Measurements of Th & U in Acrylic for SNO. E. D. Earle & E. Bonvin Sept 17, 1992 SNO-STR-92-061

Abstract:

The concentrations of ²³²Th and ²³⁸U in Stage 1 sheet acrylic purchased from Rohm and Polycast has been measured by three methods. The concentrations of radioactive daughters from these isotopes has been measured by one method. These materials satisfy the design specifications. From a radioactivity background point of view the quality of the material from both suppliers is the same i.e. at or below the 1 ppt level for both Th and U.

Section 1 Introduction:

The acrylic vessel design criteria (Ref. 1) specifies that the finished vessel should contain not more than 0.3 mg of ²³²Th and 1.0 mg of ²³⁸U and that each of the Th and U decay chains shall be in equilibrium or, failing that, any disequilibrium shall be equivalent to parent concentrations of 0.3 and 1 mg respectively. The finished vessel inventory includes the virgin sheets, the support ropes embedded in the vessel wall, the neutron detector attachments, piping introduced into the vessel, bonding syrup and any contamination added during fabrication and construction. In order to allow for the expected contamination from fabrication, construction and attachments the design criteria explicitly limits the concentrations of ²³²Th and ²³⁸U in the virgin acrylic sheet to 2 and 7 pg/g respectively. Such concentrations would amount to 0.06 and 0.2 mg in a typical 30 ton vessel (i.e. 20% of the total permitted).

This level of Th & U concentration was selected early on in the project and was a compromise between what was thought to be the minimum attainable taking into consideration the measurements of Th and U that had been made up to that time, i.e. prior to the summer of 1991 and what was calculated to be the maximum that could be accepted without seriously jeopardizing the physics of the project. A comparison of the backgrounds from all sources, i.e. the D₂O, vessel, H₂O, PMTs, PMT support, liner and norite, indicted that at these concentrations the vessel was the dominant source of background (greater than 50%) to the experiment (Ref. 2). For this reason there was and still is a strong

incentive to reduce the Th and U concentrations in the vessel to values below the design criteria.

It will be shown below that selection of acrylic, careful fabrication procedures and improved measuring techniques have indeed demonstrated that the acrylic component of the finished vessel should be significantly cleaner than specified in the design criteria. It is our intention to continue to work to obtain a vessel better than the specifications.

Section 2 Experimental Techniques:

Three techniques (MS, AS, NA) have been developed to measure Th and U in acrylic and in other plastics.

a) MS or mass spectroscopy.

Quantities of acrylic, up to 5 kg, are cut into 200g bars (each 30 cm by 2.5 cm by 2.5 cm) with a table saw dedicated to this job. The bars are cleaned in alcohol and DDD water and sequentially vaporized in a synthetic quartz tube at 500 degs C under a nitrogen gas atmosphere. The residue is rinsed out by two applications of 30 ml of HNO3/HF acid and an application of 30 ml DDD H2O. Each hot rinse contacts the tube for one hour. The 90 ml of liquid is evaporated to dryness and the contents are passed to chemists for mass spectroscopy, either by thermal ionization mass spectroscopy at CRL (Ref. 3) or by inductively coupled plasma mass spectroscopy at NRC, Ottawa (Ref. 4). Background rinses of the tube with acid and DDD H2O are done before the vaporization of the acrylic and after the rinsing of the acrylic residue. There are two background measurements for Th and U associated with each acrylic residue measurement. Ninety percent of our values have been obtained by this technique which measures only the ²³²Th and ²³⁸U in the acrylic.

b) AS or alpha spectroscopy.

