KfK 5181
April 1993

An In-Beam Compton-Suppressed Ge Spectrometer for Nondestructive Neutron Activation Analysis

R. Zaghloul, A. Abd El-Haleam, M. Mostafa,
E. Gantner, H. J. Ache
Institut für Radiochemie

Kernforschungszentrum Karlsruhe

KERNFORSCHUNGSZENTRUM KARLSRUHE

Institut für Radiochemie

KfK 5181

An In-Beam Compton-Suppressed Ge Spectrometer For Nondestructive Neutron Activation Analysis

R. Zaghloul[1], A. Abd El-Haleam[1],
M. Mostafa, E. Gantner and H. J. Ache

[1] Hot Laboratory Centre, Atomic Energy Authority, Cairo, Egypt

KERNFORSCHUNGSZENTRUM KARLSRUHE GMBH, KARLSRUHE

Kernforschungszentrum Karlsruhe GmbH
Postfach 3640, 7500 Karlsruhe 1

ISSN 0303-4003

Abstract

A high-efficiency compton background suppressed gamma-ray spectrometer by anti-coincidence counting with a NaI(Tl)-shield around a central HPGe-detector for in-beam prompt gamma-ray neutron activation analysis (AC-PGNAA) using a Cf-252 neutron source has been designed and built to provide simultaneous anti-coincidence spectrometry of natural, industrial and environmental samples. The spectrometer consists of a high-purity germanium detector as the main detector and a large volume cylindrical NaI(Tl) detector as a guard detector. The Ge detector is a closed-end coaxial detector with dimensions of 4.35 cm length, 4.8 cm diameter and a relative photopeak efficiency of 16.3 %. The guard counter has a 29 cm diameter by a 29 cm long NaI(Tl) cristal. The anti-compton (AC) array is located in a large cubic (1 m^3) polyethylene irradiation facility with a $1.5*10^8$ n/sec Cf-252 source at its axial center where the irradiated sample is in the form of a polyethylene bottle of 50 ml. The irradiation facility contains two steel-lead collimators: One for neutron beam guide with polyethylene plug for neutron thermalization; the second collimator is for gamma-emission detection where a 2.5 cm thick polyethylene box filled with Li_2CO_3 absorbers enriched in Li-6 is inserted near to the target to remove scattered neutrons within the detected gamma-beam. While the end of the collimator is further shielded by lead (20 mm thick) which had a conically shaped collimating aperture, its smallest opening had a diameter of 16 mm to protect the front part of the NaI(Tl) assembly from direct gamma-radiation from the target.
The AC-arrays are further shielded from the surrounding background by housing it in a lead-shield of 5 cm thickness lined with cadmium and copper sheets of 1 mm each. The associated electronics working in conventional fast-slow mode have been designed to operate independently and simultaneously in the anti-coincidence mode as well as in the coincidence or in the normal passive mode. When operating in the anti-coincidence mode the compton edge of Cs-137 is reduced by a factor of 7.9 to provide a peak-to-compton edge ratio of 500/1. The assembly has the ability to measure instantaneously, simultaneously and nondestructively bulk samples up to about 50 cm^3. Major constituent elements in several rocks and mi-

nerals such as H, B, N, Na, Mg, Al, Si, Cl, K, Ca, P, S, Ti, Fe, Sm , Nd, Mn and Gd can be determined, while oxygen cannot be measured due to its small capture cross section (0.27 mb). Several important minor and trace elements such as B, Cd an Hg beside the low residual activity, rare earths and short-lived isotopes could be detected.

The sensitivity of the AC-PGNAA technique is limited by the available neutron flux at the target matrix and the neutron absorption cross section of the elements of interest. PGNAA has the advantage to estimate the constituent elements which are difficult to be measured through the delayed gamma-ray measurements such as B, Bi, C, H, P, Tl, Be, Cl and S in industrial and reference materials and those elements which are transformed into other stable isotopes when undergoing neutron capture. The design of the spectrometer assembly, its properties and performance are described.

Ein Ge-Spektrometer mit Compton-Unterdrückung für die prompte nichtdestruktive Neutronenaktivierungsanalyse

Zusammenfassung

Ein hochempfindliches γ-Spektrometer mit Antikoinzidenz-Compton-Unterdrückung, bestehend aus einem "High Purity" (HP)-Detektor mit ringförmigem NaJ(Tl)-Anti-Compton-Zähler und einer Cf-252-Neutronenquelle, wurde für die "Anti-Compton Prompt Neutron Activation Analysis" (CA-PGNAA) von mineralischen, industriellen und Umweltproben konzipiert und aufgebaut. Der Ge-Detektor ist ein 4,35 cm langer "Closed End"-Koaxialdetektor mit einem Durchmesser von 4,8 cm und einer relativen Photopeak-Efficiency von 16,3 %. Der Anti-Compton-Ringdetektor ist ein 29 cm langer NaJ(Tl)-Kristall mit einem äußeren Durchmesser von 29 cm. Das Anti-Compton-Detektorsystem befindet sich unter einer würfelförmigen Bestrahlungseinrichtung aus Polyethylen von 1 m^3 Volumen mit zentraler Bestrahlungsposition für Probengefäße mit 50 ml Inhalt, vor der die Cf-252-Neutronenquelle (Quellstärke: $1,5*10^8$ n/sec) angeordnet ist. Die Bestrahlungseinrichtung besitzt zwei Stahl-Blei-Kollimatoren: Einen auf die Probe gerichteten und von der Neutronenquelle ausgehenden konischen Kollimator mit Polyethylenstopfen zur Neutronenthermalisierung; der zweite verläuft zur Kollimierung der γ-Strahlung vertikal von der Probe zum Ge-Detektor und enthält direkt unter der Probe eine 2,5 cm dicke Absorberschicht aus Li_2CO_3 (angereichertes Li-6) zur Absorption der in Richtung auf den Detektor an der Probe gestreuten Neutronen. Das untere Ende des Kollimators ist innen mit einer 20 mm dicken, konischen Bleiabschirmung zum Schutz des NaJ(Tl)-Detektors gegen die direkte γ-Strahlung von der Probe ausgelegt und hat zum Ge-Detektor hin eine Öffnung von 16 mm.

Das gesamte Detektorsystem ist zusätzlich durch 5 cm dicke, innen mit je 1mm dickem Cd-und Cu-Blech verkleidete Bleiwände um den Unterbau des Polyethylenwürfels gegen den Umgebungsuntergrund abgeschirmt. Die zugehörige fast-slow-Koinzidenzelektronik erlaubt unabhängig voneinander die Messung sowohl im Antikoinzidenz - als auch im Koinzidenz - bzw. im "Passive Mode"-Betrieb. Im Antikoin-

zidenzbetrieb wird die Compton-Kante der Cs-137 - Linie um den Faktor 7,9 reduziert, wodurch ein Peak-zu-Compton-Verhältnis von 500 erreicht wird. Mit dem System können Proben mit bis zu 50 ml Volumen prompt und zerstörungsfrei analysiert werden. So lassen sich wichtige Bestandteile von Gesteins- und Mineralproben wie z. B. die Elemente H, B, N, Na, Mg, Al, Si, Cl, K, Ca, P, S, Ti, Fe, Sm, Nd, Mn und Gd bestimmen, während Sauerstoff wegen seines kleinen Neutroneneinfangsquerschnitts (0,27 mb) nicht gemessen werden kann. Außerdem konnten neben B, Cd und Hg im Spurenbereich auch verschiedene Elemente nachgewiesen werden, bei denen nur kurzlebige Aktivierungsprodukte entstehen.

Die Empfindlichkeit der AC-PGNAA-Technik wird durch den am Probenmaterial verfügbaren Neutronenfluß und die Neutroneneinfangsquerschnitte der interessierenden Elemente begrenzt. Andererseits bietet die PGNAA die Möglichkeit, auch solche Elemente in Proben verschiedener Herkunft zu analysieren, die wie B, Bi, C, H, B, Tl, Be, Cl und S mit der "klassischen" verzögerten Neutronenaktivierungsmethode nur schwierig oder gar nicht nachgewiesen werden können, weil sie beim Neutroneneinfang direkt in stabile Isotope umgewandelt werden. In dieser Arbeit werden der Aufbau des Spektrometers sowie seine Eigenschaften und Leistungsdaten beschrieben.

Table of Contents

1. INTRODUCTION

The determination of radionuclides in their mixture samples by gamma-ray spectrometry is one of the most direct and powerful methods. Usually a mixture of many different gamma-emitting isotopes formed in the irradiated sample can be analyzed simultaneously with no radiochemical preparation necessary. These mixtures may contain fission products from the presence of thorium and uranium isotopes, particle activation products and other natural activities. Further, the samples may take a variety of chemical and physical forms, including biological and geological samples, reactor fuel and effluent as well as various environmental samples. The normal objectives of the activation analysis techniques using γ-ray spectrometry analyses are to obtain accurate measurements of the concentrations of all radionuclides in the samples rapidly, economically and with minimal effort. These objectives can be realized by using a coincidence shielded, high resolution gamma-ray spectrometer system. Although these types of gamma-ray spectrometers greatly enhance the selectivity and sensitivity for multiple radionuclides measurements, problems remain, where full energy peaks from less abundant radionuclides are obscured by compton distributions associated with full energy peaks of major radionuclides, of that the presence of higher background which cannot be tolerated in the measurement of any specific sample.

Since, we are of special interest for determining the constituents of certain elements with high accuracy due to their significant roll in the application fields, like B, Bi, C, H, P, Cl, Tl, Be and S where such elements are difficult to be estimated by delayed gamma-ray neutron activation analysis technique. Therefore, Prompt Gamma-ray Neutron Activation Analysis (PGNAA) coupled with the Anti-coincidence (AC) -shielded Ge gamma-ray measurement is a preferable technique to fulfill these investigations. A Cf-252 as neutron source is sufficient for the analyses to be performed which will strongly reduce the cost of this facility.

During the past few years (1-10), many laboratories have been developed sophisticated Compton Suppression gamma-ray spectrometers using solid-state detectors to reduce compton interference and scattered background. The theoretical basis and design consideration of the PGNAA facilities using either the internal target geometry or the external target geometry for the target-detector assembly have been reviewed recently by different authors [11-15]. In internal target geometry, the target is kept near the neutron source and observe the gamma-rays at a distance from them. While in external target geometry, the neutron beam is brought outside to a sample mounted close to the detector; this geometry is only reasonable with strong neutron source like a reactor. The internal geometry has the advantage of high neutron flux but poor counting geometry; which is applicable with limitted neutron source like Cf-252.

The method of reducing compton effect for possible detection of low energy gamma-peaks below 1 MeV, is to surround the Ge detector -as the main measured detector- with a scintillation shield; NaI(Tl) crystal or plastic phosphor; such that the compton radiation will interact in the shield producing pulses of light which in turn are converted into an electrical current by photomultiplier tubes mounted on the surface of the shield crystal. This electrical current may be used as a "Logic" signal to control the recording of the output signal from Ge detector. Thus, if a photon is fully absorbed, the corresponding signal from the Ge detector is recorded and truly represents the total gamma-ray energy. However, if instead the photon deposit only part of its energy in the Ge detector and the remainder escapes as a degraded gamma-ray and is captured in the shield, then the resulting signal can be used to block on route the Ge detector signal. In this way only photoelectric interactions are recorded. Actually, in practice this is only partially achieved, where a more detailed discussion of this technique may be found in literature (16-22).

This paper describes the design, construction and evaluation of high efficiency compton background suppression gamma-ray spectrometer by anti-coincidence counting with a NaI(Tl)-shield around a central HPGe-detector for in-beam prompt gamma-ray neutron activation analyses (AC-PGNAA) using a ^{252}Cf neutron source in internal target geometry to provide simultaneous radionuclides measurements of natural, industrial and environmental samples. The system is designed to eliminate the major cause of background and compton continuum interfering spectra which improves the sensitivity for the measurement of both the coincident and noncoincident gamma-ray emitting radionuclides. The system's performance as an anticoincidence gamma-ray spectrometer is evaluated and discussed, and examples of its application to Boron measurement and geological problems are illustrated.

