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NEUTRON INTERROGATION OF URANIUM BY A 4 MeV LINAC

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Abstract

For revealing illicit trafficking of nuclear materials, a neutron interrogation method was developed. A 4 MeV linear accelerator served as a pulsed neutron source for assay of highly enriched U samples. Produced in Be or heavy water by bremsstrahlung, neutrons subsequently induced fission in the samples. A neutron collar was built for detecting delayed fission neutrons. A PC controlled multiscaler served as a time analyzer, triggering the detector startup by the beam pulse. A lower sensitivity limit of the order of 10 mg ^{235}U was determined in a 20 s measurement time with a mean electron current intensity of 10 μA.

Keywords: *Non-destructive assay, Illicit trafficking, Be and D_2O converter, Delayed neutron counting.*

1. INTRODUCTION

For preventing spread of nuclear materials (NM), a non-destructive assay (NDA) method has long been needed, which is suitable for revealing smuggled NM at border checkpoints. Even a thin metallic shielding hinders detection of uranium-containing material by direct methods, i. e. by passive γ-ray detection. However, active methods may be promising, by irradiating NM by neutrons („active interrogation"). Neutrons can readily penetrate high-Z 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. The advantage of accelerator-driven neutron sources is that they can be switched off. Neutron generators relying on D(d,n)^{3}He, T(d,n)^{4}He [1-3], and ^{9}Be(d,n)^{10}B reactions [4,5], or linear electron accelerators (linacs) are common as pulsed neutron sources. Conversion of electron energy into bremsstrahlung is followed by subsequent neutron production via photonuclear reactions. Heavy metal (Ta, W) is common as target, while electron energies of 7-20 MeV are needed for photoneutron production [6]. Even 50-150 MeV beams are frequently used for achieving higher assay sensitivity, using photonuclear reactions and additional photofission of the NM [7], the latter even by 10-15 MeV linacs [8,9]. It was estimated [10] that a 4 MeV linac can produce a neutron yield of 5×10^7 n/s/μA in a 1 cm thick heavy water, while a yield of 4×10^8 n/s/μA can be reached by a 10 MeV linac. With 70 cm^3 heavy water, a 10 MeV linac was calculated to provide ideally ~10^9 n/s/μA neutron yield

[11]. A thermal neutron beam of 1.23×10^8 n/cm^2/s/mA intensity was achieved by a 5 MeV linac at 50 cm distance from a BeD_2 target [12].

A photoneutron interrogation method was developed using our 4 MeV linac as a neutron source, to induce fission in highly enriched uranium samples. Prompt and delayed neutrons are produced in the fissile material. Distinction from interrogating neutrons can be made using time discrimination via detection of delayed neutrons, but prompt fission neutrons are equally lost as in this case. Typically only about 1 % of fission neutrons are beta-delayed, emitted by fission products. In the absence of the huge background of interrogating neutrons, however, the sensitivity may still be high enough for performing the assay.

2. METHOD

A small linac of 4 MeV electron beam (Tesla LPR-4, 100 W at maximum) was installed at the Institute of Isotopes 24 years ago. For production of neutrons with such low energy electrons, attention has been focused on beryllium and deuterium, owing to their low (γ,n) reaction thresholds of 1.67 and 2.22 MeV, respectively. While these thresholds are quite low compared to those of heavy elements, the cross sections for the (γ, n) process are also smaller by about two orders of magnitude than the giant resonance cross sections for heavy nuclei. However, the mean free-path in a heavy element for γ-rays is limited by the atomic cross section. For the lighter targets of Be or D, the atomic cross sections are also much smaller, whereas the ratio of atomic to nuclear cross sections is not too different from that of heavy targets. Therefore, in those experiments, where target size is not an important factor, the lighter targets of Be and D might compete [13]. Actually, neutron yields from these light elements can be roughly an order of magnitude higher than those from heavy W or Pb even at 10 MeV [6]. At our 4 MeV linac, this is not the case, but a feasibility study still seemed worthwhile to try.

Neutron production is due to (e,γ) and (γ,n) double conversion. Both beryllium and heavy water was tried as photoneutron converter. The neutron energy available from the $^9Be(\gamma,n)^8Be$ and D(γ,n)H reaction is up to 1.5 and 0.9 MeV, respectively, at 4 MeV electron energy. 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 is 200 mA, while the mean current intensity is 26 μA at maximum. The energy distribution of the electrons is about 0.67 MeV FWHM at 4 MeV. Energy stability is about 4 %.