Quantities of acrylic, up to 25 kg, are cut into bars, vaporized and rinsed out as in the MS technique. About 10% of the residue is separated off for mass spectroscopy. The remainder is dissolved in acid, a ²²⁹Th tracer added and the solution passed through ion exchange columns which selectively separate the Th, U and Ra (Ref. 5). These elements are then electroplated onto stainless steel discs for alpha counting. Three separate discs are made for each acrylic sample. The intention is to measure both the parents and daughters in the Th and U chains and thereby check for disequilibrium. The last long lived daughter in the Th chain is ²²⁸Th and in the U chain is ²²⁶Ra. There is also a potential

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problem with the Th chain since ²²⁸Ra, which has a 5 year half-life, is a ²²⁸Th parent and so acrylic recently contaminated with ²²⁸Ra would not be identified by measuring the ²²⁸Th. However significant quantities of ²²⁸Ra should be associated with significant quantities of ²²⁶Ra and in those cases after a number of months a build up of ²²⁸Th in the Ra disc should be observable. The recovery efficiencies for the ion exchange and electroplating procedures are known from the measured intensities of the ²²⁹Th tracer (for the Th radioisotopes) and of the ²²⁹Th daughter ²²⁵Ra (for the Ra radioisotopes), via 225Ac ingrowth.

c) NA or neutron activation.

Up to 800 g samples of acrylic have been neutron activated in the NRU reactor and the ²³³Pa and ²³⁹Np counted with a gamma detector. The handling of the irradiated sample prior to measuring the ²³³Pa and ²³⁹Np concentrations has varied. a) Direct counting of the samples have been performed. This technique has also been used at Guelph on samples irradiated at the McMaster reactor (Ref. 6). b) Samples have been oxidized in a strong perchloric acid solution, the Pa removed by filtration and precipitation and then counted.(Ref. 7) c) Recently, and most relevant for this report, the surfaces of 800 g irradiated blocks have been milled away, the cores vaporized and the residue counted.

Section 3 Measurements:

a) Introduction:

The first measurements of Th and U in acrylic were reported in 1987 as part of the collection of annexes which accompanied the "white book" (SNO-87-12). Since then many samples have been measured on pieces provided gratis by the manufacturers of acrylic. Most of the measurements on these pieces indicated levels of Th and U above the design specifications. In some cases, the high values were due to contamination during handling since handling techniques were being developed at the same time. SNO reports have been submitted on the results of these measurements and are available upon request.

In the summer of 1991 we purchased a large 5 cm thick sheet of cell cast acrylic (Stage 0 material) from each of the two potential suppliers, Rohm and Polycast, and subsequently we purchased 2-5 cm thick sheets and 1-11 cm thick sheet, all 180 cm by 180 cm, (Stage 1 material) from each supplier. The suppliers were advised that supplier selection would be based on the quality of the Stage 1 material. Some of the Stage 0 material was sent to the company (Reynolds Polymer

Technology or RPT) chosen by SNO to fabricated the vessel so that RPT could go through their proposed fabrication procedures and SNO could check for increased contamination. This report will contain only the results of measurements on the Stage 0 and 1 material.

b) MS measurements:

- i) Recovery Efficiencies. Table 1 lists the recovery efficiency of the vaporization, acid rinse and mass spectroscopy on samples spiked with known amounts of Th and U. The first four samples were prepared by depositing a solution containing a known amount of Th and U onto the surface of the acrylic. The solution was then evaporated before the acrylic was vaporized. The other 18 samples were from pieces of an acrylic rod made at Los Alamos. The monomer was spiked with Th before the acrylic was polymerized so that the Th was uniformly distributed throughout the rod. In some cases the residue from the entire sample was measured by mass spectroscopy whereas in most cases only a fraction was used for mass spectroscopy with the remainder going for alpha spectroscopy. The two low recoveries (#6 & #8) resulted when parts of the rod containing no spike were inadvertently used. The average recovery efficiency is 93%. The operation of preparing the residue for the mass spectroscopy was independently measured to have a 97% recovery efficiency (Ref. 3) and so the vaporization and subsequent rinsing of the synthetic quartz tube would appear to have a recovery efficiency of about 95%.
- ii) An example of the data file. As an example of how the results have been tabulated the results of the mass spectroscopy measurements on the Rohm Stage 0 material are shown in detail in Table 2. Column 1 contains the date of the vaporization and the mass spectrometer used, either inductively coupled plasma or thermal ionization. Col. 2 is the weight. Col. 3 - the quartz tube used for the vaporization. Col. 4 - details of the acid mixture used, the acrylic material identification and the number of sequential rinses to remove the acrylic residue. Col. 5 - the mass of U measured. Col. 6 - the calculated U concentration in pg/g. Cols. 7 & 8 - the mass and concentration of Th. Cols. 9 & 10 - details of cutting and cleaning. Col 11 - other relevant comments. A total of 18 samples, adding up to 60 kg of acrylic, were analyzed with some of them thermoformed on felt or rubber material in a furnace at CRL. The average ²³²Th and ²³⁸U levels are 0.9 and 1.9 pg/g respectively with no indication of contamination due to thermoforming. Similar results were obtained for the Polycast Stage 0 material. In particular, 12 samples totaling 45 kg contained 0.5 and 0.3 pg/g of Th and U.

