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Technical note

INAA and DNAA for uranium determination in geological samples from Egypt

A. El-Taher *

Physics Department, Faculty of Science, Al-Azher University, Assuit Branch, 71542 Assuit, Egypt

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ABSTRACT

Uranium isotopes found in soil, rock, water, plants, air, etc., contribute to the natural radiation exposure of the population. U concentrations in some Egyptian environmental samples like Toshki soil, Aswan iron-ore, and phosphate samples from El-Sibayia in the Nile Valley and El-Quseir in the Red Sea coast were determined using instrumental neutron activation analysis (INAA) and delayed neutron activation analysis (DNAA) in the Mainz TRIGA research reactor. The results showed that the phosphate rocks are rich natural sources of uranium among the other minerals forming the earth crust.

1. Introduction

^{238}U is a naturally occurring long-lived radionuclide, which along with its daughter products are the sources of exposure of man to radioactivity. By natural processes, these are getting transferred to people through different pathways and need to be monitored to assess the possible hazards. Environmental studies are generally carried out to trace the pathway of radionuclides/radiotoxic elements. Environmental monitoring and meaningful interpretation of data from man made pollution are more complicated without adequate knowledge about the natural abundance of radioactive elements in the environment. Uranium content in geological samples provides ample information for geological considerations, and the analytical determination of uranium concentrations in such samples is of great importance of solving numerous problems in the field of geochemistry (Taylor, 1964). The U content in soils is usually in the range from 1 to 8 ppm. In fact, the general distribution of U in soils is very similar to that in stream sediments (Durrance, 1986).

There are several methods, which are applicable for determining uranium in geological samples. Each method has its advantages and disadvantages. Considering the various sensitivities of the analytical methods that can be used for determining uranium, instrumental neutron activation analysis (INAA) is of particular interest. This method is simple, fast, non-destructive and can be used for samples with relatively low content. The neutron activation analysis method also allows overcoming many of the limitations encountered in other methods such as matrix effect, internal and external interference. The INAA method has been used to determine the uranium concentration from ^{238}U by neutron capture and successive β-decay, the activation convert ^{238}U into ^{239}Np

$$^{238}\mathrm{U}(\mathrm{n},\gamma)^{239}\mathrm{U} \xrightarrow{\beta^-} {}^{239}\mathrm{Np}\,\mathrm{E}_\gamma = 106\,\mathrm{keV}$$

The characteristic γ-rays can be detected using γ-spectroscopy.

However, the world-wide increase in uranium exploration has increased the demand for uranium analysis; activation analysis has proven to be a suitable method. The present work is concerned with determining uranium concentrations in some environmental samples such as Toshki soil, Aswan iron-ore, and phosphate samples from Upper Egypt by INAA and DNAA (El-Taher et al., 2004).

2. Materials and methods

The samples under investigation were collected from different locations in Upper Egypt: 10 soil samples from Toshki area nearly 280 km from Aswan city; 5 iron-ore samples collected from Aswan iron-ore northeast of Aswan city and 10 phosphate samples collected from El-Sibayia in the Nile Valley and El-Quseir in the Red Sea coast. The samples have been prepared into finely ground homogenous material. They were crushed to a diameter range of less than 125 μm and greater than 63 μm. The crushed samples were dried at 105 °C to constant weight.

* Present address: Physics Department, Faculty of Science, Qassium University, Buraydah 51452, Kingdom of Saudi Arabia.
E-mail address: Atef_Eltaher@hotmail.com

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2.1. Instrumental neutron activation analysis (INAA)

Polyethylene capsules were filled with 100 mg of powder samples and irradiated with standard reference material using thermal neutrons at the University of Mainz Triga research reactor (100 kWth) for 6 h with a flux of 7×10^{11} n/cm^2 s. The data were collected for cooling times up to 2 days (Kernchemie Report Triga Mainz, 1989). The applied low-level gamma-ray spectrometer consists of a HPGe detector with its electronic circuit. The detector has the following specifications: energy resolution (FWHM) at 1.33 Mev Co-60 is 1.70 keV, Peak to Compton ratio Co-60 is 65.2, relative efficiency at 1.33 MeV Co-60 is 29.2%, energy resolution (FWHM) at 1.22 MeV Co-57 is 686 eV, bias voltage is +2000 dc. The detector is connected to the following components: pre-amplifier, amplifier, ADC converter and MCA. The measurements were performed and analysed using the Intergamma Software produced by Intertechnique Deutschland GmbH, Mainz, Germany. Elemental contents were determined relative to certified reference materials, which were irradiated and counted under the same conditions. The electronic dead time in all measurements was less than 10% and was automatically corrected by the Intergamma software (El-Taher et al., 2003; El-Taher, 2007, 2010).

