Measurement of Strontium-90 in Environmental Samples by Using Rapid Sr Sampler

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Introduction

Strontium-90 is produced through nuclear fission. It is an important radionuclide in the environment and mainly comes from atmospheric nuclear tests. However, the operation nuclear facilities may also release strontium-90 and contaminate the environment. Therefore, analysis of strontium-90 in the environment has been an important task in the routine environmental radiological monitoring. Strontium-90 and its daughter, yittrium-90, both are pure beta-emitter radionuclides. It is necessary to isolate strontium-90 from environmental sample mixtures before determining the beta activity. (1)

Analyses of the strontium-90 in the environmental samples are important in the routine environmental radiation monitoring. A lot of techniques such as coprecipitation, ion exchange, solvent extraction, and extraction chromatography have been described for the determinatin of radiostrontium in environmental samples. In the traditional method of strontium analysis is to separate the alkaline-earth elements (Ca, Sr, Ba, Ra) from the other matrix elements through a carbonate precipitation. Calcium is eliminated through precipitation of strontium nitrate by use of concentrated nitric acid. Barium and radium are separated as the chromates. Then, the daughter of strontium-90, yttrium-90 is eliminated through a hydroxide precipitation. The purified strontium-90 is allowed to stand for growth of yttrium-90. Obviously, the traditional are costly, time-consuming and hazardous.

Recently, the most valued method is to extract strontium using crown ethers. In the research of J. T. Chuang, DC18C6 is very effective for separating strontium and yittrium. Scarpitta et al. compare four methods of analyzing strontium for underground water. Among them, two commercialized products, 3M Strontium Empore™ Rad disk and EIChroM Sr-Spec Resin method include strontium-specific crown ethers, and MDL is able to reach 37mBq/l (2, 3, 4,). In this study, a fast, safe, reliable and easy-handling method for strontium selective extraction in environmental samples has been established by using Rapid Sr Samples. Rapid Sr Sampler also includes crown ether. Its capacity of extracting strontium is more than 3.6mg strontium and with an efficiency of nearly 100%. (As the product is still in an experimental stage, the manufacturer's name will not be released here.) A variety of environmental samples and a NIST standard reference material have been interested in this study. Since Rapid Sr Sampler is properly packed, it can be used to elute the daughter Y-90 produced from strontium 90 over and over again, thus is extremely helpful to the analyzing work.

Experimental

Samples and pretreatment

Environmental water samples were taken from the area close to INER include shihmen dam water, ditch water, deep well underground water. 2ml of 6M HNO₃ is added into water sample while sampling. It is filtered an evaporated to decrease volume of 100 ml before being transformed into a 2M nitric solution. It is ready to be extracted by Rapid Sr Sampler. The vegetation sample is a kind of tea and taken form shihbon-mountain in Taiwan. Soil sample is lake sediment of NIST SRM 4354. Both soil sample and vegetation sample are incinerated at 400°C by incineration furnace for more than 8 hours to eliminate organic interference before dissolution. The dissolutions of sample is made with HCl (Merck GR 37%), HNO₃ (Merck GR 70%) and pure water. The sample solution is heated on hotplate and keeps boiling for one hour. Then filter it and wash with 100ml water. Oxalic acid and NH₄OH (Merck GR 25%) are added to form oxalate precipitate. The supernatant is decanted. The oxalate precipitate is dissolved in HNO₃ and evaporated to form strontium nitrate precipitate. Finally, add water and HNO₃ to transform sample into a 2M nitric solution. Then the sample solution is ready to be extracted by Rapid Sr Sampler. The scheme of analytical procedures in this study is shown in figure1. The pretreatment recoveries of the samples are estimated by strontium measured by ICP-AES.

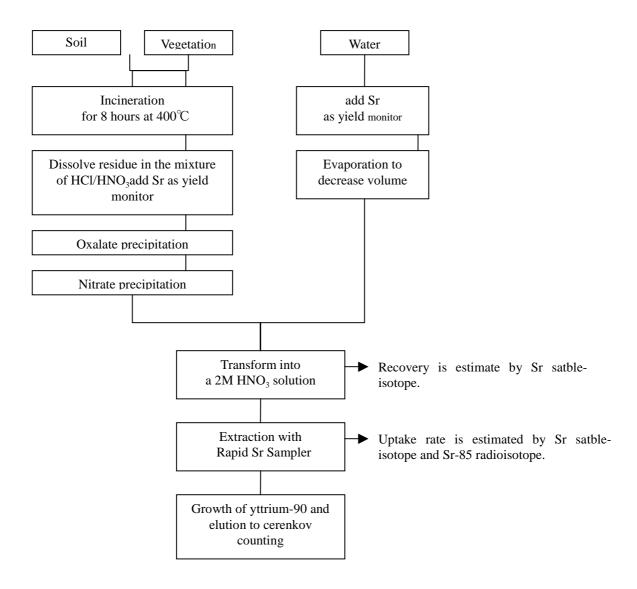


Figure 1. Scheme of strontium-90 analytical procedures

Conditioning of Rapid Sr Sampler

Rapid Sr Sampler should be conditioned before using to extraction. 2ml of Methanol (Merck GR) is injected into Rapid Sr Sampler and waits two hours for conditioning before extraction. After methanol conditioning, Rapid Sr Sampler is connected with a pump and a tube. 20ml of a 2M HNO $_3$ solution is added to the tube and flows through Rapid Sr Sampler by pump with a flow-rate of 5ml per minute. Stop the pump before Rapid Sr Sampler becomes dry. Then Rapid Sr Sampler is ready to sample extraction. Ethanol is also able to be a conditioning agent, but its efficiency is lower.