2. DESIGN OF THE SPECTROMETER SYSTEM

A newly designed anticoincidence-shielded HPGe gamma-ray spectrometer assembly using a Cf-252 neutron source is photographed in figures (1-a) and (1-b). It consists of three main parts; the prompt gamma-ray neutron activation analysis facility (PGNAA), the anti-compton gamma-ray detection array (AC), and the electronic setup.

2.1 Prompt Gamma-Ray Neutron Activation Analysis Facility (PGNAA):

It consists of a cubic container of polyethylene moderators of volume 1 m^3 with two steel-lead collimators, one for the neutron source and the other for emitted gamma-ray spectra. At the axial center of the container a Cf-252 neutron source of 50 μgm activity 1.8×10^8 n/s intensity, and the irradiated sample which is in the form of a 50 ml polyethylene bottle is placed. Two rectangular polyethylene holders for the neutron source and the sample with dimensions of 200mm, H.x200mm, W.x500mm, L., for each,

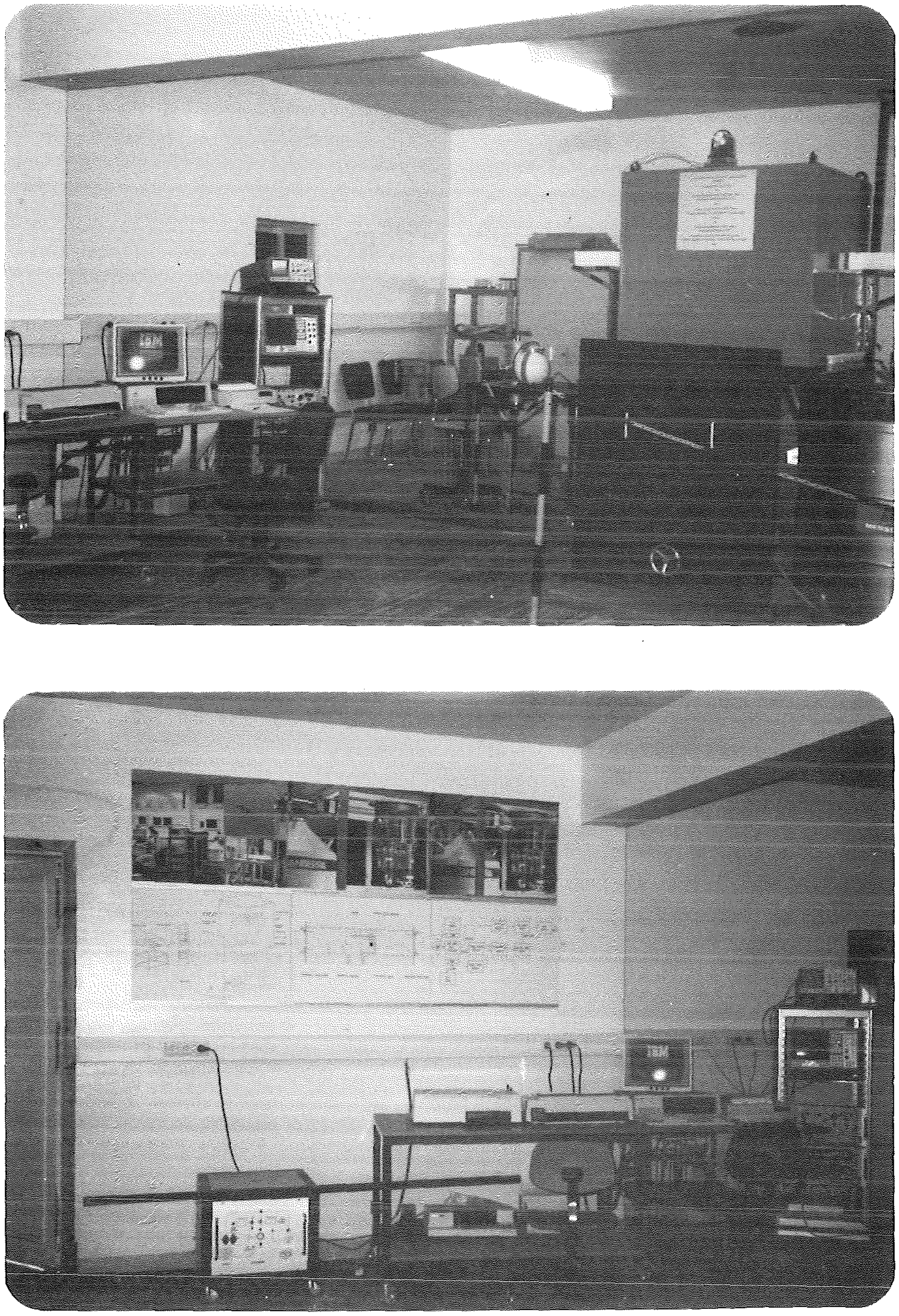

Fig. (1-a): Photograph of the In-Beam Anti-Compton Prompt Gamma-ray Neutron Activation Analysis (AC-PGNAA) spectrometer system.

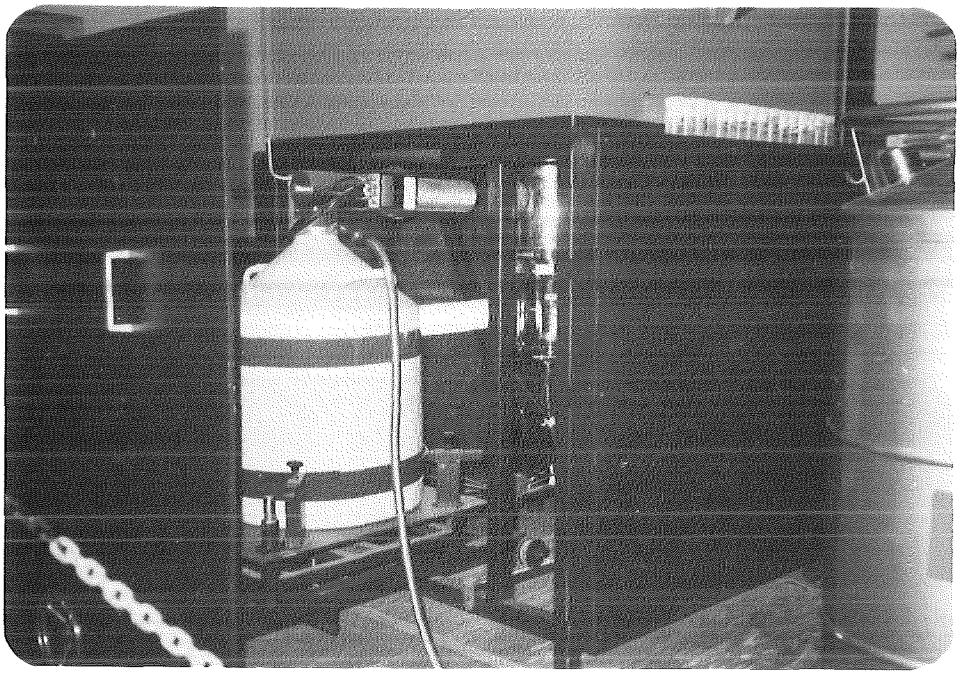

Fig.(1-b) Photograph of the Anti-Compton (AC) Gamma-Ray Detection Array Showing the HPGe-NaI(Tl) Anticoincidence gamma-ray spectrometer.

are prepared to slide easily at the axial center of the container assembly. Figure (2) illustrates a plan view of the sample and the Cf-252 neutron source holders. The neutron source in its collimator is fixed at the inner end of the source-holder. The neutron beam collimator made from steel in conical shape of 50mm long, lined with 5mm lead sheet-to attenuate the fission gamma-rays emitted within the Cf-252 neutron source- and containing a 40mm thick polyethylene plug for the sake of neutron thermalization. At the narrow end of the collimator, there is a hole of 12mm diameter for holding the Cf-252 neutron source. The sample-holder contains a hole with diameter 30mm at its inner end for placing the sample bottle. The source-, sample-holder assembly and their positions inside the polyethylene container are chosen in order to increase the neutron flux density at the target position as well as to prevent the gamma-detector from the direct neutron beam. Changing the sample under investigation or removing the neutron source is easily accomplished by sliding smoothly the two holders on both sides.

Prompt gamma-rays emitted from the investigated sample due to thermal neutrons captured are collimated at right angle to the gamma-ray detector array to minimize the neutron source background effect. The gamma-ray lead collimator is designed in a cylindrical form with dimensions 50mm, ϕ x 400 mm L., ended by an aperture of 16mm at the Ge detector to protect the NaI(T1) shielded -crystal from facing directly the measured gamma-rays. At the upper end of

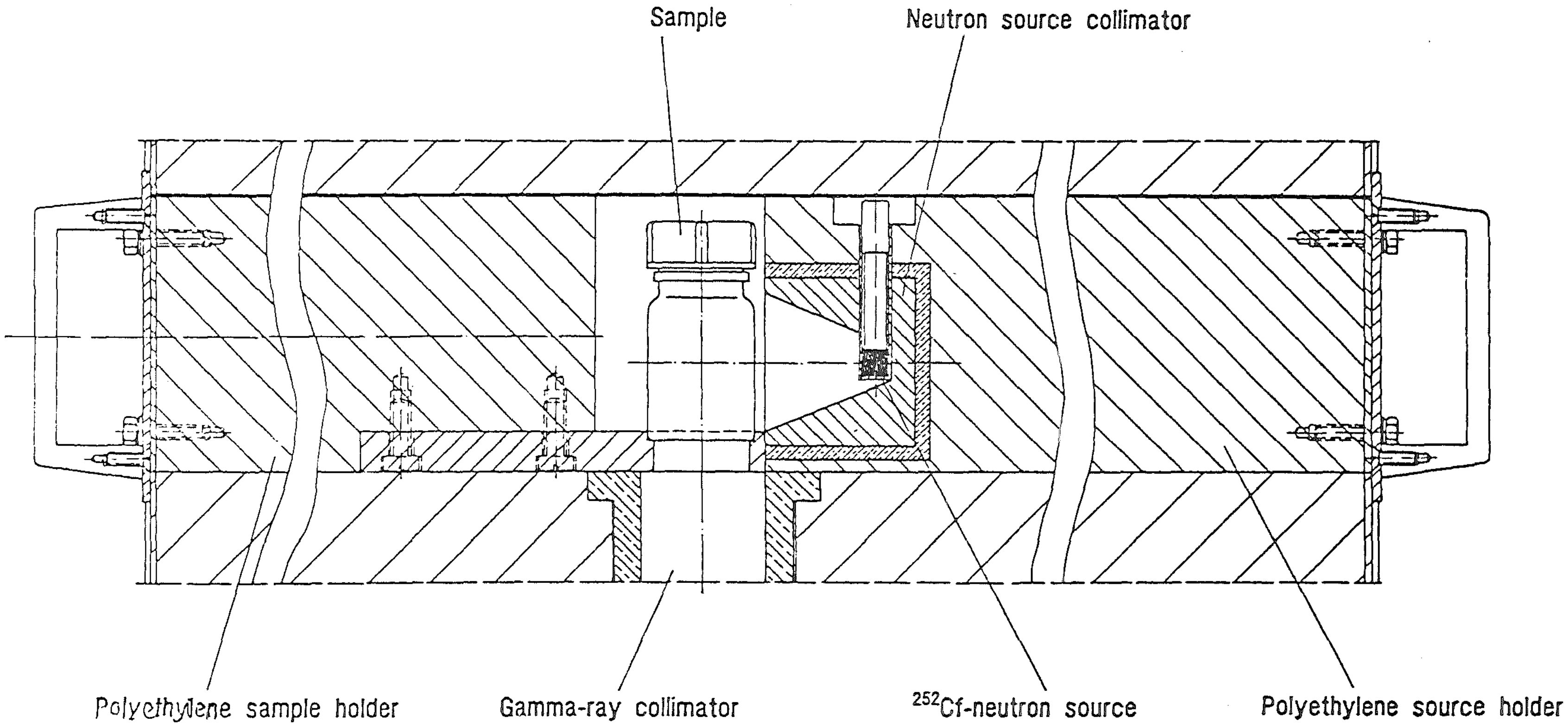

Fig.(2): Planview of the sample and the ^{252}Cf - Neutron Source holders.

the collimator, near to the irradiated sample, a polyethylene box with 50mm ϕ x 25 mm L., filled with Li_2Co_3 absorbers enriched in Li-6 is placed to absorb the scattered thermal neutrons from reaching to the Ge-detector. As well as to absorb the intense beta-radiations emitted from concerned radionuclides in the irradiated sample such a ^{20}F, ^{27}Mg, ^{28}Al, ^{31}Si, ^{32}P, ^{37}S, ^{38}Cl and ^{42}K which could generate Bremsstrahlung background during the de-acceleration process and impair the detection limits of gamma-rays below 500 keV. This background radiation cannot be suppressed by the anticompton counting. At the same time tremendous gamma-rays background arising from the interaction of the irradiated neutron flux with the polyethylene medium of the system could be reduced to permecible level by covering the front part of the NaI(Tl) crystal with a 10 cm thick lead cap. Fig.(3) shows an overall sketch of the AC-PGNAA spectrometer system.

