REPORT NO. IAEA-R-1893-F

TITLE

Determination of contaminants in nuclear materials by measuring the capture gamma rays of thermal neutrons in a reactor internal geometry

FINAL REPORT FOR THE PERIOD

1976-11-01 - 1979-06-30

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DATE January 1980

FINAL REPORT

CONTRACT NUMBER: 1893/R1/RB

PROJECT: DETERMINATION OF CONTAMINANTS IN NUCLEAR MATERIALS BY MEASURING THE CAPTURE GAMMA RAYS OF THERMAL NEUTRONS IN A REACTOR INTERNAL GEOMETRY.

INSTITUTO DE PESQUISAS ENERGÉTICAS E NUCLEARES

CHIEF SCIENTIFIC INVESTIGATOR: DR. ACHILLES A. SUAREZ

ASSOCIATE INVESTIGATOR: DRA. BRIGITTE R.S.PECEQUILO

TIME PERIOD COVERED: DECEMBER 1976 TO DECEMBER 1979.

DESCRIPTION OF RESEARCH CARRIED OUT:

a) Objective of the Research

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

The objective of this research was to search a new method of analysis of fuel material impurities using prompt gamma rays following thermal neutron capture.

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

To show this we have measured samples of 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 emited by the uranium matrix[2,3].

b) Experimental Arrangement

The experimental set up for this experiment was previously installed in the lower tangential irradiation tube of the IEA-R1 reactor of S.Paulo (fig. 1).

It was choosed that geometry with internal target because the signal to background ratio in this situation is higher than with external geometry. In this way the sample to be analysed is positioned close to the reactor core and is seen by a single Ge(Li) spectrometer placed outside of the reactor biological shielding.

The gamma rays resulting from the target are collimated by a saw shaped collimator internal to the reactor shielding and a final 0.3 cm diameter colimator close to the detector. The resulting solid angle is of the order of 4.8×10^{-9} sr. The target area seen by this collimator system is a circle of 3.5 cm diameter and the collimation is made so that the internal walls of the reactor tube are not seen by the Ge(Li) spectrometer.

In order to decrease even more the possible background coming from the reactor tube, we lined internally it with a graphite pipe of 4" internal diameter and a wall thickness of 1/2". The tube is also evacuated by a rotary pump to less than 10^{-3} torr so that scattered neutrons be not directed to the Ge(Li) spectrometer neither capture gamma rays from the nitrogen present in the air.

To filter thermal neutrons which eventually could reach the Ge(Li) detector, a $^{6}Li_2CO_3$ filter of $1.4 g/cm^2$ is placed in the gamma beam. The transmission of this filter for the 4060 keV transition of ^{239}U is about 97%.

To choose the best position for the target, measurements were made of the neutron flux as well of the signal to background ratio along the axis of the through tube. The best position coincided with the position of higher thermal neutron flux which was $3.7 \times 10^{12} n/cm^2.s$.

As neutron source we used the 2Mw IEA-R1 research reactor of S.Paulo and the gamma rays were detected either with a single Ge(Li) spectrometer or with a pair spectrometer composed by a 42.5cc true coaxial Ge(Li) detector and two optically separated 6"x6" NaI(Tl) crystals.

Until the beginning of the reform of our reactor it was possible to complete some measurements with the Ge(Li) single crystal spectrometer which ended with the conclusion of the doctor thesis of one of our colaborators B.R.S.Pecequilo and which was enclosed as Apendix of a previous Progress Report of the Project. In that thesis it was observed the general limitations of the actual technique utilizing the single crystal spectrometer and some of that data was used in the paper "Fuel Material Analysis using Radioactive Thermal Neutron Capture"(3).

It was verified however that the present technique could be of no help for the determination of trace quantities of low cross section elements in the presence of poisons of a resonable cross section and with intense high energy gamma lines. So, in order to simplify the spectra and to decrease the background we installed a pair spectrometer in order to try to increase the sensitivity and at the same time to simplify the data analysis.

During the period of December 1977 to September 1978 the Reactor IEA-R1 was in reform. All the ceramics of the swimming pool was substituted by an inox stainless steel cover and some of the irradiation tubes were eliminated. According to the Nuclear Regulatory Commission Recomendation one of the through tubes (the lower one) of the IEA-R1 reactor, which was lower than the middle level of the reactor core should be eliminated.

For this reason, our experiment had to be removed from that through tube and re-installed, after the reform, in the upper one. Several of the experimental set-up had to be modified:the final colimator as well as the detector support and shielding.

