SNO-STR-90-4

Collection of notes on acrylic radioactivity

[illegible]

1) Mass Spec. results during 1989

2) Radiochemical procedure and neutron activation results at CRNL

3) Gamma counting neutron activation result from LBL

under separate cover } 4) Gamma counting-neutron activation result from Queen's

5) Summary of neutron activation results

draft → 6) Summary of alpha-counting results at CRNL

E. D. EARLE

Acrylic Mass Spect. Results during 1989.

Summary of mass spectrometry measurements made at NRC by Jim McLaren and at CRNL by Floyd Miller. All acrylic evaporations and washings were done by Ron Deal at CRNL. Numbers are usually recorded in ng and to get the acrylic concentrations one must divide by the acrylic weight. In some cases the wash was divided into two samples which were then measured by NRC and CRNL mass spec.

Set 1

This set was measured at NRC. The tube was washed with HNO3 only. The acrylic was Polycast 1" material supplied to us several years ago.

Sample	NRC (May 26/89)		ppt
	Th	U	
Water	0.1	0.02	
Acid	0.1	0.02	
1 st blank	0.1	0.05	
2 nd blank	0.1	0.02	
3rd balnk	0.21	0.06	
4th blank	0.1	0.02	
P2 0.88 kg 1" material	0.94	1.44	0.95/1.6
Rinse	0.1	0.04	

			NRC (Jun 21/89)		
Sample			Th in ng	U in ng	ppt
1	#5 tube		0.25	0.36	
2	P2 0.88	1" material	1.26	0.53	1.1/0.4
3	#5 tube		0.43	0.08	
4	#5 tube		0.49	0.05	
5	P2 0.90		1.55	0.41	1.1/0.4
6	#5 tube		0.54	0.08	
7	#5 tube		0.40	0.02	
8	P2 0.87		1.38	0.41	1.1/0.4
9	#5 tube		0.35	0.12	
10	#5 tube		0.34	0.45	
11	P2 0.89		0.93	1.92	0.7/1.8
12	#5 tube		0.24	0.18	

sample		NRC (Aug 17/89) Th in ng	U in ng	ppt
1	unopened clean bottle	<0.1	0.05	
2	tube bkgd rinse	0.37	0.35	
3	P2 acrylic rinse 0.89 kg	1.45	0.44	1.1/0.2
4	tube bkgd rinse	0.47	0.12	
5	cleaned bottle (pyrex)	<0.1	0.05	
7	reagent blank Jim evap.	0.39	0.10	
8	reagent blank Ron evap.	0.55	0.11	
9	cleaned bottle (plastic)	<0.1	<0.05	
10	bottle left standing	0.52	0.11	
1	cleaned (days) bottle	<0.1	<0.05	
2	tube bkgd rinse	0.30	0.09	

Set 2.

These were measured by both NRC & CRNL and the acrylic is 2.5" material thermoformed in Calif. before it was shipped to CRNL. We looked at both Polycast and Rohm material.

Bottle ID	Contents	CRNL		NRC	
		Th	U	Th	U
	Tube #1 Aug 30	0.35	0.45		
1	Acid from a new bottle	0.06	0.44	0.1	0.05
	H2O	0.09	0.37		
2" delivered	Mar 88	ng	ng	ng	ng
R1C-A	Bkgd wash of tube #1	0.06	0.18	2.64	3.17
R1C-B	1.2 kg Rohm 1 C acrylic	2.02	2.38	0.17	0.05
R1C-C	2nd rinse after R1C-B	0.49	0.30	1.15	0.16
2" delivered	Oct 88				
R1D-A	Bkgd wash of tube #1	0.76	0.39	0.25	0.07
R1D-B	0.87 kg Rohm 1 D acrylic	1.1	2.16	1.21	2.65
R1D-C	2nd rinse after R1D-B	0.28	0.52	0.44	0.31
2" delivered	Oct 88				
R2A	Bkgd wash of tube #5	0.08	0.27	0.13	0.05
R2A-B	0.71 kg Rohm 2 A acrylic	2.46	?	2.94	9.18
R2A-C	2nd rinse after R2A-B	0.45	1.58	0.46	5.16
R3 was thermoformed on metal 2" Oct 88					
R3A	Bkgd wash of tube #1	0.33	0.27	0.42	1.01
R3A-B	0.82 kg Rohm 3 A acrylic	67	71	72.6	91.9
R3A-C	2nd rinse after R3A-B	20.6	9.2	17.2	13.4

The concentrations of Th in these four samples would appear to be 2.6, 1.7, 4.5 and 110 ppt if the rinsings are successful.

Label	Comments	CRNL		NRC (Oct 25/89)	
		Th	U	Th	U2
2 2 5/8 " Oct 88		ng	ng	ng	ng
P6A-A	Tube Bkgd	0.38	5.9	0.42	4.65
P6A-B	0.92 kg T. #5	5.5	7.8	10.5	60.2
P6A-C	2nd rinse	3.2	1.2	3.2	1.4
P7 was thermoformed on rubber(Oct 88)					
P7A-A	Tube Bkgd	14.2	3.5	3.8	2.5
P7A-B	0.82 kg T. #1	11.2	12.7	20.9	17.0
P7A-C	2nd rinse	11.2	0.88	14.5	1.4
P8 was thermoformed on metal(Oct 88)					
P8A-A	Bkgd	1.82	2.15	3.5	2.8
P8A-B	0.85 kg T. #5	4.43	5.23	4.6	5.5
P8A-C	2nd rinse	3.48	0.43	4.3	0.3
2 5/8" delivered Jun 88					
P4C-A	Bkgd	1.05	0.34	1.3	0.2
P4C-B	0.92 kg T. #5	29.6	8.64	55.9	11.6
P4C-C	2nd rinse	13.3	1.26	20.5	1.2

The concentration of Th in these samples would appear to be of the order of 12, 40, 8 & 68 ppt if the rinsings are successful

Bottle #1	Bkgd of suprasil boat to be used for liquid moner vaporization	0.27	0.12
Bottle #2	2nd rinse of boat.	0.32	<0.05

Following these measurements we became concerned about the completeness of the rinsing proceedures and so we embarked on a program to check the effectiveness of the proceedure. Since we were about out of Polycast material we used Cyro material, of which we had plenty. In the notes that follow we are concentrating on the history of the tube in which the evaporation was done so as to track the cleaning effectiveness and we have relisted the results of the analysis of the thermoformed material.

