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NAA OF Se WITH A GAMMA–GAMMA COINCIDENCE AND COMPTON SUPPRESSION SYSTEM

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ABSTRACT

The paper describes a method of selenium determination by neutron activation analysis using gamma–gamma coincidence spectrometer with Compton suppression. The analysis results of some standard reference material from NBS or IAEA are reported. The differences between our data and reported values are discussed.

Keywords: Selenium NAA Gamma–gamma coincidence spectrometer Compton suppression Standard reference material

I . INTRODUCTION

It has been affirmed that selenium is one of the essential trace elements to human health. However, human or animal will suffer from diseases when excessive or insufficient amounts of Se are taken from the envinronment. It has become a very significant subject in the field of evinronment and life science to determine precisely selenium in environment and human body.

Selenium exists naturally in several oxidation states, some of which are volatile. In determination of Se, it is very important to avoid the loss of Se owing to volatility. Therefore, non–destructive neutron activation analysis is preferred. However, selenium content in the environmental and biological samples is mostly within a range of 0.001 ppm to several ppm, and activation products of other elements in the samples can cause very strong interference making result with large deviation.

For the sake of surmounting these problems the gamma–gamma coincidence method with anti–coincidence shield/Compton suppression system was used in the paper. A quite fine result was obtained in our laboratory.

II . EXPERIMENT

1. Preparation and irradiation of samples and "standards"

The samples to be determined were soil–3 and soil–5 from IAEA and coal–1632a from NBS of USA. It should be noted that among these standard reference materials the only one which has been very well determined is just the coal–1632a. The samples were firstly allowed to dry at 105°C for 20 hours as recommended in Ref.[1]. Then, the coal sample was kept in a vacuum drier for 20 hours. Forty mg sample for each was wrapped up successively in one layer of cellulose paper and two layers of aluminium foil of high purity.

"The standard" was prepared as follows: a superpure standard solution (20 μl) with a known selenium content was dropped onto six layers of cellulose paper of 9 mm in diameter. The filter paper was dried in a drier, then wrapped in aluminium foil.

All the samples and "the standard" were irradiated for 8 hours in the same conditions on a reactor of China Institute of Atomic Energy. The neutron flux was about 3×10^{13} n/(s · cm^2). After cooling for 15 days, nuclide ^{75}Se in the samples was measured by a HPGe–NaI (Tl) gamma–gamma coincidence spectrometer with anti–coincidence shield.

2. Instruments

Fig.1 illustrates a electronics block diagram of the coincidence–anticoincidence gamma spectrometer used in this work. In the spectrometer, gamma–gamma coincidence is realized by using the HPGe and NaI (Tl) detectors. The coincidence signals from the coinc. circuit are used as gating signals of the HPGe main detector which is used to detect or analyse gamma spectrum.

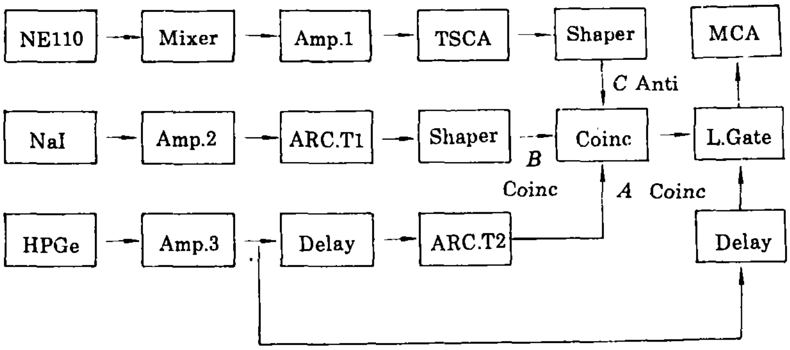

Fig.1 Electronics block diagram of the coincidence—anticoincidence spectrometer with the Compton suppression

In order to reduce interference from background and Compton scattering, a ring detector constructed with the NE110 plastic scintillator detector is used as anticoincidence shield/Compton suppression detector of the HPGe detector. The energy resolution of the HPGe detector is 1.8 keV, with 50% of a relative efficiency (with respect to the 1332 keV gamma ray of Co-60). The NaI (Tl) detector is constructed with a low background 3″×3″ NaI (Tl) crystal and a photomultiplier with a quartz window. The energy resolution of this detector for 662 keV gamma ray of ^{137}Cs is 8.7%. Other constructions and features of the spectrometer have been detailed in Ref.[2,3].

