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Determination of beryllium by use of photonuclear activation techniques

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Abstract

The possibility of using a gamma source based on ^{124}Sb for detection and determination of beryllium contents in some standards and one mine sample has been investigated. It is concluded that by using a ^{124}Sb source for bilateral irradiation of the samples and only one BF3 counter, beryllium could be measured to an absolute sensitivity of up to 0.4 mg. Such a facility would be capable of contributing to exploration of beryllium mines. A comparison with the ICP method shows good agreement between both measurement techniques.

Keywords: Beryllium; Photonuclear activation; Photo neutron sources; Antimony–beryllium; BF3 detector

1. Introduction

The principle of photoneutron sources such as ^{124}Sb–Be has been used for determination of the beryllium content in some prepared samples, as well as in an accessible mine sample.

The so-called antimony–beryllium photoneutron source (^{124}Sb–Be) has lower neutron-binding energy of 1.667 MeV (photonuclear reaction threshold) given in Mobley and Laubenstein (1950), than the incident gamma ray of ^{124}Sb (1690.980 keV 48.5% and 2090.942 keV 5.7%) given in Firestone et al. (1996). By counting the photoneutrons which are produced under the following reaction, it is possible to measure the concentration of beryllium in such samples:

$$9\mathrm{Be} + \gamma \rightarrow {}^{8}\mathrm{Be} + n.$$

We should mention that beryllium is the only element in nature that undergoes a photonuclear reaction in this gamma energy range. This method is also used for diagnosis of the beryllium-induced disease, berylliosis (beryllium accumulation in the lung). One of the preliminary studies using this technique was carried out by Ettinger et al. (1980). In this work the detection limit of 3.4 mg of beryllium has been achieved by using 72 neutron detectors (BF_3), surrounding the chest during the lungs irradiation by a ^{124}Sb photon source. In Ali et al. (1985), by using a filtered source of ^{124}Sb for bilateral irradiation of the chest, and using an array of 20 BF3 counters, beryllium has been measured with an accuracy of 0.33 mg per lung irradiation. This content corresponds to a detection limit of 0.67 mg beryllium, for a skin dose of 50 mGy of gamma rays, delivered during a period of 90 s.

The existence of beryllium in environmental samples have been proved by now. It is possible to use this technique for measuring the enrichment of this element in all kinds of mine samples, as well as other environmental pollution in nature.

In this work we used an intense ^{124}Sb gamma source (half-life 60 days). By putting this source behind the beryllium samples and counting the induced neutrons in front of the samples we succeeded in determining beryllium concentration in some compositions and in one accessible mine sample. It should be mentioned that this gamma source has been produced by neutron bombardment in the 5 MW Tehran Nuclear Research Reactor of AEOI.

The equipment which was used here consists of a heavy gamma-radiation lead-shield, which surrounded the sample and the gamma source together, in a fixed geometry. One

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BF_3 thermal neutron detector, shielded by polyethylene moderator, was used to detect the generated photo-neutrons from the samples. Inside the cylindrical lead-shield, two holes were drilled in order to keep the source and samples as close as possible. System calibration has been done, using well-known target and gamma source (standard assembly).

2. Experimental technique

The neutron yield of the above-mentioned photonuclear reactions were measured by irradiation of beryllium samples. These samples have been used in chemical form of $BeSO_4$, BeO and in standard shapes, which are given in Table 1.

In this table BeO (lab) sample was produced from $BeSO_4$ in our Laboratory. Other samples were taken from Merck Company. Some standard samples of BeO (Merck) were diluted (10%, 1%, 0.1%, 0.01%) with silicon sand (SiO_2). The weights of the beryllium content of these samples are tabulated in Table 1. The samples are put inside some quartz pipes with 8 mm diameter and 35 mm height. In this table the information of all the samples together with their calculated beryllium contents are illustrated. The weight of beryllium in the mine sample is indicated by X.

Hundred milligrams of metallic antimony in powder form, filled in a tube of quartz, was sealed and irradiated for about 100 h at 3 MW power of the Tehran Research Reactor (TRR). This tube has the same geometry as the beryllium samples. After cooling, the prepared gamma source was placed into the lead cylindrical shield, as close as possible to the beryllium samples (Fig. 1). Because of relative measurement, the activity of gamma source is irrelevant; nevertheless, it should be strong enough to deliver detectable neutrons in low-concentration samples. With the same geometry, the Be samples, mentioned in Table 1, were put on the neighboring hole and the produced photoneutrons were measured with the neutron detection system.

For counting the produced neutrons emerging from each sample, a single BF3 detector which was bought from UG company (Model: LND-INC) has been used. In order to make for higher detection efficiency, this thermal neutron detector was surrounded by a thick polyethylene shield. The distance between sample and detector is about 20 cm and the whole system (source and detection system) is surrounded again by some more polyethylene blocks of 20 cm thickness and 20 cm height, for increasing detection efficiency. A schematic diagram of this system is illustrated in Fig. 1.

The electronic measurement system consisted of a Bias supply (ORTEC-478), preamplifier (ORTEC-142PC), spectroscopy amplifier (ORTEC-575A) and a Timer/Counter (ORTEC-776) equipped with a discriminator. The discriminator level in each series of measurement must be adjusted again to separate the noise and gamma signal from neutron signals. These adjustments have been carried out using a ^{252}Cf neutron source.

The signals from the BF3 detector after amplification by preamplifier and main amplifier were introduced into the timer counter (Fig. 1).

After preparation the mine sample, first of all an ICP-method was used to determine the beryllium concentration. According to this technique the US Geological Digestion Procedure was applied and the achieved result was 4.45% beryllium and 2.00% aluminum. Then the photonuclear measurement was carried out in 6 separate runs.

Because of different gamma activities plus different position geometries and electronic setups, in each run, the system was calibrated for all samples, including the mine sample in each separate measurement. For weak, concentrated samples, counting time was increased 10–20 times or more. It should be mentioned that in each run we put beryllium samples, one by one, in a cylindrical hole in front of the gamma source and started to count. The counts from each sample were measured and gathered in Table 1.

In Fig. 2 the correlation of the count rate with the beryllium content is plotted separately for each run.

Table 1
Neutron counts and related mass of beryllium in each sample and run

Samples	Sample weight (mg)	Be in sample (mg)	1st run counts/100 s	2 run counts/100 s	3 run counts/100 s	4 run counts/100 s	5 run counts/100 s	6 run counts/100 s
BeO (Merk)	270.0	97.2	214±15	1421±38	1387±37	2413±49	1692±41	283±17
$BeSo_4$ (Merk)	490.7	24.99	55±7	335±18	–	615±25	510±23	77±9
BeO (Lab)	145.0	49.59	112±11	677±26	–	1130±34	811±28	146±12
Mine	841.6	X	90±10	533±23	–	982±11	–	122±11
%10 sample	40.86	40.86	–	–	582±24	1025±32	818±28	140±12
%1 sample	4.416	4.416	–	–	62±8	114±11	104±10	17±4
%0.1 sample	0.4016	0.4016	–	–	9±3	17±4	13±4[a]	13±4
%0.01 sample	0.004	40e-3	–	–	3±2[b]	–	3.5±2[b]	–
Calculated Be in X	841.6	39.7±6[c]	40.7±4.5 40.396	37.1±1.6 38.533	–	40.1±1.3 40.164	–	40.8±3.7 39.194

[a]Measured in 1000 s.
[b]Measured in 3000 s (could be neglected because of too big uncertainness).
[c]Calculated from mean values.