

RESPONSE CHARACTERISTICS OF A NEUTRON CAPTURE GAMMA-RAY FACILITY FOR ELEMENTAL ANALYSIS OF INDUSTRIAL MATERIALS

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الخلاصة :

تم تصميم وإعداد ومعايرة مطياف أشعة جاما الأُسري للنيوترونات بغرض تحليل الخامات الصناعية بطرق التفاعلات النووية باستخدام مصدر نيوترونات نظائري من نوع (أماريسيوم ٢٤١ / بريليوم) ذي شدة مقدارها (٥ كوري) . وقد صمم المطياف أساساً لقياس الطيف الجامي الفوري نتيجة لأُسْر النيوترونات الحرارية بنويات عناصرها ، حيث استخدم لذلك الغرض كاشف من (الجرمانيوم) بالغ النقاوة .

ولاجراء معايرة المطياف من ناحية الطاقة والكفاءة أمكن استخدام عدة خطوط جامية واضحة ومفصولة جيداً لعدة عناصر لعينات من كلوريد الصوديوم والحديد والتي أُعدت حتى (١٠) مليون الكترون فولت . وقد أمكن الاستفادة في هذه المعايرة من ظاهرتي الهروب الفردي والهروب الزوجي وخاصةً في حالة الطاقات العالية (٢ - ١٠ مليون ، الكترون فولت) الى جانب الطاقة الكلية لكل خط جامي .

ABSTRACT

A neutron capture gamma-ray spectroscopy facility has been setup for analysis of industrial materials using a 5 Ci ²⁴¹Am/Be neutron source. The facility is principally designed and calibrated for measurement of the prompt gamma-ray spectra obtained due to thermal neutron capture which is recorded by means of a hyper pure Germanium detection system.

For gamma-ray energy and efficiency calibration, some selected gamma-ray lines obtained from the neutron capture gamma-rays in NaCl and Fe ore samples are used up to 10 MeV. The response characteristics of gamma-ray full energy peak, single escape peak, and double escape peak detection mechanisms were deduced.

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INTRODUCTION

The increasing need for *in-situ* composition analysis of industrial samples has led to the development of various applied techniques. Several different methods are used for the determination of trace, minor, and major elements in these complex samples. Neutron interaction techniques using isotopic neutron sources and solid-state detection systems are extensively applied [1–13]. The advantages of these methods over others are the speed of analysis, the large sample sizes which can be used, and high reliability.

In principle, the gamma-rays emitted by an element after thermal neutron absorption fall into two classes, prompt and decay gamma-rays. The measurement of the characteristic gamma-rays emitted by particular nuclei provides information on nuclear data such as the level schemes and can provide a non-destructive analytical technique. In order to perform an analysis of the constituents of bulk samples, the mode of interaction between thermal neutrons and the nuclei in the sample can be used. The sample is bombarded by a flux of neutrons and the resulting prompt gamma-ray spectrum is recorded. Each element will emit one or more gamma-rays with energies which are unique to that element. By knowing the cross-sections for the nuclear reaction process in the different elements, and the efficiency of the detection system for gamma-rays, the percentage of each of the elements making up the bulk material can be determined. The precision of the analysis depends mainly on the number of counts obtained for each gamma-ray line, as well as the precision to which the cross-sections are known.

Also, the net number of counts in the peak areas of selected gamma-ray lines can be plotted against known concentration of the elements under investigation. This could be an aid for the determination of the concentration of the elements in unknown samples under the same conditions.

In this work a description is given of a $^{241}\text{Am}/\text{Be}$ isotopic neutron source together with a hyper-pure germanium detection system. This system is designed as a bore-hole logging system and is calibrated in the laboratory using a NaCl solution and domestic hematite ore samples.

