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Non-destructive assay of fissile materials through active neutron interrogation technique using pulsed neutron (plasma focus) device

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ABSTRACT

Pulsed neutrons emitted from a plasma focus (PF) device have been used for the first time for the non-destructive assay of ^{235}U content in different chemical forms (oxide and metal). The PF device generates $(1.2 \pm 0.3) \times 10^9$ D–D fusion neutrons per shot with a pulse width of 46 ± 5 ns. The method involves the measurement of delayed neutrons from an irradiated sample 50 ms after exposure to the neutron pulse for a time of about 100 s in the multichannel scaling (MCS) mode. The calibration of the active interrogation delayed neutron counter (AIDNEC) system was carried out by irradiating U_3O_8 samples of varying amounts (0.1–40 g) containing enriched ^{235}U (14.8%) in the device. The delayed neutrons were monitored using a bank of six ^{3}He detectors. The sensitivity of the system was found to be about 100 counts/s/g over the accumulation time of 25 s per neutron pulse of $\sim 10^9$. The detection limit of the system is estimated to be 18 mg of ^{235}U. The system can be suitably modified for applications toward non-destructive assay of fissile content in waste packets.

1. Introduction

Non-destructive assay (NDA) of nuclear fuels is one of the important topics of research and development from the point of view of nuclear material accounting, including safeguards and waste assay. NDA of plutonium based fuels can conveniently be carried out using passive assay methods based on gamma rays and neutrons emitted during the spontaneous decay of different isotopes [1]. However, for NDA of uranium-bearing fuels, passive assay methods are not convenient owing to the low specific activity of uranium isotopes as well as negligible spontaneous fission decay.

For NDA of uranium bearing fuels and waste materials, the active interrogation method is advantageous. In this method, the sample is irradiated with neutrons from a source and the prompt or delayed fission neutrons are counted using suitable neutron detectors. Delayed neutron activation analysis (DNAA) has been used for determination of uranium in various types of samples [2–5]. Benzing et al. developed a fully automated delayed neutron counting system for analysis of large uranium-bearing samples with a detection limit of 1 μg/kg [6]. The system was meant for routine analysis of environmental samples and personnel monitoring.

A large number of literature reports exist on the assay of ^{235}U and ^{239}Pu in samples by delayed neutron activation analysis (DNAA). For example, samples have been irradiated in the pneumatic carrier facility of TRIGA reactor followed by counting of delayed neutrons [7,8]. The latter report describes uranium assay in urine samples by precipitation of uranium with $Fe(OH)_3$ followed by delayed neutron activation analysis. A. El Taher reported the DNAA of uranium in geological samples with a sensitivity of 10^{-11} g for ^{235}U [9]. DNAA was used for the first time for ^{235}U determination in 1959 by Echo and Turk [10]. Rinard developed the Shuffler system for NDA of fissile materials [11]. Raoux et al. reported an NDA method based on the measurement of prompt and delayed neutrons for assay of transuranic waste using a 14-MeV pulsed neutron generator [12,13]. Recently Lakosi et al. reported an interesting example of uranium assay by the measurement of delayed neutrons emitted in fission of uranium by photo-neutrons produced in (γ,n) reactions on Be or D_2O target by Bremsstrahlung from a 4 MeV LINAC [14].

An active well coincidence counter has been used for the NDA of uranium bearing fuels in the past. In this method, fission is induced in ^{235}U by interrogating neutrons and the delayed neutrons emitted from the induced fission are measured by neutron counters, such as ^{3}He gas filled proportional counters. The neutron sources for interrogation used in the past include ^{252}Cf [11] and a D-T LINAC [12,13]. In the case of ^{252}Cf as a source of neutrons, the source is stored behind a shielding for both gamma rays and neutrons and is brought to the sample position

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for a short period of time after which it is quickly pushed back to the shield. However, despite the shielding, the background neutrons cannot be completely eliminated leading to an increase in the detection limit. In the case of the D-T LINAC, the neutron pulse repetition rate (typically few hertz) results in the rise and fall of delayed neutron counts during irradiation and hence the de-convolution of the decay profile into individual delayed neutron groups is difficult.

Plasma focus (PF) devices provide a single or multiple neutron pulses of a very small pulse width and with very high neutron intensity [15]. A plasma focus device generating 3×10^8 neutrons per pulse has been reported [16] by Verri et al. to be used in the fast neutron activation analysis of gold by the inelastic scattering in the reaction ^{197}Au(n,n′γ)^{197}Au. Neutron scattering from a compact PF device [17], producing 2×10^8 neutrons/pulse has been reported for detecting water content of a few percent in containers placed about 8.5 cm away from the PF chamber. A recent report [18] shows a plasma focus based neutron source for a single-shot detection of the illicit materials and explosives. The use of these devices for boron neutron capture therapy was also demonstrated recently [19]. An excellent review of the new developments in the active interrogation of nuclear materials for the non-destructive assay has been published recently by Runkle et al. [20].