Extraction

The sample is extracted by through Rapid Sr Sampler with an approximate flow-rate of 5ml per minute. 20ml of a 2M HNO₃ solution is added to rinse Rapid Sr Sampler. The Sr uptake-rate of the extraction is estimated by strontium stable-isotope (measured by ICP-AES) and strontium-85 radioisotope (measured by NaI detector).

Elution and counting

The Rapid Sr Sampler is eluted by 20 ml of a 2M HNO₃ solution with 10 mg yttrium three times after the growth time of yttrium-90. The elution solutions are counted directly by Cerenkov counting with a period of 0-50 keV (5, 6).

Results and Discussion

The analytical procedures of samples pretreatment are modified from the methods of routine environment

strontium-90 analyses of Institute of Nuclear Energy Research. The recoveries of the samples are listed in table 1. The recoveries of water samples are approximate 100% without doubt. The soil sample is freshwater lake sediment (NIST SRM 4354) with abundance of organic material. It should be incinerated before dissolution.

Table 1. The strontium recoveries of the environmental samples

Sample	Soil	Vegetation	Water-1	Water-2	Water-3
No.1	51.7%	71.2%	~100%	~100%	~100%
No.2	51.2%	75.5%	~100%	~100%	~100%

^{*} Soil: NIST SRM 4354; Vegetation: tea form shihbon mountain;

Water-1: drinking water; Water-2: fresh water from shihmen dam; Water-3: underground water from deep well.

Manufacturer suggests that Rapid Sr Sampler may be conditioned by methanol. Ethanol is also tested in this study. Different amounts of strontium are added to test solutions. Strontium-85 is added as an uptake tracer and measured by NaI detector. The strontium uptakes of Rapid Sr Samplers for both conditioning agents are evaluated by strontium-85 activities and listed in table 2.

Table 2. The comparison of uptakes of Rapid Sr Samplers for different conditioning agents

Sr spiked	Methanol	Ethanol
3 mg	100 %	66.6 %
1 mg	100 %	75.6 %
0 mg	100 %	100 %

The yttrium-90 elution solution is 20 ml of a 2M HNO $_3$ solution with 10mg yttrium. It is effective to elute yttrium-90 from Rapid Sr Sampler. Yttrium-90 is measured by cerenkov counting. Each sample is eluted by the solution three times. As data in table 3 indicate, yttrium-90 is almost eluted in the first time. Strontium is catch firmly by Rapid Sr Sampler. Only 1.03% of strontium is eluted with 1200ml of a 0.1M HNO $_3$ solution as shown in figure 2. Therefore Rapid Sr Sampler could be eluted many times to produce yttrium-90. The measurement of yttrium-90 activity is repeatable. The analysis result of strontium-90 of NIST SRM 4354 is 1.12Bq/g in this study. It is within the tolerance limit of NIST (1.09 Bq/g \pm 23%). All of analysis results for environmental samples interested in this study are listed in table4.

Table 3. Examples of net cerenkov counts of elution solution in different times

Elution	Sample 1 (cpm)	Sample 2 (cpm)	Sample 3 (cpm)	Sample 4 (cpm)
1st	28.17	14.87	21.06	10.80
2nd	0	0.84	0	0.60
3rd	0.37	0	0.24	0

^{*} The samples are counted in Parkard 2260XL with a period of 0-50 keV.

^{* ~100%} in the table means that the recovery is approximate 100%.

Table 4. Analysis results of environmental samples

Sample	Measured	Expected
SRM 4354	1.12 Bq/g	$1.09 \text{ Bq/g} \pm 23\%$
Tea	9.73 mBq/g	14.95 mBq/g
Drinking water	1.57 Bq/l	1.32 Bq/l
Shihmen dam	1.26 Bq/l	1.32 Bq/l
Deep well	5.51 Bq/l	5.03 Bq/l

^{*} The expected value of SRM 4354 is the tolerance limit of NIST.

1.00% 0.90% 0.80% 0.00% 0.00% 0.00% 0.30% 0.20% 0.10% 0.00% 1 2 3 4 5 6 7 8 9 10 11 12 ×100ml

Figure 2. Amount of the Sr loss in elution

Conclusion

Rapid Sr Samplers are easy-handle, fast, safe, efficient, and reliable for the analysis of strontium-90 in environmental samples. Its capacity of extracting strontium is more than 3.6mg strontium and with an efficiency of nearly 100%. The strontium-90 analysis of NIST SRM 4354 with Rapid Sr Sampler consists with the tolerance limit of NIST reported in the certificate.

Reference

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^{*} The expected value of tea is the value of analysis result measured by the routine method in our laboratory.

^{*} The expected values of water samplers are the amount of strontium-90 spiked in samples.