

# Determination of U, Th and K in Sediments and Fossil Collected from Mae Moh Mine Using Gamma-Ray Spectrometry and Neutron Activation Analysis (NAA)

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## ABSTRACT

The annual environmental dose in sediment and fossil shells matrices, collected from Mae Moh mine, Lampang province, Northern Thailand has been determined by using gamma spectrometry together with neutron activation analysis technique (NAA). This analysis technique was adopted by activating NBS standard reference materials (NBS SRM 2709: San Joaquin Soil) and sediment matrices at the same condition before measuring them at the specific time schedule. High Purity Germanium (HPGe) well type detector and spectrometer were calibrated to get efficiency ratio by reference standard sources. Most of natural radioactive elements are members of uranium, thorium and from non series nuclides, mainly potassium. The result concentration of uranium, thorium, and potassium in samples can be achieved by this technique and found to be in the range of 0.02 to 3.05 ppm, 0.04 to 11.00 ppm, and 0.01 to 1.53% respectively. The annual environmental dose is between 87-3000 mGy, depended on type of metrics, and can be specifically calculated and used for dating around the Middle Miocene time scale.

**Key words:** HPGe, NAA, Gamma-spectrometry, Uranium, Thorium, Potassium, Annual Dose

## INTRODUCTION

Most of natural radioactive elements are members of one of three radioactive series uranium, actinium and thorium and from non series nuclides, mainly potassium. These are natural sources of alpha, beta and gamma radiation that represent a continuous exposure of minerals (Hubert, 2001). If a mineral receives the natural radiation, some paired electrons are ionized, trapped by some impurities and the lattice defects are created which known as unpaired electrons.

The total unpaired electrons in matrices which were accumulated for a long time since they were formed, measured by electron spin resonance technique, (ESR) and using gamma ray doses as additive method are given to the sample to obtain the total dose by extrapolating the dose response of the signal intensity to the zero ordinate (Shimada, 2002). The annual environmental dose which is the most important factor in ESR dating can be obtained from the contents of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in archaeological and geological materials.

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Several methods are employed to determine the contents of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in archaeological and geological materials and to assess the annual dose rate. Each method has its own merits and convenience. One of them is gamma-ray spectrometry and neutron activation analysis (NAA) (Ikeya, 1993).

In Mae Moh mine, as well known for its lignite mining in the Lampang Province, northern part of Thailand, was a stratigraphic bedding plane in the steady sediment in geological time, about 3-31 million years ago (Ratanasthien, 2002). A research using magnetostratigraphic dating method (Benammid and *et al.*, 2002) has revealed that the age of sediments and fossils around coal zones was between 12.1-13.5 million years. Due to the available research reactor, facilities and gamma spectrometry with HPGe well type detector and GammaVision-32 V 3.2 Gamma-Ray Spectroscopy Software at Office of Atoms for Peace, (OAP) we could apply to determine the concentrations of U, Th and K in archaeological samples with ease and lead to evaluate the annual environmental dose for ESR dating. Our result then may be used as a reference data that can lead to ESR dating analysis within the Middle Miocene of the geological time scale.

## GEOLOGY

The Mae Moh mine is located in the Mae Moh District of Lampang Province, which is about 26 kilometers east of Lampang City. It is distributed in the area of about 135 square kilometers, 7 kilometers in east-west and 16 kilometers in north-south as shown in Figure 1. The basin floor is about 320-340 meters above mean sea level. The basin is an intermountain fault bound basin of a graben type containing Tertiary and Quaternary sediments underlain by basement rocks, including the Mae Moh Group. The Tertiary sediments have been named as the Mae Moh Group. The group consists of three formations,

namely the Huai King, Na Khaem, and Huai Luang formations, in ascending order. The total thickness is nearly 1,000 meters in ascending order (Figure 2) (Songthama, 2005).



**Figure 1** Map of Northern Thailand showing location of the Mae Moh basin with respect to locations of some provincial cities.