SOURCE-DETECTOR ASSEMBLY AND MEASUREMENT GEOMETRY

The source-detector assembly is in the form of a bore-hole probe positioned in the center of a wooden box of 0.5 m^3 volume and wall thickness of 20 mm as shown in Figure 1. It consists of a polyethylene tube

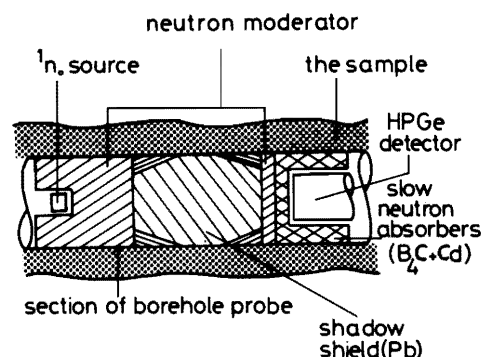


Figure 1. A Schematic Diagram of the Borehole Assembly.

with dimensions of 800 mm length, 120 mm inner diameter, and wall thickness of 2 mm. A hyper-pure germanium detector (HPGe) is located at one of its ends. The detector is surrounded by a screen containing B_4C in a plastic jacket with a thickness of 25 mm, lined with a 2 mm thick cadmium sheath. This screen is designed to protect the detector from scattered neutrons. A block of lead of 300 mm length and diameter of 140 mm, in a cylindrical form with two conical ends, as shown in Figure 1, is used as a shadow shield between the neutron source and the HPGe detector. It is designed to protect the detector from direct radiation from the neutron source. A cylinder of paraffin wax of 300 mm length and 140 mm diameter with a hole 120 mm in depth and 80 mm in diameter is used as a container for the neutron source positioned at the other end of the polyethylene tube (Figure 1).

A $5\text{ Ci }^{241}\text{Am}/\text{Be}$ neutron source doubly encapsulated in a vacuum tight stainless steel cylinder and surrounded by a cylindrical lead container of 60 mm diameter and 100 mm length, is placed in the hole in the paraffin wax cylinder (Figure 1). The distance between the neutron source and the detector, about 650 mm, was chosen so the neutron flux density at

the detector position would be negligible. Measurements by the foil activation technique verified that the detector was exposed to a suitably small number of scattered neutrons. The paraffin wax cylinder is used for neutron thermalization and the wooden box is used as a sample container.

SAMPLE PREPARATION

The wooden sample container is filled with the samples under investigation. Polyethylene bottles or polyethylene bags are used for liquid and solid samples, respectively. Solid samples should be ground to about 0–1 mm granule size and mixed very well. For liquids, the material used should be dissolved completely in order to have a homogeneous distribution of the elemental constituents.

Different amounts of NaCl samples were well dissolved in tap water in order to prepare solutions with the following chlorine concentration values, 0.0, 0.925, 1.850, 3.700, and 7.400 percent by weight. About 200 l of each sample was used for the measurement of the prompt gamma-ray spectra using the facility described above.

A domestic hematite ore sample from the Shumaysi formation which lies in the valley called Wadi Fatima and around the village of Shumaysi (20 miles east of Jeddah), was used in this study [14]. It was of sufficient size to permit total attenuation of the incident neutrons. The wooden box sample container, shown in Figure 1 was filled with the hematite sample in polyethylene bags surrounding the source-detector tube.

GAMMA-RAY DETECTION SYSTEM

The prompt gamma-ray spectra obtained are detected by using a hyper-pure germanium detector of 15% relative efficiency. The energy resolution is 2 keV for the 1.33 MeV gamma-ray line of ^{60}Co . Signals from the detector were passed through an FET preamplifier and amplified by a spectroscopy amplifier. The amplified signals are fed directly to the analyzer in a personal computer system of a 8192 channel spectrum.

ENERGY CALIBRATION

A group of well resolved and intense prompt gamma-ray lines of full energy, single and double escape peaks of ^{36}Cl from the reactions $^{35}\text{Cl}(n,\gamma)^{36}\text{Cl}$ and $^{23}\text{Na}(n,\gamma)^{24}\text{Na}$ using NaCl sample as well

as some other gamma-ray lines of Fe from the hematite ore sample, in addition to the gamma-ray lines of H and Pb observed as background in the spectra obtained, were used to establish the low and high energy scales up to about 10 MeV.