Tube #1 History

		Th ng CRNL-NRC	U ng CRNL-NRC		
July 17	R1B	for Gwen			8.72 kg
Aug 30	bdg	0.35-	0.45		
		0.06-2.64	0.18-3.17	Bottles mixed up here?	
Sept 12	R1C	2.02-0.17	2.38-0.05		2.42 kg total weight
		0.49-1.15	0.30-0.16		
		0.76-0.25	0.39-0.07		
Sept 18	R1D	1.10-1.21	2.16-2.65		1.75 kg
		0.28-0.44	0.52-0.31		
		0.33-0.42	0.27-1.01		
Sept 20	R3A	67.0-72.6	71 - 92		1.64 kg
		21 - 17	9 - 13		
Sept 21	**Spiked run in tube #2**				
		?	?		
Sept 26	R4A	?	?		1.73 kg
		?	?		
Oct 2		14 -3.8	3.5 - 2.5		
	P7A	11.2-20.9	12.7-17		1.65 kg
		11 - 14.5	0.9 - 1.4		
Oct 24 HF introduced.					
Oct 24		71			
		46			
Oct 30		19			
		29		HF here	
		18			
Nov 2		14		longer rinse & heat	
		12			
		10		18 hr rinse & heat	
		2	1.25	20 mins	
		1	0.96	20 mins	
Nov 7		2.19	1.25	1 hr	
		1.03	0.96	1 hr	
Nov 21		0.57	0.22	after 96 hr soak to clean	

Where did the 71 ng obtained on Oct 24 come from? How long was this Th in the tube? Are all the earlier results suspect? Or was the Th introduced during or after the Sept 20 vaporization?

History of Tube #5

		Th in ng CRNL-NRC	U in ng CRNL-NRC	
		0.08-0.13	0.27-0.05	
Sept 19	R2A	2.46-2.94	? -9.18	1.42 kg
		0.45-0.46	1.58-5.16	
		0.38-0.42	5.9-4.65	
Sept 27	P6A	5.5 -10.5	7.8 -60.2	1.84 kg
		3.2 - 3.2	1.2 - 1.4	
		1.82-3.5	2.2 - 2.8	
Oct 3	P8A	4.43- 4.6	5.23- 5.5	1.78 kg
		3.5 - 4.3	0.4 - 0.3	
		1.1 - 1.3	0.3 - 0.2	
Oct 6	P4C	29.6-55.9	8.64-11.6	1.85 kg
		13.3-20.5	1.3 - 1.2	
Oct 18	P2G	?	?	0.66 kg to McLaren
Dec 6	Bkg	85.5	2.7	HNO3,HF 3 heat lamps
		10.8	1.0	
Dec 15	Bkg	4.4	1.7	HCl, HNO3, & HNO3, HF
		3.0	0.72	HNO3, HF
Dec 20	Bkg	5.9	1.6	HNO3, HF, HCl 1 hr
		0.8	0.65	HNO3, HF

As with tube #1, when was the 85 ng measured on Dec 6 introduced? On or after Oct 6 or before that date?

History of tube #6

	Th in ng CRNL-NRC	U in ng CRNL-NRC	
Nov 8	0.25	0.95	
	0.26	25.6	
	0.08	0.21	
	0.10	0.29	
	0.11	0.52	Tube heated in oven for 30 hrs
	0.10	0.45	
Nov 16	0.15	0.52	
C1F 2.01 kg	91.3	40.1	1 hr
	18.3	0.62	1 hr
	10.8	0.31	2 hr
Nov 23	3.5	0.71	1 hr
	1.4	0.42	1 hr
Nov 30 Bkg	0.65	28.1	
C1G 2.05 kg	40.7	28	HNO3,HCl
	16.5	0.8	"
	15.3	0.9	HNO3,HF
	2.3	7.0	"
	0.8		"

History of tube #7

Nov 20 Bkg	0.11	0.07	
Dec 7 Bkg	0.15	0.68	HNO3, HF 3 heat lamps
C1H 0.451 kg	1.8	1.4	HNO3
	0.62	0.74	HNO3
	0.70	1.2	HNO3, HF
Dec 13	0.22	0.59	HNO3, HCl

History of tube #8

		Th in ng	U in ng	Measurements at CRNL
Nov 22	Bkg	0.11	0.19	
Dec 8	Bkg	0.15	0.67	
CII 0.440	kg	1.7	1.9	HNO3, HCl
		2.7	1.4	HNO3, HF
		0.16	0.75	"

History of tube #11

Dec 3	Bkg	0.71	0.78	
		0.05	0.14	
Dec 22				
15.55 kg C1F		73	130	HNO3,HCl,HF
	Bgd	0.77	2.7	

Solution bkgs:

Dec 22	Bldg 115 DDD	0.016	0.53
	Bldg 107 DDD	0.063	0.61
	DD H2O	0.06	0.58
	isopropo	0.01	0.54

Boat

Dec 14	Bkg	0.41	0.68	HNO3, HF 1/2 hr
		0.40	2.3	"

Results on Cyro material:

Over the period of 1 1/2 years there have been a number of measurements done on some Cryo material which we have on hand. Four of these as itemized on the previous two pages but three more measurements were done in 1988. A complete list of the Th in these eight measurements is as follows in pg/g:

	Th		U	
	CRNL	NRC	CRNL	NRC
Jul 26/88	11	15		
AUG 10/88	6	10		
Sept 21/88	6			
Nov 16/89	62		20	
Nov 30/89	36		27	
Dec 7/89	7		6	
Dec 8/89	10		7.5	
Dec 22	4.7		8.3	

Preliminary Conclusions:

1) Within a factor of two the ICPMS and TIMS results on a common rinse agree.

2) At CRNL there continues to be a U background level of about 0.5 ng.

3) The results suggest that rinsing with just HNO3 is not very successful and that at least several times a lot of Th may have been left behind. Extra effort with HF and heat lamps appears to be more successful then just HNO3. It would appear that the first rinse may remove about 50% of Th and over 90% of the U. The recovery efficiency has not yet been quantified and may have vary from time to time.