- iii) Stage 1 results. The Th and U concentrations in the various sheets of Stage 1 material are listed in Table 3 and shown graphically in Fig. 1. The measurements are grouped into lots according to the sheet from which the material was taken. A total of seven Rohm and three Polycast sheets were checked. The two 5 cm sheets of each supplier were from the same batch of monomer. The first two Rohm 10 cm sheets were from different batches and failed because of bubbles and water damage. The relatively high concentrations of U in two of the four measurements on the water damaged sheet may be from the water damage. The last three Rohm 11 cm sheets were made with different recipes, but the monomer may have been from a single batch. Clearly the material is well within specifications for both Th and U. All these sheets were visually inspected for inclusions, dust etc and a separate report written (Ref. 8). The concentration of inclusions and dust varies significantly from sheet to sheet and within each sheet but the sheets with the most dust are not correlated with the sheets with apparently higher Th/U. We believe that any apparent differences in Th/U concentration between sheets and/or suppliers are not significant. These results are available in a tabular form similar to Table 2 should they be required.
- iv) Results following fabrication qualification. While not relevant to the supplier selection decision, samples of Stage 0 material were thermoformed on rubber and felt, with and without the protective paper, were machined, sanded, annealed and bonded by RPT. The Th/U concentration was measured after each stage by mass spectroscopy. A total of 26 samples were measured and the details are available in tabular form, similar to Table 2. Visually one can see an increase in the dust concentration on the sanded surfaces and those samples may have higher Th/U concentrations also. We say "may" because the measurement uncertainty is large due to the small sample size, 0.5 kg. In any event the integrated increase in a sheet due to the sanding in less than 0.2 pg/g for the worst case of 15 samples and so is not significant. In addition, we have determined that the rubber backing is satisfactory and that it is not necessary to keep the protective paper on the sheet during thermoforming.

c) AS measurements:

Fig. 2 is an alpha spectrum of the electroplated Th radioisotopes from the residue of 14 kg of Rohm - 5 cm Stage 1 acrylic. Eight peaks are identified - ²²⁹Th, from the tracer added to the sample, and its four daughters ²²⁵Ac, ²²¹Fr, ²¹⁷At and ²¹³Po, and ²³²Th with its daughters ²²⁸Th and ²¹⁶Po. The channel width of the peaks vary because in some cases several alpha particles contribute to the decay. For example, ²¹⁶Po, ²¹³Po and ²¹⁷At each decays by a single alpha particle and we sum over

2.5 channels whereas ²²⁹Th decays by eight alpha particles and we sum over 6.5 channels. We know the amount of ²²⁹Th added to the sample and thereby know the recovery efficiency for the Th radioisotopes. The ²²⁹Th daughters are much weaker than the parent because they have not had time to reach equilibrium after the chemical separation of the Th, U and Ra. (Recall that the ²²⁹Th daughter, ²²⁵Ra (which decays by beta emission to ²²⁵Ac), has a 15 day half life.) It would appear from this spectrum that ²²⁸Th may not be quite in equilibrium with its parent ²³²Th. The ²¹⁶Po peak is noted because it is due to a single alpha particle and has an energy well away from other alphas. It can be a confirmation of the strength of the ²²⁸Th peak. We concluded from these data that the ²³²Th in the sample was 1.6 +- 0.4 pg/g and the ²²⁸Th (normalized to the equivalent ²³²Th) was 2.6 +- 0.5 pg/g. The mass spectroscopy ²³²Th value for this sample was 1.3 pg/g.