The relative yields of the six main groups of delayed neutrons from ^{235}U fission induced by fast neutrons are shown in Table 1 [14]. Fast neutrons are considered, because thermalization is rather imperfect. Relative intensities are also given, obtained by multiplying the former values by the respective decay constants and normalized their sum to 100 %. Uncertainties are in parentheses. It is seen that contribution of the three first groups predominates, i.e. delayed neutron intensity practically goes into saturation in the first 10 - 15 seconds of irradiation. This means that after electron beam is turned on, measurements can be started after pulsing for 10-15 s.

The relative intensity decay of delayed neutron groups after a single pulse is shown in Fig.1*a*. 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.1*b*, as an example.

Table 1. Relative contribution of the delayed neutron groups at saturation

Group	$T_{1/2}$ (s)	Relative yield (%)	Rel. 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)	1.69(0.04)
6	54.51 (0.94)	3.8 (0.3)	0.11(0.01)

More importantly, saturated amplitudes get successively halved, of course, whenever pulse frequencies are 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. 2*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. Note that the resulting saturated total amplitude is more than 110 times higher at 50 Hz than that of a single pulse (see later), while still about 14 times higher at the lowest rate, 6.25 Hz. Fig.2*b* shows saturation curves of the individual delayed neutron groups at 25 Hz.

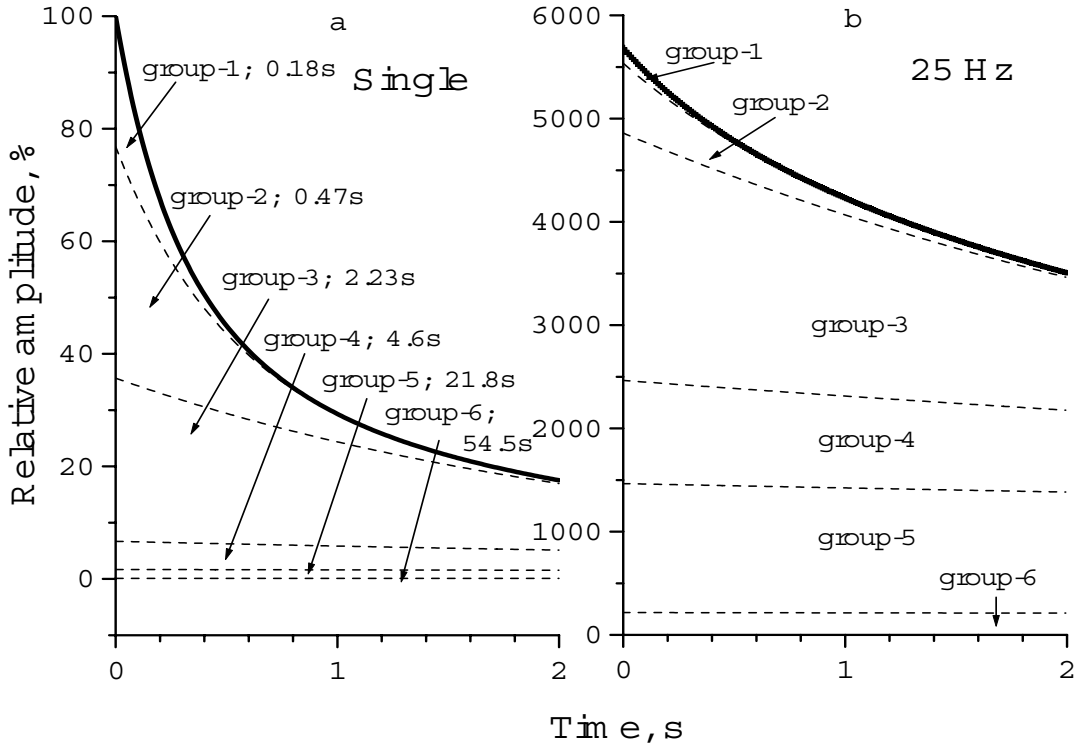

Fig.1. Decay of delayed neutron groups: after a single pulse (a); after irradiating to saturation at 25 Hz (b)