Due to this new configuration the neutron flux in the target position decreased from $3.7 \times 10^{12} n/cm^2.s$ to $5.0 \times 10^{11} n/cm^2.s$ (fig. 2).

The signal to background ratio with the pair spectrometer was improved in average by a factor of six(6) in respect to the single crystal spectrometer. The pair spectrometer installed is composed of two NaI crystals 6HSW8/(2)5A1 , 6"x12" crystals from the firma Bicron and the same Ge(Li) detector(42.5cc) utilized in the previous experiments. They were installed at a distance of 654cm from the internal target.

Due to the fact that our multichannel is rather old(Hewlett Packard-out of line) we have had several problems with it, delaying the measurements several times. The stabilization of all the spectrum was also a problem for us during the measurements mainly due to temperature fluctuations of the reactor hall.

The electronics comprise standard coincidence hardware (fig. 3). Fast output signals from 3 linear amplifiers trigger timing single channel analysers and are subsequently fed into a fast coincidence module operating with time resolution 2τ ~100ns. Coincident events generate a gate pulse for a gated biased amplifier allowing the linear signal from the unipolar output of the Ge(Li) amplifier to be processed. Selection of 511 keV annihilation radiation in the scintillators is acomplished by a proper adjustment of discriminator levels. Variations of photomultiplier and amplifier gain are offset by analog stabilization. The energy signal from the Ge(Li) detector is digitally stabilized at the ADC level in order to maintain energy resolutions of ~7 keV at 4.1 MeV during long measuring periods.

c) Targets

The samples to be analysed were placed in a container of nuclear pure graphite due to its low capture cross section and quite simple spectrum.

That container had a wall thickness of less than 0.1 cm and were installed close to the reactor core by means of an aluminium rod which is removed afterwards. Actually the containers permits to handle targets of 2 cm diameter x 0.3 cm thick. The attenuation of neutrons in the target is less than 1.3% in this arrangement.

The targets were prepared mixing UO_2 or ThO_2 with the proper contaminants with a mortar and pistil of agate. The uniformity of the samples were checked by analysing it with a scanning electron microscope from the firm Johnson Mattey & Co. Ltd., USA.

d) Results

Some exploratory measurements were done with samples of UO_2/ThO_2 contamined with Dy, Gd, Sm, Eu, V, Fe, Mn, Ti, Ni and Hg. The total weight of the samples utilized were around 4g. Examples of the spectra obtained with some samples were shown in the Ph.D. thesis of Dr. B.R.S.Pecequilo enclosed as Apendix in another report submited to AIEA. Some of these spectra utilizing the pair spectrometer are contained in the paper(reference 4) "Prompt neutron capture gamma ray analysis of nuclear fuel materials with a pair spectrometer" to be presented in the IV International Conference on Nuclear Methods in Environmental and Energy Research to be held in Columbia-USA April 14-17, 1980(The abstract of that publication is enclosed in the summary of 500 words).

The data analysis were performed by using the GAUSS V program develloped by R.G.Helmer from the Nuclear Reactor Testing Station of Idaho Nuclear Corpo ration which is in operation in our computer center.

The energy calibration of our system was done by putting nitrogen gas in the reactor tube and using the capture gamma lines of it. With them, a non--linearity table of the system was constructed so that more precise energy determinations could be done. The areas of the measured peaks were then auto matically evaluated by the computer code and a relative efficiency curve for the spectrometer was constructed.

With the installation of the pair spectrometer the obtained resolution of the system in the routine measurements were around 7 and 9 keV for 4 and 6.5 MeV gamma rays respectively.

Unfortunatelly the reduction of the neutron flux in the upper through tube decreased the possibility for minor component analysis in spite of the fact that in the new configuration it was possible to increase the solid angle seen by the Ge(Li) detector from 4.8×10^{-9}sr to 1.03×10^{-7}sr giving an increase in the product $\phi\Omega$ of 2.9 and an improvement in the signal to background ratio of a factor of 6.

References

1- "Analytical Methods in the Nuclear Fuel Cycle" - Proc.Symposium Vienna, 1971. IAEA, Vienna, 1972, p.137.

2- B.R.S. Pecequilo, Ph.D.Thesis, IEA-DT-078(1978).

3- "Fuel material analysis using radioactive thermal neutron capture",B. R.S.Pecequilo and A.A.Suarez in "Neutron Capture Gamma-Ray Spectroscopy" ed. by R.E.Chrien and W.R.Kane, Plenum Press, 1979 p.710--13.