Table 1 shows a list of the gamma-ray energies used for energy calibration. A simple computer program, prepared for this purpose with an accuracy of 0.10 keV, determined the energy of the peaks.

Table 1. Gamma-Ray Energies Used for Energy and Efficiency Calibration Curves (Taken from References [5, 6, and 13]). The Single and Double Escape Peaks are Labeled by ' and '' Respectively.

Source	E_γ , keV	Source	E_γ , keV
Annihilation	511	Fe'	5508
Cl	518	Cl	5715
Cl	789*	Fe	5921
Na	870*	Fe	6019
Cl	1165*	Cl	6111
H''	1201	Pb''	6346
H'	1712	Fe''	6610
Fe	1725*	Pb'	6857
Cl	1959*	Cl	6978*
Na	2027	Fe'	7121
H	2223	Fe'	7134
Cl	2676*	Pb	7368
Cl	2864	Cl	7414
Cl	3062*	Fe	7646*
Fe	3268	Cl	7790*
Fe''	4219*	Fe''	8276
Fe''	4899	Cl	8579
Fe''	4997	Fe'	8787
Fe	5410	Fe	9298*

*The gamma-ray lines used for the efficiency calibration.

EFFICIENCY CALIBRATION

As mentioned above it is necessary to measure the relative efficiency for elements being analyzed accurately. Accordingly, some selected gamma-ray lines of Cl, Na, and Fe obtained in these measurements were used to plot the relation between the relative full energy peak efficiency as a function of gamma-ray energy using the peaks labeled by asterisks in Table 1. The following relation could be applied for determination of the relative efficiency for the full energy peaks with the HPGe detection system [12]:

$$\mathcal{E}_{(i)}^{(n)} = (A_{(i)}/A_{(n)})/(\bar{I}_{(i)}/\bar{I}_{(n)}) \quad (1)$$

where:

- $\mathcal{E}_{(i)}^{(n)}$ is the efficiency at energy $E_{(i)}$ relative to that at $E_{(n)}$;
- $A_{(i)}$ and $A_{(n)}$ are the full energy peak areas belonging to the i th and n th transitions respectively;
- $\bar{I}_{(i)}$ and $\bar{I}_{(n)}$ are the weighted average intensities of the i th and n th transitions respectively.

The error in $\mathcal{E}_{(i)}^{(n)}$ includes the errors of $\bar{I}_{(n)}$ and $I_{(i)}$, the statistical errors of $A_{(n)}$ and $A_{(i)}$ and the estimated error under the peaks i and n . The full energy peak relative efficiency curve for the (HPGe) detector derived from the gamma-ray lines of Na, Cl, and Fe up to 10 MeV is shown in Figure 2.

For high energies (above 3 MeV) the single-escape and double-escape peaks are more intense

