PHOTONEUTRON INTERROGATION OF HIGHLY ENRICHED URANIUM SAMPLES BY A 4 MeV LINAC

László Lakosi and Cong Tam Nguyen

Institute of Isotopes, Hungarian Academy of Sciences, P.O.BOX 77, Budapest, Hungary, 1525, lakosi@alpha0.iki.kfki.hu

For revealing illicit trafficking of nuclear materials, a non-destructive method using photoneutron interrogation described before was further developed. A 4 MeV linear accelerator was applied as a pulsed neutron source for active interrogation of highly enriched U. Produced in Be or heavy water by bremsstrahlung, neutrons subsequently induced fission in the samples. Delayed fission neutrons were detected by a newly designed neutron collar built up of 14 ^{3}He counters embedded in a polyethylene moderator. Significant progress was also achieved in controlling (synchronizing) a time analyzer by the electron beam pulse and 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 established in a 20 s measurement time with a reasonable amount of Be (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.

I. INTRODUCTION

A non-destructive assay (NDA) method (a "portal monitor" as an ultimate goal) has long been needed, suitable for revealing smuggled nuclear material (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. Neutrons can easily 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 used, see references in [1]. Development of a method of photoneutron interrogation was started previously as a feasibility study[1], results of which encouraged us to pursue our development efforts. So we continued using our 4 MeV LINAC as a photoneutron source, to induce fission in highly enriched uranium samples, while low enriched ones were examined previously. Distinction of fission neutrons from interrogating ones was made via time discrimination between prompt and delayed neutrons, as was earlier.

II. EXPERIMENTAL LAYOUT

The previous experimental setup[1] consisted of a neutron collar with 4 circularly arranged ^{3}He gas-filled proportional counters, embedded in a polyamide moderator. In a very similar cylindrical setup, the newly built neutron collar[2,3] consists of 14 ^{3}He tubes in a polyethylene moderator. Outer dimensions of the two concentric polyethylene cylinders are ∅20x42 cm, the diameter of the cavity (inner cylinder) for the material to be assayed is 5.5 cm. A Cd shielding on the inner side of the counter ring was also inserted, as shown in Fig. 1. Moreover, a Be photoneutron converter of a mass of 170 g was also applied, alternatively with a 100 g heavy water only used earlier.