2.2 Anti-Compton Gamma-ray Detection Array:

A high-efficiency compton background suppression gamma-ray spectrometer by anti-coincidence counting with a NaI(Tl)-shield around a central HPGe-detector for in-beam prompt gamma-ray neutron activation analysis (AC-PGNAA) using a ^{252}Cf neutron source has been designed and built. The spectrometer consists of a high-purity germanium detector as the main detector and a large volume cylindrical NaI(Tl) counter as a guard detector. Hence, a coaxial one open end and closed end facing window HPGe detector (Canberra) with a

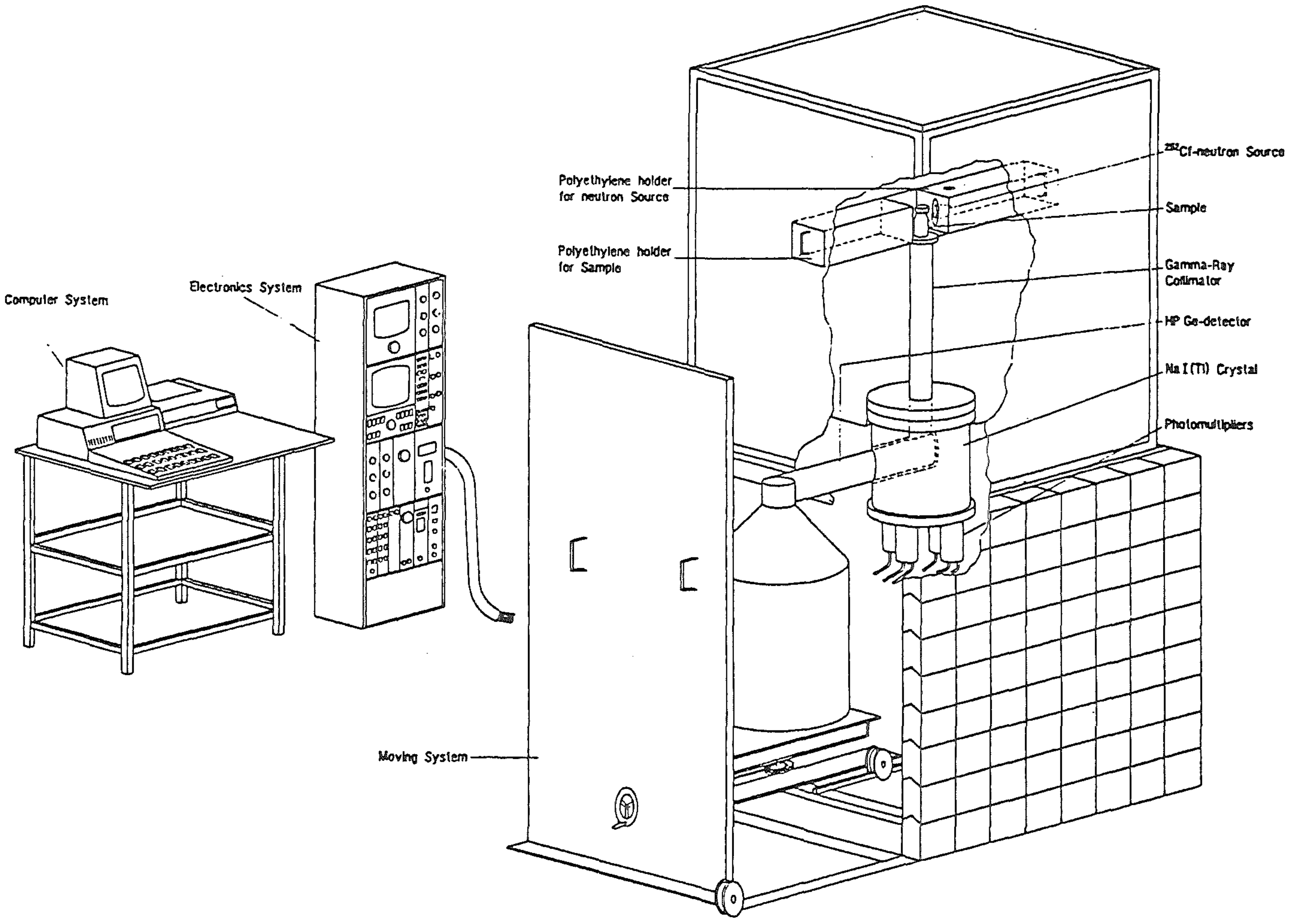

Fig.(3): Compton-Suppressed Non-destructive Neutron Activation Analysis System AC-PGNAA.

relative efficiency of 16.3% was selected. It has a volume of 79 cm^3 (4.8 cm diam. by 4.35 cm long) and an active area facing window of 18.1 cm^2 and is mounted in a cryostat of the horizontal dipstick type with 20 cm long aluminum end cap. The distance of the HPGe crystal to the 0.5 mm thick entrance window is 5mm. An energy resolution of 1.69 KeV (FWHM) and a peak-to-compton ratio of 51.3: 1 for the 1332. KeV line of Co-60 was measured. To prevent disturbances during long-term measurements a 30-liter Dewar of long holding time was assembled. The Dewar-detector assembly was kept on a moving carriage covered with a foam pad, to easy aligned the detector with its NaI(T1)-shield and to isolate it from direct contact with the floor and surrounding shield, for providing measurements against vibrations leading to micro phonic noise.

The guard counter of a 29 cm x 29 cm well-type Nal(T1) crystal with a 7.6-cm-diam. by 18-cm-long well (Bicron) which accommodates the HPGe detector cryostat. The NaI(T1) crystal is coupled to four 5.8-cm-diam.x 7.6 cm-length phototubes, and its energy resolution (FWHM) is 9.8% for Cs-137. A schematic diagram of the cross-section of the anticompton (AC) array is shown in figure (4). The AC-array is fixed with the irradiated container in matching position with the gamma-ray collimator. It is further shielded from the surrounding background by housing it in a 5cm low-background lead shield chamber lined with 1 mm cadmium and copper sheets as well as plexiglas plates.

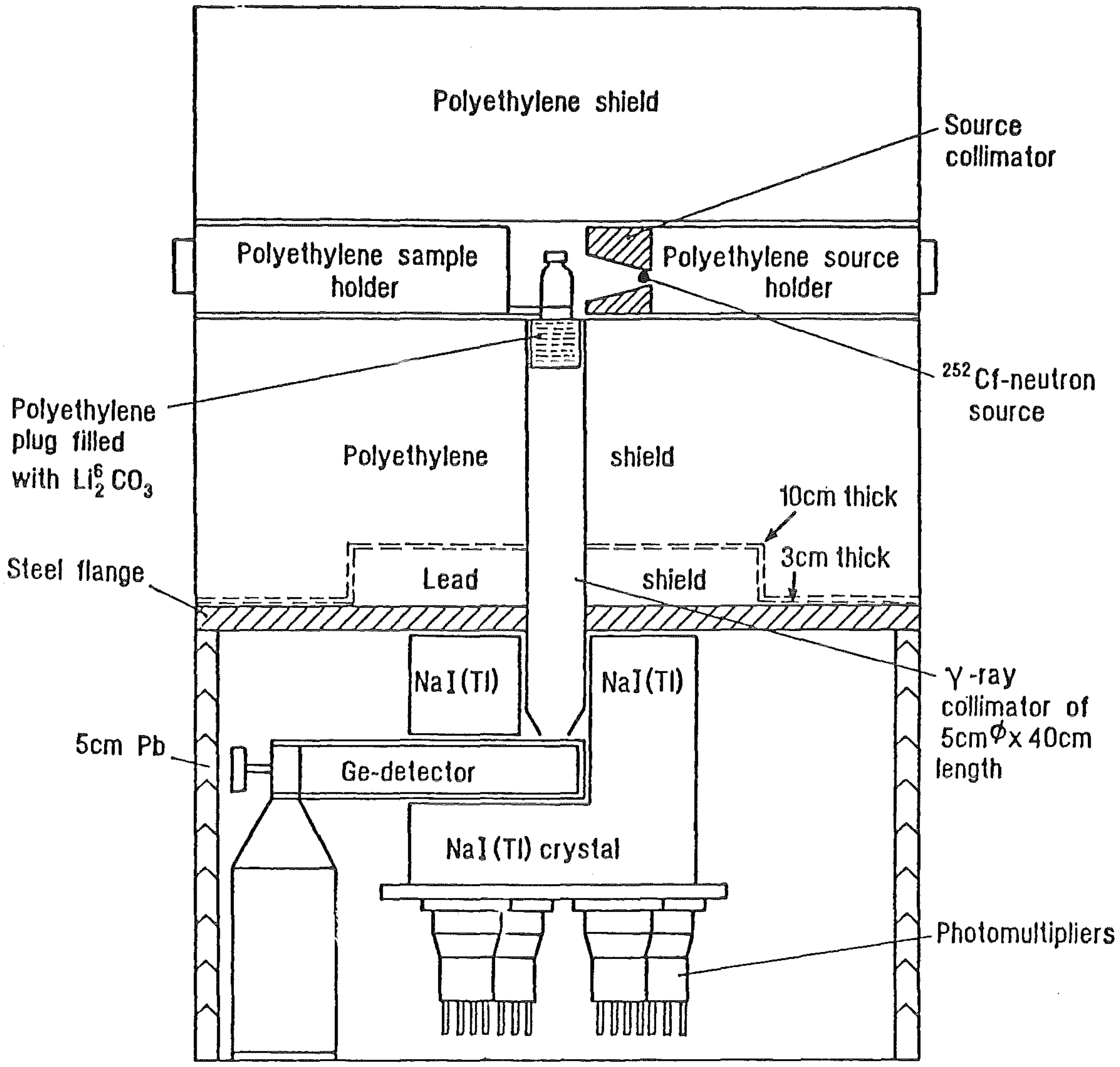

Fig.(4): Cross-sectional view of AC-PGNAA spectrometer facility.

2.3 Electronic Set-Up :

The associated coincidence electronics (canberra) working in conventional fast-slow mode, is designed to operate independently and simultaneously in the anti-coincidence mode as well as in the coincidence or normal, passive shield mode. The coincidence circuit is used to reduce the accidental coincidence rate between the HPGe detector and the NaI(Tl) counter. The block diagram of the electronic set up operated in the anti-coincidence mode is clarified in Figure (5).

The electrical signals from the four multipliers viewing the NaI(Tl) crystal are fed into four preamplifiers connected in series in the alternative way (1 & 3, 2 & 4) to sum up the signals forming the "logic" signal, which is fed to the amplifier-timing single channel analyzer (Amp/TSCA2015 A Canberra) to be used for controlling the recording of the output signals from the Ge-detector, in such a way that events which will be recorded at the same time in both Ge and NaI(Tl) detectors are cancelled. The discriminating level in the timing single-channel analyzer (TSCA) is usually set at minimum energy ($\simeq$50 KeV).