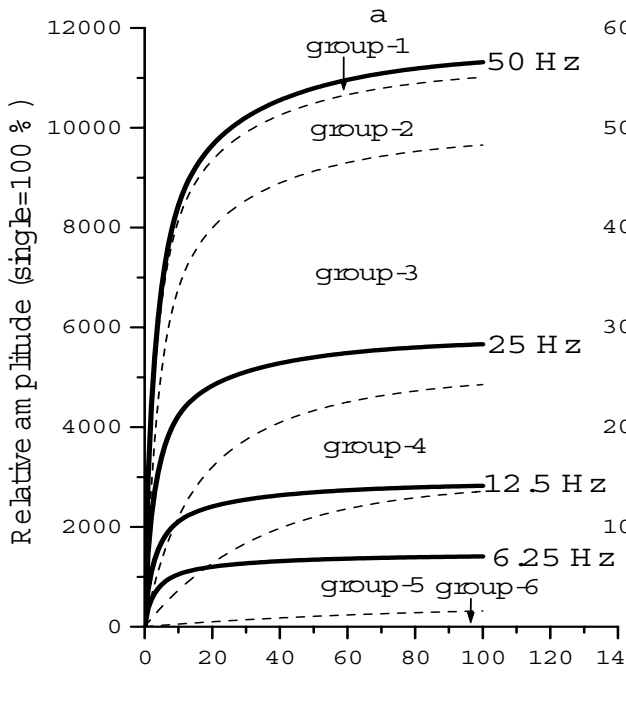

Fig.2a. Saturation curves of delayed neutrons at various pulse frequencies and those of individual groups at 50 Hz