4) The thicker acrylic from both Rohm and Polycast seems to have much higher levels of Th and U then the 1" Polycast. The Rohm material seems to be better than the Polycast. Additional measurements are required to verify these conclusions however.

5) A significant variation was found in Cyro material. The results from five pieces of the same sheet varied from 5 to 62 ppt of Th and from 5 to 20 ppt of U.

Significant uncertainties remain in this analysis method. The fact that we have to rinse out a large tube expecting to find less than ng quantities of Th and U may be a continuing problem. We are unable to state at the present time that this is a reliable method of measuring the Th in the acrylic by mass spectromotry.

We now believe that rinsing the tube first with aqua regia and then with HNO3 and HF may be successful but it will require several more weeks of work to verify this belief.

Should we be looking into the Re filament TIMS method as done by Kelly at NBS?

E. D. EARLE

PROCEDURE FOR THE RADIOCHEMICAL NEUTRON ACTIVATION ANALYSIS OF LUCITE FOR URANIUM AND THORIUM

Summary

The radiochemical neutron activation analysis (RNAA) of polymethylymethacrylate is carried out by first irradiating the sample with slow neutrons to a total dose of about 3×10^{17} neutrons/cm^2 to form the activation products ^{239}Np and ^{233}Pa. This is followed by dissolution of the sample in a mixture of Perchloric and Nitric acids (Liquid Fire Reaction) and then precipitation of the activation products as fluorides with HF, using lanthanum as a co-precipitant. The precipitate is converted to the sulfate, and passed through an anion exchange column in HCl solution to retain the ^{239}Np and ^{233}Pa, which are later eluted from the column.

Irradiation Procedure

A 30-35 g sample of lucite is sealed into a cylindrical polyethylene holder. The holder has a cavity 3.8 cm in diameter with a depth of 12.7 cm. The base of the cavity is sealed with a 3.8 cm polyethylene plug by heat welding. Above the cavity the holder continues about 3 cm as a solid cylinder with a 2.54 cm diameter polyethylene rod inserted into an axial blind hole and held with an aluminum pin. Prior to assembly a second blind hole is drilled in the solid top of the sample cylinder parallel to but off axis and clear of the support rod. Two duplicate standards are placed in this hole. These standards are small quartz tubes each loaded with about 1 microgram of Uranium and one microgram of Thorium from the evaporation of a few micro liters of a standard solution. These tubes have been flame-sealed and cleaned externally with a nitric acid wash before insertion. The cavity containing the standards (about 0.6 cm diameter) is then sealed with a heat-welded polyethylene plug.

Fig. 1 is a construction diagram of this container. The support rod and sample cylinder are lowered into a water-filled irradiation facility of the NRU reactor and rotated to average the flux direction for 6 hours. The entire irradiation structure is thus made of polyethylene except for the aluminum pin holding the support rod to the sample cylinder. After the 6 hour irradiation the assembly is retrieved and the cylinder containing the sample and standards separated from the support rod by extracting the aluminum pin. The sample cylinder is then returned to the laboratory for opening and the support discarded.

Sample Recovery and Dissolution

The irradiation container is washed and dried to remove any external activity from the irradiation facility and cut open just below the sealed end. The holder is then emptied into a beaker for weighing. At this point the sample has shattered into pieces of a variety of sizes from centimeter-sized chunks to coarse powder. However there is very little (< 1%) loss in weight from the effect of the radiation on the methylmethacrylate. Any gas that has formed from radiation decomposition remains trapped in the plastic, at the temperature of the irradiation facility (≈ 85°C).

The weighed material is emptied into a 3 liter Bethge reaction apparatus (see Fig. 2) containing the oxidation solution.This solution is composed as follows:

Perchloric Acid (70%)....	210 mL
Nitric Acid (conc.)......	90 mL
Potassium Dichromate.....	15 mg
Vanadium Pentoxide.......	10 mg

The vanadium is present as an oxidation catalyst while the dichromate serves as a color indicator of the oxidation completeness.

With the Bethge apparatus set to return the refluxate to the pot, heating of the mixture is begun at about 2°C per minute, continuing until the temperature of the solution reaches 140-145°C at which point the temperature increase stops. Gas is evolved from the sample from about 110°C onward. This is not an oxidation process but represents radiolytic gas formed during irradiation; it does not occur with test samples of unirradiated material.

The temperature is held at the 145°C point for about 15 minutes after it is confirmed visually that all traces of solid have disappeared and solution is complete. While the methacrylate has been dissolved at this point it is not greatly oxidized and would precipitate out if the solution were cooled and diluted. The color of the solution has changed from the original orange color of the dichromate to a deep green of the reduced chromium ion.

The upper stopcock on the apparatus is now closed so that the refluxate cannot return to the pot (see Fig 2.). This permits the temperature to continue rising as the low boiling components distill away. At about 170-180°C the reaction mixture begins to foam strongly,accompanied by an increase in the rate of temperature rise from the exothermic oxidation reaction. The foam is originally a slightly brownish color from radiation degradation products but turns colorless prior to the end of the reaction. Heating is continued after the foam subsides until at about 200°C the green chromium is abruptly converted to a red chromate solution indicating total oxidation of all organics. The full temperature cycle is shown if Fig. 3.

After cooling, the solution is decanted into a polytetrafluorethylene beaker and the flask washed with water which is added to the same beaker. Sufficient water is added (about 150 ml are lost during the oxidation period) to bring the volume back to about 280 ml. At this time 100 mg of $La_2(NO_3)_3$ is added and the solution divided among 6 polythene centrifuge tubes.

Five ml of 48% Hydrofluoric Acid are added to each tube and the tubes centrifuged to separate the LaF_3 precipitate. The precipitate is collected and the precipitation repeated with fresh lanthanum carrier. All the fluoride precipitates are combined and washed with water. At this stage the recovery of ^{233}Pa and ^{239}Np is about 80±10 %.

Ion Exchange Separation

The fluoride precipitation provides some chemical separation of the activation products from the more soluble active contaminants such as ^{24}Na, and also serves to concentrate the activation products. However numerous other impurities, including active lanthanides, are also carried down. These other activities interfere in the subsequent γ-spectrometry and limit the sensitivity of the method. To remove the impurities and leave essentially only ^{233}Pa and ^{239}Np we employ a conventional ion exchange technique for the actinides.