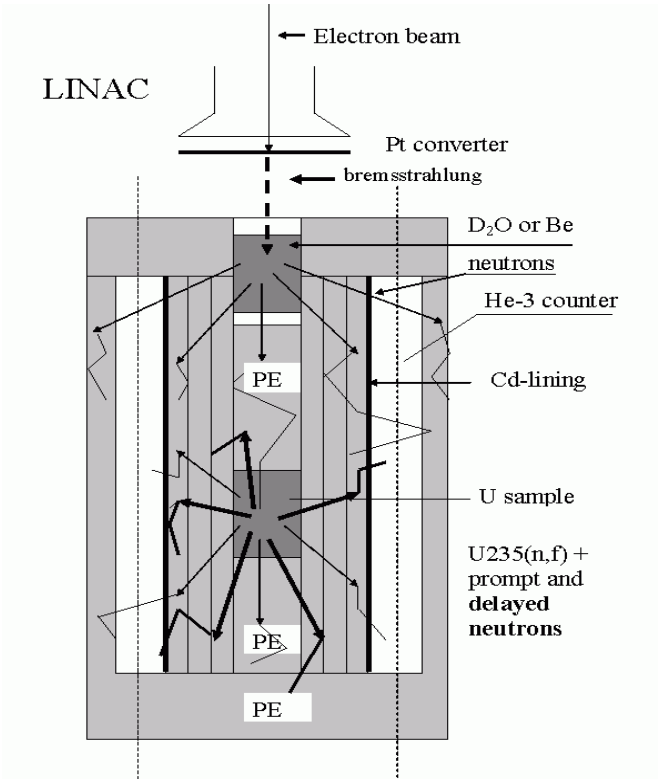

Fig.1. Experimental layout

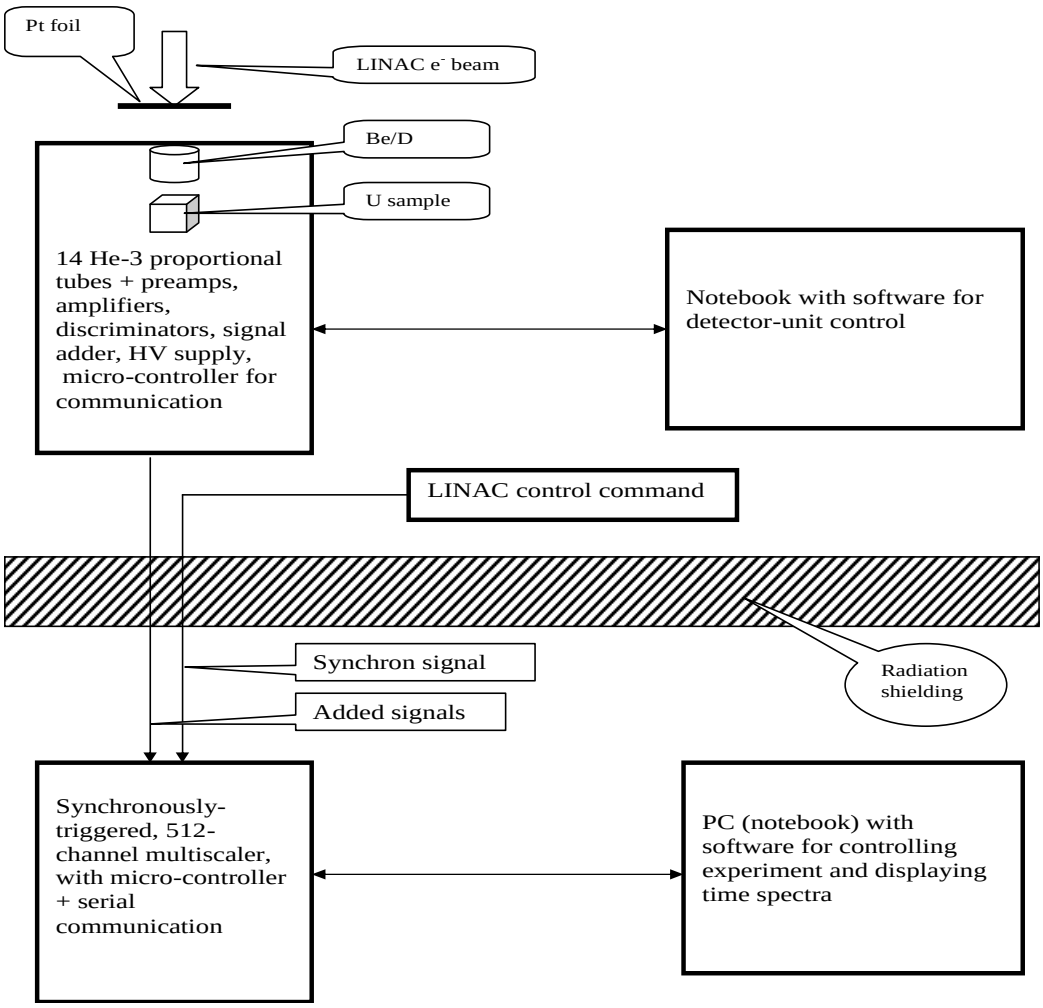

Fig.2. Schematic block diagram of signal processing

Parameters of the LINAC were the same as earlier; pulse rate of the 4 MeV energy electron beam can be 50, 25, 12.5, and 6.25 Hz. Single pulses can also be shot. The electron pulse length is 2.6 μs, with nominal amplitude of 200 mA at maximum.

The schematic block diagram of the signal processing electronics is shown in Fig. 2. It consists of a 512 channel analyzer in a 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 in a range of 25.6 μs/channel to 65.5 ms/channel. The whole time interval thus covers a period extending from the fastest 13 ms to the slowest range of about 1.6 s.

III. INTENSITY OF DELAYED NEUTRONS DURING AND AFTER 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 [4].

TABLE I. Relative yields (*R*) and intensities (*I*) of delayed neutron groups

Group	$T_{1/2}$ (s)	Rel. yield *R* (%)	Rel. intensity *I* (%)
1	0.179	2.6	23.1
2	0.496	12.8	41.1
3	2.23	40.7	29.1
4	6.0	18.8	5.0
5	21.84	21.3	1.6
6	54.51	3.8	0.11

The relative intensities are also given, obtained by multiplying the former values by the respective decay constants and normalized their sum to 100 %, according to the formula

$$I_i = \frac{R_i \ln 2}{T_{1/2_i}} \frac{100}{\sum_i \frac{R_i \ln 2}{T_{1/2_i}}} \qquad (1)$$

where R_i–s are relative yields and $T_{1/2i}$-s are the respective half-lives.

