PROMPT NEUTRON CAPTURE GAMMA RAY ANALYSIS OF NUCLEAR FUEL MATERIALS WITH A PAIR SPECTROMETER*

B.R.S. Pecequilo, G.C. Vandenput**, C.R.S. Stopa*** and A.A. Suarez
Instituto de Pesquisas Energéticas e Nucleares
C.P. 11049 - Pinheiros, 01000 São Paulo, S.P. - Brasil

ABSTRACT

Prompt gamma rays following thermal neutron capture are used to determine fuel material impurities in a reactor internal geometry. In order to increase the detection sensitivity for weak lines and simplify the analysis of the spectra, a 3-crystal coincidence spectrometer was mounted and the gain in peak-to-background ratio calculated for several spectra was about a factor of 6. The calibration of the experimental arrangement was performed with UO_2 targets contamined by controlled amounts of Gd and Sm.

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** Now at SCK/CEN B-2400 MOL, Belgium.
*** Now at CTA, São José dos Campos, Brazil.

INTRODUCTION

Man's interest in atomic power has increased with the world power crisis.Thus, special care has been devoted to the improvement of techniques of purification and analysis of the nuclear fuel materials. Some typical contaminants of those materials are Al, B, Be, Cd, Ni, W, V and rare earths like Gd, Sm, Dy and Eu[1].

There exist already several techniques which are quite sensitive for the determination of trace impurities present in nuclear materials, however some of them loose precision when measuring quantities greater than 30 ppm. Spectrographic analysis have high sensitivity only for trace element analysis and needs a previous chemical separation. Neutron activation analysis relies on the properties of the product nucleus and in numerous cases the product nucleus may have a short half-life, a long half-life or a stable composition or it may not be a gamma emitter at all. In the specific case of nuclear materials (like uranium, for example) the neutron activation analysis of the contaminants is difficulted by the production of fission fragments in the matrix.

In view of the increasing interest and necessities of new techniques for determinations of impurities in nuclear fuel materials, like uranium compounds, we developed and presented in previous papers[2,3] an alternative method of analysis of fuel material impurities using prompt gamma rays following thermal neutron capture.

Prompt gamma-ray spectrometry offers advantages over many of other techniques in that it provides a relatively rapid and nondestructive trace and major multielement analysis requiring a minimum of physical preparation and no chemical processing.

Prompt gamma-rays are typical of each isotope. Thus, the analysis of gamma-ray spectra obtained for a certain sample permits the identification of the contaminants of the sample.

The main advantage of this technique proceeds from the relatively low neutron binding energy(~ 4.8 MeV) of nuclear fuel materials which allows all high energy capture gamma-rays belonging to the impurities to be detected in relatively low background conditions.

We showed the viability of this method by measuring with a single coaxial Ge(Li) detector samples of uranium oxide(UO_2) with controlled amounts of rare-earth impurities and the prompt high energy gamma rays of these elements were easily seen since above 4.8 MeV there are no more prompt gamma rays emitted by the uranium matrix[2,3].

However, neutron capture γ-ray spectra of isotopes in the medium and heavy mass region, obtained with Ge(Li) detectors, tend to become very complex due to characteristically high level densities of the product nuclei. They are further complicated by the presence of single and double escape peaks, apart from the one corresponding to full energy absorption, for every gamma ray detected with energy E_γ>1022 keV and unresolved multiplets are therefore a common feature.

Moreover, Compton scattering and subsequent escape of scattered gamma-rays from the detector are the source of an important continuous background distribution on which lower energy peaks are superposed.

This complicates the analysis of the spectra, limits the detection sensitivity for weak lines and reduce the precision of peak area estimates.

Nevertheless, greatly simplified and significantly "cleaner" spectra can be obtained by a slight sofistication of the detection system, namely by introducing a three-crystal coincidence technique. The double escape peaks result from the (e^+-e^-) pair production in the detector followed by positron annihilation and the subsequent escape from detection of both 511 keV γ-rays produced in the process. If this annihilation radiation is allowed to strike two additional detectors surrounding the central Ge(Li) crystal, then single escape and full energy peaks can be eliminated by imposing triple time coincidence. The only peaks retained in the spectra result from double escape and, it should be emphasized, are free of Compton distributions.

