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Alpha Spectrometric Report, SNO

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In addition to carrying out further analyses for direct comparison of alpha spectrometric and mass spectrometric measurements, my work in the past quarter has concentrated on improving the reproducibility of radium recovery and counting efficiency.

A full elemental analysis by neutron activation of 395 grams of acrylic, performed at Guelph in 1988 but only recently brought to my attention, has indicated that Ba may exceed Ca in this material by a factor of 6. Even if a large error is assumed, this is still a most surprising discovery given the usual ratio of these two elements in nature. The likelihood of ^{226}Ra contamination in excess of its parent ^{238}U is consequently much enlarged. A recent analysis by myself of spec-pure BaCO_3 has shown this compound to contain as much as 0.13 dpm $^{226}\text{Ra}/100$ μg . This information puts a considerably higher priority on obtaining reliable Ra data.

Several analyses of spiked acrylic have been performed in such a way as to identify the procedures causing substantial losses.

A. Purification

1) Ignition Tube washout and removal of uranium and thorium on anion exchange resins.....total loss <5%

2) Separation from interfering ions remaining in solution:
i) Coprecipitation with BaSO_4

If substantial amounts of barium are present in the sample, then a coprecipitation step will provide the necessary decontamination from other elements in solution. Tests with 100-200 μg Ba carrier showed good reproducibility of radium recovery (~90%). Unfortunately in order to obtain good energy resolution, Sill (1987) has recommended that no more than 100 μg of Ba be present: quantities in excess of 400 μg would be considerably too large for good source preparation.

ii) Uptake of Ra on MnO_2 -coated beads.

This procedure is considered to be highly specific for radium in the presence of Na, Ca, etc in seawater concentrations. However recent tests using 100-200 μg Ba and ^{133}Ba tracer have shown that under the conditions chosen (very low ionic strength, $\text{pH} > 7$ and 4 hour contact time) barium is also strongly adsorbed by the beads and/or the test tube walls. Since losses of radium during this step were averaging >35%, and Ra -

Ba separation was slight. this procedural step is of little benefit.

iii) Cation Exchange Separation

Since Ba and Ra are very closely related in the periodic table, any procedure designed to separate these 2 elements quantitatively should provide adequate decontamination from the remainder of the residue. The cation exchange step already in place in our radium procedure has been given further study to optimize this separation. Column size was increased somewhat to handle potentially larger quantities of Ba than previously anticipated, and trial runs were carried out using ^{133}Ba tracer. The best conditions determined to date are as follows: the sample is dissolved in 0.01M NaEDTA at pH 6.4, and transferred to a column already prepared in that form. Ba is eluted at pH 8.5, and NaEDTA is washed from the column with 0.5M HCl until no residue remains in the dried effluent. Ra is subsequently eluted in a small volume of 3M HNO_3 . Recovery of Ra through this step has averaged 82%.

Average recovery through 2 anion and 1 cation exchange separations.....77%

B. Source Preparation

i) Precipitated Ba(Ra)SO_4 , mounted on a 0.1 micron filter.

These sources show good peak resolution, though losses due to self absorption reduce counting efficiency to ~25%. However a much more serious consideration is the substantial increase in background introduced by the Tuffryn filters used, despite overnight soaking in dilute acid (0.024 dpm in the region of interest). Other possible filters should be investigated, though the anticipated variability in barium from sample to sample makes this procedure less attractive as a routine method.

ii) Dried Source Preparation

While it is possible to prepare an almost massless source by drying a tracer solution directly on a counting disc, it seems to be extremely difficult to do so once the solution has been transferred from one container to another, even in a Class 100 fumehood. This situation is further compounded by passing the acid solution through a small cation exchange column immediately prior to source preparation. Consequently some energy degradation seems inevitable. However by flaming the disc following drying, it is possible to achieve a counting efficiency of 26% on a routine basis.

iii) Electroplating

This technique provides an excellent source, with very good peak resolution and correspondingly small background correction. Unfortunately recoveries have proved to be extremely sensitive to the presence of other ions in solution. However I have recently found that repeated refluxing of the final column eluate with HNO₃-H₂O₂ has sharply reduced this problem, and further improvement beyond the 65% plating yields recently obtained are anticipated.

In summary, then, recent recoveries for a full separation procedure: a) -obtained with dried source.....77%
 (counting efficiency 26%)
 b) -obtained with electroplated source..49%
 (counting efficiency 30%)

Recent Results

The last two large samples vapourized have been subdivided, following ignition tube washout, with approximately 95% being used for alpha spectrometric measurement, and 5% used for mass spectrometry. Agreement in both cases has been well within 2σ error.

Supplier	TIMS (pg.g ⁻¹)		ALPHA SPEC. (pg.g ⁻¹)	
	Th	U	Th	U
CY/RO	4.7	8.3	(232)7.0±1.0	(238)6.7±.79 (234)
			(228)6.0±1.0	(230)0.5±.15
				(226)0.22±.22
Stores	2.8	0.98	(232)3.2±.53	(238)0.71±.17 (234)2.0±.28
			(228)3.0±.49	(230)0.38±.09
				(226)0.82±.29

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