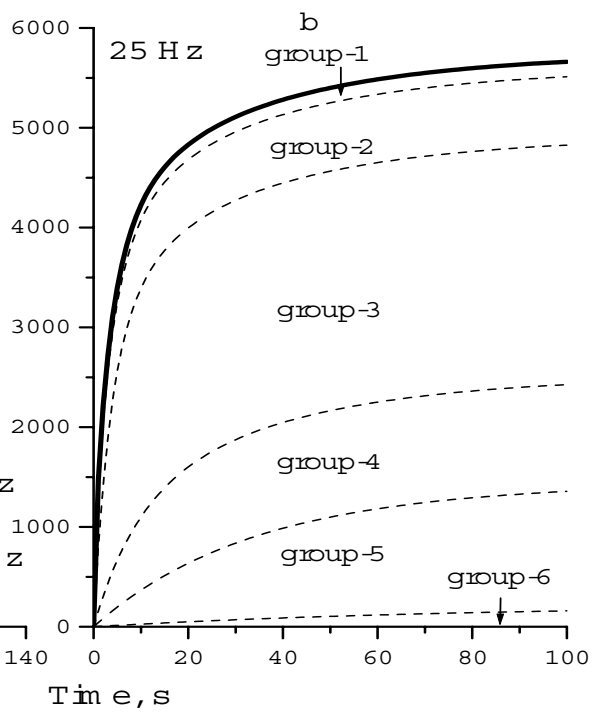

Fig.2b. Saturation curves at 25 Hz

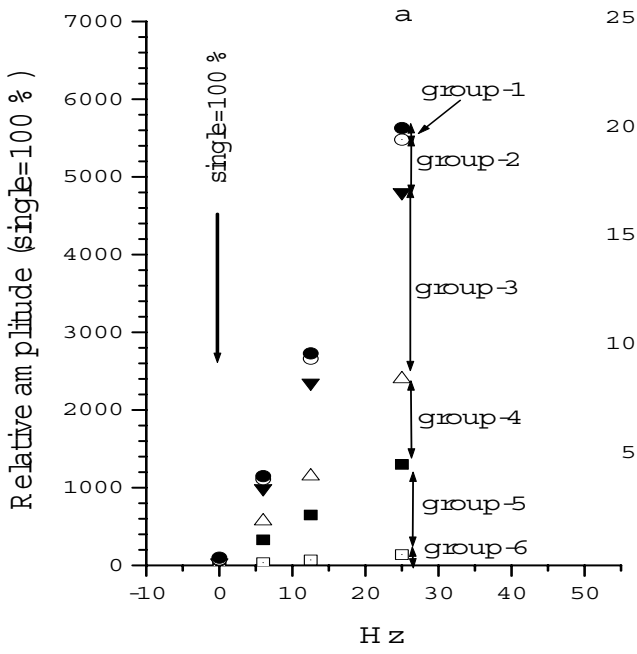

Fig.3a. Saturation amplitudes of delayed neutron groups at various frequencies

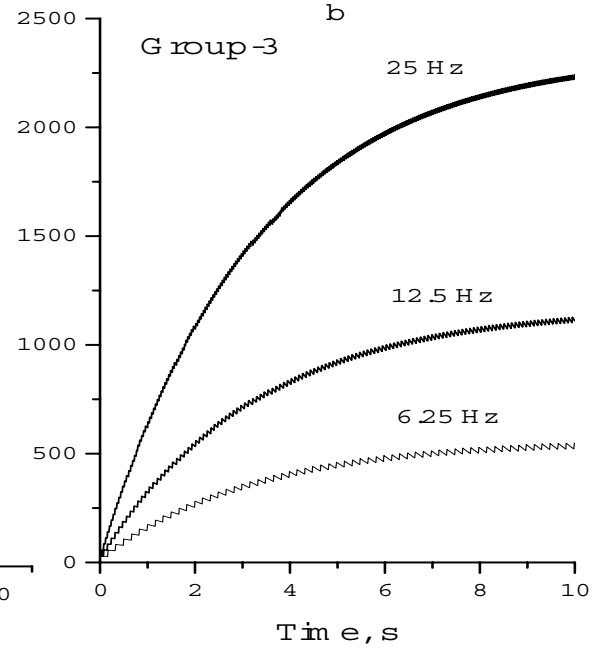

Fig.3b. Saturation curves of the delayed neutron group 3 for various frequencies

Saturation amplitudes of different delayed neutron groups are depicted in Fig.3*a* as a function of pulse frequency. In Fig.3*b* saturation curves of the delayed neutron group "3" are shown for various frequencies, as an example. The case of 50 Hz was not considered, because this frequency was not used, see below.

3. EXPERIMENTAL SETUP

Bremsstrahlung was generated on a 20 by 30 mm size Pt converter positioned at 3 cm distance from the exit window of the linac. The diameter of the electron beam was about 2 cm at converter distance. The 0.9 mm thick converter provided complete stopping of the incident electrons. The total bremsstrahlung output with energy above 1 MeV was calculated to be about $2.4(0.3)\text{x}10^{13}$, about $1(0.2)\text{x}10^{13}$ above 1.67 MeV, and $7.3(1.1)\text{x}10^{12}$ photon/s above 2.22 MeV at full intensity to forward directions (2π solid angle) [15].

A neutron collar was built in our laboratory [16], consisting of two concentric polyethylene cylinders of an outer size of 200 mm diameter and 420 mm length (Fig.4). The inner cylinder of 55 mm diameter forms a measurement cavity of wall thickness 10 mm for the material to be assayed. In between the two cylinders, proportional counters filled with ^{3}He gas to a pressure of $4\text{x}10^5$ Pa (4 atm) serve as neutron detectors. A Cd shielding on the inner side of the counter ring was also inserted, as shown in Fig. 4.