4- "Prompt neutron capture gamma ray analysis of nuclear fuel materials with a pair spectrometer", B.R.S.Pecequilo, G.C.Vandenput, C.K.S. Stopa, A.A. Suarez, to be presented in the IVth. International Conference on Nuclear Methods in Environmental and Energy Research (to be held in Columbia-Missouri-USA April 14-17, 1980).

TITLE: DETERMINATION OF CONTAMINANTS IN NUCLEAR MATERIALS BY MEASURING THE CAPTURE GAMMA RAYS OF THERMAL NEUTRONS IN A REACTOR INTERNAL GEOMETRY.

RESEARCH INSTITUTE: INSTITUTO DE PESQUISAS ENERGÉTICAS E NUCLEARES (IPEN-ex-IEA).

CHIEF SCIENTIFIC INVESTIGATOR: DR. ACHILLES A.SUAREZ

ASSOCIATE INVESTIGATOR: DRA. BRIGITTE R.S.PECEQUILO

PERIOD OF CONTRACT : DECEMBER 1976 to DECEMBER 1979.

SCIENTIFIC BACKGROUND AND SCOPE OF THE PROJECT

EXPERIMENTAL METHOD

RESULTS OBTAINED

CONCLUSIONS

All these informations are included in the 500 words summary submitted to the IVth. International Conference on Nuclear Methods in Environmental and Energy Research to be held in Columbia, Missouri-U.S.A., next April, 1980.

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

In previous papers[1,2] we presented a new method of analysis of fuel material impurities using prompt gamma rays following thermal neutron capture. This method is relatively rapid and nondestructive and permits trace and major component analysis for several elements without previous chemical separation.

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

To show this we have measured samples of UO_2 with controlled amounts of rare-earth imputities 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[1,2].

As neutron source we used the IEA-R1 research reactor of São Paulo and gamma rays were detected with a single Ge(Li) spectrometer.

However, neutron capture γ-ray spectra by 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 γ-energies > 1.022 MeV and unresolved multiplets are therefore a commun feature.

Moreover, Compton scattering and subsequent escape of scattered γ-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. Greatly simplified and significantly "cleaner" spectra can be obtained with a 3-crystal coincidence technique. Double escape peaks results from (e^+-e^-) pair production in the detector followed by positron annihilation and escape from detection of both 511 keV γ-rays produced. If this annihilation radiation is allowed to strike 2 additional detectors surrounding the central Ge(Li) crystal, single escape and full energy peaks are 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 developed for our measurements consists of a 42.5cc true coaxial Ge(Li) detector and two optically separated 6"x6" NaI(Tl) crystals mounted at the tangencial tube of the IEA-R1 reactor at a distance of 654 cm from an internal target. The neutron flux is typically about $5x10^{11} cm^{-2}.s^{-1}$ at the target position.

The electronics comprise standard coincidence hardware. Fast output signals from 3 linear amplifiers trigger timing single channel analyzers and are subsequently fed into a fast coincidence module operating with time resolution $2\tau \sim 100$ns. Coincident events generate a gate pulse for a gated biased amplifier allowing the linear signal from the unipolar output of the Ge(Li) amplifier to be processed. Selection of 511 keV annihilation radiation in the scintillators is acomplished by a proper adjustment of discriminator levels. Variations of photomultiplier and amplifier gain are offset by analog stabilization. The energy signal from the Ge(Li) detector is digitally stabilized at the ADC level in order to maintain energy resolutions of ~ 7.0 keV at 4.1 MeV during long measuring periods.

With this systems we measured several UO_2 samples with rare earth impurities (Sm, Gd). We observed that although the coincidence requirement reduces the detection efficiency by a factor of about 4.5 in comparison with the single mode, a very substancial decrease in the background is obtained, so we could detect lower intensity peaks than in the single mode and the peak area estimates also became more accurate.

The gain in peak-to-background ratio calculated for several spectra was about a factor of 6.

With those samples we also have determined calibration curves of the experimental system.

We would like to point out that our method of analysis can be used not only for the nuclear fuel materials, but also for biological and geological applications.

PAPERS PUBLISHED ON WORK DONE UNDER THE CONTRACT

1- B.R.S.Pecequilo, Ph.D. Thesis, IEA-DT-078(1978).
2- "Fuel material analysis using radioactive thermal neutron capture", B.R.S. Pecequilo and A.A.Suarez, in "Neutron Capture Gamma Ray Spectroscopy",ed. by R.E.Chrien and W.R. Kane, Plenum Press, 1979, p.710-13.
3- "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, to be presented at the 4th International Conference on Nuclear Methods in Environmental and Energy Research to be held in Columbia, Mo, USA, next April, 1980.

