Determination of Strontium in Biological Materials by Charged Particle Activation Analysis using the Stable-isotope Dilution Method

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In a previous paper[1], we have been proposed a new method named the stable-isotope dilution activation analysis. The outline of the method can be written as follows. When an element to be determined consists of at least two stable isotopes which are converted easily to the radionuclides through particular nuclear reactions, quantity x (g) of the element in the sample can be determined by irradiating simultaneously or separately the duplicated sample containing y (g) of either enriched isotope, and by using the following equation.

$$x = y(M/M^*)[(R^*/R)(\theta_2^*/\theta_2) - (\theta_1^*/\theta_1)] \;/\; [1 - (R^*/R)] \qquad (1)$$

where M and M* are atomic weights of the element to be determined and the enriched isotope used as a spike, θ_1 and θ_2 are natural abundances of two stable isotopes in the element, θ_1^* and θ_2^* are isotopic compositions in the enriched isotope used, and R and R* are counting ratios of gamma-ray emitted by two radionuclides produced in the sample and the isotopic mixture.

The most striking characteristic of the above method is that the self-shielding effects and inhomogeneities of the flux between the sample and the isotopic mixture are corrected simultaneously and completely, because one of the stable isotopes is used just as an internal standard. When the method is applied to charged particle activation analysis, it should be noted that the above self-shielding effects of the sample and the isotopic mixture are given rather in term of dE/dx, which is well-known as the stopping power of the material.

In the present study, further application of the above method was examined by proton activation through determination of strontium in biological standard reference materials.

The standard reference materials of tomato and citrus leaves were purchased from the National Bereau of Standards (NBS SRM-1572 and -1573), and used as unknown samples in the present experiments. On the other hand, isotopically enriched ^{86}Sr to be used as a spike was obtained from the Oak Ridge National Laboratory. Percentage isotopic compositions of this enrichment and isotopic abundances of natural strontium are shown in Table 1 together with their atomic weights. This was used after an exactly known amount of $^{86}SrCO_3$ was dissolved in dil. HNO_3, and then diluted with re-distilled water to give the des' concentration.

Possible proton induced reactions leading to radionuclides on strontium are listed in Table 2 together with their nuclear characteristics. Among them, the $^{86}Sr(p,n)^{86}Y$ and the $^{87}Sr(p,n)^{87m}Y$ reactions were chosen as suitable reactions to determine strontium, because both radionuclides have reasonable half-lives and high probabilites of gamma-ray emission.

On the other hand, in charged particle activation analysis, it is strongly required that all irradiation sources should be prepared in the same chemical and physical forms in order to cancel out the stopping powers of the sample and the comparative standard. Hence, the sample and the isotopic mixture in the present experiments were processed according to the method proposed by Mitchell et al.[2] after strontium was separated from the matrix elements together with almost of calcium as nitrates by adding fuming HNO_3, respectively. The resulting powders as silica gel were then pressed into smooth-surface pellets, which are thick enough to stop the incident protons, and each pellet was wrapped in a pure aluminium foil for irradiation. Irradiations were carried out with 1-4 μA beam of 13 MeV protons for 2 hours by the use of a rotating target assembly, and the redionuclides produced were measured at the same time a half-day after the end of irradiation using a high-resolution Ge(Li) detector connected to a multichannal pulse height analyser. Gamma-ray of 1077 keV due to ^{86}Y was used for a sensitive determination of ^{86}Sr, while that of 381 keV due to ^{87m}Y was also used for a susceptible determination of ^{87}Sr. The R and R* in eq. (1) were then obtained as the ratios of the net photopeak countings due to 381 keV line (^{87m}Y) to those due to 1077 keV line (^{86}Y) in the sample and the isotopic mixture, respectively.

When the method is applied in practice, the quantity of y to be added to the duplicated sample is also of very importance. Therefore, the optimum quantity of isotopically enriched ^{86}Sr to be added to each duplicated sample was determined by the preliminary experiments. As the results, 13.65 and 27.28 μg of ^{86}Sr were added to each 2 g of tomato and citrus leaves as spikes, respectively. The concentrations of strontium found in these standard reference materials are shown in Table 3 together with the certified values given by NBS. On the basis of the values obtained in three replicate analyses, the method was also found to be accurate and reliable even when it is applied to the charged particle activation analysis.

References

1) Masumoto K. and Yagi M., J. Radioanal. Chem. 78 (1983) 233.
2) Mitchell J. W., Blitzer L. D., Kometani T. Y., Gills T. and Clark L. Jr., J. Radioanal. Chem. 39 (1979) 355.

Table 1. Isotopic composition of enriched ^{86}Sr and natural abundance of strontium.

Isotope	Enriched isotope Isotopic composition,%*	Element Natural abundance,%
84	>0.05	0.56
86	96.89±0.10	9.8
87	0.99±0.05	7.0
88	2.13±0.10	82.6
Atomic weight	86.012	87.58

*As reported by ORNL.

Table 2. Proton induced reactions on strontium.

Target	Reaction	Product	Half-life	Principal γ-ray (keV)
^{84}Sr	(p,n)	^{84}Y	39 m	793, 974, 1040
^{86}Sr	(p,n)	^{86}Y	14.74h	443, 628, 1077
^{86}Sr	(p,n)	^{86m}Y	48 m	208
^{87}Sr	(p,n)	^{87}Y	80.3 h	485
^{87}Sr	(p,n)	^{87m}Y	13 h	381
^{88}Sr	(p,n)	^{88}Y	106.6 h	898, 1836

Table 3. Concentrations of strontium in the standard reference materials.

Sample	Concentration of strontium (ppm) Present Work	Certified
Tomato leaves	45.3	44.9±0.3
	44.4	
	44.8	
	Av. 44.8±0.5	
Citrus leaves	95.4	100 ±2
	102.1	
	100.7	
	Av. 99.4±3.5	