The fluoride precipitate is first metathetized to the sulfate form by treatment with 0.25 ml of concentrated Sulfuric Acid in a 10 ml 'Teflon' beaker. The sample is heated to drive off hydrofluoric acid and convert the precipitate to sulfates. The beaker can be heated sufficiently to fume off excess sulfuric acid (≅300°C) but will not stand the temperatures required to break down the sulfate salts to oxides. The converted salt is dissolved in 3 l of HCl with the concentration adjusted to leave a final HCl concentration of 9.5 mol/L.

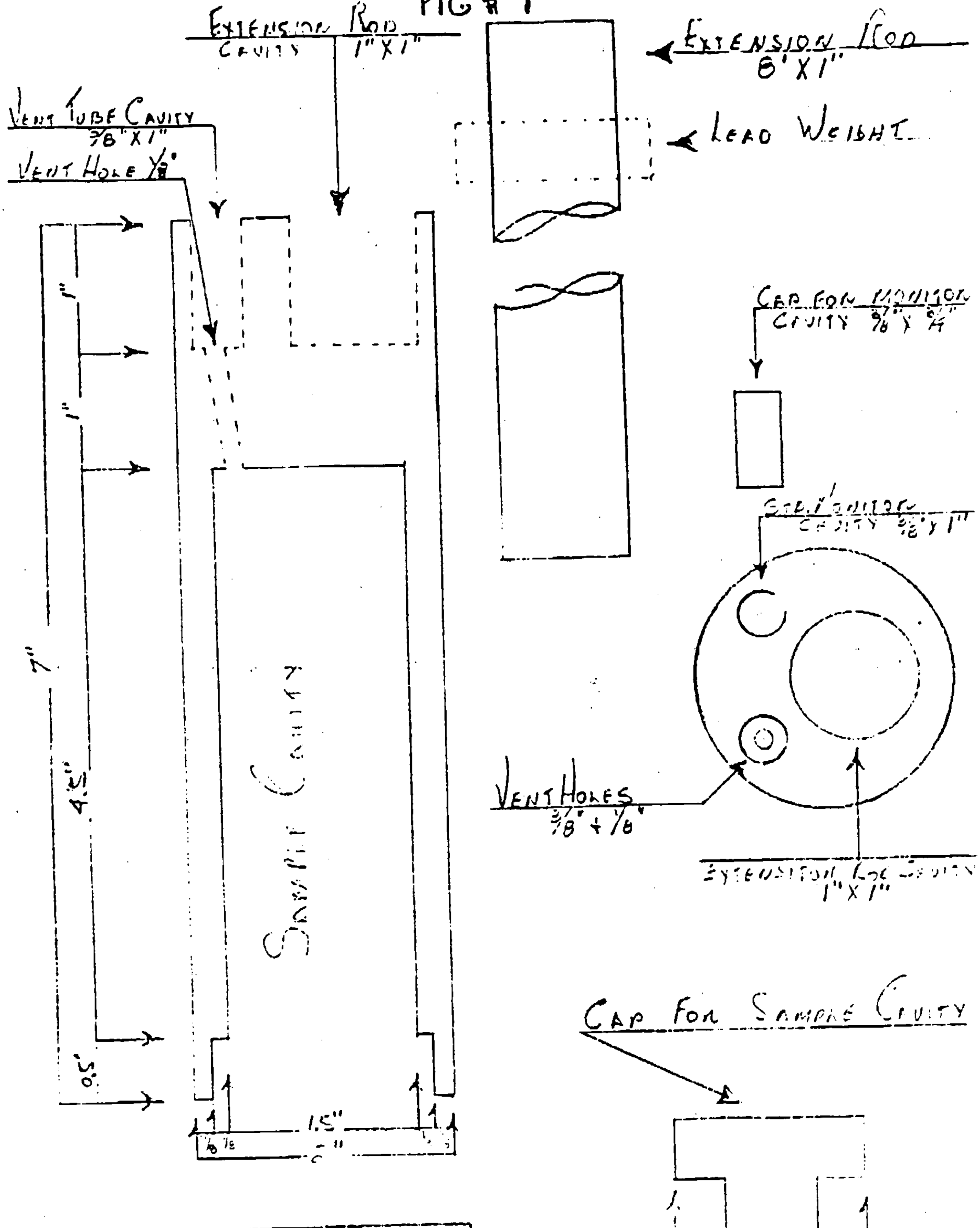
An anion exchange column of approximately 1 ml volume and 15 cm length was previously prepared from AGMP1 200-400 mesh resin, and converted to the chloride form with 9.5 M hydrochloric acid. The HCl salt solution is added to the column and the various cations eluted with 9.5 M HCl, while the protoactinium and neptunium remain on the column as anionic chloride complexes.

Neptunium and Protoactinium are then eluted from the column in dilute HCl/HF solution (≅10% HCl-1% HF). The eluant is collected in a 20 ml glass bottle and evaporated to dryness prior to γ-spectrometric measurements.

Procedure for Standards

The Uranium and Thorium standards are removed, still sealed in their quartz tubes, to another laboratory and added (in separate runs) to a slightly smaller Bethge apparatus. A piece of unirradiated lucite is added to the perchloric mixture and the standard is then processed exactly as the irradiated sample.