The pair spectrometer used in our measurements consists of a Ge(Li) detector and two optically separated NaI(Tl) crystals. With this system we measured several UO_2 samples with controlled samarium (Sm_2O_3) and gadolinium (Gd_2O_3) impurities and we observed that although the coincidence requirement drastically reduces the detection efficiency, a very substancial gain in peak-to-background ratio is obtained, so we could detect lower intensity peaks than in the single mode and the peak area estimates also became more accurate.

EXPERIMENTAL FACILITIES AND TARGETS

The experimental set up for this experiment is installed at the tangential tube of the IEA-R1 reactor of São Paulo and the basic features of our internal target facility are illustrated in fig. 1.

The internal target geometry is definitely superior to the external one in sensibility (higher signal-to-background ratio). In the external target geometry there are two solid angles: one for the neutrons at the target and other for the gamma rays at the detector, whereas in the internal geometry there is only one solid angle from the target to the detector. So, despite the difficulty in handling the samples we choose the internal target geometry.

The samples to be analysed are positioned close to the reactor core and the neutron flux is typically about $5 \times 10^{11} cm^{-2}.s^{-1}$ at the target position.

The γ-beam is collimated by a set of internal graded lead collimators and a 4mm diameter final collimator focusing on the compensated annular area of a Ge(Li) crystal situated outside of the reactor biological shield at a distance of 654cm from the target. In this conditions, the solid angle seen by the Ge(Li) detector is 1.03×10^{-7}sr.

A $^6Li_2CO_3$ thermal neutron absorber is installed in front of the Ge(Li) detector to protect it against probable thermal neutron existing in the γ-beam.

There are also some other aspects observed in order to reduce the background:

a - The lead collimators inside the aluminium tangential tube of the reactor are conical behind the target in order to prevent that no radiation from the beam tube wall close the core reach the detector directly.

b - The reactor beam tube was covered with a nuclear pure graphite tube of 3 7/8" internal diameter on the whole inner surface where there are no collimators. Since gamma rays from the aluminium tube can be scattered at the target, we covered the wall with graphite which it is a material emitting very few thermal neutron capture gamma rays.

c - The reactor beam tube was evacuated by a rotary pump to less than 10^{-3}torr in order to decrease the scattered neutron to the Ge(Li) spectrometer as well the capture gamma rays from the nitrogen in the air.

d - The target holder was of nuclear pure graphite due to its low capture cross section and rather simple gamma spectrum.

The calibration of the experimental arrangement was performed with UO_2 targets contamined by controlled amounts of Gd and Sm. The total weight of the samples utilized was around 5 grams.

THE PAIR SPECTROMETER

The pair spectrometer developed for our measurements consists of a 42.5cc true coaxial Ge(Li) detector which fits into a cylindrical opening in the common housing of two optically separated 6" x 6" NaI(Tl) crystals facing the central Ge-crystal from opposite sides (Fig. 2). The electronics also showed in fig.2 comprise standard coincidence hardware. The fast output signals from three linear amplifiers trigger 3 timing single channel analyzers and are subsequently fed into a fast coincidence module operating with a time resolution $2\tau \sim 100$ns.

The coincident events generate a gate pulse for a gated biased amplifier (for choosing high energy ranges) allowing the linear signal from the unipolar output of the Ge(Li) amplifier to be processed. The selection of 511 keV annihilation radiation in the scintillators is accomplished by a proper adjustment of the discriminator levels at the single channel analyzer. By using analog stabilization the variation of photomultiplier and amplifier gain are offset. The energy signal from the Ge(Li) detector is digitally stabilized at the ADC level and this is essential for maintaining energy resolutions of ~7.0 keV at 4.1 MeV during long measuring periods.