The outputs of the Amp/TSCA units for the Ge and NaI(Tl) detectors are tuned, that they are in time coincidence, which are connected to two inputs of the coincidence unit (2144 A). The coincidence positive output signal will be delayed for a few microseconds ($\simeq$5 μs) by means of a delay unit (2055). This delayed signal will be fed into gate input of the linear gate and stretcher unit

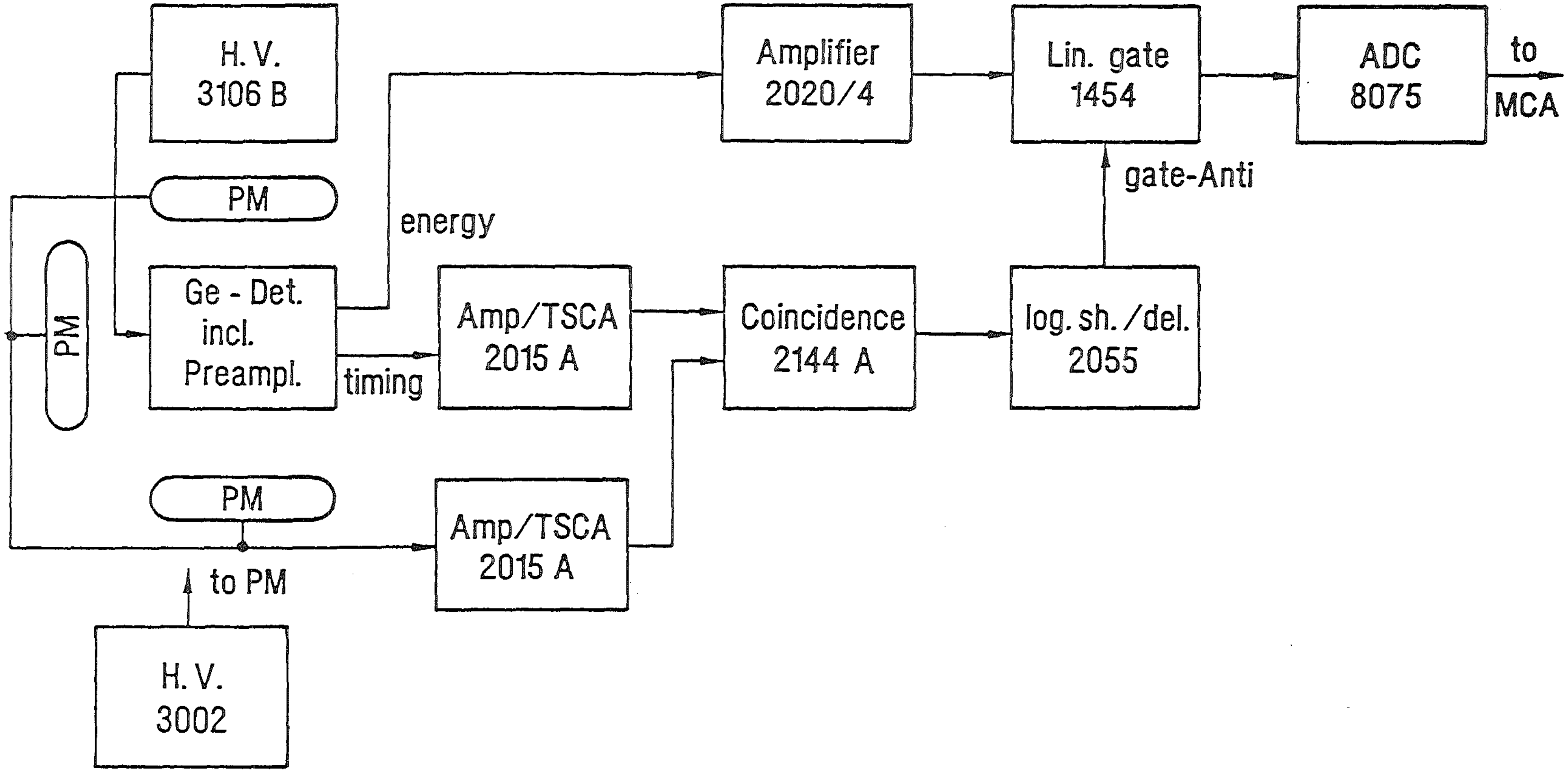

Fig.(5): Block diagram of the electronics set-up in anticoincidence-mode.

(1454), while the unipolar output signal of the main amplifier (2020) of the Ge-detector is fed into signal input of the linear gate, which is switched at anticoincidence set. Using the oscilloscope, the delay time has been adjusted so that the output signal of the delayed unit (2055) overlaps with the leading edge of the unipolar amplifier signal. When the two inputs of the linear gate are in coincidence, the linear gate will be gated off, while when the unipolar amplified signals are in anticoincidence with the delayed coincidence signal (gated signal), the linear gate will allow the Ge-amplified signals (anticompton gamma-ray spectra) to be recorded at the analog-to-digital converter (ADC/8075). The recorded gamma-ray spectra are analyzed and data are treated, and printed out by a 4096 multichannel analyzer (MCA, canberra series 35 plus) in conjunction with IBM personal computer model XT/286 with fixed disk drive that can store up to 20 Mb of formatted data.

Such technique results in a reduction of the compton background since all compton events occurring in the Ge detector in which the compton scattered gamma is detected by the NaI(Tl) counter are rejected. This arrangement has the flexibility that, the NaI(Tl) spectroscopy branch may be operated independently and in coincidence as well as in anticoincidence mode.

3. RESULTS

3.1. Performance of the system:

In order to evaluate the energies and the intensities of the measured spectra, the system has to be calibrated. The energy calibration has been performed by fitting a polynomial function to all reference points. Fig.(6) illustrates a typical energy calibration curve for energy range up to about 2000 KeV. Using standard radioactive sources; ^{241}Am, ^{133}Ba, ^{137}Cs, ^{203}Hg, ^{57}Co, ^{60}Co. Fig.(7) shows the same typical energy calibration curve for the higher range up to about 7000 KeV. Using the energy peaks of the prompt γ-rays emitted through the (n,γ) reaction for the elements H, Fe, Cl, Mn.

The energy resolution of such spectrometer described here, is represented by following equation

$$(\mathrm{FWHM}) = 0.6378 + (7.1 * 10^{4}) * E + (1.76 * 10^{7}) * (E)^{2}$$

Also, the photopeak low energy tailing can be represented by the following equation

$$(\mathrm{TAILING}) = 0.5758 + (2.6190 * 10^{-4}) * E$$

The energy resolution of the present AC-array spectrometer, was found to be 1.87 KeV at 1332.46 KeV gamma-line of Co-60, and 1.17 KeV at 661.61 KeV of Cs-137. Also, it was found that the resolution is equal to 2.04 KeV at 1461.1 KeV of K-40 photopeak. The (FWHM) and (TAILING) are determined for each calibration peak by making a weighted least-squares fitting of a modified gaussian function to be the pulse height data for each peak, by using the computer.

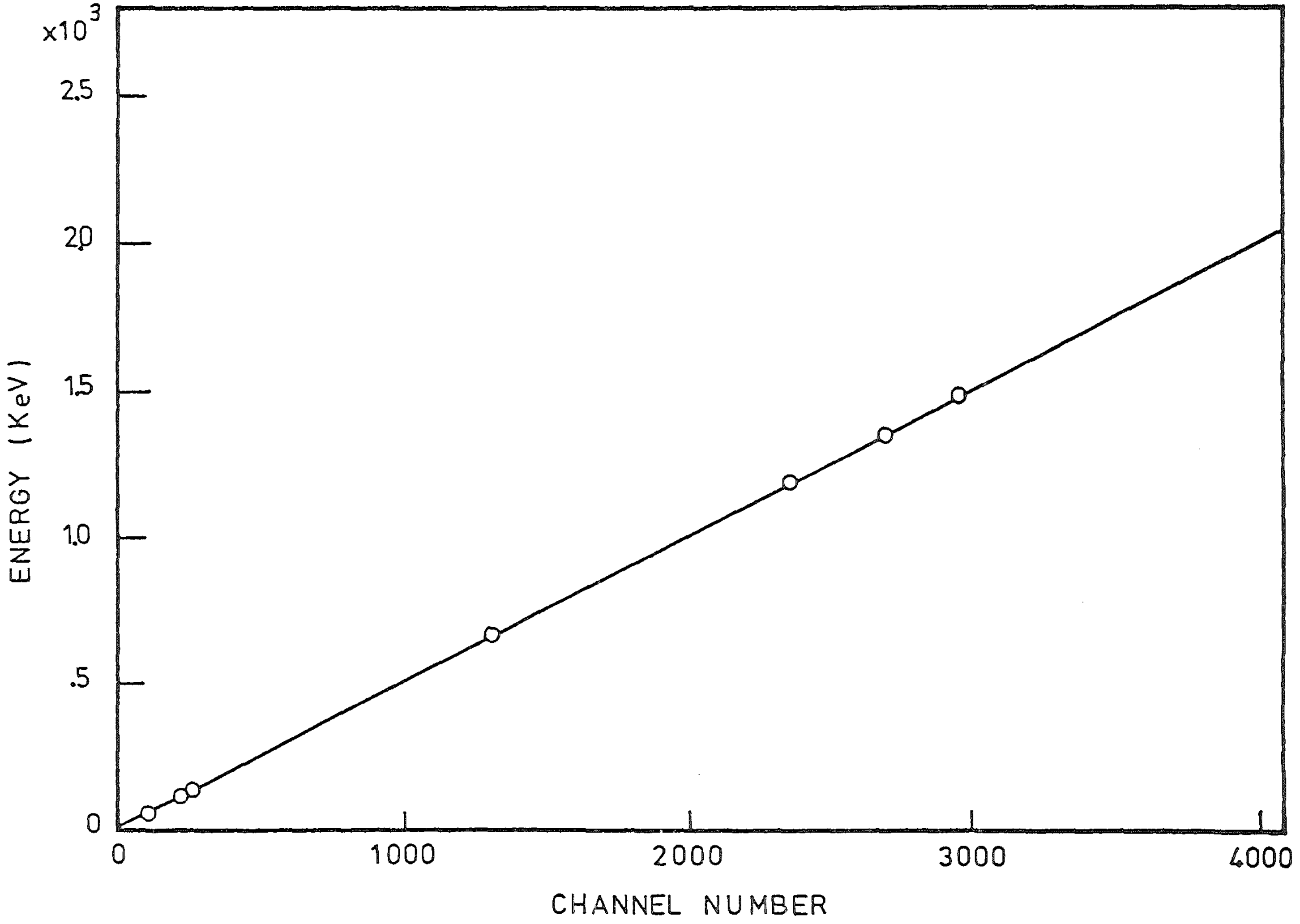

Fig.(6):Typical low energy calibration curve up to~2000 KeV.

