

PROMPT GAMMA-RAY ACTIVATION TECHNIQUE FOR IN-SITU ANALYSIS OF MERCURY POLLUTION IN WATER

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ABSTRACT

Industrial and mining pollutants discharged into water are in general distributed homogeneously and we investigated a prompt neutron activation technique for the in-situ analysis, to start with of Hg content in water. The laboratory test employed a ^{252}Cf neutron source (of $\sim 3 \times 10^6$ n/s fluence) submerged in a test tank of water of ≈ 500 litres, and to monitor the gamma-ray emission a $4'' \times 4''$ NaI (Tl) detector system was employed. In 3000 s time interval trials, for a 46 ppm contamination level of Hg, we observed an excess of counts of $\sim 9.2 \sigma$ significance in the energy range of 4000-6500 keV, which can be attributed to the presence of mercury. This test system for a 10 hour monitoring can provide a minimum detectable sensitivity (MDS) at 4.78 ppm. In the future experiments, we propose to replace the NaI(Tl) detector by a HPGe detector to facilitate simultaneous analyses of pollutants such as cadmium, chlorine, chromium etc for detection at few tenths to tens of ppm levels or better.

INTRODUCTION

In-situ analysis of elemental composition by nuclear techniques has important applications in many areas of geochemical exploration. Especially, Prompt Neutron Gamma Activation (PNGA) techniques are being increasingly utilized in bore hole exploration in the oil-industry, the mineral industry and in the environmental monitoring [1-3]. In oil and mineral resources investigations the elemental distribution is uneven in the matrix, but in the case of industrial pollutants in water they are essentially distributed homogeneously and these techniques permit simple interpretation.

We investigated a prompt neutron-gamma technique by making measurements of the gamma-rays which result from the interactions of thermal neutrons with the trace elements under investigation. The γ - ray flux measured by the detector is dependent on the spatial and energy of the detector relative to the source, and the trace element content and distribution in the matrix of the material. A simple test detection system utilizing a ^{252}Cf neutron source and a NaI(Tl) detector was employed for the analysis of mercury trace component, so as to permit a design of a PNGA probe for the analysis of Hg, Cd, Cl, Cr, Mn, Fe and Cu elements.

ACTIVATION EXPERIMENT

In the PNGA experiment, our aim was essentially to concentrate on the emission of γ - rays above 3 MeV due to the presence of trace elements. The advantage is that the γ - rays of ≥ 3 MeV, due to natural radioactivity in the laboratory or in the field operation are negligible,

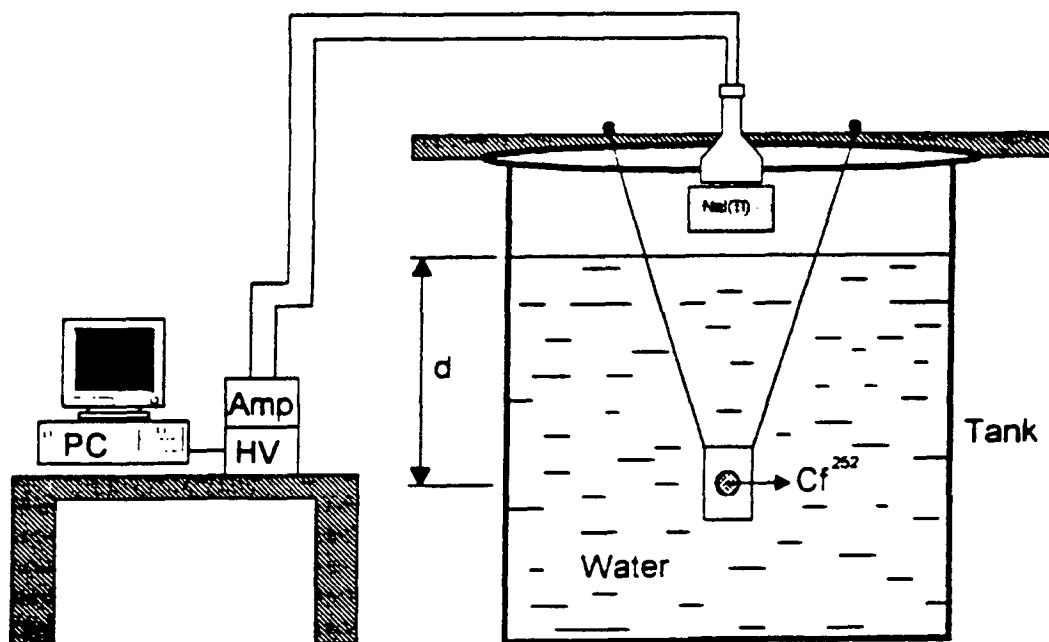


Figure 1: Schematic of the Test Detection System for Detection of Mercury.

and the event count background is practically due to the neutron source emission. Further, we adopted a simple experimental configuration, where in we exploited the water column itself as a shield against the neutrons and the γ - rays (≤ 500 keV) of the source reaching the detector. This represents for the test-system, better statistics and lesser dead-time for the electronic processing of the counts.