Fig. 3 is an alpha spectrum of the Ra radioisotopes from the residue of a 10 kg Rohm - 10 cm Stage 1 piece of acrylic. The ²²⁹Th tracer is absent but its daughters which have had time to grow in from the decay of ²²⁵Ra are all evident i.e. ²²⁵Ac, ²²¹Fr, ²¹⁷At and ²¹³Po. In addition, there may be some ²²⁶Ra from the ²³⁸U chain. As with Fig. 2 the peaks are marked; ²²¹Fr is particularly wide. The Ra radioisotopes recovery efficiency can be determined from the intensity of the ²²⁵Ac peak, knowing the ingrowth time between the chemical separation and the alpha counting. The ²²⁶Ra concentration (normalized to the equivalent of ²³⁸U) is 0.3 +- 0.1 pg/g. The ²³⁸U valve for this sample from mass spectroscopy was measured to be 0.6 pg/g, indicating no significant disequilibrium in the U chain.

The AS results on Stage 1 material are summarized in Table 4 where they are compared to the MS values on the same sample. Daughter concentrations are converted to parent concentrations assuming equilibrium. Procedural blanks have also been done from time to time to confirm the background conditions. Two 10 cm thick samples (#7 &10), one from each supplier, showed high Th concentrations when first measured but the second samples from these same sheets (#14 & 15) did not. Otherwise the measurements are consistent with the MS measurements and disequilibrium is not more than a factor of two. The AS spectroscopy measurements are few, there are large statistical uncertainties in the measurements and the potential for background contamination during the chemical separation is greater than with the MS measurements. Despite these caveats we conclude that the acrylic is below the design criteria of 2 pg/g Th/U and in equilibrium.

d) NA measurements:

Fig. 4 is a gamma ray spectrum taken with a well-Ge detector of the residue from the vaporized core of 677 g of neutron irradiated Rohm 11 cm Stage 1 material. The sample was irradiated in the Si J-rod facility at NRU for 30 mins in a 1.5 x 10¹³ n/cm² flux. Neutron capture on ²³²Th produces 27 day half-life 233Pa which has a 312 keV gamma ray in its decay. No peak at that energy is observed. The detector resolution is evident in the adjacent 320 keV peak from 51Cr. The 2 sigma limit on ²³²Th in this sample is 0.05 pg/g. Concentrations of ²³²Th below 0.3 pg/g have been measured by this method in three samples of Polycast acrylic and in two other samples of Rohm acrylic (Table 5). Neutron activation of ²³⁸U produces 2.4 day ²³⁹Np which has a 278 keV gamma ray. No peak at that energy is observed in Fig 4. The 2 sigma limit on ²³⁸U in this sample is 1 pg/g. The activity in several other irradiated Rohm samples, even after the surfaces were milled away, was two orders of magnitude higher, giving us a significant handling problem and preventing us from making similar measurements on these samples. From the spectrum shape we believe this to be a beta activity due to some trace impurity in the Rohm acrylic, not to gamma rays from Pa and Np. The apparent variation in Rohm samples is being investigated in more detail.

Two pieces of acrylic spiked at LANL with ²³²Th were also irradiated, counted and, then, vaporized and counted again in order to verify that we were not losing the ²³³Pa in the vaporization stage. The recovery efficiency was found to be 100% or similar to that for ²³²Th. (See Section 3(b) above)

Section 4 Conclusions:

The apparent absence of ²³²Th at the 0.1 ppt level in the core of a number of samples analyzed by neutron activation suggests that the acrylic from both suppliers may be much cleaner than the < 2 ppt measured by both mass and alpha spectroscopy. We suggest that either the bulk of the Th and U contamination is on the surface of the acrylic or we are adding trace amounts of contamination during the handling of the material. In any event the acrylic from both suppliers meets the design criteria and, in addition, there is no experimental evidence in these radioactivity measurements to prefer one supplier over the other.

