Letter to the Editors

A New Possibility for Simultaneous Determination of Radioactive and Stable Isotopes in Environmental Samples by the Substoichiometric Method†

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1. Introduction

The determinations of both the activity of a radionuclide and the amounts of its stable element are often necessitated in the studies on the behaviors of environmental radioactive species. It seems very convenient and advantageous, if the simultaneous determination of these two amounts could be possible.

In this paper, a possibility of application of the substoichiometric method is studied for this purpose, and the proposed principle is verified experimentally in the determination of ^{60}Co activity and the amounts of its stable isotope.

2. Principle

Two equal portions of the solution of the sample are taken. Each portion contains unknown amounts of the radionuclide (activity: A_x) and the stable element (weight: W_x) to be determined. To each portion, a known but different amount (W_1 and W_2 , respectively) of the stable element is added and mixed. From each solution, a definite part of the element to be determined is separated by using the definite but substoichiometric amount (w) of the reagent which reacts with the element, and its activity (a_1 and a_2 , respectively) is measured. As the

specific activity of the radionuclide in the separated part is unchanged from that in the solution before separation, the next equations hold.

$$\frac{A_x}{W_x + W_1} = \frac{a_1}{w} \quad (1)$$

$$\frac{A_x}{W_x + W_2} = \frac{a_2}{w} \quad (2)$$

By solving the simultaneous equations, A_x and W_x are obtained as follows:

$$A_x = \frac{W_2 - W_1}{w} \cdot \frac{a_1 a_2}{a_1 - a_2} \quad (3)$$

$$W_x = \frac{a_2 W_2 - a_1 W_1}{a_1 - a_2} \quad (4)$$

As the amounts W_1 , W_2 and w can be maintained constant throughout a series of experiments, Eqs. (3) and (4) can be written as follows:

$$A_x = \alpha \cdot \frac{a_1 a_2}{a_1 - a_2} \quad (3)'$$

$$W_x = W_1 \cdot \frac{k a_2 - a_1}{a_1 - a_2} \quad (4)'$$

where $\alpha = \frac{W_2 - W_1}{w}$ and $k = \frac{W_2}{W_1}$.

Thus, A_x is proportional to $a_1 a_2 / (a_1 - a_2)$ and independent of W_x , while W_x is proportional to $(k a_2 - a_1) / (a_1 - a_2)$ and independent of A_x . Accordingly, plotting A_x vs. $a_1 a_2 / (a_1 - a_2)$ or W_x vs. $(k a_2 - a_1) / (a_1 - a_2)$ should give a straight line on the linear graphic paper.

3. Experimental

A 100 ml portion of the mock sample solution, in which 200 - 2000 cpm of ^{60}Co and 20 - 200 μg of stable Co(II) were contained, was taken in

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a 200 ml separatory funnel and 5 g of magnesium chloride was added. After addition of 510 μg Co^{2+} (W_1) of cobalt chloride and the aqueous solution of 1.50 mg (w) of sodium diethyldithiocarbamate, the solution was shaken with 20 ml of benzene. The activity (a_1) of the organic layer was measured with a liquid scintillation counting system.

The second measurement of activity (a_2) was carried out with another 100 ml portion of the mock sample solution in the similar way, by adding 850 μg Co^{2+} (W_2) of cobalt chloride solution.

4. Results

An example of the linear relationship between activity A_x and $a_1 a_2 / (a_1 - a_2)$ is shown in Fig. 1. In the similar way, the weight of stable cobalt W_x shows fairly good linearity with respect to $(ka_2 - a_1) / (a_1 - a_2)$.

By using these calibration curves, some mock samples containing picocurie level of ^{60}Co and 100 μg level of stable cobalt were analysed. The results are shown in Table 1. Considering that significant figures are 2 digits, the observed values are in fairly good concordance with taken ones.

The advantages of this method are: 1) The procedure is simple and rapid. 2) By using the calibration curves or the constant values of the slope for each straight line, the accurate amounts of W_1 , W_2 and w are not necessary to be known. Only necessary are to keep the volumes of the stable element solution added and reagent solution constant throughout a series of experiments, and to measure the activities a_1

Table 1 Simultaneous determination of ^{60}Co and stable cobalt

Amounts taken		Amounts observed	
^{60}Co (Bq)	Stable Co (μg)	^{60}Co (Bq)	Stable Co (μg)
23	170	25	160
34	57	37	69
46	110	50	120

Fig. 1 Linear relationship between A_x and $a_1 a_2 / (a_1 - a_2)$.

and a_2 correctly. 3) The measurements of the chemical yield of the stable carrier are unnecessary.

The method seems suitable for the routine and many samples analyses for radioactive and stable cobalt in such environmental samples as marine deposits and biological ashes, etc.

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