FIG # 1



Note
Material Polyethylene
W.J.F. + H.A.H. Oct 16-89

FIG. #2
BETHGE REACTION APPARATUS

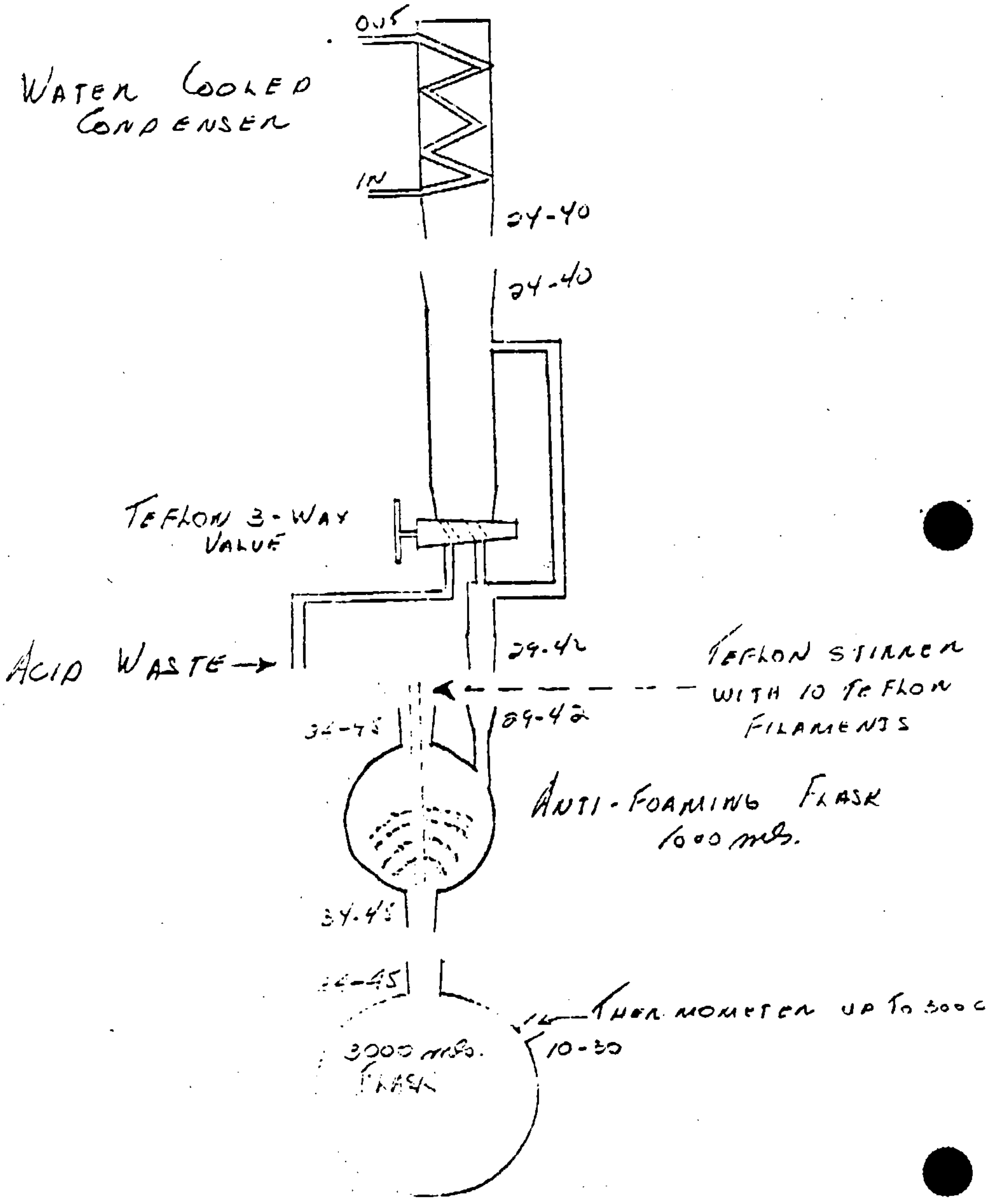
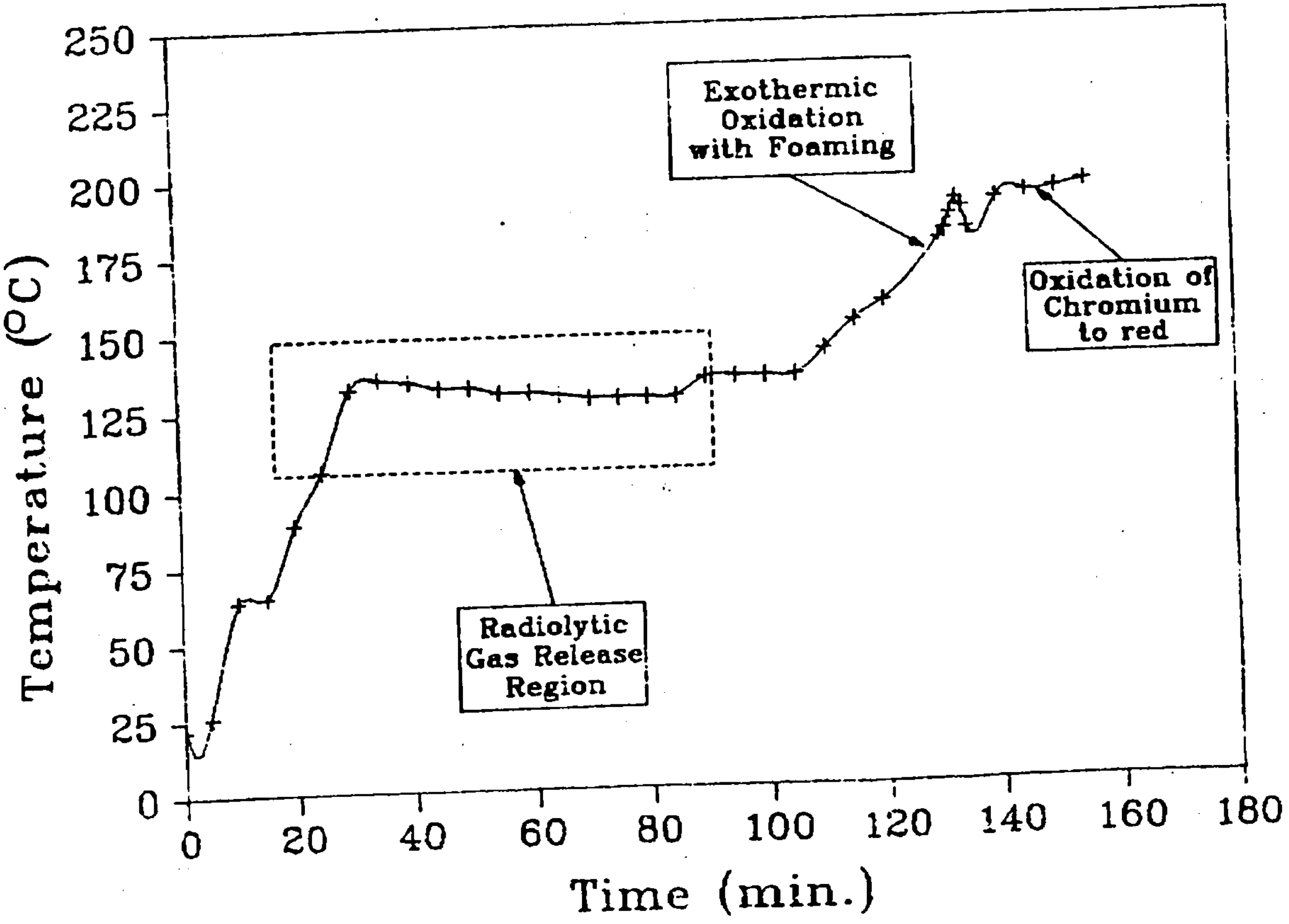
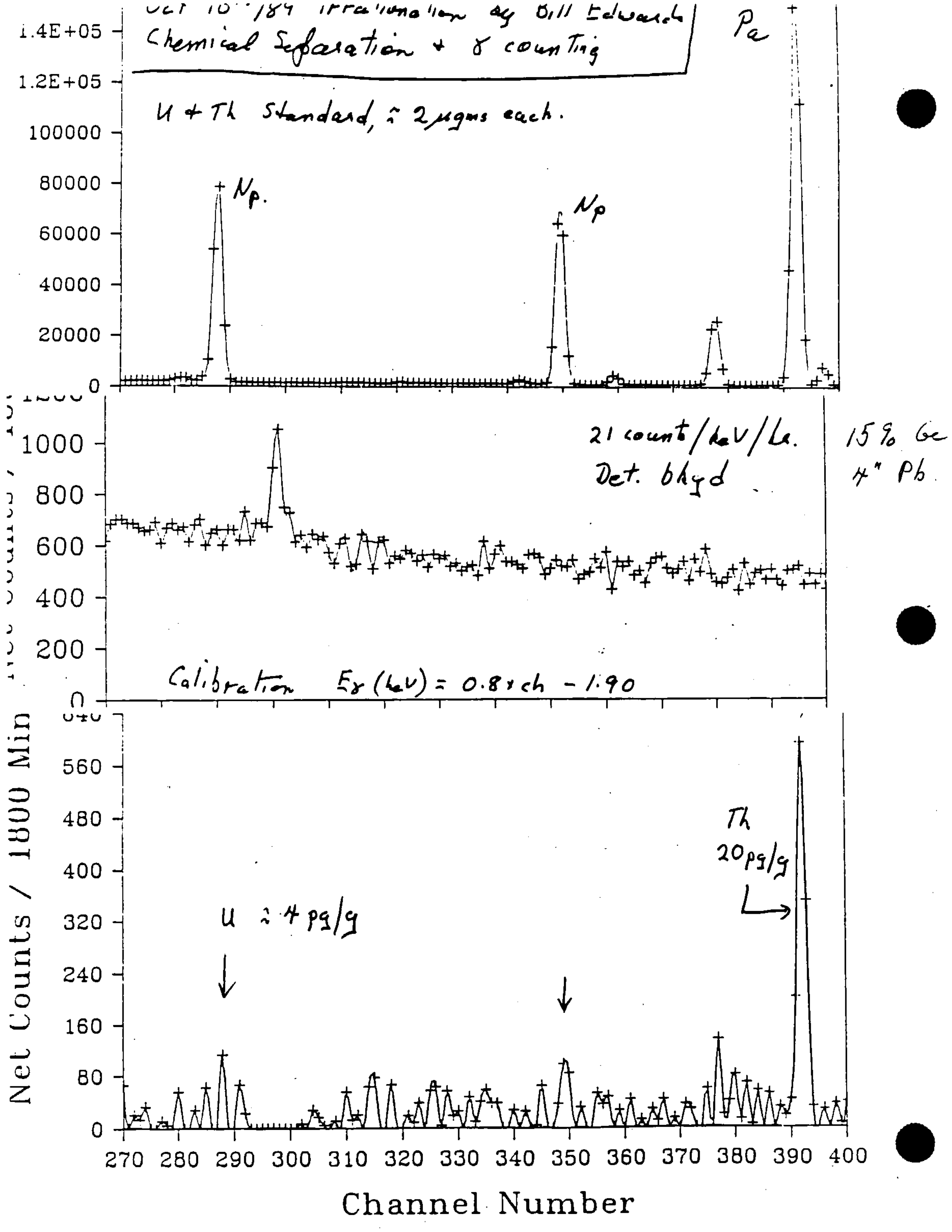