Section 5 Acknowledgements:

The work summarized in this report has been performed with dedication and determination by many people. Ron Deal has been indispensable, working on this project from the beginning. He has directed the flow-through of acrylic and developed the vaporization technique. He, with Mark Gauthier and Earl Gaudette, have done all of the vaporization. The mass spectroscopy at CRL has been competently done by Nancy Elliot, assisted by Monique Maillet. At NRC, Dr Jim McLaren has provided much advice and with Joe Lam performed the ICPMS measurements. Dr Gwen Milton has directed the alpha spectroscopy work, ably assisted by Sheila Kramer and Bert Risto. Dr Bill Edwards with Marceal Hurteau and Rozanne Collins have performed the neutron activation and early on some sophisticated chemistry to dissolve irradiated acrylic in attempts to separate the protactinium. Dr Ted Clifford was very much involved at the beginning in developing techniques and setting up equipment.

References:

- 1) P. Cumyn, Acrylic Vessel Design Criteria, DC-17-310.01.1
- 2) I. Stairs, B. Robertson and P. Skensved, Beta-Gamma Backgrounds from ²¹²Bi, ²²⁸Ac, and ²³⁴Pa in the heavy water and acrylic, STR-92-066
- 3) N. Elliot, TIMS Analysis for the SNO Project, STR-92-059
- 4) ICPMS at NRC
- by J. McLaren
- 5) G. Milton et al, Uranium Series Disequilibrium in Rock-Water Systems of the Canadian Shield, PhD Thesis, Univ. of Waterloo.
- 6) Jagam et al., SNO Acrylic Radioactivity, Guelph prep. GWP2-NP-20, submitted to Jour. Radioanalytical and Nuclear Chemistry.
- 7) M. Hurteau & W. Edwards, Neutron Activation of U & Th in Lucite followed by Dissolution and Oxidation using the Liquid Fire Process. Text of a conference talk on file and available upon request.
- 8) D. Earle, Visual Inspection of Stage 1 Acrylic. Report on file and available upon request.

Tables:

- 1) Recovery efficiencies of spiked samples by MS.
- 2) Details of MS results on Rohm Stage 0 material.
- 3) Th and U content of Stage 1 material by MS.
- 4) Concentrations of Th, U and Ra isotopes in Stage 1 material by AS.
- 5) Two sigma limit of Th/U in acrylic core.

Figures:

- 1) Weight distribution of Stage 1 material as a function of measured Th/U concentration..
- 2) Example of an alpha spectrum after Th separation.
- 3) Example of an alpha spectrum after Ra separation.
- 4) Example of a gamma spectrum from irradiated acrylic residue.
- 5) Two sigma limits on Th/U in acrylic core.

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				Table 1						
				Recovery Effic	iencies	of s	piked	sampi	es	
Date)	wt.	tube		U ng	Meas.	Th ng	Meas.	% rec.	
				Elliot spike						
	Apr-90				27.3	26.2	36.7	34.6	95	
					48.7	44.7	65.3	59.8	92	
					0	1.2	0	0.62		
					76.3	74.8	102	100	98	
				LANL spike at 1	32 ng/	g		Th ng	% rec.	
Nov	7-90	3.47g	9	18.6 % of #1				72.1		
	19-91	2.3		#2				234		
	30-91	2.47		#3				310		
Dec	12-91	3.15		#4				397		
Dec	13-91	2.45		#5				289	89	
		4.5		3.85 % of #6				9.16	43	
	1-91	0.84		#7				105		
	6-91	4		9.6 % of #8				6.9		
	12-91	3.75	_	6.1 % of #9				30.6	102	
	4-91	3.92		#10						
	5-91	9.36		5.3% of # 11				68		
	17-91	11.27		11.6 % of #12				173		
	30-91	2.6		12.2% of #13				46.8		
	30-91	3.26		13.4% of #14				58.5		
	23-91			12% of #15				44	82	
	19-91	3.81		9.8% of #16				41	83	
	18-91	4		15.7% of #17				79		
Jun	5-92	5.8	V1	6.4% of #18				43	86	77.20.