Ⅲ. ANALYSIS

In this work, standard reference materials coal 1632a and the soil-3 and soil-5 samples were analysed. Selenium content in these samples ranges from 0.1 ppm to several ppm. It should be emphasized that there were over ten elements which would interfere with the determination of selenium in the samples. The main results are given in Table 1.

Table 1

Content of some elements in the NBS 1632a

Element	Concentration	Unit
Se	2.6	ppm
Ta	410	ppb
Eu	527	ppb
Hf	1.68	ppm
Hg	146	ppb
Co	6.4	ppm
Lu	176	ppb
W	820	ppb
Fe	1.12	%

NAA determination of selenium is currently based on two selenium isotopes, ^{77m}Se (half-life 17.6 s) and ^{75}Se (half-life 120 d). They are produced by (n,γ) reactions of nuclides ^{74}Sr and ^{76}Se, respectively. The nuclide ^{75}Se is used in most cases. Because of the relatively long half-life of ^{75}Se, the irradiated samples need not be measured immediately after activation. This is of great advantage for laboratories far away from neutron sources.

The determination of ^{75}Se in the samples was carried out by gamma-gamma coincidence of 135.8 and 264.5 keV gamma rays from the cascade radioactive decay of ^{75}Se, because both of the lines are not only cascade, but also have higher intensity than other ^{75}Se gamma lines. The 135.8 keV gamma rays were detected by the NaI (Tl) detector, and the 264.5 keV gamma rays by the HPGe detector. The detection efficiency of the NaI (Tl) for 135.8 keV gamma rays is higher than for 264.5 keV gamma rays. The detection efficiencies of the HPGe for both rays are close to each other. As shown in Table 1, there are ^{152}Eu, ^{181}Hf, ^{182}Ta, and ^{203}Hg in the irradiated samples emiting 121.8, 133.05 and 136.25; 264.07 and 279,17 keV gamma-rays respectively which would interfere with ^{75}Se gamma rays. On the other hand, Compton scattering from gamma rays with higher energies from other activated products would make the spectrum even more complicated. The gamma-gamma coincidence technique enables one to select desired radioactive events from such a number of gamma-rays. And the Compton suppression technique decreases accidental coincidence events, especially those from Compton scattering events caused by the gamma rays with higher energies. All these have improved the sensitivity and the precision in Se determination.

The selenium content in samples is then given by

$$C = AT_sM_s/A_sTM \qquad (1)$$

where A is the net-peak area of 264.5 keV gamma ray in the sample spectrum; T and M are collect time of the spectrum and sample mass, respectively; and A_s is the net-peak area of 264.5 keV gamma ray in "standard" spectrum, T_s and M_s are collect time of the spectrum and selenium mass in "standard", respectively.

IV. RESULTS AND DISCUSSION

The results of the analysis are listed in table 2. Table 2 also shows those results obtained with gamma-gamma coincidence only. It can be seen that the results for sample coal-1632a[4] obtained with coincidence-anticoincidence method are in conformity with the literature values.

Table 2

Selenium content in samples (ppm)

Sample	Literature		This work	
	$X \pm S(n)$*	Range	$X \pm S(n)$	Method
NBS coal–	2.6 ± 0.2 (7)	2.4–3.1	2.65 ± 0.19 (4)	Coinc–anti
1632a			2.79 ± 0.25 (3)	Coinc
IAEA		0.14* *	0.19 ± 0.03 (4)	Coinc–anti
Soil–3		1.32–2.37* * *	0.22 ± 0.10 (3)	Coinc
IAEA	1.3 (1)		0.48 ± 0.35(4)	Coinc–anti
Soil–5			0.54 ± 0.09 (3)	Coinc

* *n* shows the number of the values to be joined average * * RNAA method * * * INAA method

It should be pointed out that there are large differences between results we obtained and literature values for soil-3 and soil-5. In the literature[5], for soil-3, the Se concentrations were obtained by Instrumental Neutron Activation Analysis (INAA) and Radiochemical Neutron Activation Analysis (RNAA). There were large differences between the data obtained by INAA and RNAA, and the former was more than ten times greater than the latter. Our results for soil-3 lay between them, and close to the result obtained by RNAA very much. We think that the data obtained by INAA, perhaps, contain quite many spurious counts from the interfering nuclides, such as ^{152}Eu, ^{181}Hf, ^{182}Ta, *etc*. And in the datum obtained by RNAA perhaps some counts were lost during the radiochemical processes. Only one datum is reported for selenium of soil–5[1] and no analytical method is shown. So, it is very difficult to make a correct interpretation for the difference. But, according to our results for coal–1632a, we think that our data obtained by coincidence–anticoincidence method should be more believable than those in Ref.[1,5].

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