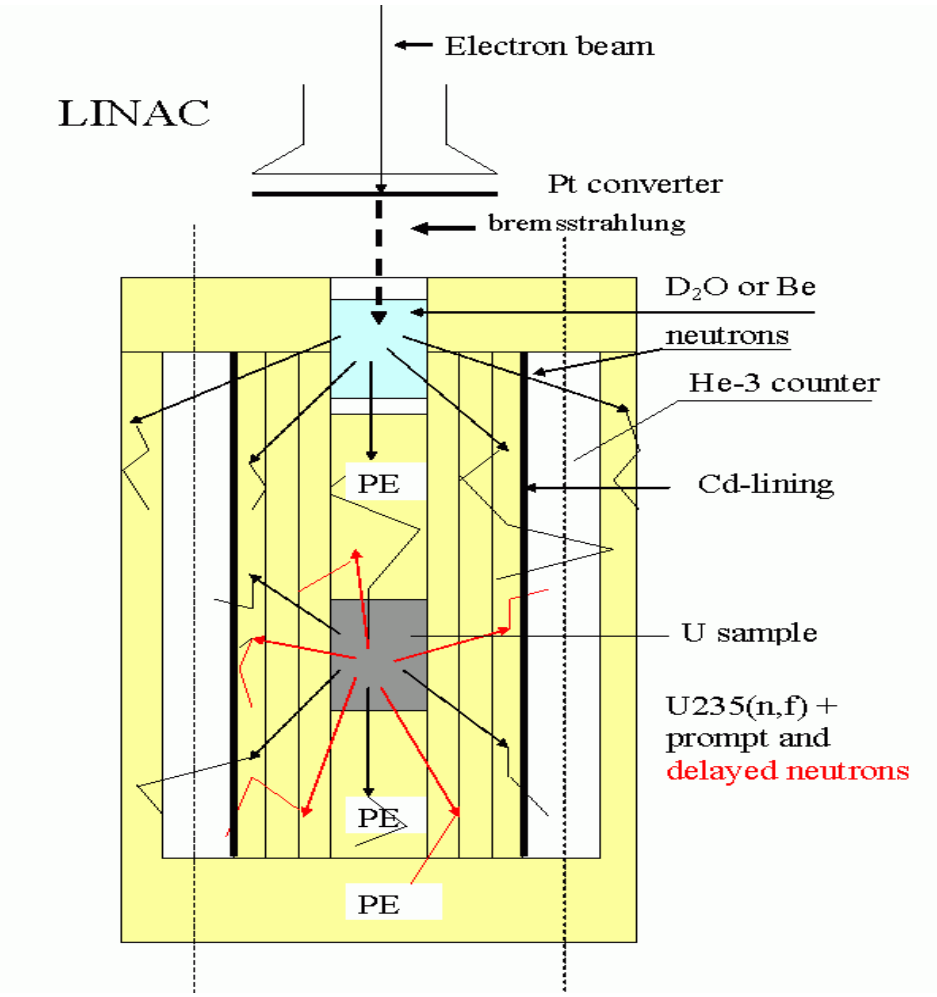

Fig.4. Experimental setup

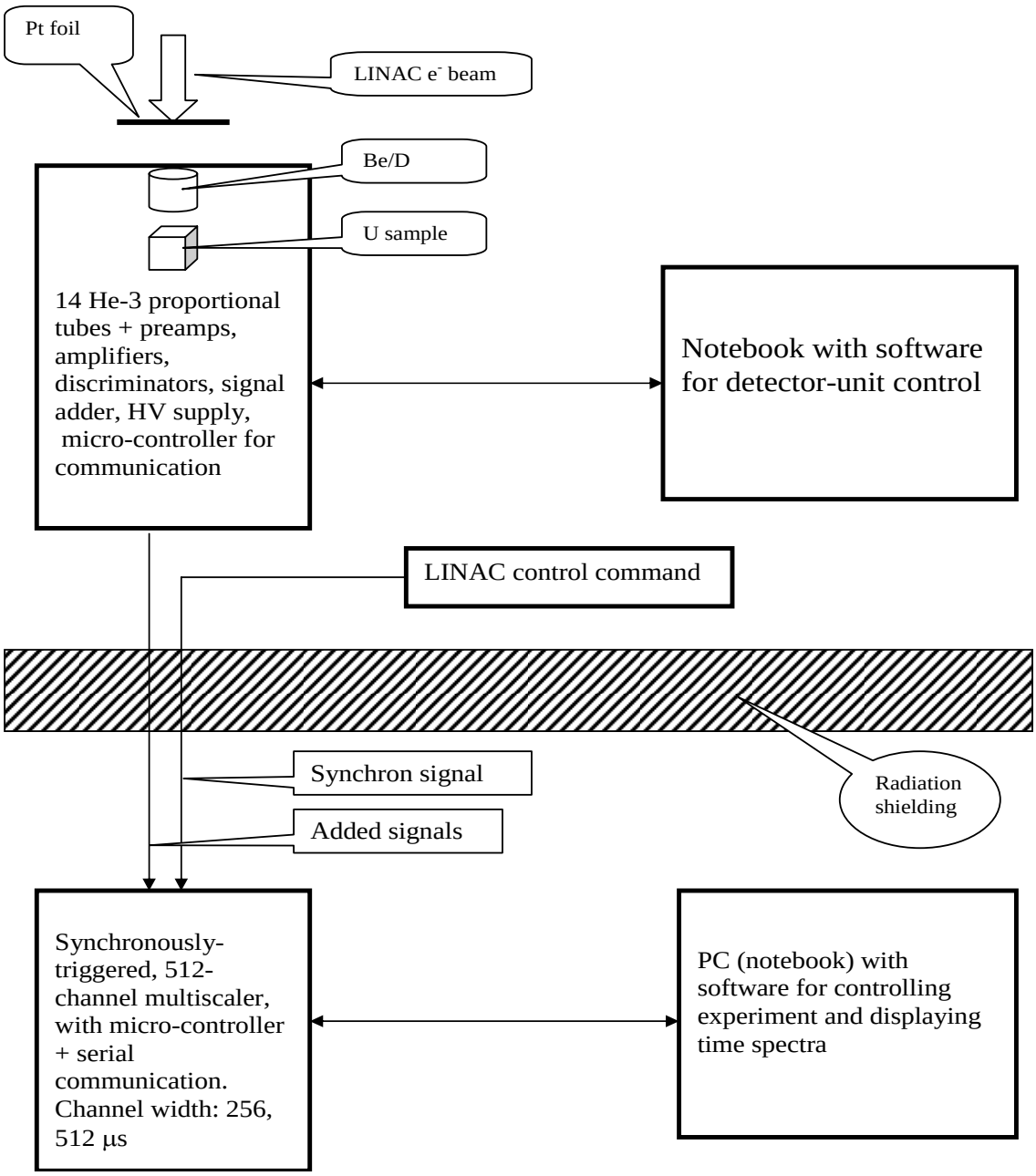

Fig.5. Schematic block-diagram of signal processing electronics

A beryllium photoneutron converter of a mass of 170 g was applied at the top of the collar, alternatively with 100 g heavy water. The total (interrogating) neutron output was estimated to be about 10^7 neutron/s/μA/g heavy water. U oxide samples were inserted in the central cavity for irradiation and measurement.

The schematic block diagram of the signal processing electronics is shown in Fig.5. It consists of a 512 channel analyzer in multiscaler mode of operation as a time analyzer. The multiscaler receives commands from a PC through a micro-controller. Triggering the analyzer was synchronized with the linac control command pulse. The time scale can be varied from

25.6 μs/channel to 65.5 ms/channel. The whole time spectrum thus covers a period extending from the fastest 13 ms to the slowest range of about 1.6 s.

4. DELAYED NEUTRON COUNTING

U oxide samples enriched to 36 and 90 %, of mass up to 16 g were assayed, by applying Be or heavy water converter.

Time spectra of the detected neutron pulses are seen in Fig.6. All the spectra were acquired during 1000 subsequent cycles of irradiation and counting, by using the Be converter. Spectra taken up by heavy water look quite similar. Time analyzer channel widths of 256 and 512 μs were selected. In order to reach a sufficient degree of saturation, 10 s irradiations were uniformly carried out before starting cyclic delayed neutron measurements.

Detected neutron pulse shape at 50 Hz repetition rate without U sample is shown in panel *a*. A 1000 cycles measurement at 50 Hz lasts for 20 s. Exponential decay with a time constant of about 2 ms can be observed, with a total pulse length of 20 ms. In panel *b*, the time spectrum of a 16 g, 36 % enrichment U