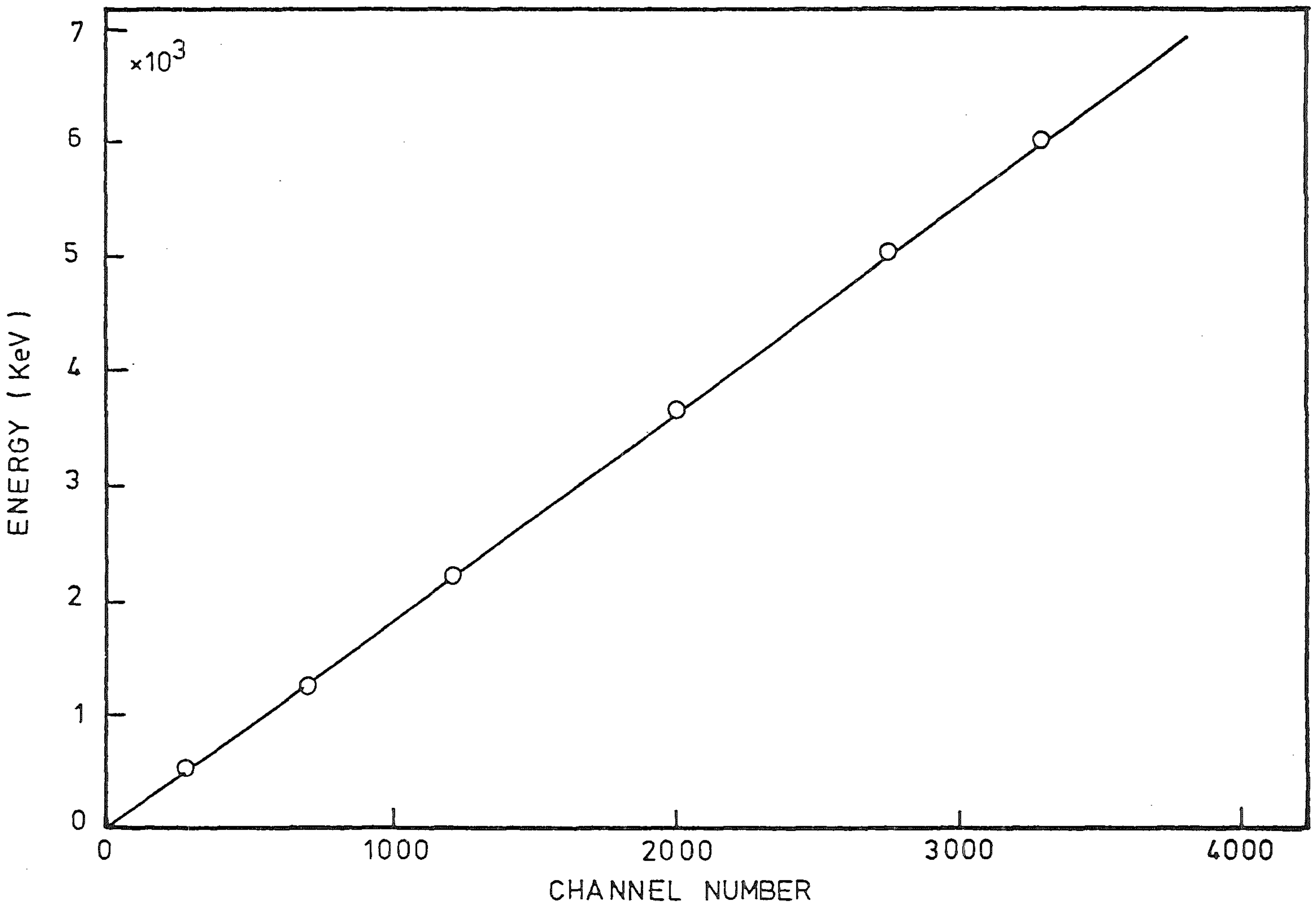

Fig. (7):Typical high energy calibration curve up to ~ 7000 KeV.

In large volume detectors like these which are used in the pressent system, and for energies greater than 200 KeV, compton interaction events are the most common contributions to the full energy peak, in which the present AC-array spectrometer plays the essential part to reduce its effect. The efficiency of the present compton suppressed non-destructive neutron activation analysis described in Fig.(4) has been well-determined, through a prolonged method, in the energy range (80 KeV-10000 KeV). Fig.(8) shows the obtained efficiency calibration curve as a function of energy, with a crossover energy (E_c) at 136.5 KeV. The equations which fit the low and high energy ranges was found to be as follow:

$$\text{Ln (Eff.)} = -1.281 * (\ln E)^2 + 12.407 * (\ln E) - 38.461$$

(for low energy range $E<E_c$)

$$\text{Ln (Eff.)} = -0.106 * (\ln E)^2 + 0.483 * (\ln E) - 8.241$$

(for high energy range $E > E_c$)

Normal and anticoincidence spectra of Ca-137. Fig.(9) shows the normal as well as anticoincidence mode for the gamma-ray spectra of Cs-137 source. It is clear from the figure that the anticoincidence mode reduces the compton edges by a suppression factor of 7.97 to give peak-to-compton ratio (P/C) from (63.1 :1) to (500 : 1) with loss in photopeak count of less than 0.5%. Also, the anticoincidence mode reduces the 185 KeV backscatter peak by a factor of 6.5. A comparison measurements between the normal and anticoincidence mode is expressed in table (1).

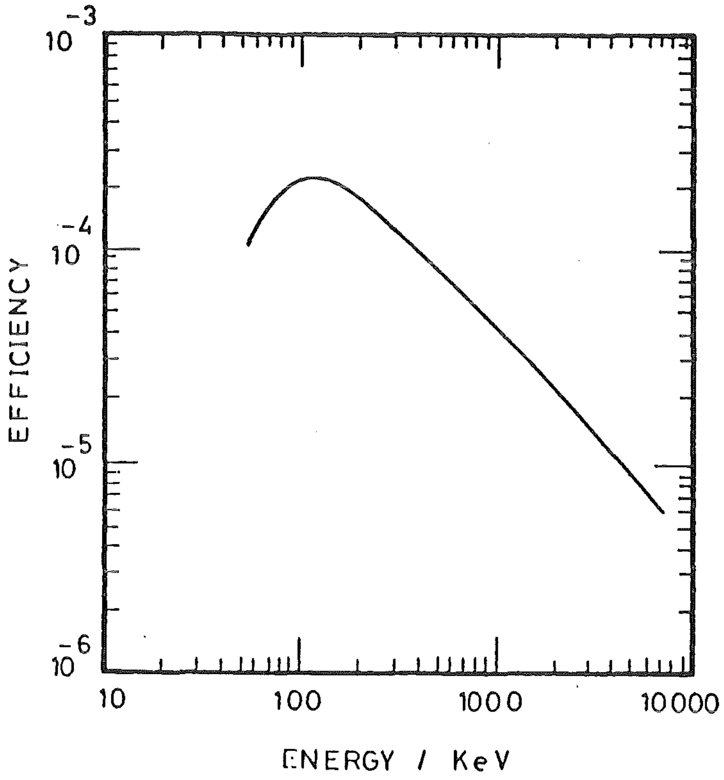

Fig.(8): Efficiency calibration curve of HPG.e detector at the AC-array geometry.

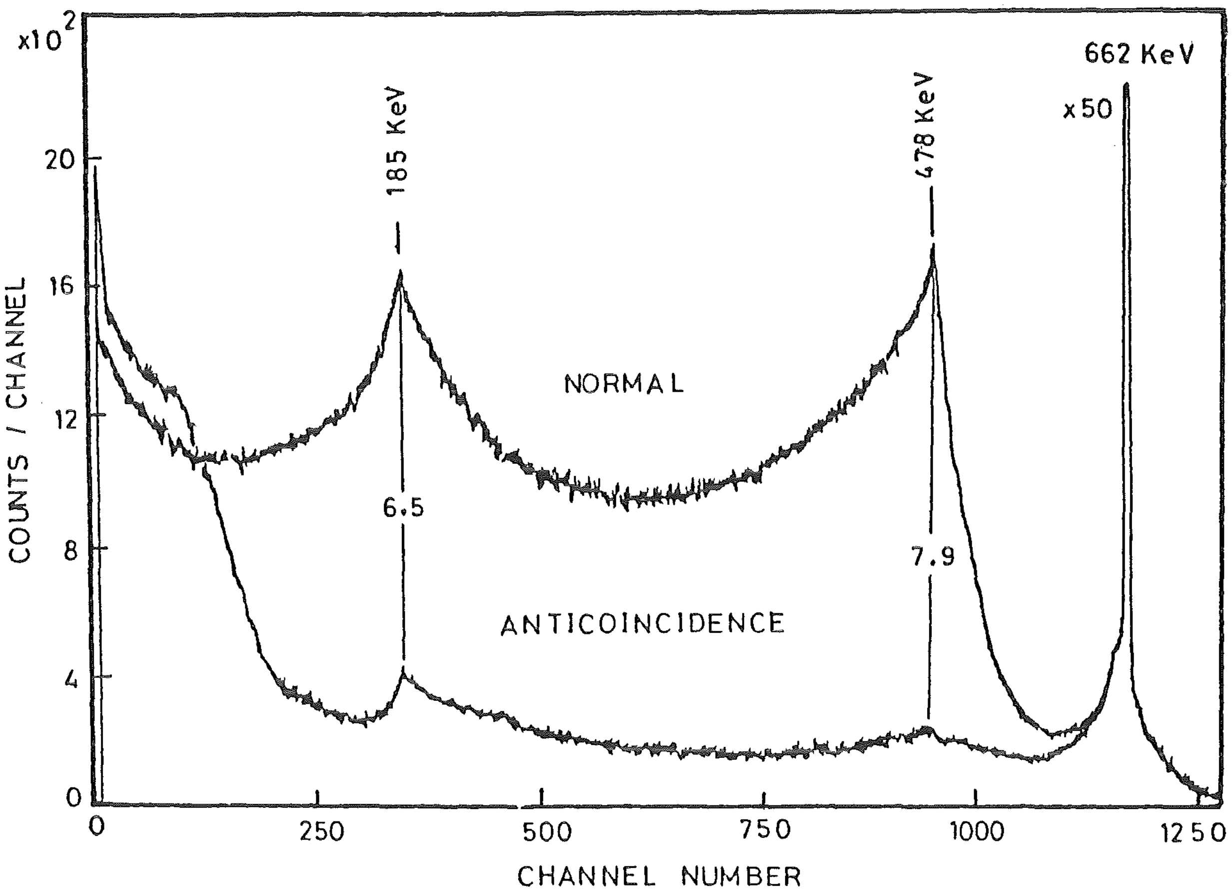

Fig.(9): Normal and Anticoincidence spectra of CS -137

Table (1)

Data measurements for normal and anticoincidence modes.

Data	Without suppression	with suppression
* Mean counting rate per channel at compton edge.	1730	217
* Cs-137 peak max.	1.09×10^5	1.08×10^5
* The ratio (P/C)	63.1/1	500.0/1
* Count rate loss in Cs-137 Peak.	---	0.5%

Fig.(10) shows the normal and anticoincidence spectra of the gamma-rays of Co-60 radioactive standard source. It can be seen that AC-mode reduces the compton edge by a factor of 8.8 to give (P/c) ratio changes from (60:1) up to (528:1). Also, the 550 KeV peak has been reduced by a factor of 8.2, while the 210 KeV peak has been reduced by a factor of 6.2.

Fig.(11) shows the normal as well as anticoincidence mode for the gamma-rays spectra of (Cs-137 and Co-60) together. It can be seen that the anticoincidence mode spectrum reduces the compton background by a factor of 7.8 at 960 KeV peak, by a factor of 7.1 at 478.0 KeV peak, and by a factor of 6.3 at 185.0 KeV peak.