RESULTS AND DISCUSSION

The data analysis of the γ-ray spectra was done by using the GAUSS V program[4] in operation in our computer center. In the program, the peak is represented by a Gaussian function and the underlying spectral background by a linear function. The basic mathematical operation is the nonlinear least-squares fit by which the parameters of the Gaussian (height, width and position) are determined. For the energy calibration we used the quite well known capture gamma rays of nitrogen[5] (for more details, see reference[6]).

Some of the UO_2 targets with rare earth impurities was measured even in the single and in the coincidence mode and the direct and pair spectra obtained are showed in figs. 3 and 4.

We observed that although the coincidence requirement reduces the detection efficiency by a factor of about 4.5 in comparison with the single mode, the gain in peak-to-background ratio calculated for several spectra is about a factor of 6. So, we obtained a very substantial decrease in the background and we could detect lower intensity peaks.

For example in fig. 3 some peaks of lower intensity like the double escape of the 6670 keV(I_γ = 0.08%) and 6913.4 keV(I_γ = 0.07%) γ -transition of ^{158}Gd are more proeminent in the pair spectrum. Also, the 6913 DE peak is no more overlapped by the 5902.9 keV γ transition, visible only in the direct spectrum.

Moreover, in fig. 4 the single escape of the 6750 keV γ transition of ^{158}Gd in the direct spectrum difficults the double escape peak of the 7283 keV γ transition of the ^{156}Gd. However, in the pair spectrum the 7283 keV DE peak is quite "clear", and the spectral background is also lower. It's interesting to point that the detection probability of the more intense γ-transition of the

^{156}Gd(7283 keV) is lower by a factor of 14 comparing with that one of the more intense γ-transition of the ^{158}Gd(6750 keV), i.e, σ = 61000 barns for ^{155}Gd and I_γ = 0.4% for ^{156}Gd and σ = 250000 barns for ^{157}Gd and I_γ = 1.32% for ^{158}Gd.

The peak-to-background ratio gain calculated for several spectra was about a factor of 6, but in the lower energy region this gain is more significant. In those regions, the background radiation is increased by Compton effect in higher energies and the peak-to-background ratio in the direct spectrum is lower than 1. As an example, for the double escape peak of the 4060 keV γ-transition of the ^{239}U we have a 0.58 ratio in the direct spectrum and a 9.19 ratio in the pair one. So, in the lower energy region the gain in identifying weak transitions is about a factor of 15.

With the UO_2 targets with controlled rare earth impurities of Sm and Gd we also have determined calibration curves of the experimental system. The impurity content is proportional to the peak area and is expressed in ppm(ratio of micrograms of impurity to grams of uranium oxide).

The calibration curves are presented at fig. 5. The impurity content in the uranium oxide expressed in ppm plotted against the peak area ratio of impurity to uranium (strongest transitions) results in a straight line which has a slope proportional to the sensibility of the arrangement to that particular impurity.

The reproducibility of this method of analysis using prompt neutron capture gamma-rays was checked previously and the results are presented in previous papers[2,3]. The observed deviation was less than 2%.

The multielement capability of analysis, the rather convenient sensibility for impurities contents greather than 20 ppm and the gain obtained in peak-to-background ratio by using a three-crystal pair spectrometer permits to say that this method of analysis acts as a complementary method to the existing techniques.

Also, we would like to point out that the analysis of impurities using radioactive thermal neutron capture could be used not only for the nuclear fuel materials, but also for biological and geological applications.