sample, acquired at 50 Hz, shows that the pulse length of prompt (interrogating and fission) neutrons covers the whole 20 ms time interval again, available for measurement. It means that by applying 50 Hz frequency, no time remains for delayed neutron counting, before the subsequent pulse arrives. Repetition rate was set therefore to be lower, with the consequence of accordingly lower saturation amplitude of delayed neutrons.

By selecting 25 Hz, the effective time remaining for delayed neutron measurement is about 20 ms, a half of the 40 ms interval between two pulses. In this way, 1000 cycles last for 40 s, as displayed in panel *c* (plus an additional 10 s for initial irradiation prior to starting measurement).

Determined by the material and size of the moderator, i. e. by the neutron spectrum ultimately, the pulse length cannot be influenced electronically, and it is practically the same for Be and for heavy water.

It has been expected that applying Cd shielding on the internal side of the counter ring would diminish neutron life time. This is why a 0.8 mm thick Cd foil was initially inserted. It turned out by now, however, that it does not result in noticeable decrease of the pulse length, but does cause a substantial decrease of the count rate. These features are illustrated in Fig.6*d* through *f*, where spectra taken up after removing Cd can be compared with those shown in the first three panels, where Cd was applied.

Performing a 25 Hz experiment without Cd resulted in an efficiency enhanced by a factor 4.67, as seen in panel *d*. Turning the frequency to 12.5 Hz (panel *e*), 1000 cycles measurement time expanded to 80 s (60 s effective), with accordingly higher total number of counts, but with lower count rate, due to halving the saturation amplitude. The result was fully analogous at 6.25 Hz frequency, with a measurement time of 160 s (out of which 140 s was effective).