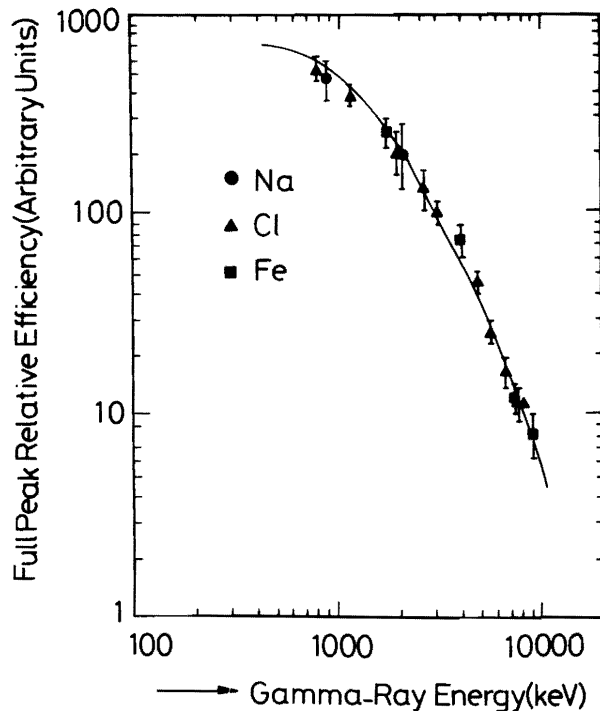


Figure 2. Full Peak Relative Efficiency for the (HPGe) Detector Derived From the Prompt Gamma-Ray Spectra of NaCl and Hematite Ore Samples in the Arrangement Shown in Figure 1. Some selected gamma-ray lines of NaCl and Fe are used. The gamma-ray line at 3062 keV is taken as 100.

than the corresponding full energy peak. Accordingly, the use of these peaks permits analysis with greater sensitivity. To establish the relative efficiency curves of single-escape peaks and double-escape peaks, a study of the ratio of their counts to full energy counts at different energies was made; these ratios are shown in Figure 3. By utilizing the full energy peak relative efficiency, the single-escape/full energy peak ratio and the double-escape/full energy peak ratio as a function of energy, it was possible to construct the relative efficiency as a function of energy for the double and single escape peaks as shown in Figure 4.

QUALITATIVE AND QUANTITATIVE ANALYSIS

(a) NaCl Solution Sample Investigation and Chlorine Determination

Different amounts of NaCl were dissolved in tap water to prepare solutions with chlorine concentration of 0.0, 0.925, 1.85, 3.7, and 7.4 percent by weight. A quantity of 200 l of each sample was used for measurement of the prompt gamma-ray spectra using the facility described above for 3600 s counting time. Figure 5 shows a portion of a typical spectrum for a sample with 7.4 percent chlorine by weight in the energy region from 780 to 2230 keV. The spectra recorded for the different samples under investigation could range up to 9000 keV. The single and double escape peaks at high energies (above 3 MeV) are very prominent. The main background lines of H, C, Fe, Pb, and Cd are evident in addition to chlorine and sodium gamma-ray lines. More than 30 resolved gamma-ray lines of chlorine were observed. This was due to the ^{35}Cl isotope which has an abundance of 75.7% in natural chlorine and a high cross-section for thermal neutron capture (43 b). The ^{36}Cl gamma-ray lines are easily observed in NaCl spectra because sodium has a low capture cross-section (0.534 b). Gamma-ray lines emitted through the reaction $^{23}\text{Na}(n, \gamma)^{24}\text{Na}$ were observed in the spectra but with very low statistics. Only the strongest sodium lines, at 870 and 2027 keV, were used in the energy and efficiency calibration of the system.

The well-resolved lines of chlorine at 1165, 4980, and 6978 keV were used to establish the set of calibration curves shown in Figure 6 which could be used for determination of chlorine under the same experimental conditions. Due to self-absorption, a saturation region appeared at high concentration values (more than 5%).