Fig. 3

OXIDATION OF LUCITE IN PERCHLORIC ACID



TEMPLCTE.DRW



Oct 10 '84 irradiation by Bill Edwards
Chemical Separation + γ counting

U + Th Standard, ≈ 2 μgms each.

21 count/keV/hr.
Det. bkgd

15% Ge
4" Pb

Calibration Eγ (keV) = 0.8 × ch − 1.90

U ≈ 4 pg/g

Th
20 pg/g

Units
pg/g

SUMMARY OF LUCITE ANALYSES IN 1989

SAMPLE	COMMMENTS	Ur-238	Th-232
CYRO	Mechanically cut. Slight water leak observed	21	5
CYRO	Better quality mechanical cut. No leakage seen.	32	13
CYRO	Laser Cut Material	15	16
	Ion Exchange Secondary Separation for following		
P2	One of 6 pieces supplied by R. Deal (One piece irradiated and shipped to Berkley and 3 irradiated and lost from water leaks)(One returned to R. Deal, unirradiated)	8	48
P2	Last of above pieces	4	20
???	Irradiated Oct 29/89. No I.D.	19	34
CYRO	Irradiated Dec 3/89	76 (60)*	149 (143)*
Polycast	Dec 10/89 irradiation. Sent to Queen's University for direct counting with a Compton suppression spectrometer before dissolving.		

* Calculated directly on the raw spectrum before chemical processing

Two samples were γ-counted with Compton suppression at Queens by Ted Clifford.

The Dec 3/89 sample gave U < 105 ppt, Th - 182±7 ppt

The Dec 10/89 ~ before chemical separation gave U 80±11 ppt Th - 174±7 ppt

These are not consistent with [illegible] ppt.