It is seen that the contribution of the three first groups predominates, i.e. the delayed neutron intensity practically goes into saturation in the first 10 - 15 s period of irradiation. Thus, after turning on the electron beam, measurements can follow after pulsing for 10-15 s.

Examining more precisely the situation, decay of the delayed neutron groups after a single pulse was calculated and is shown in Fig. 3. However, after continuous pulsing, the decay curves look somewhat different, i. e. relative intensities differ from those prevailing after a single pulse, depending on the pulse rate. Similarly, the relative contribution of various groups upon saturation depend on pulse rate again. More importantly, saturated amplitudes get successively halved, of course, whenever turning pulse frequencies 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. 4 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 about 110 times higher at 50 Hz than that of a single pulse, while still about 14 times higher at the lowest rate, 6.25 Hz.

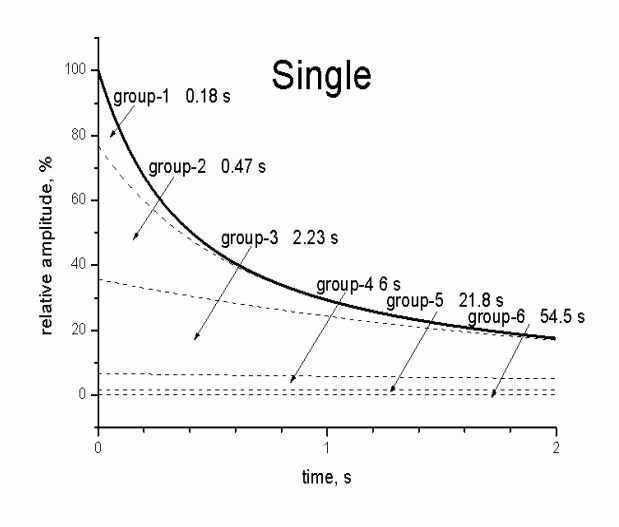

Fig.3. Decay of delayed neutron groups after a single pulse

IV. 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 converters.

Neutron pulse shapes as a function of time are seen in Fig. 5, acquired during 1000 subsequent cycles of irradiation and counting, by using Be converter. Those 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 counting-irradiation-counting cycles.

The pulse recorded at 50 Hz repetition rate without U sample is shown in the first panel. A 1000 cycle measurement at 50 Hz lasts for 20 s. Exponential decay can be observed, with a total pulse duration of about 20 ms.

In the second panel, pulse shape 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 has 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 LINAC pulses. In this way, 1000 cycles last for 40 s, as displayed in the next panel (plus an additional 10 s for initial irradiation prior to starting measurement).

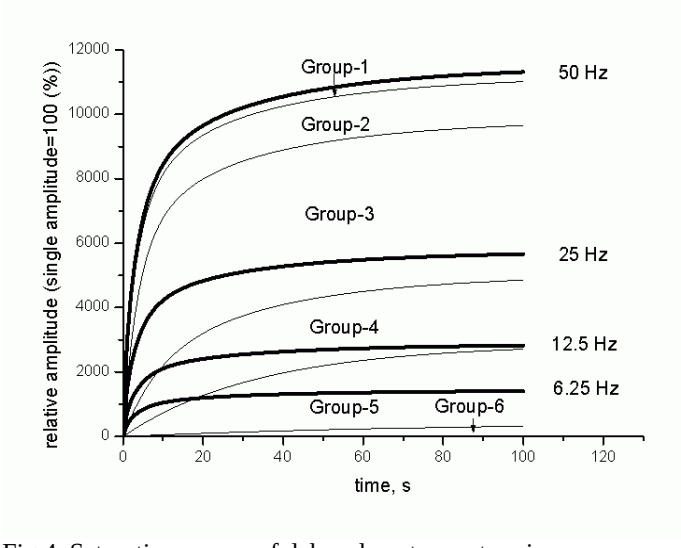

Fig.4. Saturation curves of delayed neutrons at various pulse frequencies and those of individual groups at 50 Hz