Systematic measurements were carried out with other U samples of 10.5, 5.5, and 2.3 g mass of 36 % enrichment, as well as a 0.53 g sample of 90 % enrichment. In Fig.6*f* the time spectrum of the latter sample is seen at 25 Hz interrogation. The ratio 0.084 of the

corresponding number of delayed neutron counts 1332 to that of 15836 of the 16 g sample is in good agreement with the ratio of the ^{235}U contents of the two samples, 0.477/5.76= 0.083.

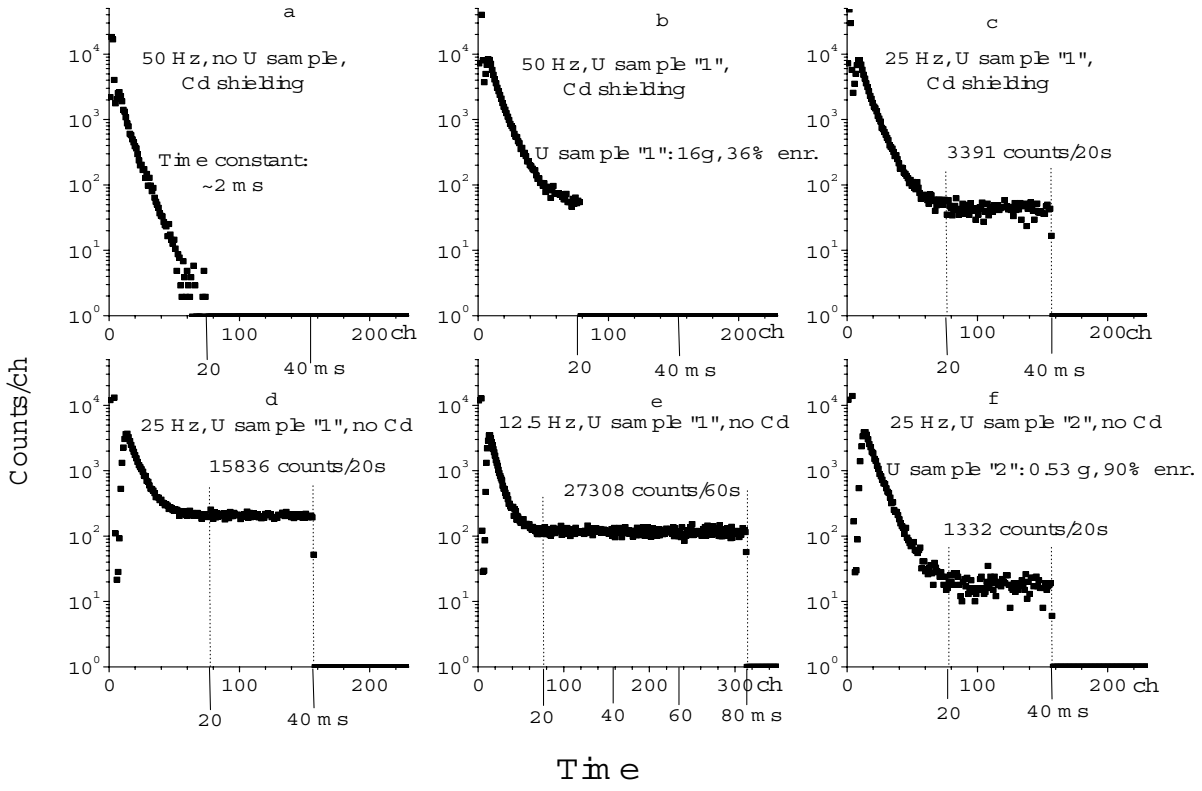

Fig.6. Time spectra taken upon interrogating U samples with Be photoneutron converter by 1000 linac pulses

5. RESPONSE TO ELECTRON CURRENT INTENSITY AND TO U CONTENT

The response of the system was studied as a function of the electron current intensity and mass of the U content. Results obtained without Cd shielding are reported here. In Fig.7*a* the count rate of delayed neutrons using Be converter is plotted as a function of electron current intensity, at various U masses as a parameter. Very similar results were obtained with heavy water, too, as displayed in Fig.7*b*. The count rates were obtained by dividing the number of counts by the effective measurement time. Note that by halving the pulse repetition rate, current intensity gets halved as well.