LUCSUMRY

E. D. EARLE

Measurement of U and Th in Acrylic via Neutron Activation Analysis

E. B. Norman, B. Sur, and K. T. Lesko
Lawrence Berkeley Laboratory

On October 17, 1989, Davis Earle had a 34.9-gram sample of acrylic irradiated with a flux of about 2×10^{13} neutrons/cm^2 /sec for a period of six hours in a reactor at Chalk River. Two separate U/Th standards, containing 1 microgram each of U and Th, were irradiated at the same time and in the same flux as the acrylic sample. Following the irradiation, the samples were shipped to Lawrence Berkeley Laboratory for gamma-ray analysis.

The samples arrived in Berkeley on the afternoon of Friday, October 20. The acrylic sample was gamma-ray singles counted using a germanium detector. The total counting rate observed from the sample was fairly low and almost all of the observed activity was from ^{24}Na ($t_{1/2}$ = 15 hours). The acrylic sample was removed from the plastic bag in which it was shipped to us and placed into two plastic bottles for counting. The U/Th standards were placed in similar plastic bottles in order to count all the samples in as nearly the same geometry as possible. The samples were allowed to cool over the weekend and then were gamma-ray singles counted using a 110-cm^3 high-purity germanium detector shielded with 5 cm of lead. Data was accumulated in 16384 channels at 0.1 keV/channel and was recorded on an IBM PC/AT. The full gamma ray spectrum observed in 2.45 days of counting the acrylic sample is shown in Figure 1. Figures 2 and 3 show expanded regions of this spectrum around the expected positions of the strongest gamma rays from the decays of ^{239}Np (produced by neutron capture on ^{238}U followed by the beta decay of ^{239}U) and ^{233}Pa (produced by neutron capture on ^{232}Th followed by the beta decay of ^{233}Th). Figures 4 and 5 show the same expanded regions of the spectra observed from counting the U/Th standards. Clear peaks are observed in the acrylic spectra at 106 and 228 keV attributable to the decay of ^{239}Np. Similarly, a clear peak is observed at 312 keV attributable to the decay of ^{233}Pa. The sample was counted several times over the course of about two weeks. During this time, the 106- and 228-keV peaks were observed to decay away at a rate consistent with the known 2.35 day half life of ^{239}Np. To address the question of whether the peak we observe at 312 keV could be the double escape peak of the 1332-keV line from ^{60}Co, we placed a ^{60}Co source on the front face of our detector and counted for about 15 minutes. A clear peak did show up at 310 keV with an intensity of 3.3×10^{-3} as compared to that observed in the 1332-keV photopeak. Using this ratio and the number of 1332-keV gammas observed from the acrylic sample, we conclude that less than two percent of the gamma rays in the 312-keV peak could be due to ^{60}Co decay.

The net numbers of 106- and 228-keV gamma rays extracted from the spectrum shown in Figures 1-3 are 483 ± 108 and 244 ± 87, respectively. The net number of 312-keV gamma rays observed in the same spectrum is 203 ± 89. Using the observed counting rates of these same gamma rays from the U/Th standards and the measured masses of U and Th in these standards, we then obtain the following results for the U and Th contents of the acrylic sample:

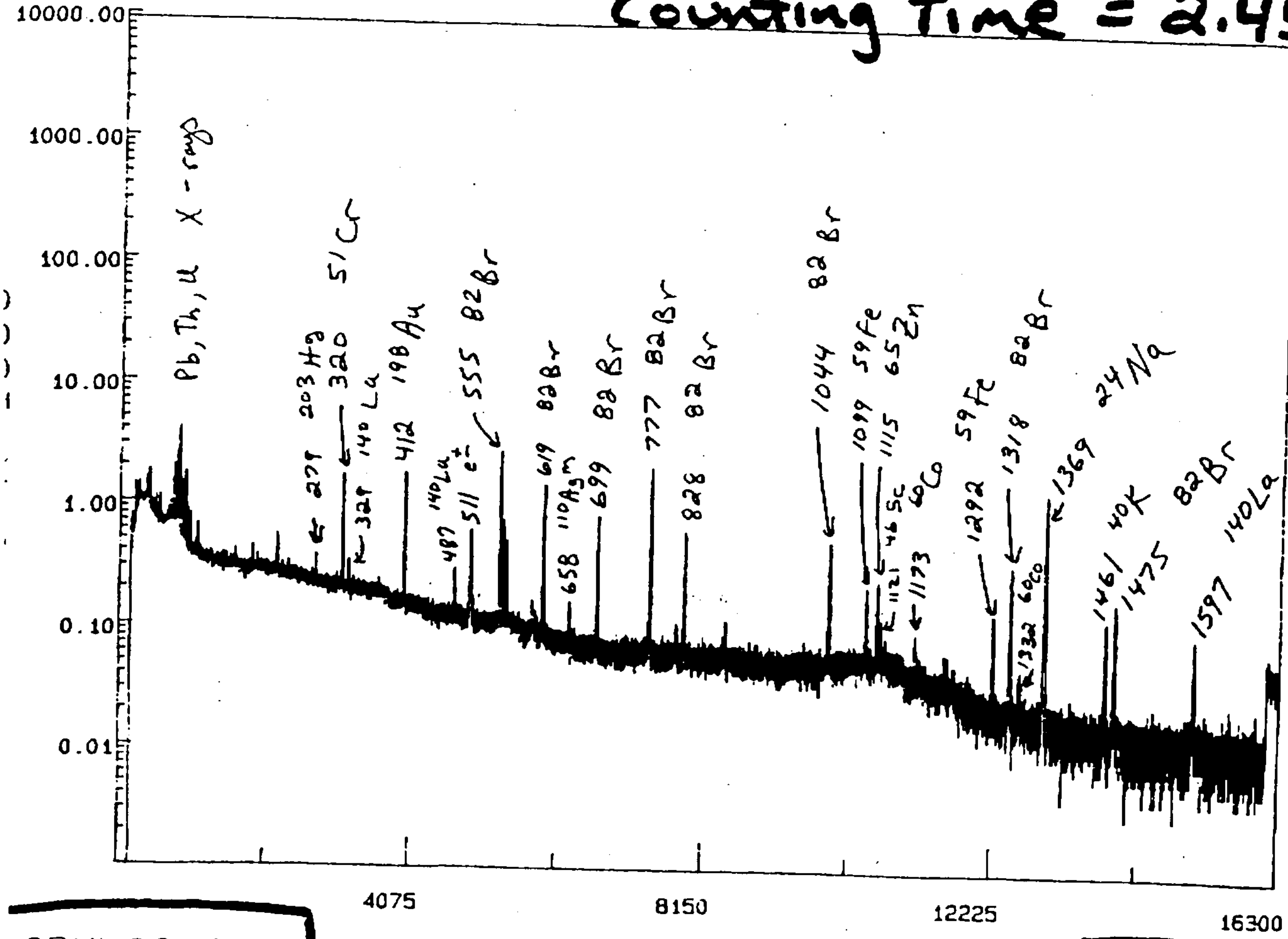
$$[U] = (12.5 \pm 0.5) \times 10^{-12} \text{ grams/ gram}$$
$$[Th] = (2.8 \pm 0.9) \times 10^{-12} \text{ grams/ gram.}$$