In this paper we report the results of our study on the delayed neutron measurements produced in the fission of enriched ^{235}U samples irradiated with a pulse of neutrons from a plasma focus device [21], which provide a single pulse of neutrons having width of 46 ± 5 ns. The methodology for assay of ^{235}U content in uranium oxide as well as metal has been demonstrated successfully for the first time using the plasma focus device. This is a unique source of neutrons and is ideal for activation of short lived radionuclides [22]. The methodology for NDA of ^{235}U in pure uranium samples has been demonstrated over a large dynamic range.

2. Experimental

2.1. Plasma focus device

A Mather-type [15] transportable medium energy (11.5 kJ/24 kV) plasma focus device providing $(1.2\pm0.3)\times10^9$ D–D fusion neutrons per pulse with a pulse width of 46 ± 5 ns has been developed in our laboratory. The device is driven by a capacitor bank (40 μF capacity) of four capacitors. The filling gas is deuterium at 5 mb pressure. A schematic diagram of the system is shown in Fig. 1. The details of the setup are available elsewhere [21,22]. The basic components of the system are the capacitor bank, the power supply and the plasma focus unit. The diagnostics consists of a miniature Rogowsky coil for monitoring the current derivative, the silver activation detector for total (time integrated) neutron yield measurements and the plastic scintillator detector coupled to the photomultiplier tube for recording the temporal profile of neutron emission from the source. The delayed neutrons were measured by a ^{3}He detector [23] bank coupled to 8 k channel analyzer operating in the multichannel scaling (MCS) mode.

2.2. Sample preparation

U_3O_8 samples of mass varying from 0.1 to 40 g, enriched with ^{235}U (14.8%), were doubly sealed in polyvinyl chloride (PVC) bags of dimensions 5×5 cm^2. The enrichment was determined before use in the present experiments, using γ-ray spectrometric measurements of liquid samples on a well type 7.6×7.6 cm^2 NaI(Tl) detector coupled to a 2048 channel analyzer. The count rate of the 185 keV γ-ray peak had been calibrated with the ^{235}U amount in the samples, total uranium content of which had previously been

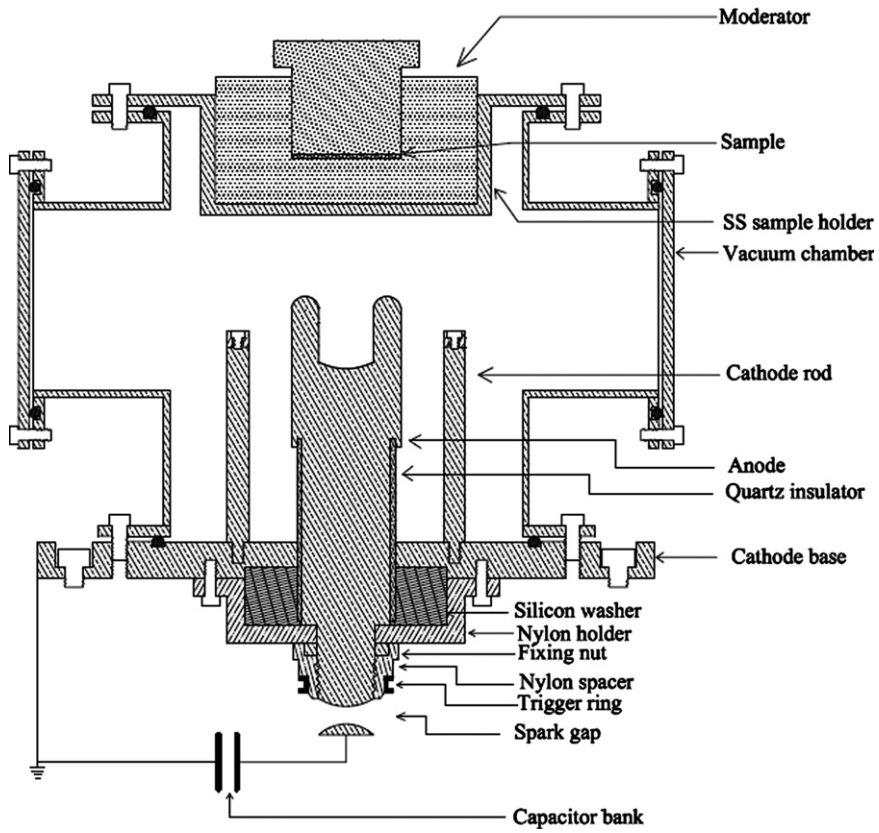

Fig. 1. A schematic of the plasma focus device and irradiation assembly.