3.2. Suppression Factor:

Compton suppression factor is a good indicator for the detection system efficiency, which improves the sensitivity of the experimental measurements. Fig.(12) shows the measured suppression factor of the AC-array under the optimum different parameters of the accompanied electronic system over a wide range of energy. It is clear from the figure that the best suppression factor obtained is between the value 6 and 9 for the energy range (200 KeV - 1000 KeV). Fig.(13) represents the natural background spectrum for 1000 min. collecting time by anticoincidence mode. It is shown from the figure that only small photopeaks of K-40 and the natural daughter products of Uranium and Thorium originating from materials which are used in the housing and shields of

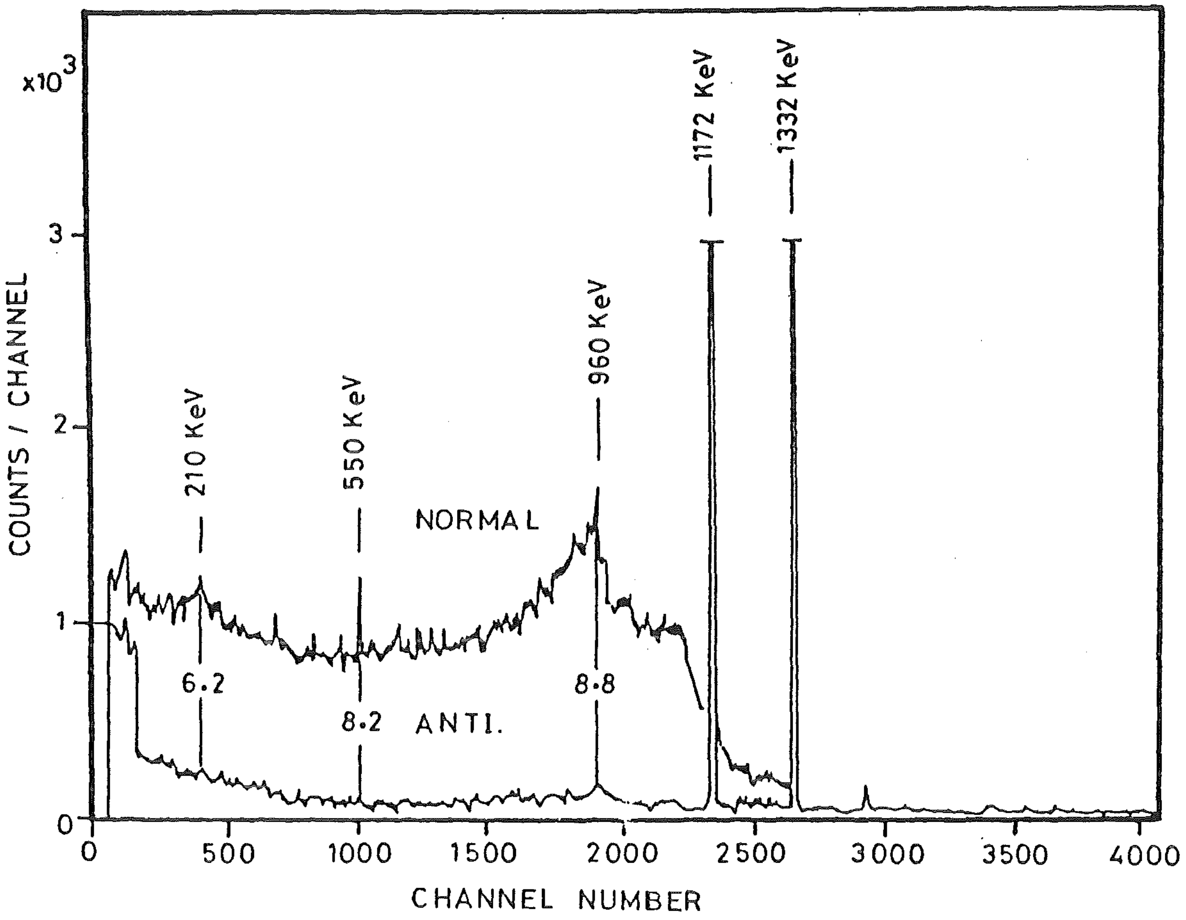

Fig. (10): Normal and Anticoincidence spectra of Co-60

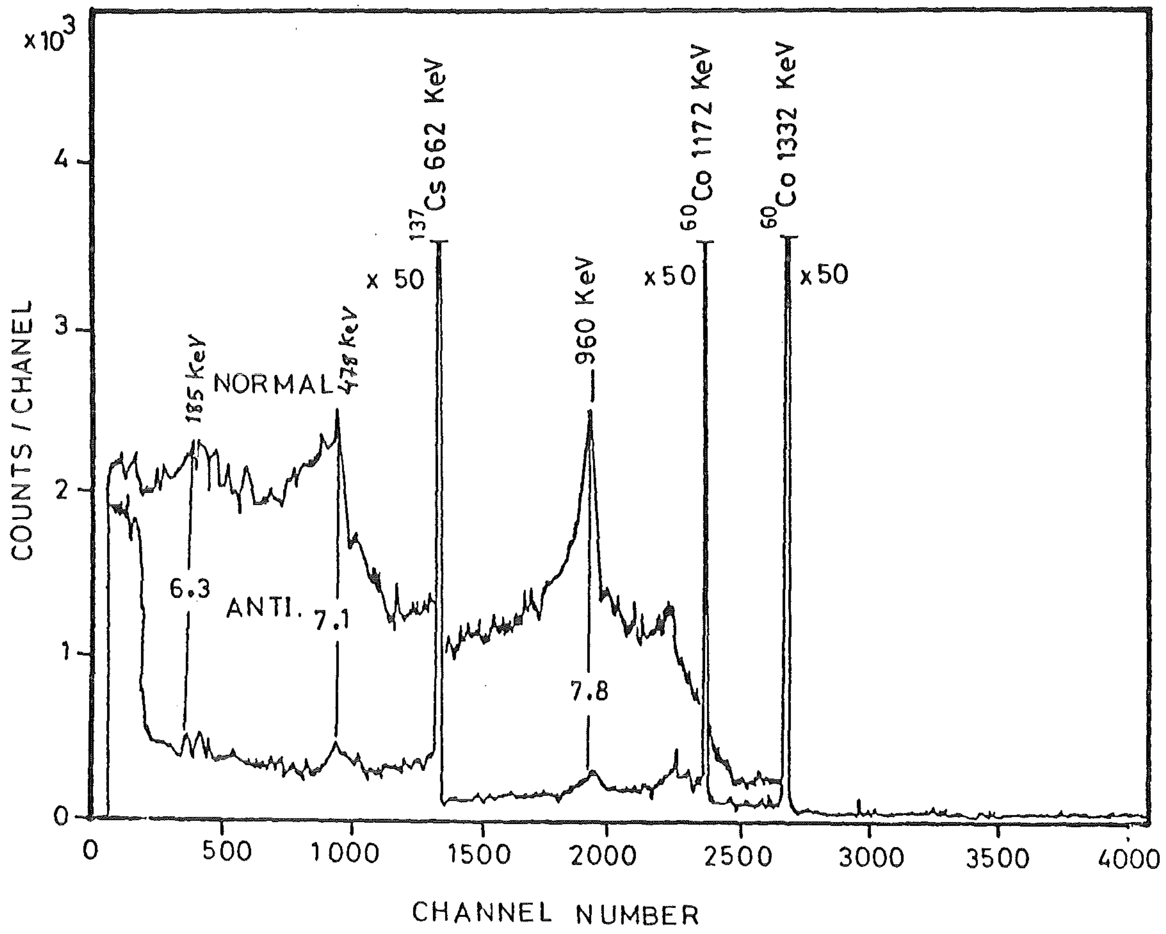

Fig.(11): Normal and Anticoincidence spectra of ^{60}Co and ^{137}Cs

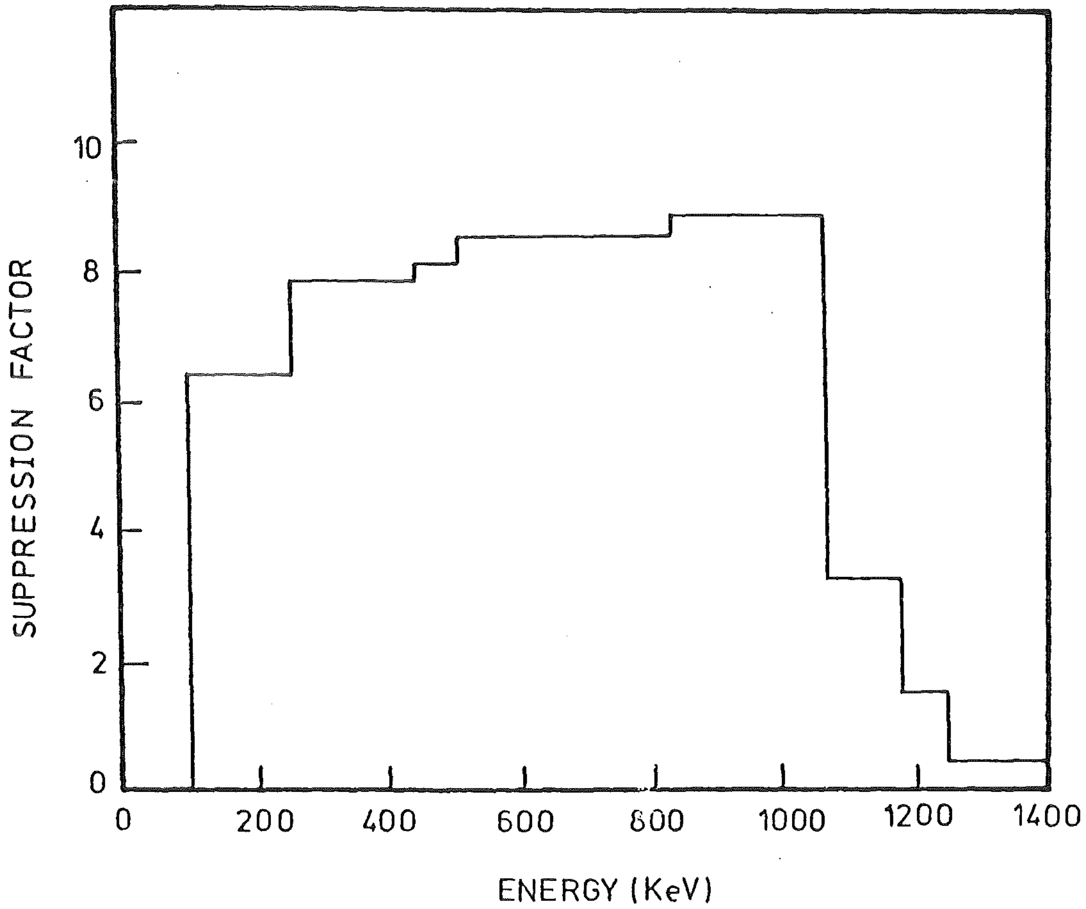

Fig. (12): Measured performance of AC - array .

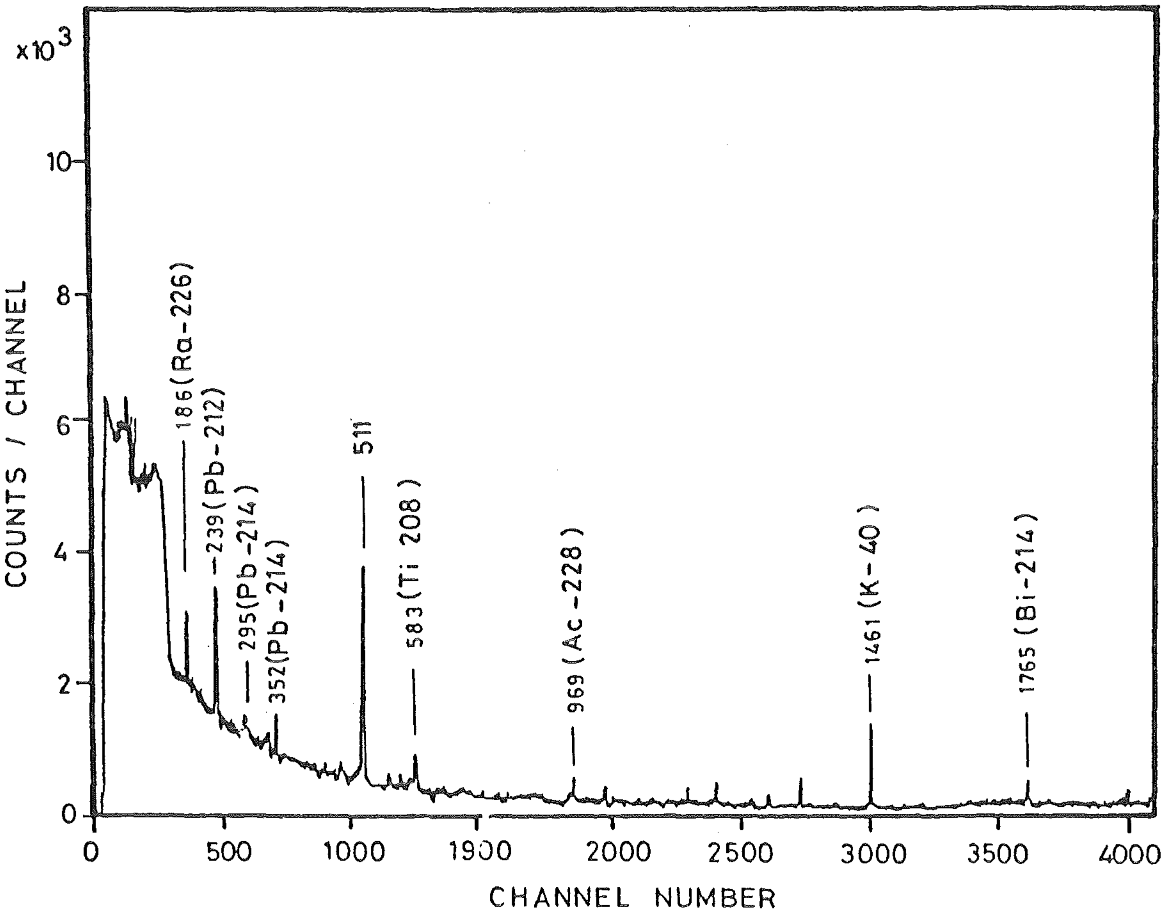

Fig.(13): Anticoincidence spectra of the natural back ground.

the system. Also, it may be from the environment around the laboratory.

