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Neutron interrogation of high-enriched uranium by a 4 MeV linac

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Abstract

For revealing unauthorized transport (illicit trafficking) of nuclear materials, a non-destructive method reported earlier, utilizing a 4 MeV linear accelerator for photoneutron interrogation, was further developed. The linac served as a pulsed neutron source for assay of highly enriched uranium. Produced in beryllium or heavy water by bremsstrahlung, neutrons subsequently induced fission in the samples. Delayed neutrons were detected by a newly designed neutron collar built up of 14 ^{3}He counters embedded in a polyethylene moderator. A PC controlled multiscaler served as a time analyzer, triggering the detector startup by the beam pulse. Significant progress was achieved in enhancing the detector response, hence the sensitivity for revealing illicit material. A lower sensitivity limit of the order of 10 mg ^{235}U was determined in a 20 s measurement time with a reasonable amount of beryllium (170 g) or of heavy water (100 g) and a mean electron current of 10 μA. Sensitivity can be further enhanced by increasing the measurement time.

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1. Introduction

For preventing spread of nuclear materials (NM), a non-destructive assay (NDA) method (a “portal monitor” as an ultimate goal) has long been needed, suitable for revealing smuggled NM. Even a thin metallic shielding hinders detection of uranium-containing material by direct method, i.e. by passive γ-ray detection. However, active methods may be promising, by irradiating NM by neutrons. Neutrons can readily penetrate non-hydrogeneous shielding material, induce subsequently fission in the NM and fission neutrons are to be detected. For producing interrogating neutrons, a lot of methods have widely been applied. In the fields of safeguards and illicit trafficking of NM, pulsed D-T neutron generators (14 MeV) represent a sensitive and versatile variant of active interrogation systems, by counting delayed fission product neutrons [1,2], or gammas [3]. Larger and more effective systems designed basically for inspecting sea cargo rely on photoneutron interrogation by – up to 24 MeV – linacs [4,5]. For more references see [6].

Our photoneutron interrogation project was started a few years ago [6], results of which encouraged us to pursue development efforts. So we continued exploiting our linac as a photoneutron source, to induce fission in highly enriched uranium samples, while low enriched ones were assayed previously [6]. Prompt and delayed neutrons are produced in the fissile material. Distinction of induced neutrons from interrogating ones can be made using time discrimination via detection of delayed neutrons, but prompt fission neutrons are equally lost in this case. Typically only about 1% of fission neutrons are beta-delayed, emitted by fission products. Nevertheless, in the absence of the huge background of interrogating neutrons, the sensitivity may still be high enough for performing the assay.

By utilizing our small 4 MeV linac, neutrons can be produced by (e,γ) and (γ,n) double conversion. Both beryllium and heavy water was tried as photoneutron converter. The

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neutron energy available from the ^{9}Be(γ, n)^{8}Be and D(γ, n)H reaction is up to 2.33 and 1.78 MeV, respectively, at 4 MeV electron energy. However, the yields abruptly vanish above around 0.9 MeV neutron energy, whereas maximum intensity of the spectrum of evaporated neutrons is at about 0.5 MeV, even at much higher linac energies (15–50 MeV) [7,8]. Electron pulses can be produced in one of the two basic modes of operation; either single pulse is fired by an external trigger, or continuously with a repetition rate of 50, 25, 12.5, or 6.25 Hz. The normal pulse duration is 2.6 μs. The peak intensity was 200 mA at maximum (with a mean current of 26 μA) in the beginnings, but now the available intensity is much less.

2. Time structure of delayed neutrons during and after pulsed interrogation

The relative yields of the six main groups of delayed neutrons from ^{235}U fission induced by fast neutrons are shown in Table 1 [9]. Relative intensities are also given, obtained by multiplying the former values by the respective decay constants and normalized their sum to 100%. They are relevant to the intensity after a single linac pulse. The relative intensity decay of delayed neutron groups after a single pulse is shown in Fig. 1.

However, after continuous pulsing the decay curves look different, i.e. relative intensities of individual groups differ from those prevailing after a single pulse, depending on the pulse rate. Decay after irradiating to saturation by 25 Hz repetition rate is plotted in Fig. 2, as an example. The difference can well be seen in the insert, where the decay curve in the beginning 2 s can be compared with that of a single pulse (Fig. 1). The amplitude is more than 50 times higher than that of a single pulse. Decay was traced experimentally in a longer run, as Fig. 2 shows. The starting point was the number of counts acquired during the measurement of the 10.5 g uranium sample at 25 Hz pulsing. Then the numbers of counts were read out after linac stop in consecutive counting times. Measurement points (normalized to the relative amplitude) fit well the calculated curve (details about the measurement method follows below).