Thickness (m)	Formation	Log	Lignite Zone
100	Huai Luang		
200		RB	
300			I-Zone
400	Na Khaem	RB	
500			J-Zone
600		OB	K <sub>1</sub>
700		IB	K <sub>2</sub> Zone
800		UB	K <sub>3</sub> Q-Zone
900	Huai King		R-Zone
			S-Zone
	Lampang Group		

**Figure 2** Schematic lithostratigraphic units of the Mae Moh group.

The Huai King formation unconformable overlies the basement rock, Lampang group. It consists of a sequence of upward grading conglomerate, sandstone and pebbly sandstone, siltstone and finally fining upwards into interbedded red and gray claystone. A thin layer of coal named the S coal zone marks the uppermost part of the formation. The Na Khaem formation is a coal measure comprising three main coal zones, Q, K, and J assigned to Middle Miocene age. This formation has been divided into three members as Member III, Member II, and Member I, in ascending order. The Member III, or the so-called underburden (UB), is a greenish gray-to-gray claystone with a thin layer of coal, named the R coal zone. The Member II is composed of two main coal zones, which are the Q coal zone in the lowermost part and the K coal zone in the uppermost part, and are intercalated by an interburden (IB) of clay stone. A nearly twelve meter-thick freshwater fossil shell bed was discovered between coal zones K-3 and K-4 in 2003 by a mine staff of the Mae Moh coalmine. From our investigations, the Member I is a thick overburden (OB) consisting of claystone with a series of coal layers named the J coal zone. The J coal zone is an intercalation between claystone and six main coal layers. The Huai Luang formation is composed of claystone and siltstone with some sandstone and conglomerate lenses. A red to brownish red color is the general characteristic of this formation with some gray layers interbedded in some horizons. There is a coal zone, named I coal zone, in the middle portion.

The Mae Moh mine was certainly a large lake and swamp where living organisms such as fish, turtle, crocodile, and various species of aquatic molluscs lived. The molluscs might have appeared and disappeared throughout time interval depending on environmental changes that altered the composition of the molluscan assemblages in each successive time slice in the Middle Miocene (Songthama, 2005).

The purpose of this paper is to determine the concentrations of U, Th and K in archaeological samples confined within Redbed deposits, (RB) from Huai Luang formation and gray claystone layer, quartz, fossils beds and surrounding sediment in coal which stay between coal zones K-3 and K-4 from Na Khaem formation.

## MATERIALS AND METHODS

### Collection and preparation of samples

The sediment and fossil samples were collected from Mae Moh mine. A sample was collected from Huai Luang formation and other six samples from Na Khaem formation. All samples were ovened dry at 60 °C for 24 hours, crushed, pulverized to a fine powder and homogenized. Notice that the preparation and handled steps must be in clean containers to avoid any contamination (Ghawi, 2005). Each powdered samples, precised weighing between 300-400 mg, in a polyethylene vials has same geometry and volume as in vials containing 600-700 mg of a standard reference material (NBS SRM 2709: San Joaquin Soil). Then the vials were sealed and left about one month to let the standard and the sample reach secular equilibrium, by following Soliman's method (Soliman, 2006).

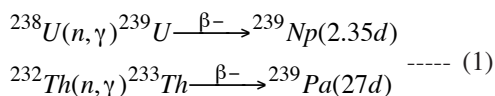
### Neutron irradiation

All samples were irradiated in the Thai Research Reactor-1/Modification (TTR-1/M1), Office of Atoms for Peace, (OAP) in two steps. First, long time irradiations for 12 hours and cooling time for 5-6 days were used in epithermal neutron flux of  $2 \times 10^9$  n/(cm<sup>2</sup>.s) to determine concentration of U and Th. Next for short time irradiations for 10 minutes and cooled for 12 hours were used to determine concentration of K with thermal neutron flux of  $2 \times 10^{11}$  n/(cm<sup>2</sup>.s).