PROJECT EXPENDITURES

List of equipment utilized in the project:

Quant.	Item	Cost of equipment (US$)
2	ORTEC 410 Linear Amplifier	1,520.00
2	ORTEC 450 Research Amplifier	1,410.00
1	401B/402D Bin and Power Supply	990.00
2	401A/402A Bin and Power Supply	695.00
1	ORTEC 448 Pulser	1,185.00
2	ORTEC 113 Preamplifier	260.00
1	ORTEC 459 Bias Supply	525.00
1	ORTEC 551 Timing Single Channel Analyser	540.00
2	CAMBERRA 1520 Analog Stabilizer	1,870.00
1	ORTEC 414 A Fast Coincidence	600.00
1	ORTEC 427 A Delay Amplifier	400.00
1	ORTEC 433 A Dual Sum Invert Amplifier	225,00
1	CAMBERRA 1462 Biased Amplifier	935.00
1	HEWLETT PACKARD Multichannel Analyser	10.600.00
2	CAMBERRA 1501 Stabilization Pulser	1,100.00
1	HEWLETT PACKARD 5586 A Spectrum Stabilizer	2,500.00
1	5050 B Digital Recorder HEWLETT PACKARD	1,900.00
1	7004 B x-y Recorder HEWLETT PACKARD	1,445.00
1	2895 Tape Punch HEWLETT PACKARD	2,810.00
1	9820 A Calculator HEWLETT PACKARD	6,525.00
1	11223 A Cassette Memory/Special Programs HEWLETT PACKARD	520.00
1	11222 A User Definable Functions HEWLETT PACKARD	520.00
1	11221 A Mathematics HEWLETT PACKARD	520.00
1	ORTEC 456 High Voltage	615.00
2	ORTEC 115 Power Supply	670.00
2	ORTEC 455 Timming Single Channel Analyser	1,280.00
1	Ge(Li)	~10,000.00
1	6HSW12/(2)5AL, 6"dia x 12" Long Crystal BICRON	9,683.00
	Sub.Total:	$ 61,843.00

OTHER COSTS:		US$
Computer Time		2,500.00
Liquid Nitrogen		2,800.00
Electronic Maintenance		2,700.00
Shielding		5,000.00
	Sub. Total:	$ 13,000.00
	TOTAL:	$ 74,843.00