3.3. Test analysis :

For further confirmation of the success of the AC-PGNAA system and before using it as a tool for multielemental analysis purpose, it was used to determine the boron content in well-known standard samples. Boron was selected as it gives good detection sensitivity due to high absorption cross section (752b) and high intensity through the reaction ^{10}B (n,α) ^{7}Li with thermal neutrons, the product Li-7 formed mainly in excited state which decays within $5x10^{-14}$ sec. to emit a line of 478 KeV which has clear Doppler broadening peak. A typical prompt gamma-ray spectrum of one of the analysed samples is shown in Fig.(14). Also, Fig.(15) shows the calibration curve obtained by ten standard samples prepared in the same way (SiO_2 ground mixed with crushed glass). Comparison between the present measured values and their standard labelled values give a percentage error of ±0.02% which indicates the success of the system as a reliable tool for elemental analysis with an error much less than the acceptable error.

4. Application of the system:

4.1. Analysis of solid sample:

A sample of 10g of black sand was taken from the accumulated solid material from the bottom of River Nile near Rashid city, North Egypt. This sample has been

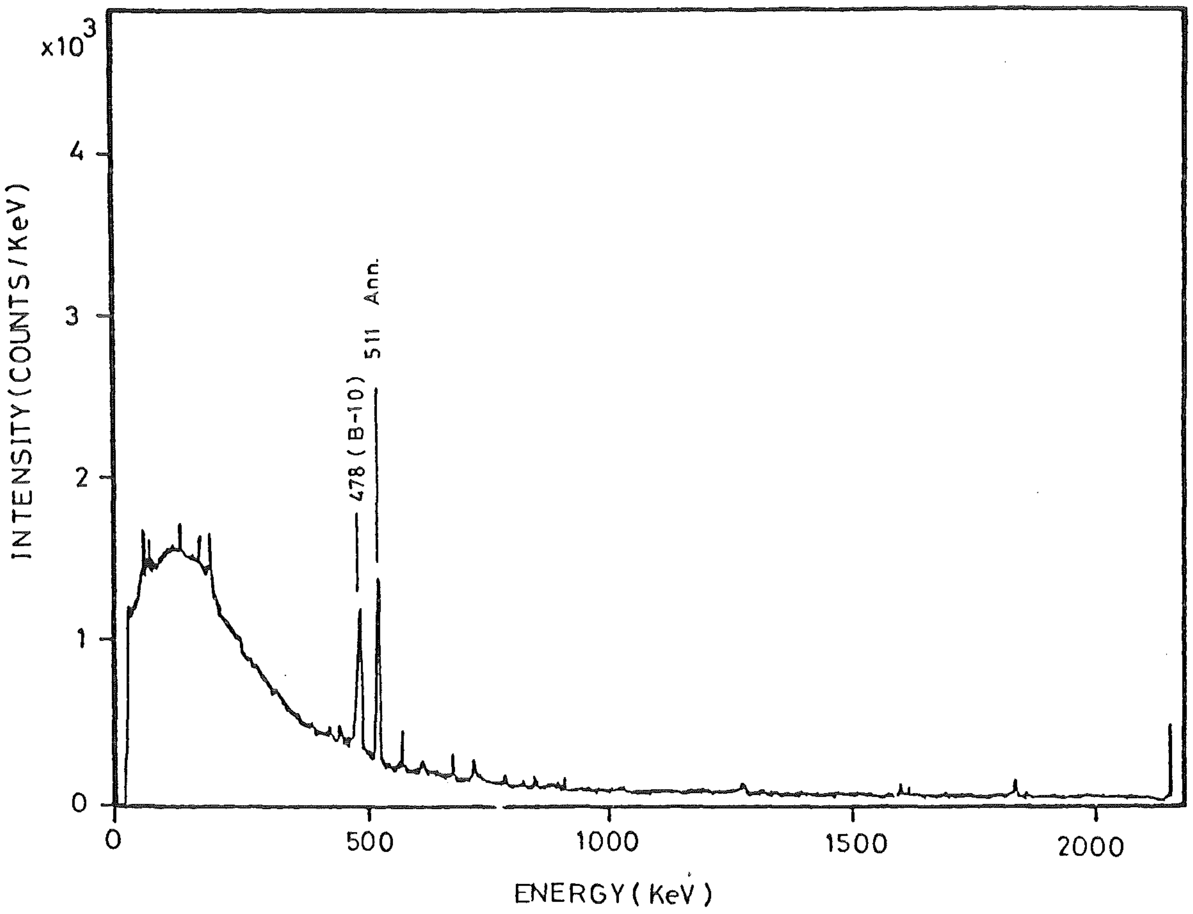

Fig.(14) Anticompton prompt neutron gamma - ray spectrum of Boron sample.

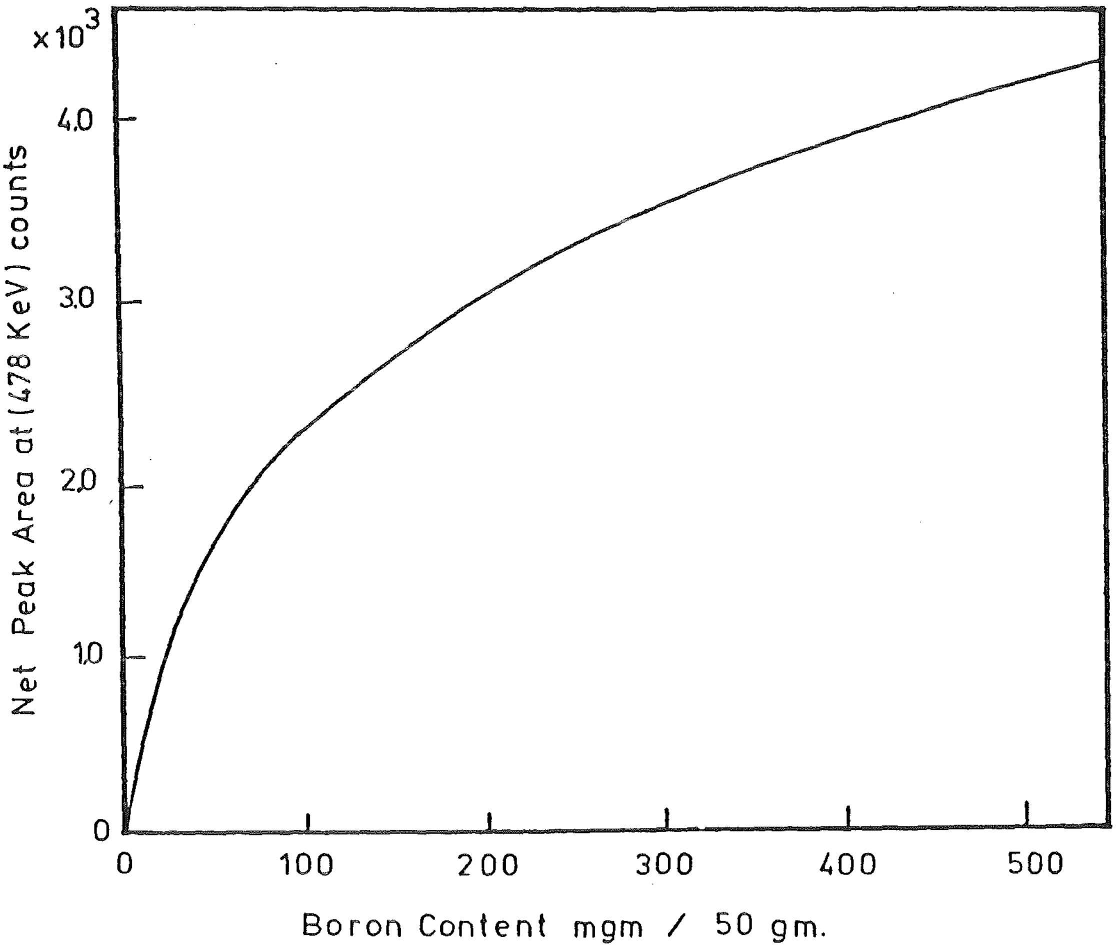

Fig.(15): Calibration curve for Boron determination.

encapsulated in a 50 ml polyethylene bottle and irradiated in the PGNAA facility for 30000 sec. Fig.(16) represents the AC-mode spectrum of the black sand sample. The result indicates that more than 15 measured elements which are Ti, Si, Th, Sm, Gd, Cd, Ba, Ca, Na, Tb, Ni, Ce, Nd, In, Fe, Pr and Au as a trace element have been found. Table(2) describes the neutron capture gamma-ray energies observed in the black sand sample and the identified elements.

4.2. Analysis of liquid sample:

Prompt gamma-ray neutron activation analysis measurement of water sample was carried out by means of AC-PGNAA system. The samples have been collected from Manzala-lake at the east north of Nile Delta, Egypt. The samples which have been properly preserved in a 50 ml polyethylene vial, has been irradiated for 30000 Sec. Fig.(17) shows the anticoincidence neutron capture prompt gamma-ray spectrum which collected by AC-array detection system. The measurement indicate the presence of more than 20 elements in the water sample, as indicated in table (3) which are Ca, K, Na, Cl, Ba, Mn, Cu, Cd, Zn, Pd, Gd, In, Pr, Ge, Ir, Sm, I, Ti, Si, and Au. From these statements of analysis, it is evident that the present system AC-PGNAA can be used for multielemental analysis for both solid and liquid environmental samples.

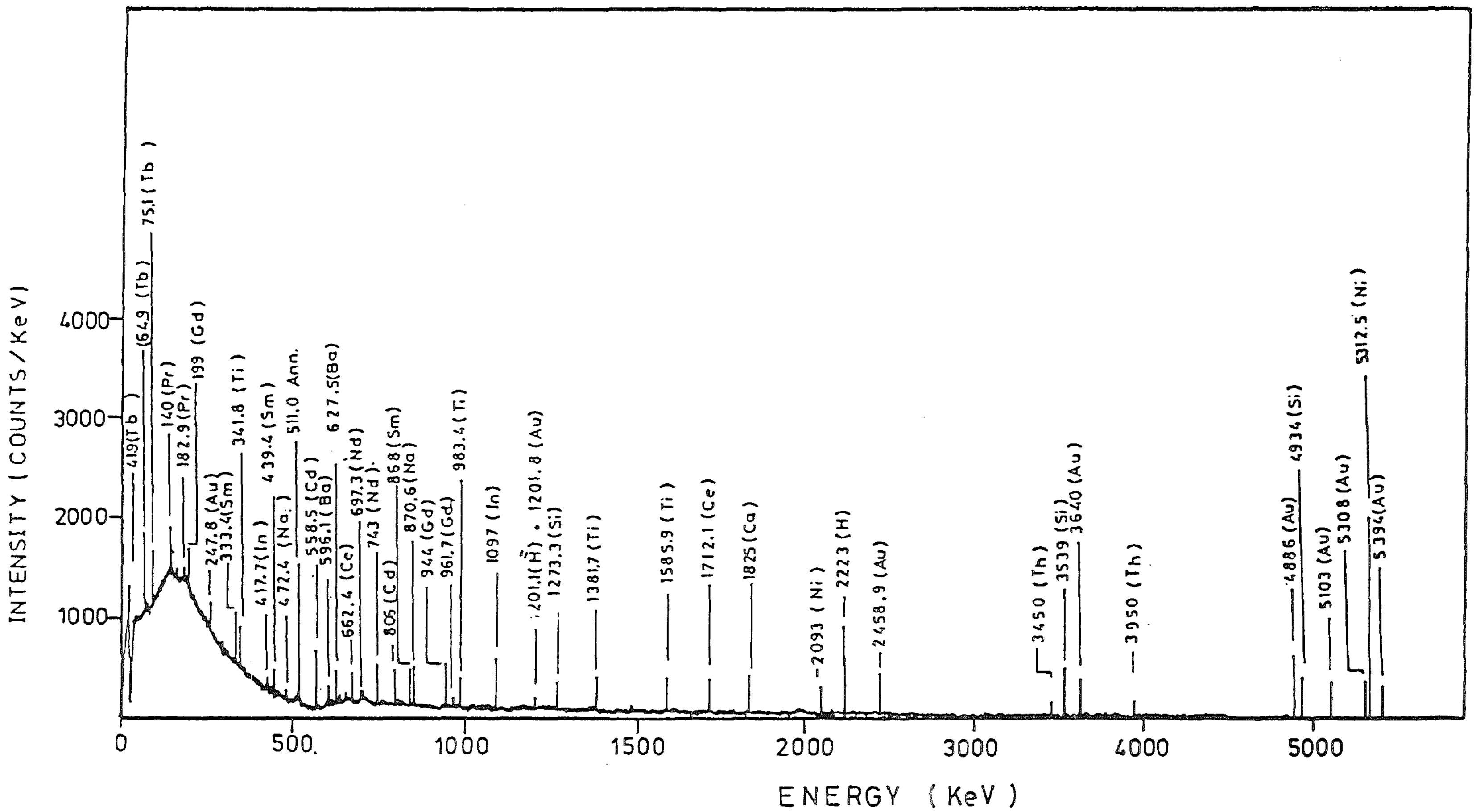

Fig. (16) Anticompton prompt neutron gamma-ray spectrum of black sand sample.