2.2. Delayed neutron activation analysis (DNAA)

Neutron emission is a relatively rare phenomenon in radioactive decay. Except for isotopes of heavy elements undergoing spontaneous fission, a limited number of radionuclides of very short half-life (ranging from a fraction of a second to one minute) emit neutrons during decay. These nuclides are known as delayed neutron precursors, most of them are fission products, which decay by β^-n processes. There are about six groups of fission products emitting delayed neutrons with half-lives from tens of milliseconds to 55.6 s such as ^{87}Br. Irradiation of samples of natural origin in a reactor will emit delayed neutrons from fission products originating from ^{235}U by thermal neutrons on ^{238}U and ^{232}Th.

Delayed neutron activation analysis (DNAA) is a special version of activation analysis and uses the counting of β^- delayed neutrons emitted from very neutron-rich fission products. Delayed neutron activation analysis is used for the fast determination of fissile nuclides, such as ^{233}U, ^{235}U, ^{239}Pu, ^{232}Th, etc. by measuring the delayed neutron emission after fission. From the nuclides produced in nuclear fission about 170 are known precursors of β^- delayed neutron emission with β^- half-lives ranging from a few milliseconds to about 1 min (Tomlinson, 1973). Delayed neutrons emitted by long lived isotopes ^{88}Br, ^{137}I, ^{87}Br are usually measured. The sensitivity is less than 10^{-11} g for ^{235}U, 10^{-6} g for ^{232}Th and 10^{-11} g for ^{239}Pu under conditions of the TRIGA Mainz research reactor (Eberhardt and Kronenberg, 2000).

Delayed neutron activation analysis (DNAA) involves irradiation of the sample in a reactor, transfer of the sample to a neutron counting assembly and counting of delayed neutrons after a certain delay (Amiel, 1981). Polyethylene capsules filled with 100 mg of the powdered samples were sent to the irradiation position near the reactor core by using a pneumatic transfer system. After an irradiation time of 2 min the samples were automatically sent to the counting station, and after a decay time of 15–20 s they were counted for 1 min with ^{3}He proportional counters in a circular arrangement. The Mainz measuring system contains 15 ^{3}He tubes placed in a cylindrical configuration. The delayed neutrons are thermalized by paraffin and polyethylene and detected with high efficiency. The detector assembly was coupled to an electronic counting system. After suitable electronic processing, pulses due to neutron interactions were measured in a counter/timer. The background was determined by irradiating and counting empty capsules. In our experiments the background was 5 counts/min. This method is routinely used for the inspection of workers from the uranium industry. Generally α-spectroscopy is employed for such investigations, but DNAA is more simple and faster.

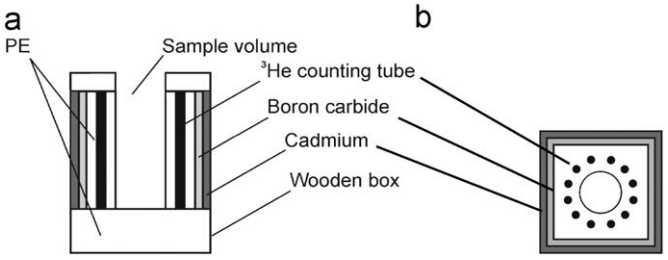

Fig. 1. Vertical (a) and horizontal (b) cross section view of the neutron detector arrangement used for DNAA at the TRIGA-Mainz facility.

Fig. 1 shows a schematic view of the neutron detector used for DNAA. Knowing the isotopic composition of uranium a screening of a large number of samples can be carried out with the precision demanded by the necessary precision. Thus, DNAA can routinely be applied for the inspection of workers in the nuclear industry.

3. Results and discussion

3.1. Uranium concentrations in Toshki soil samples

Table 1 shows the concentration of ^{235}U and ^{238}U in different soil samples collected from different places in the Toshki area in Upper Egypt. From the results it can be seen that the ^{238}U concentrations determined by INAA vary between 1.0 and 1.6 ppm and by DNAA between 1.5 and 2.5 ppm. The statistical counting error in the determination of ^{238}U in soil lies between 8% and 12%. The error for ^{238}U in the case of ordinary soil is thus rather high. More accurate results can be obtained in the case of soils having relatively high uranium contents. The extensive use of phosphate compounds or phosphate rocks as altering fertilizers could contribute to the increase of U concentration levels. The data presented in this work suggest that the phosphate fertilizers have not contributed significantly to the radioactivity levels of the region under study, because this area is a new agricultural area and the use of fertilizers is not observed.

Interpretation of the results of soil surveys can be extremely difficult, depending upon the type of terrain concerned. It should always be borne in mind that transport of elements takes place in soils. Under some circumstances, it is also possible for solid soil particles to be moved over considerable distances by the wind. These effects will vary from one soil to another and according to meteorological conditions. The concentration of ^{238}U is significantly increased in the upper layers of the soil compared with the original values. The main reason for this, over and above the deposition of fly ash, is the mixing of the original soil with the slag of coal due to industrial, building activity and some household activities of the private population (Robothan et al., 1987).

3.2. Uranium in Aswan iron-ore deposits

The concentrations of ^{235}U and ^{238}U in iron-ore samples are listed in Table 2.

From the results it can be seen that the average values of ^{238}U concentrations for Aswan iron-ore samples determined by INAA