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Evidence for a Heterogeneous Distribution of Thorium in Acrylic

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1 Introduction

This report describes a measurement of the thorium concentration in acrylic by the method of neutron activation. Heterogeneity of the thorium distribution was studied by subdividing the sample *after* irradiation.

2 Method

A 36.7 g sample of "P2" acrylic (Polycast) was placed in a polyethylene bottle which was sealed by passing hot nitrogen gas around the lid of the container. A thorium and uranium standard was prepared by placing standard solutions in a supricil quartz apoule, evaporating them to dryness, and then flame sealing the ampoule. The ampoule was then placed in a hole in the polyethylene bottle and sealed there. The standard contained 1.4668μg of uranium and 1.5584μg of thorium.

The bottle was irradiated for 6 hours in the NRU reactor at Chalk River. The polyethylene bottle was cut open and the acrylic removed for counting. After irradiation the acrylic has swelled in volume, and has broken up into a coarse powder. The standard was removed from the bottle and dissolved in aqueous solution to provide a gamma ray counting standard.

The sample was counted at Queen's University using a Compton-suppressed germanium detector. The whole sample was counted several times and then it was divided into subsamples which were counted individually to search for evidence of a non-uniform distribution of the γ-ray activity. The masses of the subsamples are given in Table 1.

Finally, the sample was returned to Chalk River for analysis by mass spectrometry. Each of two sub-samples, with radioactivity known from the γ-ray analysis, was pyrolysed in a nitrogen atmosphere in a supricil quartz tube that subsequently was washed with aqueous solutions of HNO_3, HCl and HF. These solutions were then analysed for thorium (and uranium) content.

Table 1: Thorium Content of Acrylic Samples

Run	Sample	Counts	Time min.	Mass g	Rate s^{-1}	Std. rate s^{-1}	Thorium mass (ng)	Th Concentration $\times 10^{-12}$
4	ALL	2374 ±75	818.	36.7	2.90 ±0.09	572.	7.91 ±0.25	215.±7.
7	ALL	1188 ±41	733.	36.7	1.62 ±0.06	422.	5.99 ±0.21	163.±6.
8	A	1392 ±42	720.	20.02	1.93 ±0.06	422.	7.14 ±0.21	357.±11.
9	B	244 ±23	720.	16.44	0.34 ±0.03	422.	1.25 ±0.12	76.±7.
13	A1	521 ±31	708.	9.69	0.74 ±0.04	422.	2.72 ±0.16	280.±17.
14	A2	68 ±19	471.	10.33	0.14 ±0.04	422.	0.533 ±0.15	52.±15.
16	B	159 ±26	798.	16.44	0.20 ±0.03	422.	0.736 ±0.12	45.±7.
19	A1	261 ±26	638.	9.69	0.41 ±0.04	329.	1.94 ±0.19	200.±20.
21	A1a	345 ±26	543.	5.15	0.64 ±0.05	329.	3.01 ±0.23	585.±44.
24	A1b	281 ±26	541.	5.02	0.52 ±0.05	290.	2.79 ±0.26	557.±51.
26	B	140 ±25	564.	16.44	0.25 ±0.04	290.	1.33 ±0.24	81.±14.
29	A1a	586 ±38	950.	5.15	0.62 ±0.04	290.	3.32 ±0.21	644.±41.
31	A1a+A1b	657 ±34	590.	5.02	1.11 ±0.06	290.	5.98 ±0.31	588.±31.
33	B	297 ±35	790.	16.44	0.38 ±0.04	290.	2.02 ±0.24	123.±15.

3 Results

The results of the γ-ray counting are shown in Table 1. The sample labelling is as follows: ALL indicates the full sample; this was divided into subsamples A and B; subsample A was divided into A1 and A2; A1 was divided into A1a and A1b. "Std. rate" indicates the count rate of the standard source. Most of the samples have had repeated measurements, as shown in the table.

The thorium mass, determined for each sample, is given again in Table 2, where each column corresponds to set of measurements done at the same time. Unbracketed numbers indicate independent measurements. Numbers in parentheses () have been deduced. Some measurements are shown more than once in the table, in which case the repeated value appears in square brackets []. The errors indicated are statistical only. It can be seen that some of the measurements do not repeat well, and this is probably due to geometrical efficiency problems. In particular, the measurements for samples A1a + A1b ≡ A1, in columns 3 and 4, show less thorium than either of the constituent samples, A1a and A1b. With the exception of these two columns, the overall agreement between measurements is not too bad, and gives an average of 7.7±0.5 ng. An independent measurement of the total sample at CRNL gave 8.9±1.6 ng which is in agreement.

4 Discussion

The last column of Table 1 shows large variations in the thorium concentration from one sample to another. This variation is independent of such variables as the standard, handling of the samples, and the neutron flux. The division into subsamples, in effect, took place inside the sealed container during the neutron irradiation, (*i.e.*, it was during the irradiation that the sample fractured into a powder). After irradiation there is no more production of ^{233}Pa, and so contamination of the samples after the container has been opened is unlikely.

Table 2: Thorium content (ng)

	1	2	3	4	5	6	7
A1a	3.0±.2	3.3±.2					
A1b	2.8±.4	(2.7±.4)					
A1a+A1b	(5.8±.3)	6.0±.3	2.7±.2	1.9±.2			
A2	[0.5±.2]	[0.5±.2]	0.5±.2	[0.5±.2]			
A1+A2	(6.3±.4)	(6.5±.4)	3.2±.3	(2.4±.3)	7.1±.4		
B	1.3±.2	2.0±.2	0.7±.1		1.3±.1		
A+B	(7.6±.5)	(8.5±.4)	(3.9±.3)		(8.4±.4)	7.9±.2	6.0±.2

Many of the measurements of acrylic that have been made in the past have shown large fluctuatations from sample to sample, and it has been difficult to show that the variations were due to the material rather than due to contaminations, or problems with procedure. The results of the present experiment point to heterogeneity as a plausible reason for the fluctuations.

5 Comparison with Mass Spectrometry

The two samples which were analysed by mass spectrometry, were sample A, (*i.e.*, all the A subsamples mixed back together), and sample B. the result for sample A was 0.91 ng and for sample B 1.28 ng. The result for sample B compares favourably with the neutron irradiation results but sample A shows a disagreement by a factor of about 4.

6 Conclusions

Variations in the local concentration of thorium in acrylic have been found to range over a factor of 6 within a 37g sample. This is evidence that thorium has a heterogeneous distribution in acrylic.

The disagreement between neutron irradiation and mass spectrometry results on the same sample of acrylic is disturbing, and may indicate that thorium is being lost during the processing for mass spectrometry.

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