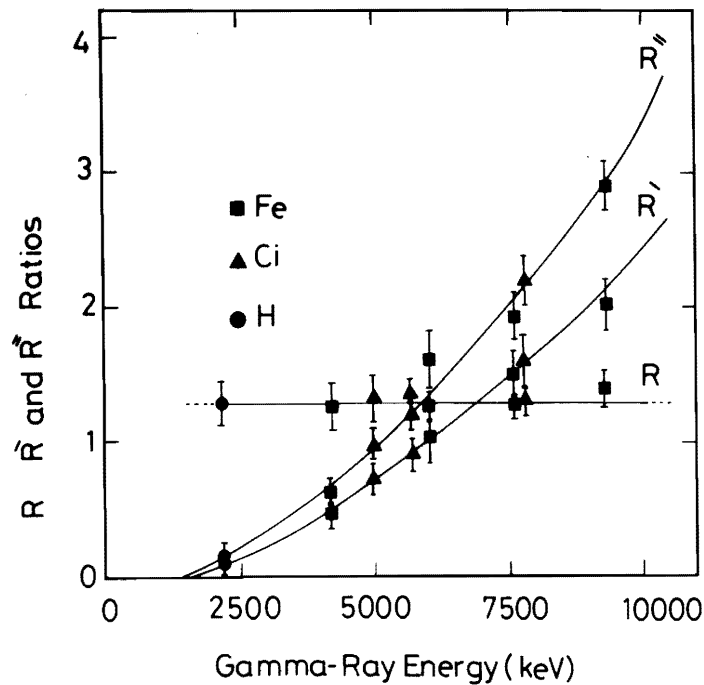


Figure 3. The Ratios (R'' , R' , and R) for the (HPGe) Detector as a Function of Energy Using the Prompt Gamma-Ray Spectra of NaCl and Hematite Ore Samples in the Arrangement Shown in Figure 1.

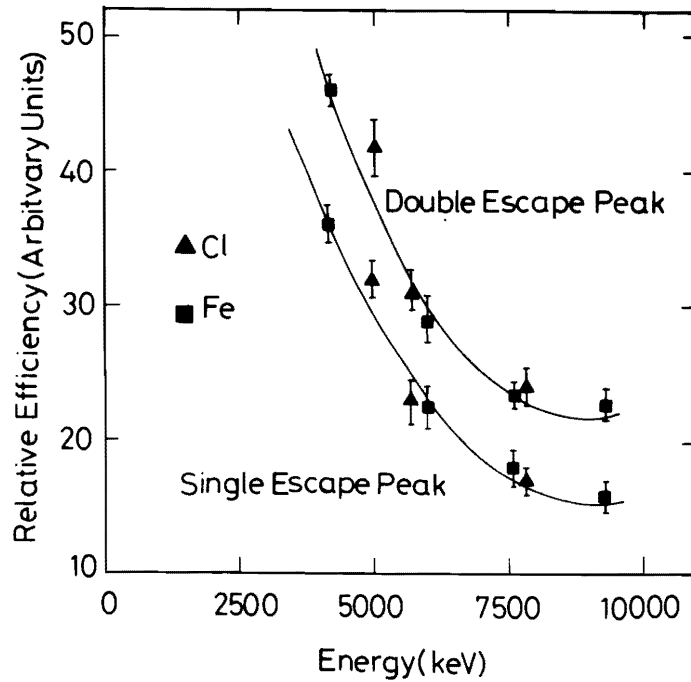


Figure 4. Relative Efficiency Curves of the (HPGe) Detector for Single and Double Escape Peaks Using the Prompt Gamma-Ray Spectra of NaCl and Hematite Ore Samples in the Arrangement Shown in Figure 1.