### Gamma-Ray measurement

The gamma spectrometer system consists of GWL series HPGe (High-purity Germanium) coaxial well with 0.5 mm aluminum absorbing layers (well wall) thickness, mounted in a vacuum tight cryostat (Model GWL-120230, crystal diameter 54.9 mm, well inside diameter 10 mm and active well depth 40 mm), a liquid-nitrogen Dewar and dipstick cryostat (Model HJ-GWL) and 1500 volts high voltage supply. A computer-based MCA, (DSPEC) and Gamma Vision-32 V 3.2 Gamma-Ray Spectroscopy Software, a graphical user interface, that ideal for manipulation and analysis of spectra with a personal computer was used. Four isotopes (Ba-133, Eu-152, Cs-137 and Co-60) were used for energy and efficiency calibrations. After irradiation, the  $\gamma$ -radiation spectra of the samples and standard soils were measured and compared by using the flux monitors. This would give the quantitative ratio correction of gamma-ray spectrum.

Because the  $^{239}\text{U}$  and  $^{233}\text{Th}$  isotopes decay in short half-life, it is convenient to measure U and Th, the  $\beta$ -decay of  $^{239}\text{U}$  and  $^{233}\text{Th}$  to  $^{239}\text{Np}$  and  $^{233}\text{Pa}$  respectively, as shown in equation 1 (Rossini, 1991).



### RESULTS AND DISCUSSION

Gamma spectra of U, Th and daughters are shown in Figure 3a and that of K is shown in Figure 3b. The concentrations of U, Th and K, obtained from 7 different samples were in the range of 0.02 to 3.05 ppm, 0.04 to 11.00 ppm, and 0.01 to 1.53% respectively. The annual environmental dose was finally calculated using the finite and infinite method (Ikeya, 1993) and found to be between 87 to 3000  $\mu\text{Gy}$  as shown in Table 2.

### CONCLUSIONS

The present experiment shows that the gamma spectrometry and NAA techniques is well suited in determining the concentration of U, Th and K in sediment from Mae Moh mine, Northern Thailand. Due to the formation of freshwater fossil and surrounding sediment, annual dose calculated using the finite and infinite methods gives a small value (about 90 mGy/a) compared to annual dose in other embedded in claystone samples (about 1000-3000 mGy/a). Since we collected these fossil shells at almost twelve meter-thick bedding plate, surrounding by a lot of small fractional freshwater fossil shell, the calculated annual dose is acceptable. These methods provide a great importance to dating techniques since the result can be used as a reference data that lead to ESR dating analysis within the Middle Miocene of the geological time scale.