			Table 2								
			etails of MS	on ROHM		Stage 0	material	=			
Date	Weight Tube ID	npe	ΩI	n	n	Th	Th	Th/U	Cut	Cleaned	Comments
	kg	1		ng	ppt	Б	ppt				
Sep 24-91	2.13	-	Bkgd, A4	<0.2		0.3					
ICP-MS			Ri2	7.1	3.1	6.3	2.7		115,tabl	alc/H20	opt., 10 pieces
			3 rd rin	0.3		0.3					
Sep 25-91	2.12	2	Bkgd, A4	<0.2							
ICP-MS			R12	5.3	2.2	2.4	0.9		115,tabl	alc/H20	opt., 10 pieces
			3 rd. rin	0.4		0.2					
Sep 25-91	2.15	က	Bkgd., A4	0.4		0.4					
ICP-MS			R12	7.1	C	2.2	0.7		115,tabl	alc/H2O	opt., 10 pieces
			3 rd rin	0.4		0.3					
				•							
Sep 26-91	2.13	-	Bkgd, A5	0.3		0.2					
ICP-MS			R12	4.3	1.7	1.5	0.5		115,tabl	alc/H2O	opt., 10 pieces
			3 rd rin	0.4		0.2					
Sep 26-91	2.13 7A		Bkgd, A4	<0.2		0.3					
ICP-MS			R12	4.9	2	4.4	1.8		115,tabl	alc/H20	opt., 10 pieces
	•		3 rd rin	0.4		0.3					
Sep 30-91	2.35	2	Bkgd., A4	0.2		<0.2					
ICP-MS			R12, A5	6.1	2.3	2.5	0.8		115,tabl	alc/H2O opt.,	opt., 11 pieces
			3rd rin	0.4		0.2					
Sep 27-91	2.33	6	Bkgd., A4	<0.2		<0.2	-				
ICP-MS			R12, A5	4	1.5	1.4	0.5		115,tabl	alc/H2O	opt., 10 pieces
			3rd rin	0.2		<0.2					
		7									

Date	WeightTubeID	pe	OI	n	n	Th	ų.	Th/U	Cut	Cleaned	Cleaned Comments
	kg			ng	ppt	Б	ppt		-		
Sep 30-91	2.32	-	Bkgd. A5	<0.2		0.4					
ICP-MS			R12	6.4	2.5	4	1.3		115,tabl	alc/H2O	opt., 11 pieces
			3 rd rin	0.4		0.8					
	100	\top	14	0		•					1
Oct 2-91	1./8 / A		BKgd., A5	0.3		-					Щ.
ICP-MS			R12 on felt	3.7	1.7	Ī	0.4		115,tabl	alc/H2O therm.,	therm., felt,
		-	3rd rin	0.5		0.5					down
Oct 2-91	1.78	6	Bkgd., A5	0.2		<0.2					opt., 9 pieces
ICP-MS			R12 on felt	3.8	1.7	1.4	0.4		115,tabl	alc/H20	therm., felt,
			3rd rin	0.5		0.5					dn
Oct 3-91	1.85	3	Bkgd,A5	0.59		0.55					therm.,
TIMS			R12,	3.3	1.5	2.3	1.1		115,tabl	alc/H2O	opt., 9 pieces
			3rd rin	0.65		0.81					rubber,up
Oct 3-91	1.68	11	Bkgd,A5	0.37		0.4					down
TIMS			R12	4.2	2.7	1.6	-		115,tabl	alc/H2O ther.,	ther., rubber,
			3rd rin	1.1		-					opt., 9 pieces
- 1				-							- 1
Oct 18-91	10.79	က	Bkgd,A5	0.7		o					50 pieces
TIMS			R12	18.5	1.7	12	-		115,tabl	alc/H20	
			3 rd rin	1.4		1.2					
Oct 24-91	1.86	2	Bkgd,A5	0.4		0.7					opt.
TIMS			R12	2.9	1.1	2.4	-		115,tabl	alc/H2O	11 pieces,
			3rd rin	0.4		0.3					felt, down
Oct 30-91	1.78 7A	ı	Bkgd, A5	0.7		0.8					opt., 11 pieces
TIMS			R12	3.3	-	2.1	0.4		115,tabl	alc/H2O felt,	felt, up side
			3rd rin	0.7		0.7					