Chief Scientific Investigator
Dr. Achilles A. Suarez

Financial Official
IPEN

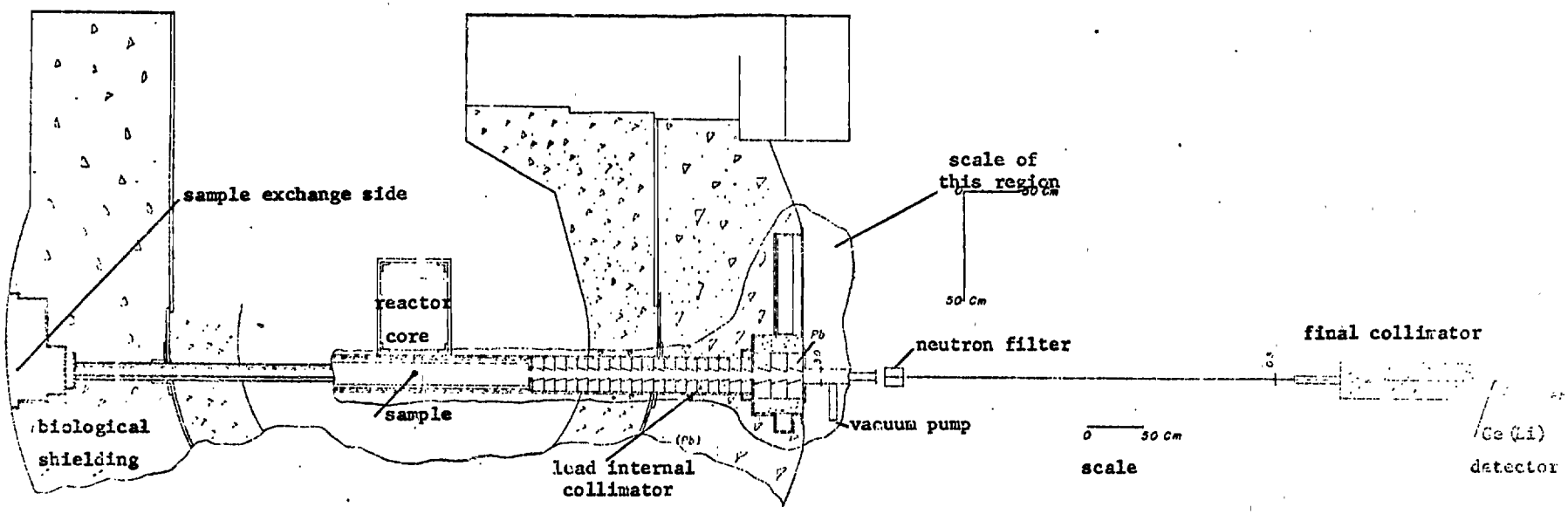

sample exchange side
scale of this region
0 50 cm
50 cm
reactor core
Pb
neutron filter
final collimator
sample
biological shielding
vacuum pump
0 50 cm
scale
Ge (Li) detector
(Pb)
lead internal collimator