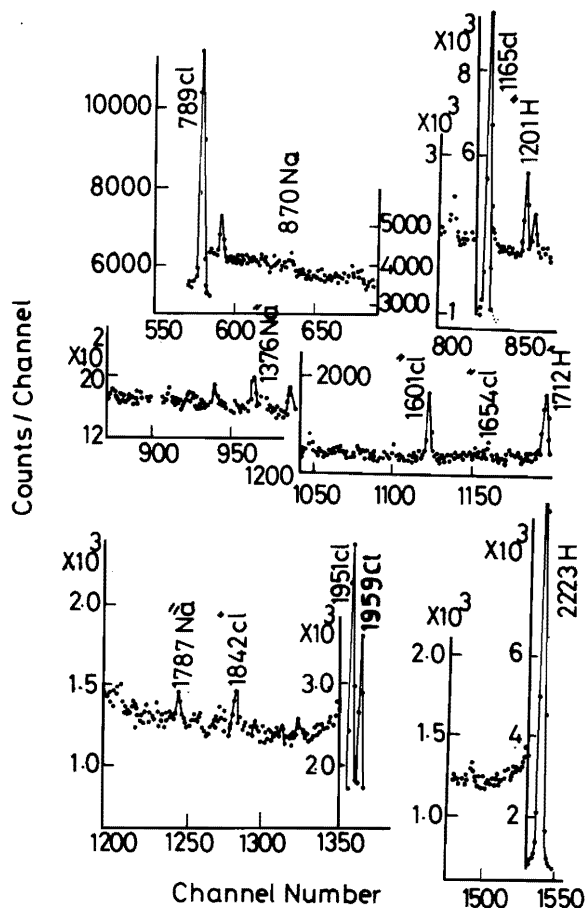


Figure 5. A Portion of Prompt Gamma-Ray Spectrum due to Thermal Neutron Capture in NaCl Sample (Concentration 7.4%) in the Energy Range 780–2230 keV. The irradiation time was 3600 s. The single and double escape peaks are labeled ' and " respectively.

(b) Hematite Ore Sample Investigation

The prompt gamma-ray spectrum of a hematite ore sample of about 160 kg was collected in 3600 s counting time. A total of about 100 gamma-ray lines could be identified for Fe, Al, Si, Cl, Ti, Mn, Mg, Na, V, Cr, P, K, Ni, Mo, Zn, S, and Cu, in addition to the background lines. Most of these elements are not monoenergetic gamma-ray emitters and they produce several gamma-rays of different energies. In the high energy (above 3 MeV) the single and double

escape peaks are evident. The large number of peaks makes the gamma-ray spectra of the samples very complex. Figure 7 shows a portion of the hematite prompt gamma-ray spectrum in the region between 6600 to 7650 keV.

In order to determine the elemental concentrations the following relationship was used [1, 9]:

$$C_j = K \frac{A_j R_{ij}}{\mathcal{E}(E_{\gamma}^{ij}) I_{(ij)} \sigma_j}, \quad (2)$$

where:

- C_j is the mass concentration of element j ;
- K is a normalization constant chosen such that $\sum C_j = 100$ and \sum refers to the summation over those elements measured;
- A_j is the atomic mass of element, j ;
- R_{ij} is the detector response (counts in the γ -ray peak to gamma-rays within a γ -ray group i from element j);
- $\mathcal{E}(E_{\gamma}^{ij})$ is the relative detector efficiency for gamma-rays of energy (E_{γ}^{ij});
- I_{ij} is the number of gamma-rays in energy group i emitted by element j per 100 neutrons captured [5, 6];
- σ_j is the neutron capture cross-section for element j .

The preliminary calculated concentrations in percent by weight for some elements in hematite ore samples are as follows: (Fe) 36.900 ± 2.130 ; (Al) 0.950 ± 0.110 ; (Mn) 0.680 ± 0.050 ; (Ti) 0.090 ± 0.0150 ; (V) 0.042 ± 0.0100 ; and (Na) 0.850 ± 0.150 .

The errors in these measurements include: the statistical error of the line areas, the relative efficiency values, the intensity of the gamma-ray lines used and the cross-section values. The complete analysis of the domestic hematite ore sample will be published elsewhere.

CONCLUSION

A neutron capture gamma-ray spectroscopy facility for analysis of industrial samples using a $5 \text{ Ci } ^{241}\text{Am/Be}$ neutron source has been designed and calibrated for energy and efficiency values up to 10 MeV. In principle this facility can be used in the laboratory and in the field as a useful tool for rapid analysis of bulk complex samples. Sensitivity curves using different concentration values of Cl have been constructed. The preliminary determination of some elemental constituents of domestic hematite ore were obtained.