Saturation amplitudes get successively halved, of course, whenever pulse frequencies are consecutively turned from 50 to 25 Hz, from 25 to 12.5 Hz and from 12.5 to 6.25 Hz, in parallel to the successive halving of the mean current intensity. In Fig. 3(a) saturation curves are seen for the four operational pulse frequencies. Contributions from individual delayed neutron groups are indicated only for the 50 Hz case. The resulting saturated total amplitude is still about 14 times higher at the lowest rate, 6.25 Hz, than that of a single pulse (see below). Fig. 3(b) shows saturation curves of the individual delayed neutron groups at 25 Hz. (The curves are not simply halves of those at the 50 Hz case, but the minor differences are not visible.)

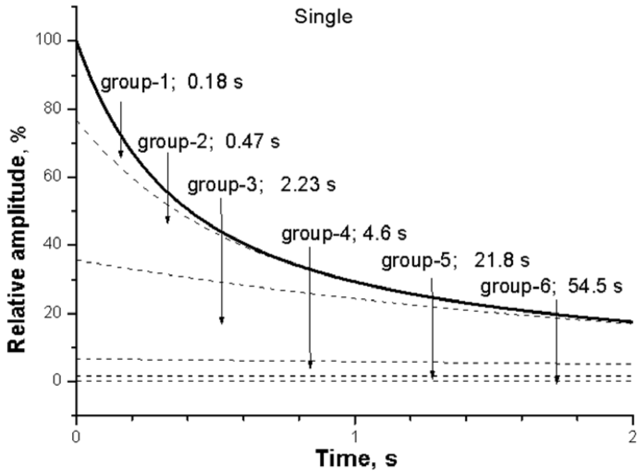

Fig. 1. Decay of delayed neutron groups after a single pulse.

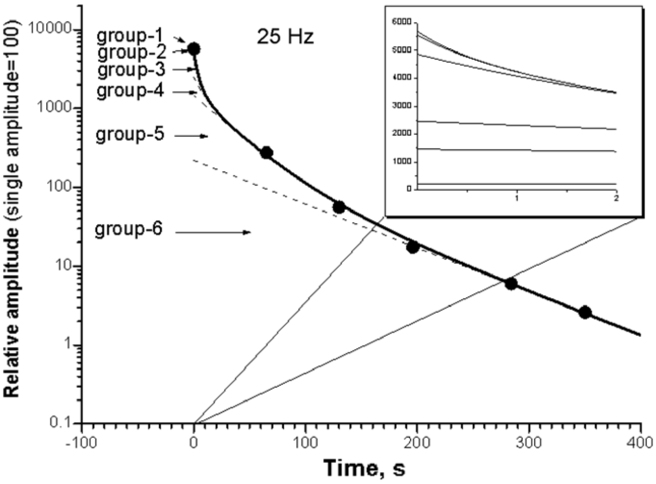

Fig. 2. Decay of delayed neutron groups after irradiating to saturation at 25 Hz.

Table 1
Relative yields and intensities of the delayed neutron groups from ^{235}U fission induced by fast neutrons

Group	$T_{1/2}$ (s)	Relative yield (%)	Relative intensity (%)
1	0.179 (0.017)	2.6 (0.3)	23.1 (2.7)
2	0.496 (0.029)	12.8 (0.8)	41.1 (2.6)
3	2.23 (0.06)	40.7 (0.7)	29.1 (0.5)
4	6.0 (0.17)	18.8 (1.6)	5.0 (0.14)
5	21.84 (0.54)	21.3 (0.5)	1.6 (0.04)
6	54.51 (0.94)	3.8 (0.3)	0.11 (0.01)

Saturation amplitudes of different delayed neutron groups are depicted in Fig. 4 as a function of pulse frequency. Group 3 gives the highest contribution to delayed neutron intensities, as seen. Fig. 5 shows what saturation curves of this group, as an example, are really like, for various frequencies, taking into account the sawtooth-like increasing-decreasing patterns during pulsing. (Smooth curves of Fig. 3. are approximations only). The case of 50 Hz was not considered, because this frequency was not applied, see below.

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