Fig. 1

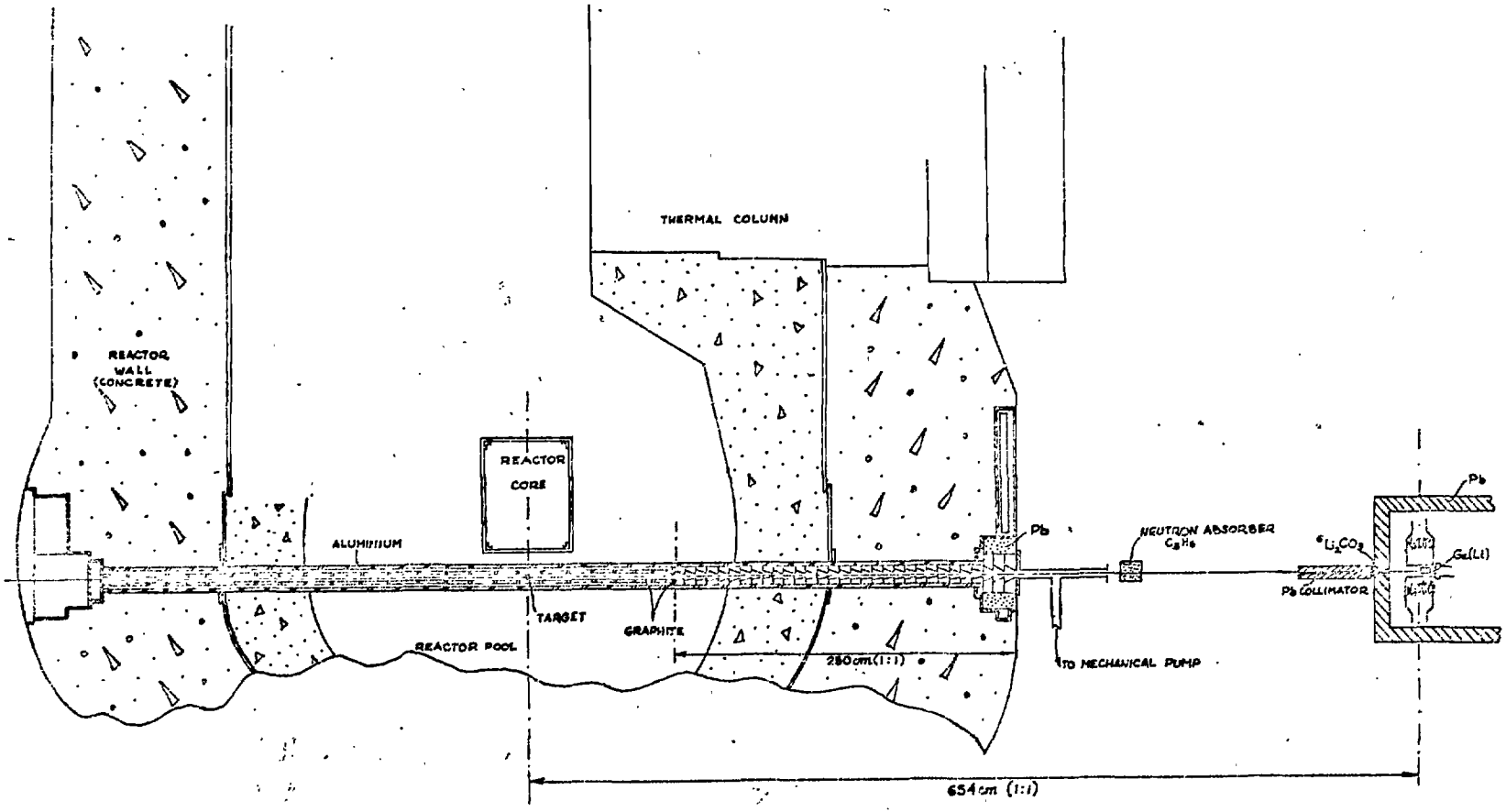
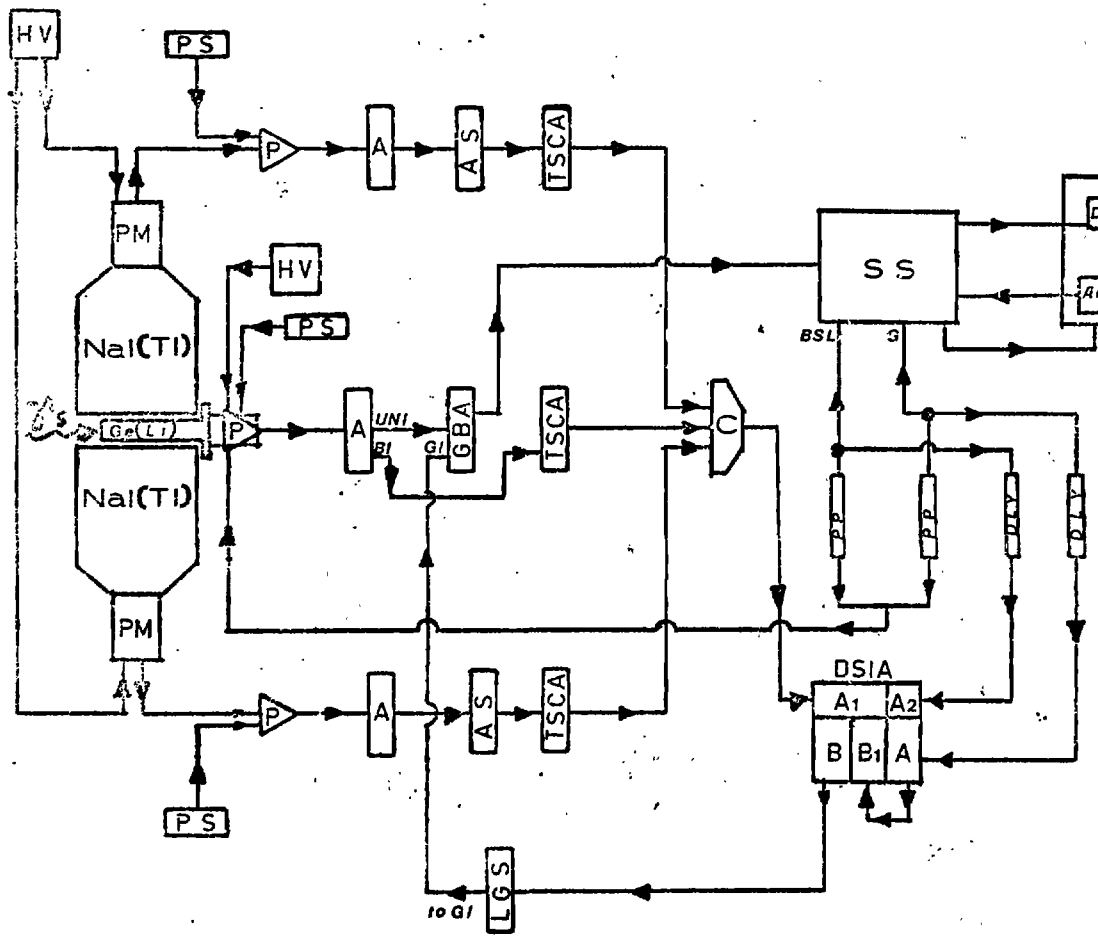

THERMAL COLUMN
REACTOR WALL (CONCRETE)
REACTOR CORE
ALUMINIUM
TARGET
GRAPHITE
REACTOR POOL
Pb
NEUTRON ABSORBER C_8H_6
$^6Li_2CO_3$
Pb COLLIMATOR
Ge(Li)
TO MECHANICAL PUMP
280cm(1:1)
654cm (1:1)

Figure 2



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

Fig. 3- Electronics of the three-crystal spectrometer.