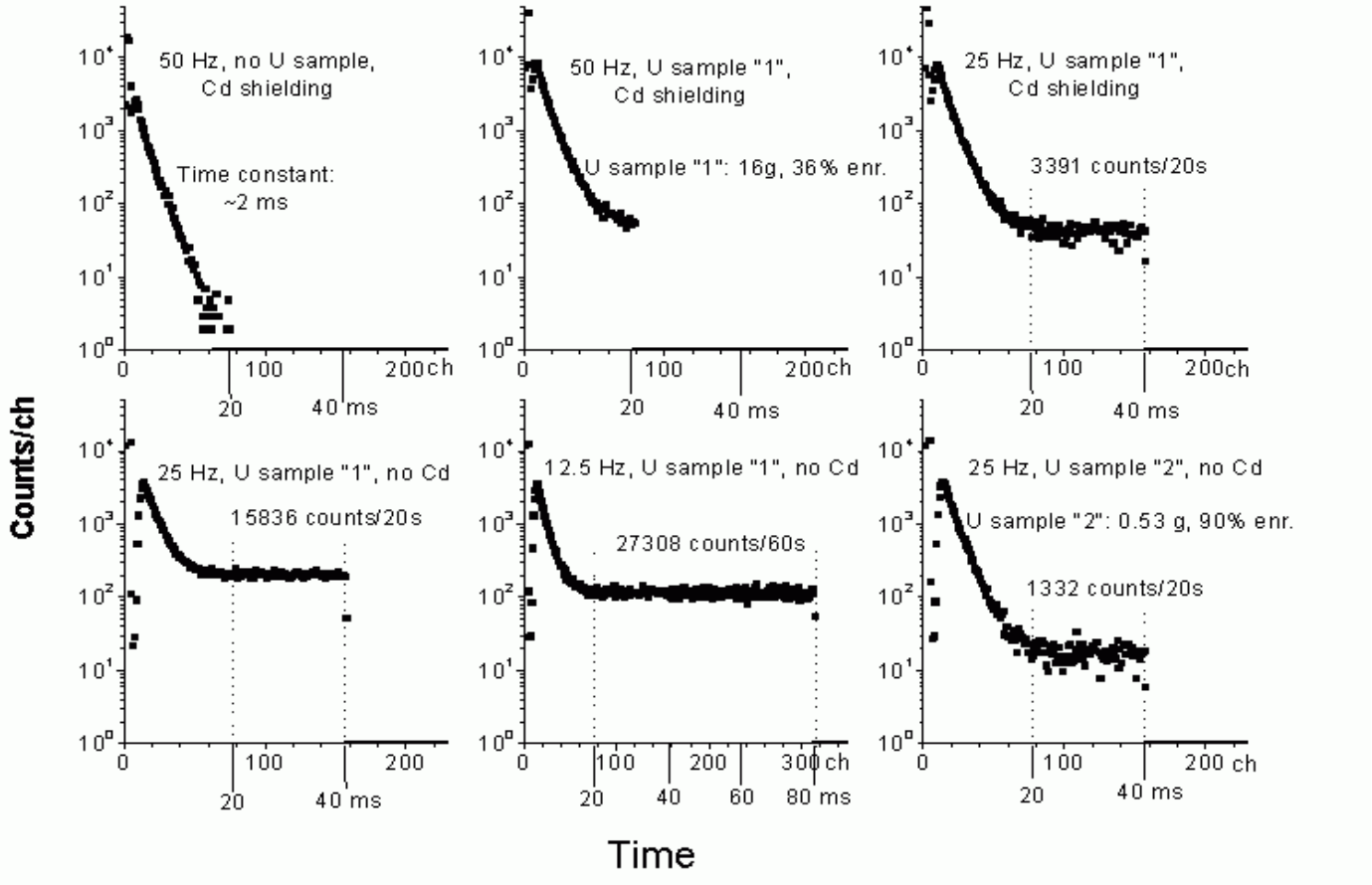

Fig.5. Neutron pulse decay curves taken with Be photoneutron converter by 1000 LINAC pulses. Time intervals for delayed neutron counting are indicated.