The same results are plotted as a function of U sample mass with Be and heavy water converter, at various mean electron current intensities, in Fig.7*c* and *d*, respectively.

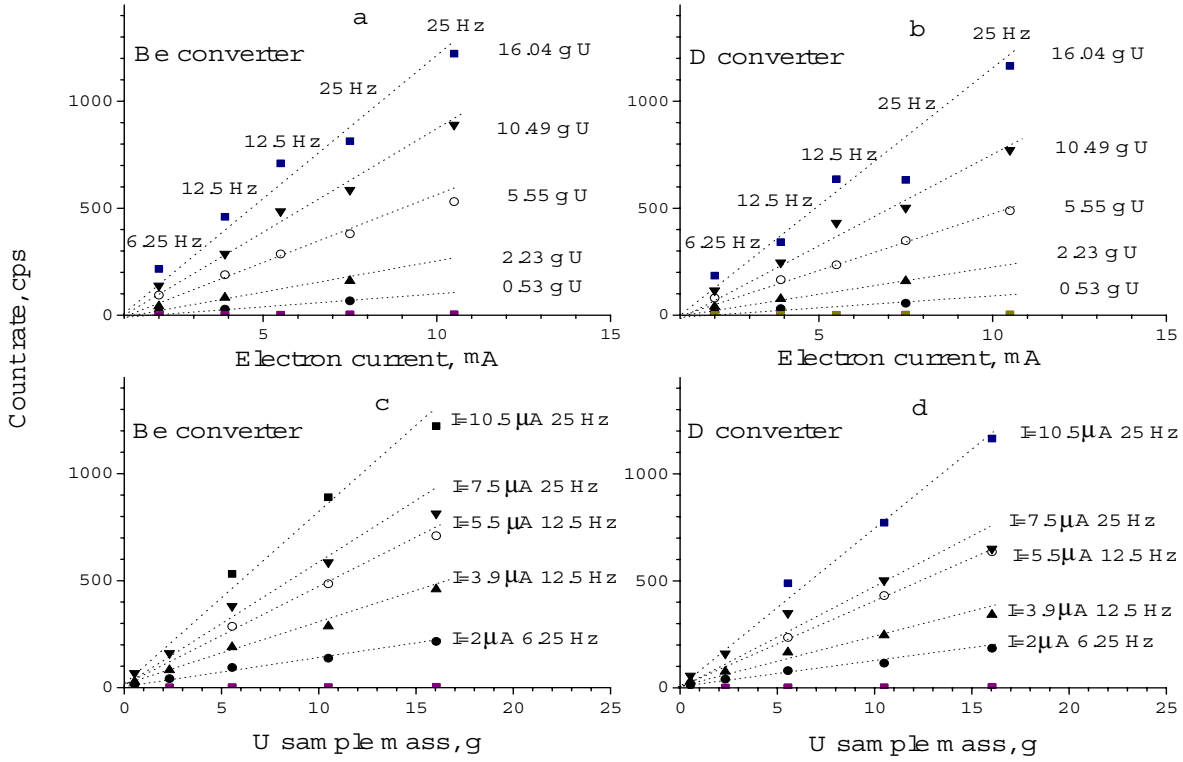

Fig.7. Count rate of delayed neutrons without Cd shielding vs. electron current, with use of Be (a) and heavy water (b). The same vs. U sample mass using Be (c) and heavy water converter (d)

6. CONCLUSIONS

As it was not possible counting delayed neutrons at 50 Hz, 25 Hz was to be chosen. Even so, half of the time interval 40 ms between pulses could only be exploited, i. e. 20 ms effective time was available for counting at this frequency.

The long pulse tail cannot be affected electronically, so it may be due to the long life time of interrogating neutrons. Nor was it possible to shorten the pulse by applying Cd shielding.

Delayed neutron signal is practically linearly related to the mean electron current intensity and also to sample mass.

A sensitivity limit of 10 mg ^{235}U was achieved at a mean electron current intensity of 10 μA in a 20 s measurement time (1000 cycles).

Interrogation time can be further reduced, if needed. While interrogating, e. g., for 250 cycles with the present system, corresponding to 10 s measurement time (+10 s initial irradiation), its capability allows about 900 counts/g ^{235}U at 10 μA current intensity. Assuming a detection limit of 1 count/s (1 cps), this corresponds to 10 mg order of magnitude ^{235}U content.

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