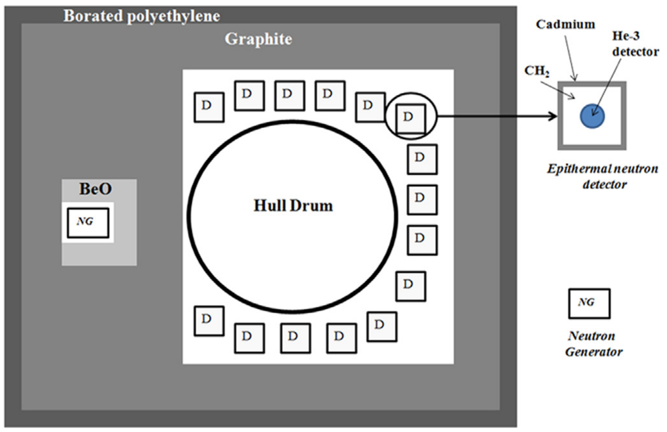

Fig. 1. Schematic of experimental set-up.

Fig. 2. Experimental set-up.

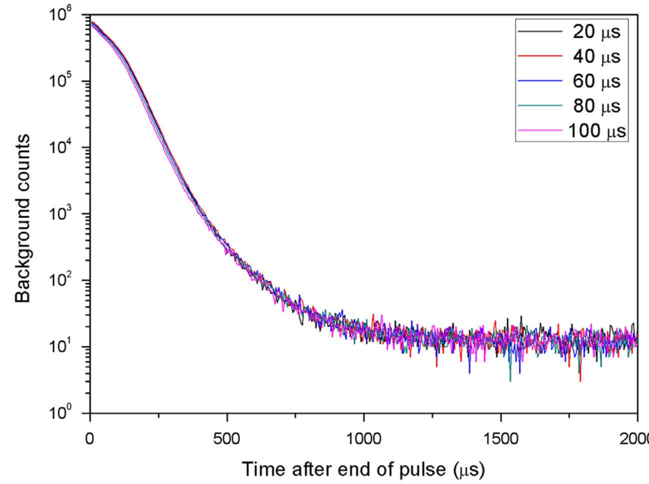

Fig. 3. Plot of background counts for different pulse width.

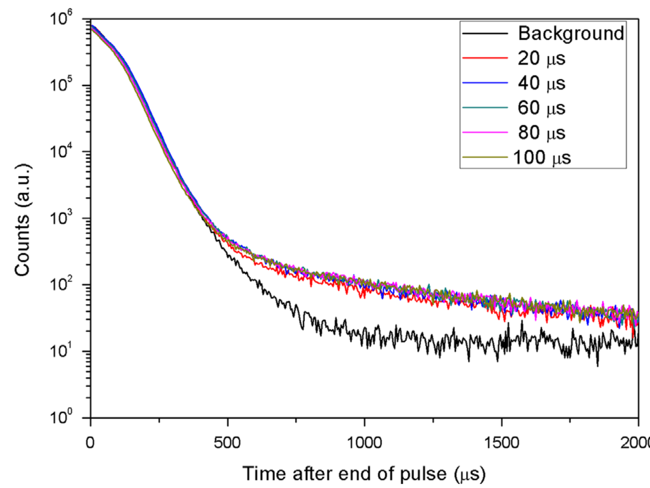

Fig. 4. Plot of signal counts for different pulse width.

2. Differential Die-Away technique

In DDA technique, the inspected medium is exposed to pulses of neutrons. An external fast neutron detector measures the time-dependent decay of neutrons from the matrix. When no fissile material is present in the medium the detector should only measure a signal representing the diffusion of the thermalized neutrons in the detector body (detector neutron "die-away" time). If fissile material is present, the detector will show, in addition to a signal decaying with the detector die-away time, an additional signal decaying with the die-away time of the inspected medium. If the latter is significantly longer than the former it will dominate the decay curve at later times and unequivocally establish the presence of fissile material in the inspected object. The DDA technique works by separating time zone of decay of primary signal of fast neutron, signal region of prompt neutrons emitted by interaction of die away thermal neutron with fissile material in the system and signal region of delayed neutron. These regions are well separated in time domain by an order of magnitude.

3. Experimental set-up

An experimental arrangement (Figs. 1 (schematic) and 2) to investigate the potential of this technique has been set up in BARC. Dry zircaloy pieces weighing approximately 150 kg have been placed in stainless steel drum of diameter 900 mm and height 1000 mm to simulate the cladding materials in hull. This assembly is surrounded by a bank of epithermal neutron detectors (^{3}He detector enclosed in high density polyethylene and covered with cadmium) from three sides. The drum assembly has been surrounded by graphite to increase the die-away time of thermal neutrons. The assembly has also been shielded from all sides by borated polyethylene so as to reduce influence of scattered neutrons in the measurement. Furthermore, as the neutron generator emits neutrons with energy 14 MeV, its energy was reduced suitably by using BeO bricks surrounding the neutron generator all around.

A pulsed D-T neutron generator with source strength of 1×10^8 n/s has been used as external neutron source for inducing fission. The neutron pulses are generated at a frequency of 400 Hz with pulse width of 80 μs. The pre-amplifier signal from 16 ^{3}He detectors is processed through amplifier and discriminator and summed TTL signal is fed to MCS for collecting the data. The total acquisition time for each data set is 300 s. The experiments to simulate various conditions as encountered in a hull matrix have been carried out as described in the subsequent discussion.

4. Experimental results

4.1. Effect of pulse width variation on signal to noise ratio

Pulse width determines the source strength and the available counting time. For our experiments, the counting gate starts with the end of the neutron pulse. A large pulse width means more source neutrons in the system which may reduce sensitivity and a small pulse means neutron flux may not be sufficient to get good counting