Such a long decay of the primary pulse was already observed in the previous experiments[1]. Determined by the material and size of the moderator, i. e. by the neutron spectrum finally, the pulse length cannot be influenced electronically, and it is practically the same for Be and for heavy water.

It has been expected[1] that applying Cd shielding on the internal side of the counter ring would diminish neutron die-away 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 the panels four through six of Fig.5, where pulses acquired 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 four. Turning the frequency to 12.5 Hz (panel five), 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 the last panel of Fig. 5 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.

By subtracting the pulse detected without a U sample, i.e. the primary interrogating neutron pulse (first panel), from that detected in the presence of the sample (second panel), the net decay curve due to fission neutrons emitted by the U sample alone was obtained, as shown in Fig. 6. In this way, by separating interrogating neutrons from

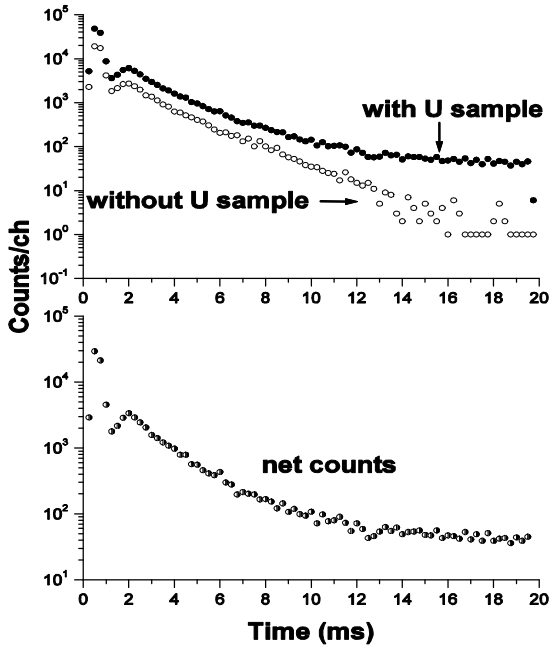

Fig.6. Neutron pulse with and without U sample (second and first panels in Fig. 5, respectively) and the net pulse due to prompt fission neutrons

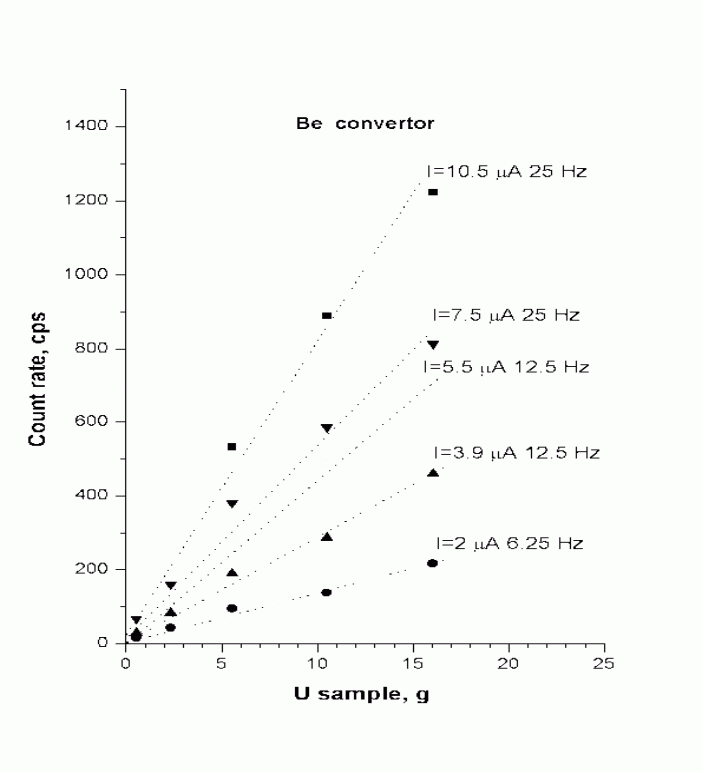

Fig.7. Count rate of delayed neutrons vs. U sample mass using Be, without Cd shielding

fission ones, prompt fission neutrons could be utilized for assay purposes as well.

Neutron life time in the medium containing the moderator can be characterized by the time constant of the exponential decay, which is 1.96 ± 0.04 ms. It is seen that time constants of interrogating and prompt fission neutrons are about the same, even though mean neutron energies of the two sources are different. It is of no surprise, because the contribution of slowing down (thermalizing) time to life time is negligible compared with diffusion time in the moderator.