Date	WeightTub	Tube	eID	n	n	Th	Th	Th/U Cut	Cut	Cleaned	Cleaned Comments
	kg			ng	ppt	ng	ppt				
Oct 30-91	1.8	5	Bkgd, A5	0.21		0.11					opt., 11 pieces
TIMS			R12	2.7	6.0	1.7	4.0		115,tabl	alc/H20	alc/H2O rubber, down
		,	3rd rin	0.45		0.45					
Oct 30-91	1.84	3	Bkgd, A5	1.2		1					opt., 11 pieces
TIMS			R12	6.3	2.6	2.6	0.5		115/tabl.	alc/H20	115/tabl. alc/H2O rubber, up
			3rd rin.	8.0		8.0					
Jun 4-92	16.9	16.9 s15	A13	0.15		0.22					
SWIL	15%		R12 Stage 0	4.6	1.7	1.4	0.5				
			1/2/2	1.4		1.5					
			85% for alpha								
			S15 bgd again	2.2		1.5					

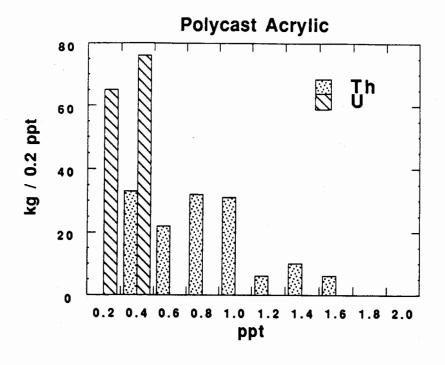
Table 3.	Mass Spe	ectroscopy	Results of	on Stage	Material.
Rohm, Wt	Th in pg/g	U in pg/g	Polycast, Wt.		U in pg/g
sheet 1, 5cm			sheet 1, 5 cm		
2.2	0.6		2.97		
2.4	0.6		2.83		
2.25					
2.28	0.8				
18.17			5.96		
2.22	1	0.6			
2.36			17.9	0.4	0.2
2.24	0.6	0.4			
2.3	0.5		sh 2, 5 cm		
2.18	0.8	0.6			
2.2	0.8	0.9	6.58		0.5
2.2	0.9				0.4
2.2	0.9	1	7.42	1	0.3
sheet 2, 5 cm			sh 3, 10 cm		
6.63	0.4	0.4	6.64		
6.62	0.5	0.3	6.7		
6.66	0.6		6.64		
6.81	0.3	0.2	6.56		
	·		10.38	1.1	0.4
sheet 1 & 2			10.12		0.4
17	1.3	0.6	23.28	0.8	
			18.99	0.9	0.3
sh. 3, 10 cm					
4.7	0.5	0.6		,	
sh 4, 10 cm					
4.8	1.5	4			
4.55	1.5	4.9			
4.63	1.3	0.8			
4.6	1.5	1.1			
9.2					
9.2					
sh 5, 11 cm	5 · -				
4.18	1.1	0.5			
4.3	1.2	0.9			
1.0	,	0.0			,
sh 6, 11 cm					
13.22	0.6	0.4			
sh 7, 11 cm	0.0	0.4			
16.88	0.5	0.5			,