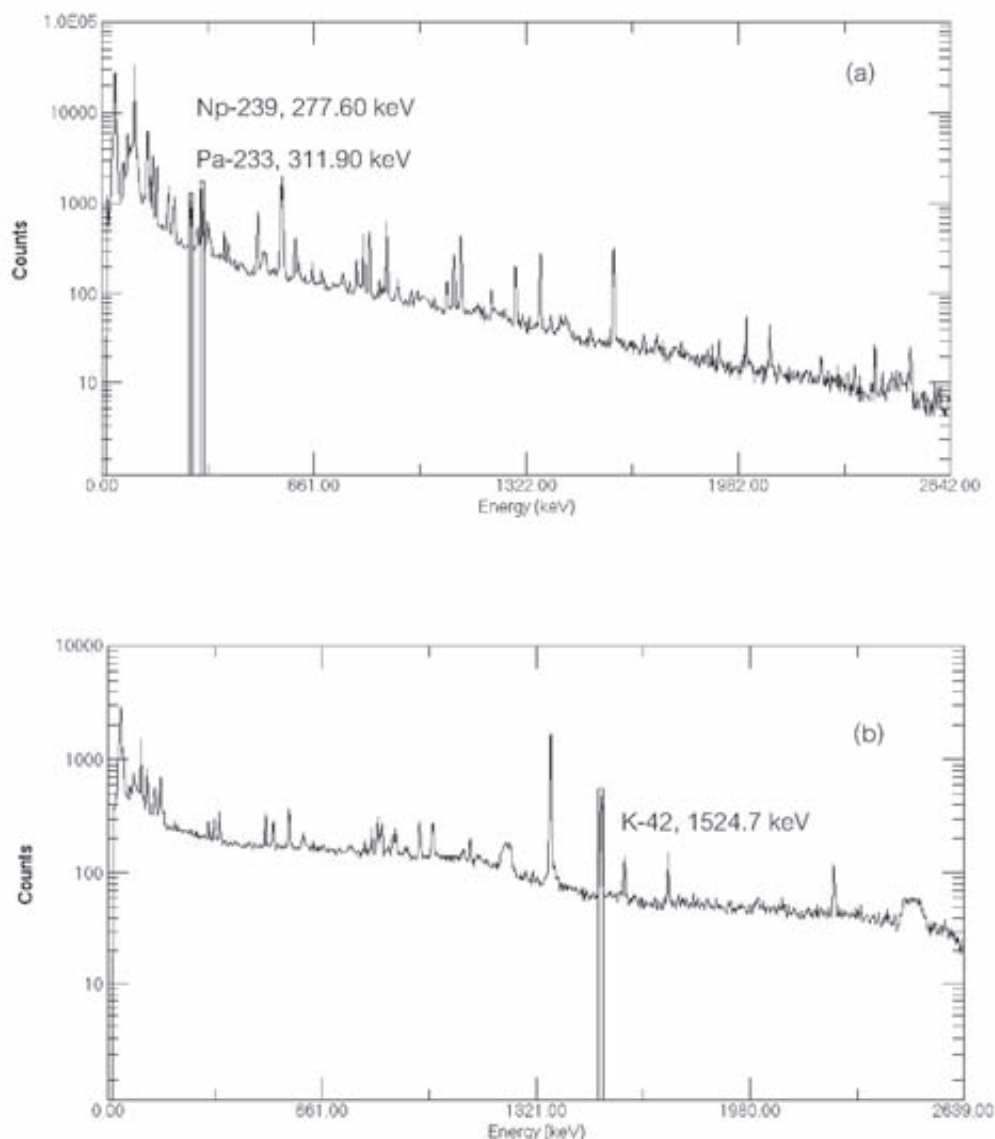
**Table 1** The used important nuclear data from neutron activation of U, Th and K (Rossini, 1991; Ghawi, 2005; Soliman, 2006).

Element	Isotope	Half-lives	Energies of emitted Gamma ray (keV)	% of emission
U	$^{239}\text{U}$	23.5 min	74.6	59.3
	$^{239}\text{Np}$	2.55 d	277.6	14.1
Th	$^{233}\text{Th}$	22.3 min	86.5	2.6
	$^{233}\text{Pa}$	27.4 d	311.9	33.7
K	$^{42}\text{K}$	12.36 h	1524.7	17.9

### ACKNOWLEDGEMENTS

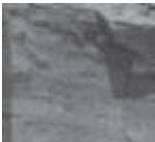




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**Figure 3** The measurement of the main part of a typical gamma ray spectrum of sample sr st4 (No.6 from Huai Luang formation). The important identified photopeaks and their associated radionuclides are shown: (a) and (b) display gamma spectra for radioactive elements U, Th and K respectively.

**Table 2** Concentrations of U, Th and K and results of annual dose in 7 different samples.

No.	sample	Characteristic of sample	Description of sample	U-238 (ppm)	Th-232 (ppm)	K-40 (%)	Dose rate (mGy/a)
1	s st1		gray claystone	0.80	6.20	1.20	1100 ± 55*
2	s st2		between coal	0.97	4.60	0.80	960 ± 50
3	s st3		K-3 and K-4	1.40	6.70	1.50	1540 ± 60
4	w st1		surrounding sediment of fossil	0.20	0.50	0.07	84 ± 3
5	sh st1		fossil between coal K-3 and K-4	0.02	0.04	0.01	
6	sr st4		claystone in RB from Huai Luang	3.10	11.00	0.86	3000 ± 90
7	q st2		quartz in coal between coal K-3 and K-4	0.04	0.11	0.27	87 ± 4

\* is the total uncertainty at 95% confidence interval

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