A ^{252}Cf isotopic neutron source, is ideal for the production of thermal neutrons in the field operations. Further, it has the highest specific neutron intensity and the lowest interfering γ - ray emission. For these thermal neutrons, Hg possess approximately an average cross-section $\bar{\sigma}_{th} \approx 376$ b and emits γ - rays in a wide energy range of 3000-6000 keV, with emission of ~ 13.85 % at 5966 keV [4].

The schematic arrangement of the test detection system for the in-situ PNGA is shown in figure 1. A water tank made of polythelene material, with a capacity to hold ≈ 500 litres, is utilized to simulate the testing in field conditions and was filled with regular tap water. The ^{252}Cf sealed neutron source of $\sim 21,1$ μg (with fluence of $\sim 3 \times 10^6$ n/s) placed in a water-tight polythelene capsule, was submerged into the water at a fixed depth of $d \sim 54$ cm from the water level. A NaI(Tl) detector of cylindrical shape of dimensions 4" x 4", was placed at a fixed distance of ~ 9 cm from the water level. For this configuration of neutron source-detector distance, essentially all neutrons (isotropical distribution inside water) travel ≈ 30 cm from the source, before suffering absorption and attenuation. A standard pulse processing equipment and a 512 channel pulse-height analyser(PHA), were employed to measure the prompt γ -rays.

RESULTS AND DISCUSSION

At the outset, the detector system was calibrated with laboratory standard sources and with the line emission at 2226 keV due to $\text{H}(n,\gamma)$ reaction inside the water. We obtained successively, γ - ray emission spectra for time intervals of 3000 s; first the background due to water alone and later with two different Hg concentrations of ~ 2 g and ~ 12 g in the water volume of ~ 380 litres. In table I, we present these observed counts in four broad energy intervals of 1400-3000 keV, 3000-4000 keV, 4000-6500 keV and 6500-8000 keV. In σ confidence levels, the excess of count rates in the case of Hg concentrations over the case of water alone are indicated. In measurements made for 3000 s time durations the same results shown in figure 2, clearly show the excess at ~ 9.2 σ significance in the energy interval of 4000-6500 keV, for the two trials with Hg content of 12 g. These excess are essentially due to γ - ray line emissions[4] at 5966 keV(13.80%), 5657 keV(6.16%), 5049 keV (5.02%), 4841 keV (5.0%) and 4739 keV(7.27%), and their Compton contributions. The lower energy intervals also had some γ - ray line emissions and Compton tail of all lines.

The two trials with Hg content of 12g in water of 380 litres correspond to Hg presence 46.05 ppm and show an average significance of 9.2 σ . This establishes for our test set-up, for the 3000 s time monitoring in the 4000-6500 keV interval, a minimum detection sensitivity (MDS)

Table I - Counts in the Experiment for the Detection of Hg.

Energy keV	Water	Water+Hg (2g)	Water+ Hg (12g) (Trial I)	Water+ Hg (12g) (Trial II)
1400-3000	2005412 ±1416	2000171±1414 (-2.62σ)	2012642±1419 (3.61σ)	2016937±1420 (5.75σ)
3000-4000	108011 ±329	108566±329 (1.19σ)	109344±331 (2.86σ)	110276±332 (4.84σ)
4000-6500	171189 ±414	172045±414 (1.46σ)	176602±420 (9.18σ)	176578±420 (9.14σ)
6500-8300	47487 ±218	46504±215 (-3.21σ)	46143±215 (-4.39σ)	47169±217 (1.03σ)

Obs: σ confidence levels indicate excess of counts over the counts due to water alone.