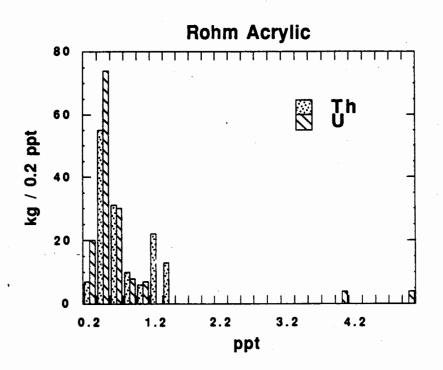
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Table 4	. Alp	ha Spectro	scopy F	Results	on Sta	age 1	Material		
Supplier	ID	Date Vap	Wt kg	ļ	MS	ppt (M		aipha S	ppt (AS)
Stage 0	# 1	Jun 3-92	16.88		232Th			232Th	0.3 (0.3)
R-2"				<u> </u>	238U	1	1.3	228Th	1.0 (0.5)
	-							226Ra	0.14
P-2.5"	#11	Nov 29-91	17.2		232Th			232Th	0.6
Stage 0					238U	(0.1	228Th	0.4
								238U	
								234U	0.2
								230Th	0.1
								226Ra	0.6
P-4"	#3	Jun 5-92	23.28		232Th	C	0.8	232Th	0.45 (0.3)
					238U	(0.5	228Th	1.3 (0.8)
							·	226Ra	< 0.05
R-2"	#4	Jan 15-92	18.17		232Th	0).5	232Th	0.4 (0.2)
					238U	().4	228Th	0.5 (0.3)
								238U	0.3
								234U	0.8
								230Th	0.2
	<u> </u>							226Ra	0.1
P-2"	#5	Feb 19-92	17.9		232Th).4	232Th	4 (4)
	· · ·	100 100			238U	·).2	228Th	6 (3)
					2000		7.2	216Po	2 (2)
	+							226Ra	<0.5
		2nd separa	ation		 			232Th	<1.3
	+	Ziid copaire					+ :	228Th	1.5
	+				 	 		230Th	0.8 (0.4)
R-2"	#6	May 22-92	17		232Th	1	.3	232Th	1.6 (0.4)
11-2	" 0	IVIAY ZZ JZ	1,		238U).6	228Th	2.6 (0.5)
					2000	-	,	226Ra	<0.15
P-4"	#7	Mar 16-92	10.12		232Th	-	.4	232Th	1.7 (0.5)
F * 4		es combined	10.12		238U).4		
	#8	Mar 16-92	10.38		232Th			228Th 216Po	6.2 (1.0)
	#8	Mar 16-92	10.38		 		.1		5.4
D 4"	440	A 10.00	0.01		238U).4	226Ra	<1
R-4"	#10	Apr 16-92	9.91					232Th	13
	-				<u> </u>			228Th	38
5 4"	-		10.00		00071			226Ra	0.3
R-4"	#13	Jun 30-92	16.88		232Th).5	232Th	0.4 (0.3)
	ļ				238U).5	228Th	0.8 (0.3)
	-							226Ra	0.06 (.06)
R-4"	#14	Jul 8-92	13.22		232Th).6	232Th	0.8 (0.3)
	_				238U	0).4	228Th	0.3 (0.3)
	<u> </u>							226Ra	<0.2
P-4"	#15	Aug 14-92	18.99		232Th	+).9	232Th	1.0 (0.3)
					238U	C).3	228Th	1.3 (0.3)
								226Ra	0.3 (0.1)

Table 5.	Two S	Sign	na Limit	of
	Th/U	in a	erylic c	ore.
Sample	Wt g		Th in pg/g	U in pg/g
Rohm-4"			< 0.05	< 1
Rohm-2" Polycast-2"			< 0.05 < 0.04	
Polycast-2" Polycast-2"			< 0.12 < 0.3	
Rohm-4"				0.5 +- 0.1

-





100 $213P_0$ lpha-spectrum from Th isotopes, acrylic residue 8 0 228Th 225Ac 221Fr 229Th 0 0 20 100 -200 counts

hannels

100 α -spectrum from Ra isotopes, acrylic residue $213P_0$ 0 0 0 80 217At . 0 9 0 225Ac221Fr 9 o 0 226Ra | 0 20 -40 8 20 Sinno

(. .)

Channels

Fig. 4