ACKNOWLEDGEMENT

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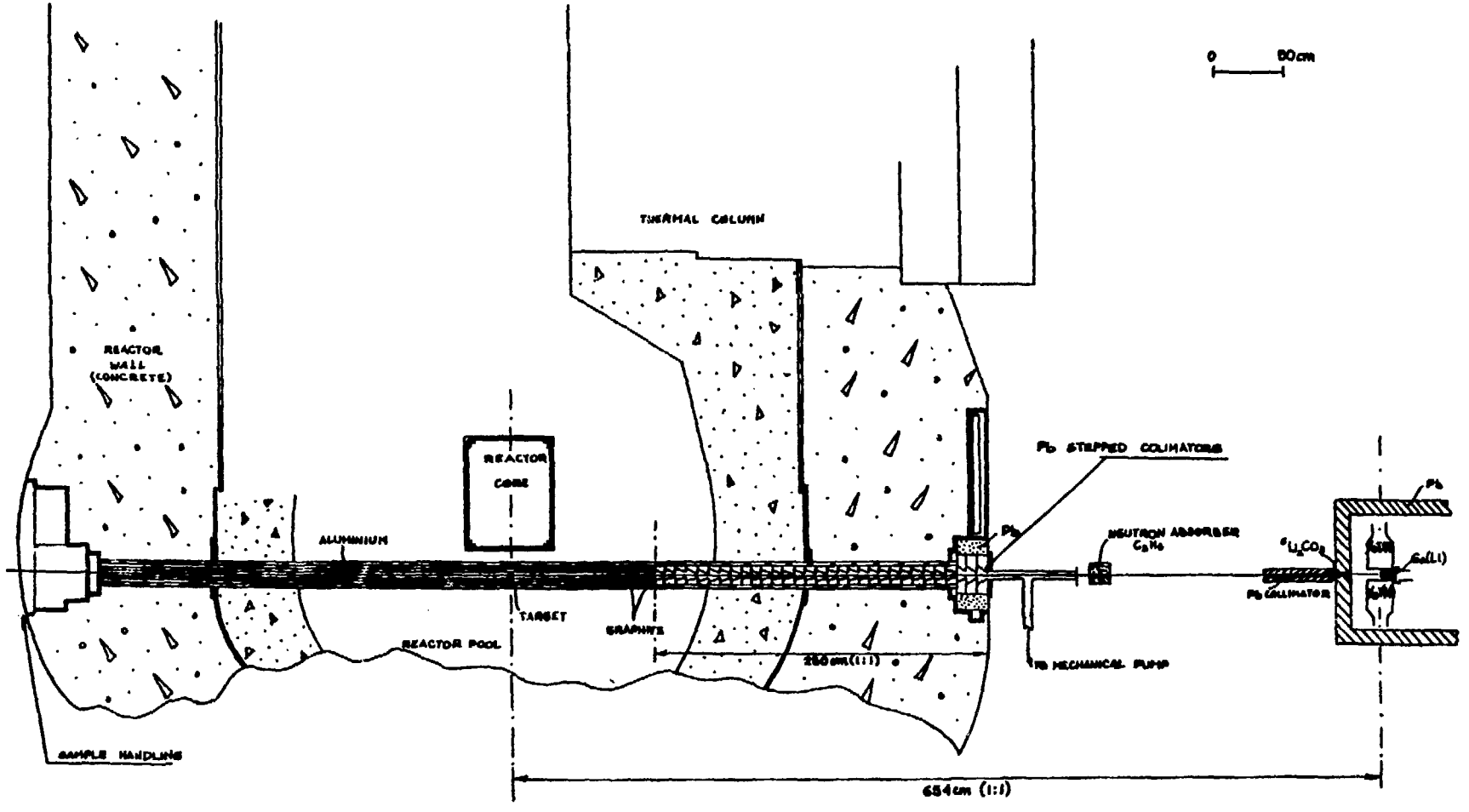
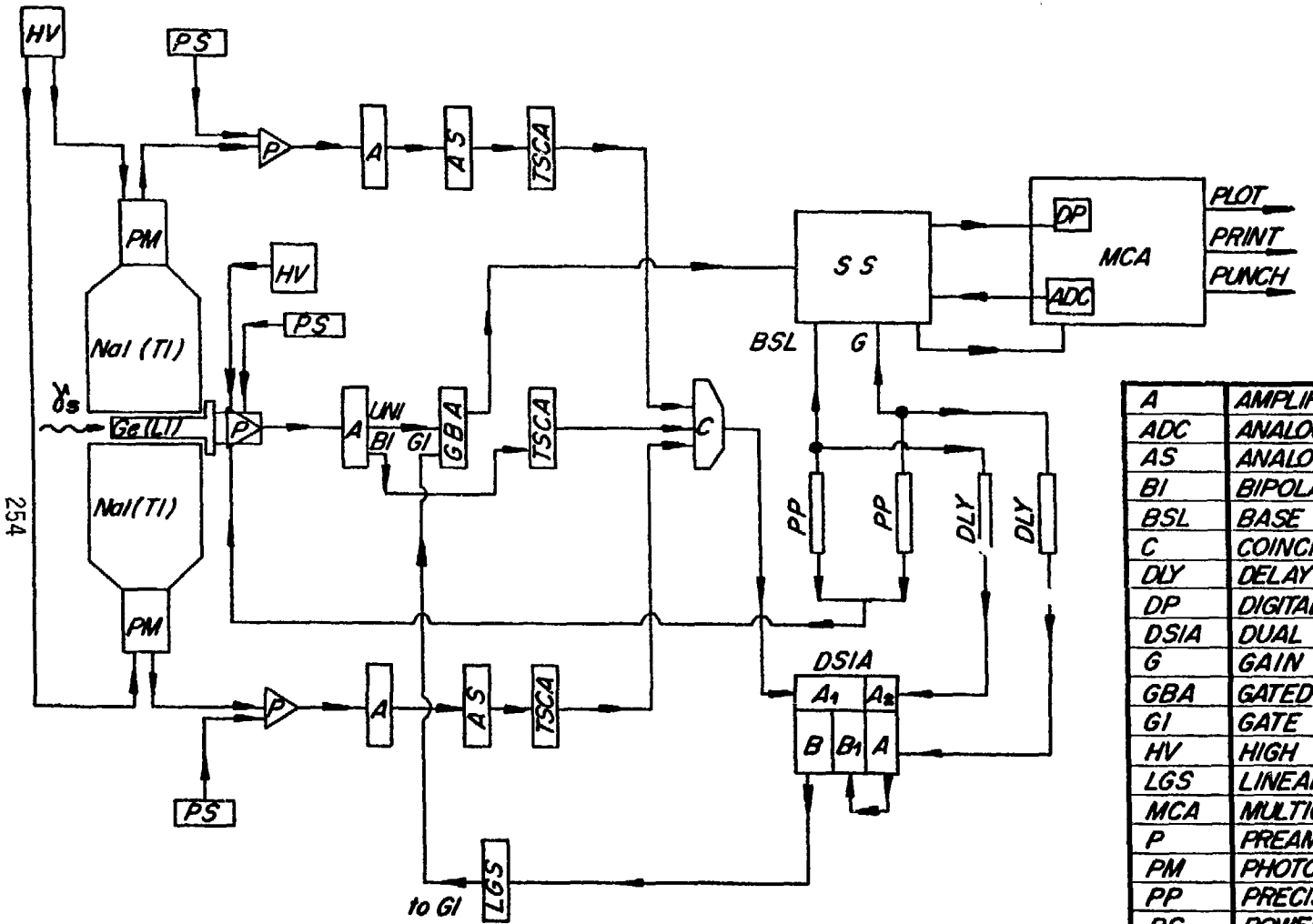