V. RESPONSE TO U CONTENT AND TO THE ELECTRON CURRENT INTENSITY

The response of the system was studied as a function of the U content and electron current. Results obtained without Cd shielding are reported here. In Fig. 7 the count rate of delayed neutrons using Be converter is plotted as a function of U sample mass, at various mean electron current intensities. The count rates were obtained by dividing the number of counts by the effective time of the measurement. Note that by halving the pulse repetition rate, current intensity gets halved as well. Very similar results obtained with heavy water are displayed in Fig.8.

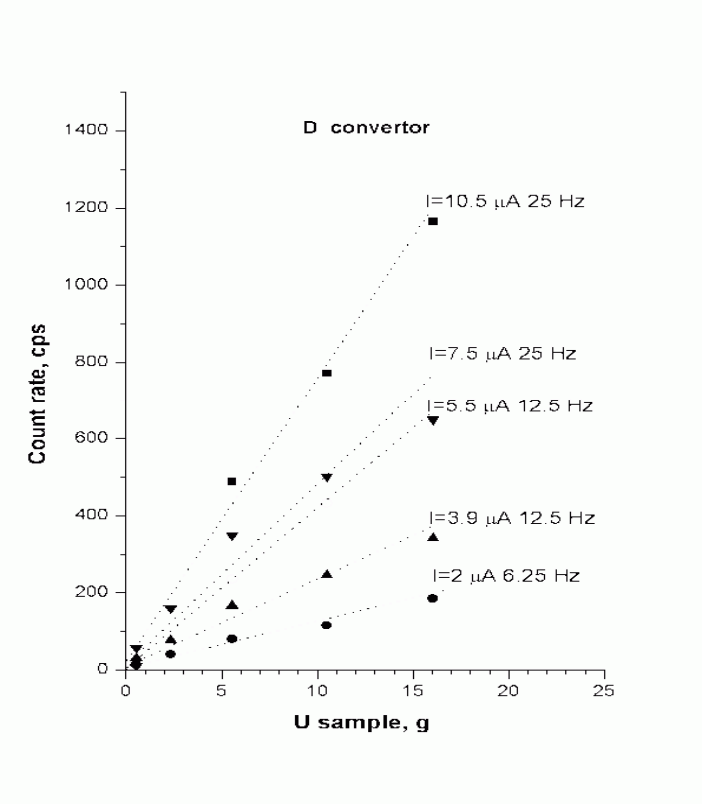

Fig.8. As the previous one, using heavy water converter

The same results are plotted as a function of electron current intensity in Figs 9 and 10, at various U masses as a parameter, for Be and heavy water, respectively.

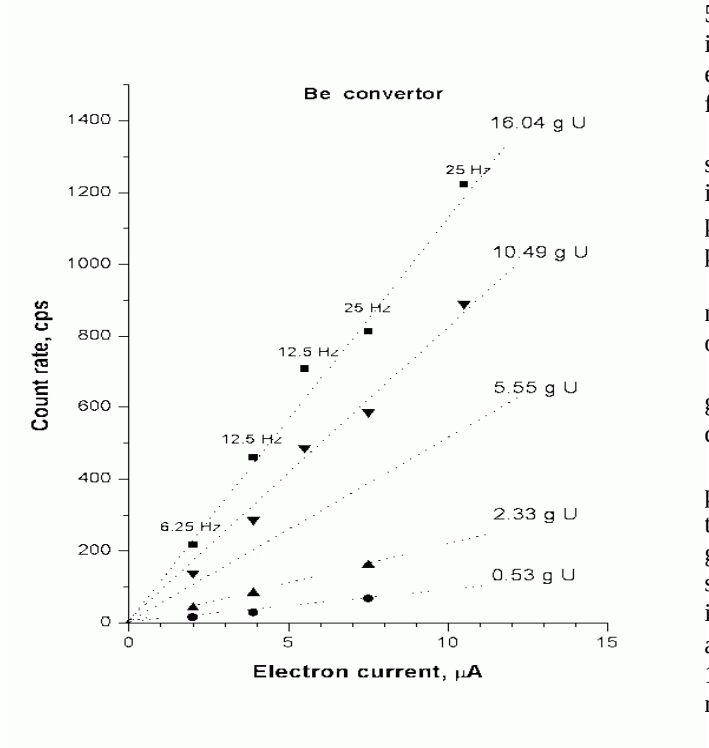

Fig.9. Count rate of delayed neutrons vs. electron current, using Be, without Cd shielding