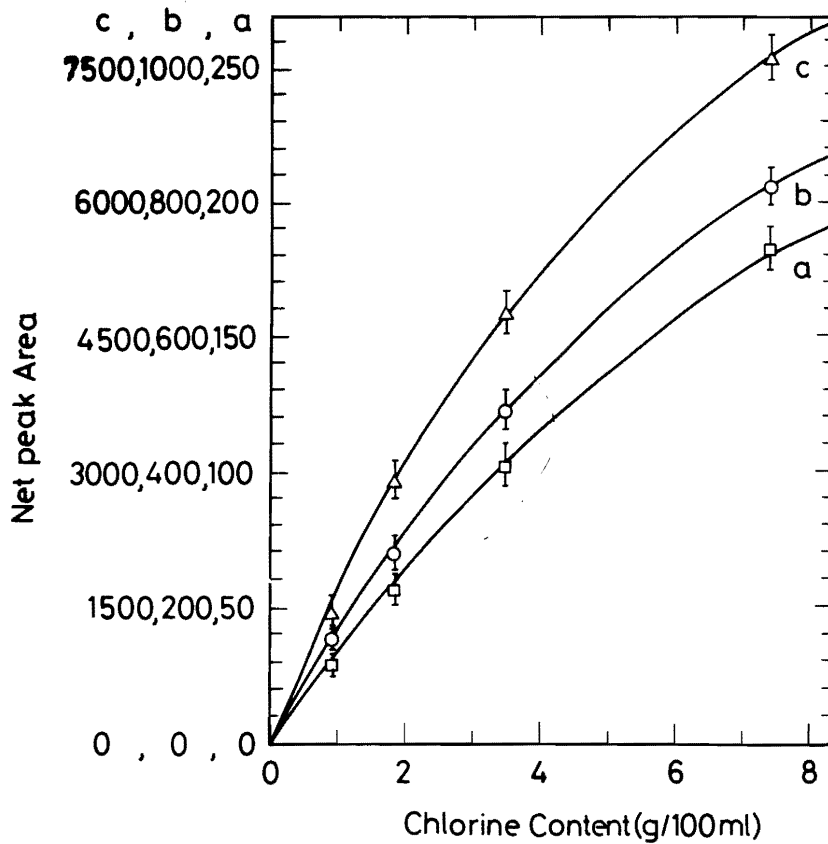


Figure 6. Calibration Curves for Chlorine Determination in NaCl Using the Net Peak Area of the Prompt Gamma-Ray Lines at (a) 1165, (b) 4980, and (c) 6978 keV Against the Chlorine Content. The data is corrected to the tap water background. The irradiation time was 3600 s.

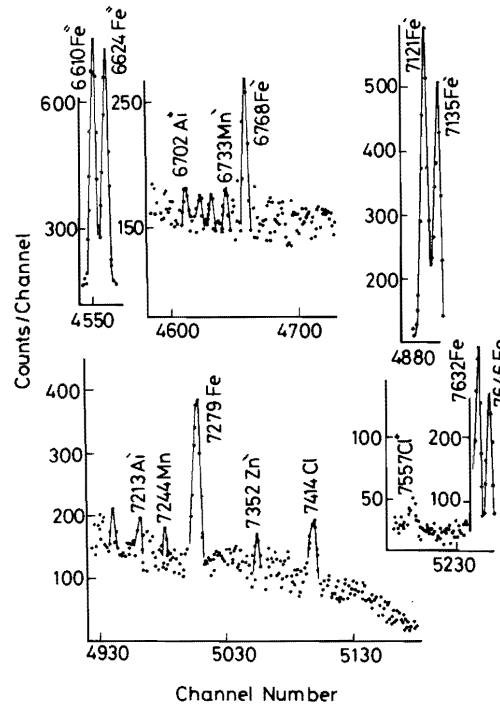


Figure 7. A Portion of Prompt Gamma-Ray Spectrum due to Thermal Neutron Capture in Hematite Ore Sample in the Energy Range (6600–7650 keV), Irradiation Time was 3600 s. The single and double escape peaks are labeled by ' and '' respectively.

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