FIG. 1 - Experimental arrangement installed at the tangencial beam tube of the IEA-R1 reactor



A	AMPLIFIER
ADC	ANALOG-TO-DIGITAL CONVERTER
AS	ANALOG STABILIZER
BI	BIPOLAR
BSL	BASE LINE
C	COINCIDENCE
DLY	DELAY
DP	DIGITAL PROCESSOR
DSIA	DUAL SUM INVERT AMPLIFIER
G	GAIN
GBA	GATED BIASED AMPLIFIER
GI	GATE IN
HV	HIGH VOLTAGE
LGS	LINEAR GATE STRETCHER
MCA	MULTICHANNEL ANALYZER
P	PREAMPLIFIER
PM	PHOTOMULTIPLIER
PP	PRECISION PULSER
PS	POWER SUPPLY
SS	SPECTRUM STABILIZER (DIGITAL)
TSCA	TIMING SINGLE CHANNEL ANALYZER
UNI	UNIPOLAR

FIG.2 - Electronics of the 3-crystal pair spectrometer

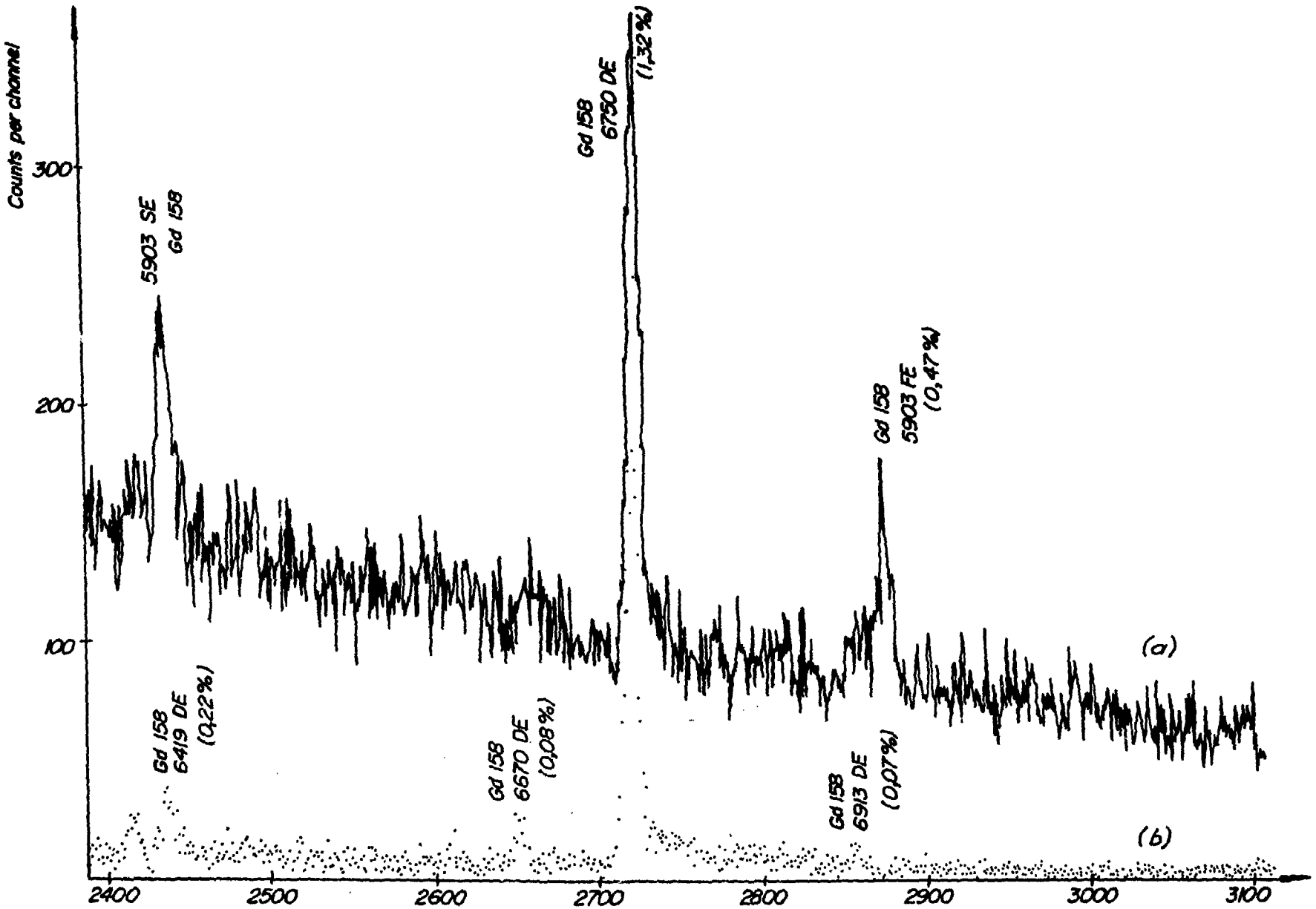


FIG. 3 - Prompt γ-ray spectrum of the UO_2+ 48ppm Gd sample in the region of 5300 to 6200 kev. FE, SE and DE are full, single and double-escape peaks. a) direct spectrum, run of 13 hs. b) pair spectrum, run of 34 hs.