Table (2)

Identified elements and its neutron capture gamma-ray energies in black sand sample

Element	Energy (KeV)
Ti	341.8, 983.4, 1381.7
Si	1273.2, 3539, 4934
Th	3450, 3950
Sm	333.4, 439, 868.4
Gd	944.2, 961.7
Cd	558.5, 651.0, 806.
Ba	596.1, 627.5
Ca	1825
Na	472.4, 870.6
Tb	41.9, 64.9
Ni	339.5, 465, 5312.3, 6583.6
Ce	439.9, 662.4
Au	247.8,2458.9, 3640, 4886
Nd	697.3, 743.0
In	417.7, 1097
Pr	140, 182.9
Fe	6018.5, 6573.1, 6762.3

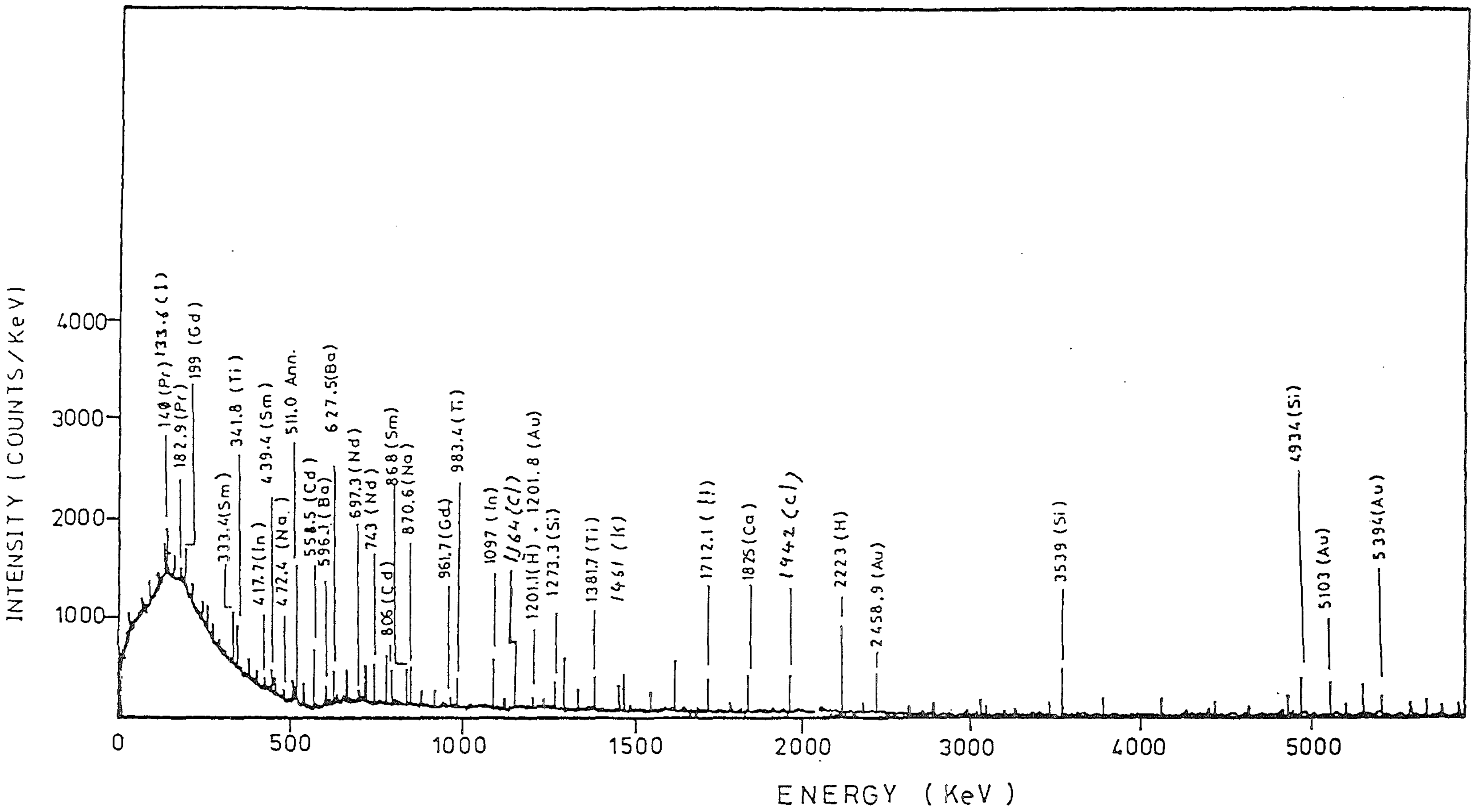

Fig.(17) Anticompton prompt neutron gamma-ray spectrum of water sample of local Manzala lake

Table (3)

Identified elements and its neutron capture gamma-ray energies in Manzala-lake water sample.

Element	Energy (KeV)
Ca	520, 726.9, 1942
K	1461, 770.2
Na	472.4, 870.6
Cl	1164, 1618, 1942
Ba	627.5, 1435
Ir	351.0,
Ge	500.2, 3028
Pr	141.2, 177.2, 4801
Mn	7058
In	1097, 1293.5, 417.7
Cu	6575., 7252., 466.0
Co	320.0, 5659
Sm	333.4, 439.4, 868.
Cd	558.5, 806.,
Zn	6655, 6910
Gd	199., 961.8,
Pb	6736, 7367.7
I	133.6
Ti	341.8, 983.4, 1381.7
Si	1273.3, 3539., 4934
Au	1201.8, 2458.9, 5103, 5394

5. CONCLUSION:

The anti-compton gamma-ray spectrometer is built in conjunction with the prompt gamma-ray neutron activation analysis (AC-PGNAA) facility to provide simultaneous anticoincidence spectrometry of natural, industrial and environmental samples. The HPGe-NaI(Tl) anticompton array is mainly to improve the sensitivity for the measurement of both coincidence and non-coincidence gamma-ray emitting radionuclides. This facility is principally constructed for the measurement of prompt gamma-ray spectra by means of the internal target geometry.

The sensitivity of the (AC-PGNAA) technique is limited by the available neutron flux at the target matrix and the neutron absorption cross-section of the elements of interest. The well shielded AC-array has succeeded in improving the sensitivity of both single and anticompton gamma-ray measurements to a considerable degree. Moreover, the peak-to-compton ratio for the gamma-ray spectrum of Cs-137 has been improved from the ratio (63:1) for single spectrum measurement to (500:1) in the case of the anti-coincidence mode with compton suppression factor of about 8 at the compton edge. While in case of Co-60 gamma-ray spectrum, the (P/c) ratio is (528:1) instead of (60:1) to give compton suppression factor equal to 8.8 at the compton edge peak. The sensitivity of the facility has been investigated quantitatively through the analysis of the boron standard samples, and it was found to be less than 5 ppm for boron in its solid samples.

PGNAA technique has the advantage to estimate the constitute elements which are difficult to be measured through the delayed gamma-ray measurements, such as B, Bi, C, H, P, Tl, Be, Cl and S in industrial and reference materials and those elements which are transformed into either stable isotopes or with very short half-lives when undergoing neutron capture. However, this new facility seems to offer interesting possibilities for the determination and measuring the elements content in different samples with high accuracy in the absence of the reactor as a powerful irradiated neutron tool. To ensure this fact, samples of black sand sample, Manzala-like lake water sample have been analysed by the system successively.

The success of planning and construction of this unit is considered as a real achievement with regards to the accommodation of nuclear technology in the scope of monitoring the nuclear fuel cycle and the early detection of criticality. Besides, services are forward for conducting micro-analyses of radioactive wastes and for the determination of tracer elements and the measurement of the degree of environmental radioactive pollution.

ACKNOWLEDGEMENT:

This work was financially supported by the International Buro, KFA, Juelich, Germany, as a part of the cooperative research program of the IRCH-KFK, Karlsruhe and Hot Lab. Center, AEA, Cairo,Egypt. Deep thanks for Dr.E.Gantner for his fruitful help and discussion during preparing this manuscript.

6. REFERENCES

1- H. Hoetzl, R. Winkler, IAEA-SM-252/59, 77 - 91, Proc. Int. Symposium, Berlin, April 6 - 10 (1981), IAEA, Vienna

2- N. A. Wogman, IAEA-SM-252/59, 15 - 29, Proc. Int. Symposium, Berlin, April 6 - 10 (1981), IAEA, Vienna

3- J. Bleck-Neuhaus, U. Boikat, R. Goedecke, W. Herzer, R. Wehrse, IAEA-SM-252/59, 31 - 40, Proc. Int. Symposium, Berlin, April 6 - 10 (1981), IAEA, Vienna

4- G. D. Dracoulis, Nucl. Instrum. Methods 187 (1981), 413

5- R. Holzmann, I. Ahmad, R. V. F. Janssens, T. L. Khoo, D. C. Radford, M. W. Drigert, U. Garg, Nucl. Instrum. Methods A 260 (1987), 153

6- G. J. Hunt, M. C. O'Riordan, P. D. J. Whetmath, Nucl. Instrum. Methods 156 (1978), 573

7- R. Beetz, W. L. Posthumus, F. W. N. DeBoer, J. L. Maarleveld, A. Van Der Schaaf, J. Konijn, Nucl. Instrum. Methods 145 (1977), 353

8- J. A. Cooper, R. W. Perkins, Nucl. Instrum. Methods 99 (1972), 125

9- J. A. Cooper, L. A. Rancitelli, R. W. Perkins, J. Radioanal. Chem. 6 (1970), 147

10- D. J. G. Love, P. J. Nolan, P. J. Smith, D. M. Todd, Nucl. Instrum. Methods A 239 (1985), 639

11- Y. Yusaka, H. Tsuji, J. Radioanal. Chem. 5 (1970), 359

12- T. C. Clark, R. P. Gardner, K. Verghese, Nucl. Instrum. Methods 193 (1982), 365

13- T. Lukander, S. Unsitalo, 3^{rd} Int. Symp. on (n,γ) and related Topics, 681 - 683, BNL, Upton, New York, Sept. 18 - 22 (1978)

14- G. Subrahmanian, G. Venkatraman, U. Madhvanath, IAEA-SR-3/8, 207 - 236, Vienna (1976), Proc. Educ. Seminar, Karlsruhe, April 14 - 18 (1975)

15- S. M. Lombard, T. L. Isenhour, Anal. Chem. 40 (1968), 1990