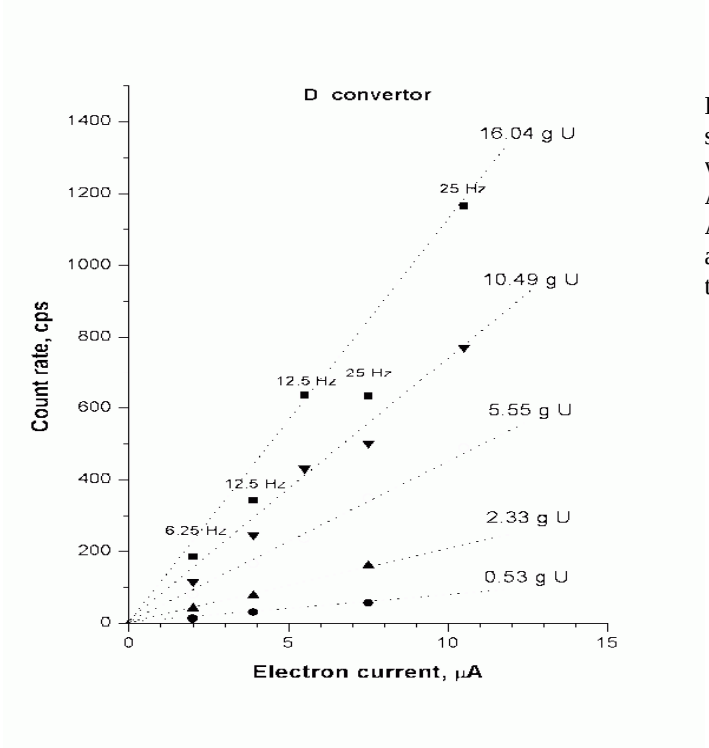

Fig.10. As the previous one, using heavy water converter

VI. 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 die-away time of interrogating neutrons. Nor was it possible to shorten the pulse by applying Cd shielding, as was expected and proposed earlier.

Practically linear relationships describe delayed neutron signal as a function of the sample mass and also of current intensity.

In the previous experiments[1] a sensitivity limit as 0.5 g ^{235}U was achieved in a 20 s measurement time (500 cycles) at 2 μA mean current intensity.

By comparing this with the results reported here, the present equipment has a detection sensitivity of about twice an order of magnitude higher than the earlier one. E. g., while interrogating for 250 cycles with the present system, corresponding to 10 s measurement (+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 0.01 g order of magnitude ^{235}U content.

ACKNOWLEDGMENTS

Thanks are due to Mr. P. Hargittai for running the LINAC, to Mr. E. Serf for designing the electronic control system and for his help in the measurements. The work was supported by the Hungarian Atomic Energy Authority under contracts No. OAH-ABA-76/00, OAH-ÁNI-ABA-05/01, -05/02, and -03/06. Also, one of the authors (L. L.) is grateful for the grant provided to him by the International Atomic Energy Agency.

REFERENCES

1. L. LAKOSI, C. TAM NGUYEN, J. BAGI, Photo-neutron Interrogation of Uranium Samples by a 4 MeV LINAC. A Feasibility Study. *Utilization of Accelerators*, Proc. Int. Conf. Dubrovnik, 5-9 June 2005, IAEA-CN-115-64, IAEA, Vienna (2006) CD-ROM.
2. J. BAGI, C. TAM NGUYEN, L. LAKOSI, Assessment of the Pu content of Pu-Be neutron sources, *Nucl. Instr. Meth.* **B 222**, 242 (2004).
3. L. LAKOSI, J. BAGI, C. TAM NGUYEN, Quantitative non-destructive assay of PuBe neutron sources. *Nucl. Instr. Meth.* **B 243,** 385 (2006).
4. G. R. KEEPIN, et al., *Phys. Rev*. **107** 1044 (1957); G. R. KEEPIN, *Physics of Nuclear Kinetics*, p.101, Addison-Wesley, Reading, Mass. (1965).