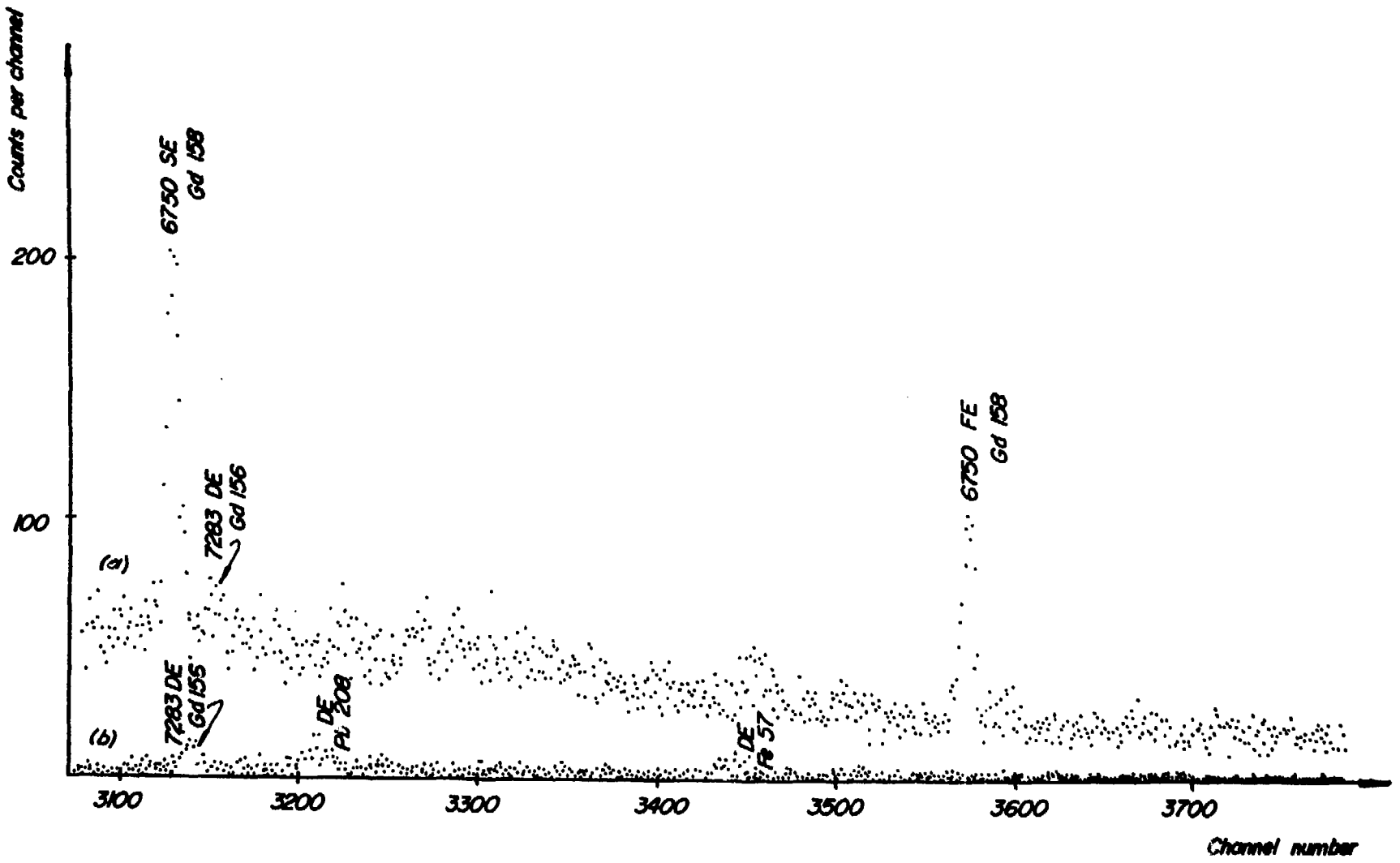


FIG 4 - Prompt γ-ray spectrum of the UO_2 + 48ppm Gd sample in the region of 6200 to 7000 kev. The iron and lead impurities are from the target holder.