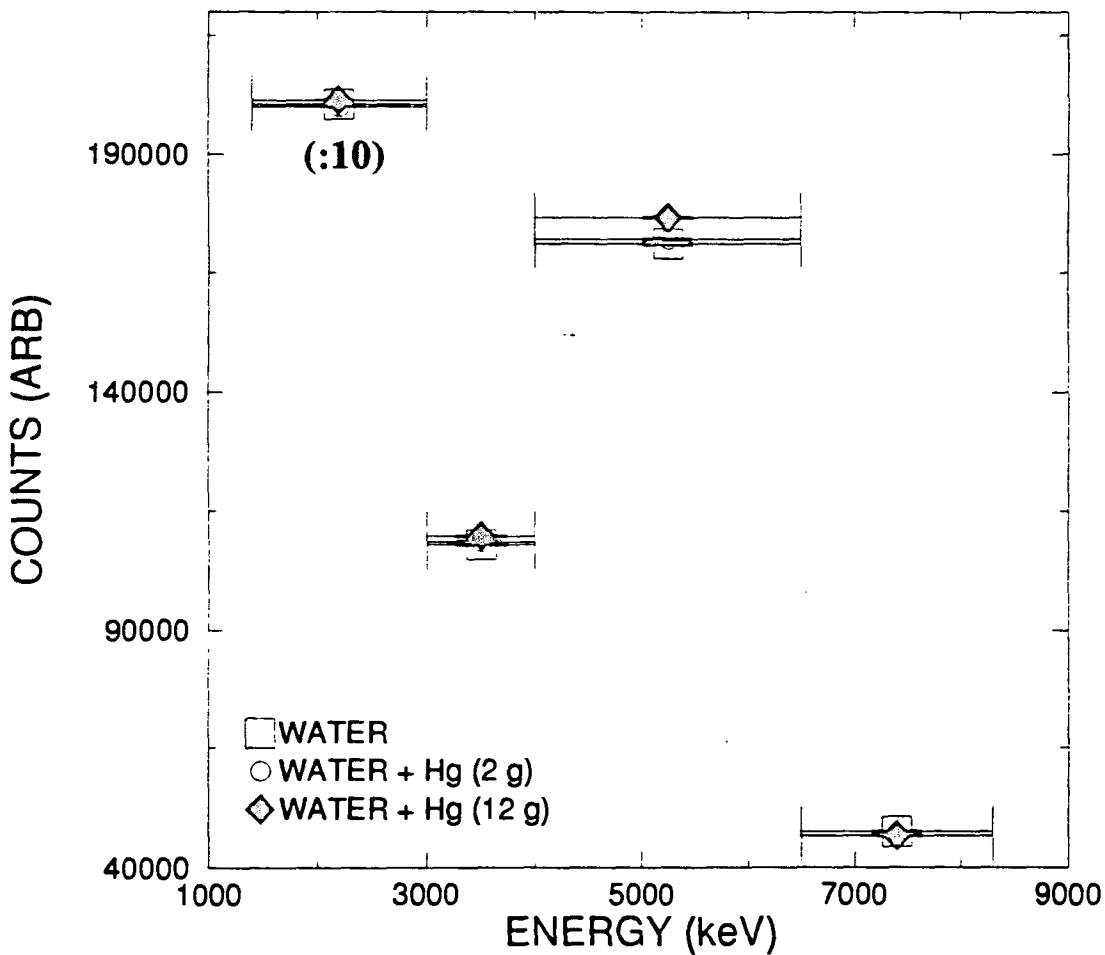


Figure 2: Gamma - Ray Fluxes Monitored in the Experiment for 3000 s. (The first energy interval counts were divided by a factor 10.)

of 14.25 ppm (for 3.28σ confidence) or for a 10 hour monitoring a MDS of 4.78 ppm.

This limit of MDS can be very much improved by increasing the strength of the source and or by increasing the time interval of monitoring. In the experimental set-up, we have not varied the detector-source distance which can also help increase the sensitivity. However, our test set-up is to help us to derive the MDS parameter and we envisage an order of magnitude of improvement by employing an n-type HPGe detector (where the damage to detector by neutrons is negligible) and monitor the lines directly. This is because, for the NaI(Tl) detector system, we employed a broad energy band of 2500 keV to define the MDS at the cost of high background statistics. If we replace the detector by a 75 cm³ HPGe, we can use line widths of 3-5 keV and consequently very less background for the counting statistics. Further, such a system permits a simultaneous detection of elements listed in table II. Taking the efficiencies of the detectors and the nuclear data parameters for the elements[5], the calculations indicate the possibility of detection of MDS at few tenths of ppm for Hg and Cd elements and few tens of ppm for Cl, Cr, Mn, Fe and Cu if present in water as pollutants.

Table II - Nuclear Data for Different Elements for Prompt Gamma - Rays .

Element	Average $\bar{\sigma}_{th}$ barns	Prompt γ - ray Energy (keV)	Prompt γ ($\gamma / (n, \gamma)$)
Cl	33.2	6111	0.20
Cr	3.1	8884	0.27
Mn	13.3	7244	0.12
Fe	2.55	7640	0.53
Cu	3.79	7914	0.31
Cd	2450	559	0.73
Hg	375	5966	0.14

REFERENCES

- [1] SCHWEITZER, J.S. Nuclear techniques in oil industry, Nuclear Geophysics, v.5, p.65-90, 1991.
- [2] EVANS, L.G., LAPIDES, J.R., TROMBKA, J.I., JENSEN, D.H. In situ analysis using neutron-capture gamma-ray spectroscopy, Nuclear Instruments and Methods, v.193, p.353-357, 1982.
- [3] JAYANTHI, U.B. GONÇALEZ, O.L., FIGUEIREDO, A.M.G., RIGOLON, L.S.Y., JAYANTHI, K.A. A neutron activation technique for the analysis of cryolite in core samples, Nuclear Geophysics, v.7, p.515-517, 1993.
- [4] LONE, M.A., LEVITT, R.A. and HARRISON, D.A. , Atomic data and nuclear data, v.26, p.556, 1981.
- [5] LEDERER, C.M., SHIRLEY, V.S., Table of isotopes. ed. Wiley, New York, 1978.