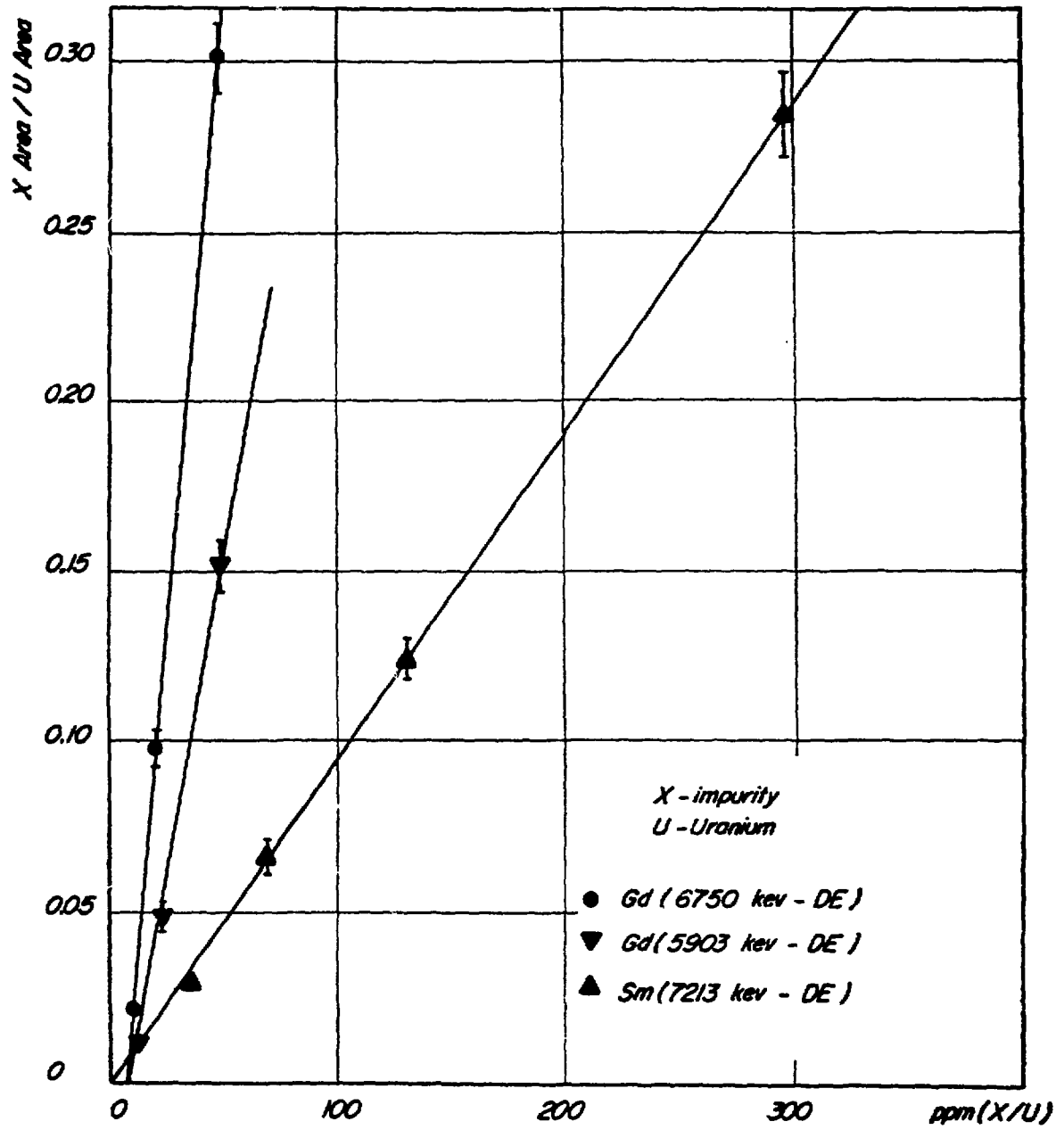


FIG. 5 - Calibration curves of the experimental arrangement with the pair spectrometer

DE = double - escape peak