These numbers appear to us to be encouraging. The relative ease in being able to perform these measurements is no doubt due in large part to the cleanliness exercised in preparing the sample and in preventing the sample from being exposed to the reactor water during the activation. Note that all of the measurements reported here are simply gamma-ray singles counting using modest passive shielding. We also did some measurements with the acrylic sample and germanium detector placed inside a nearly 4π NaI anti-coincidence shield. This method reduces the Compton-background on which the ^{239}Np and ^{233}Pa gamma rays sit by a factor of 5 - 10, and thus should provide sensitivity to subtantially lower levels of U and Th than those observed here.

6 hours with $\Phi_n = 2 \times 10^{13}/cm^2\text{-}sec$

TYPE = -1 MCA # 1 SEGMENT # 1
REALTIME = [redacted] LIVETIME = [redacted]
DATA COLLECTED AT 17:19:00 ON 23-OCT-89

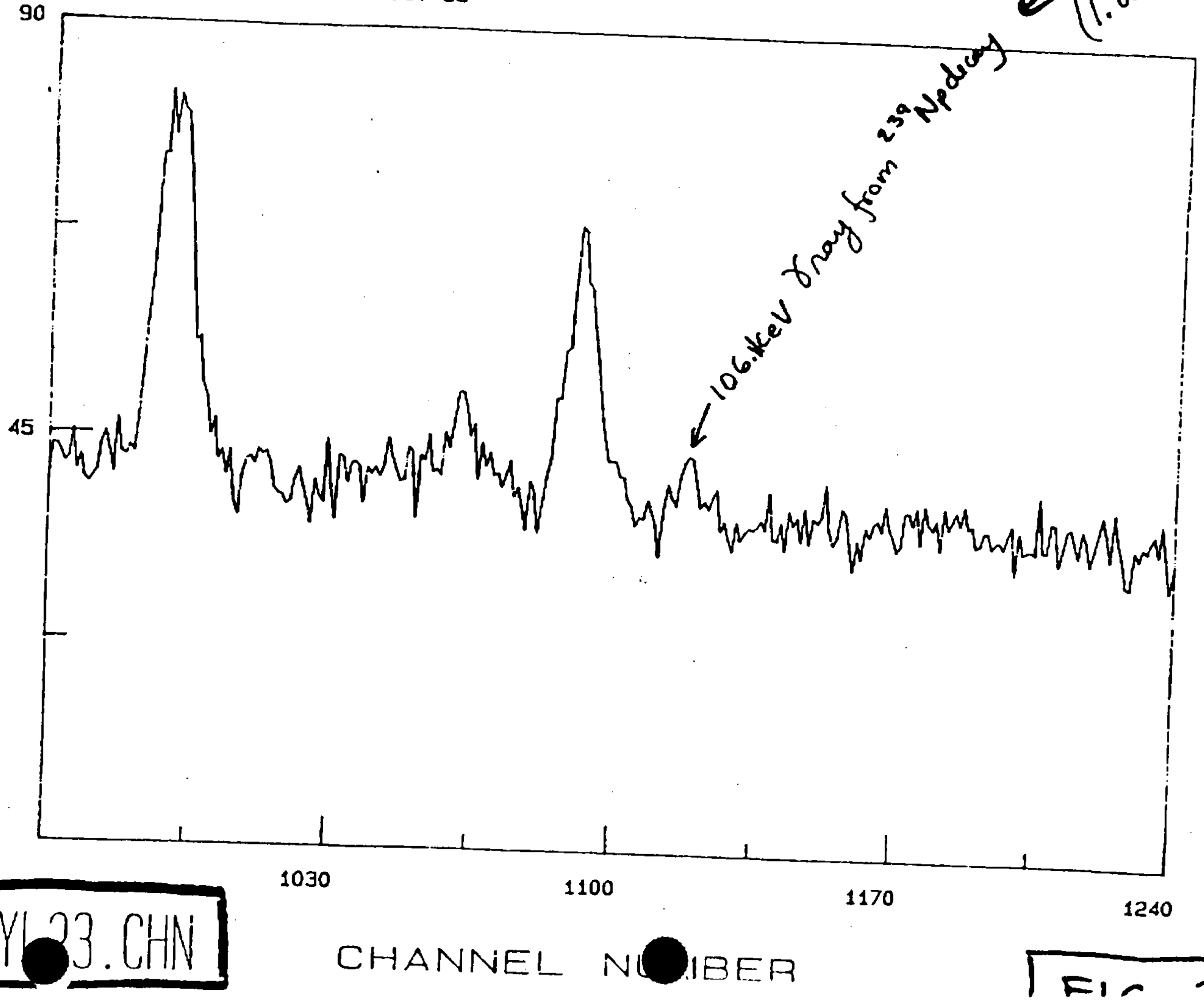
Counting Time = 2.45 days



CRYL23.CHN

FIG. 1

TYPE = -1 MCA # 1 SEGMENT # 1
REALTIME = [redacted] LIVETIME = [redacted]
DATA COLLECTED AT 17: 19: 00 ON 23-OCT-89



106.keV γ ray from ^{239}Np decay

^{238}U $(1.25 \pm 0.28) \times 10^{-11}$ g/g

CRYL23.CHN

FI

MCA # 1 SEGMENT # 1
REALTIME = [redacted] LIVETIME = [redacted]
DATA COLLECTED AT 17:19:00 ON 23-OCT-89

grams of acrylic
Irradiated for 6 hours with
$\Phi_n = 2 \times 10^{13}/cm^2\text{-}sec$

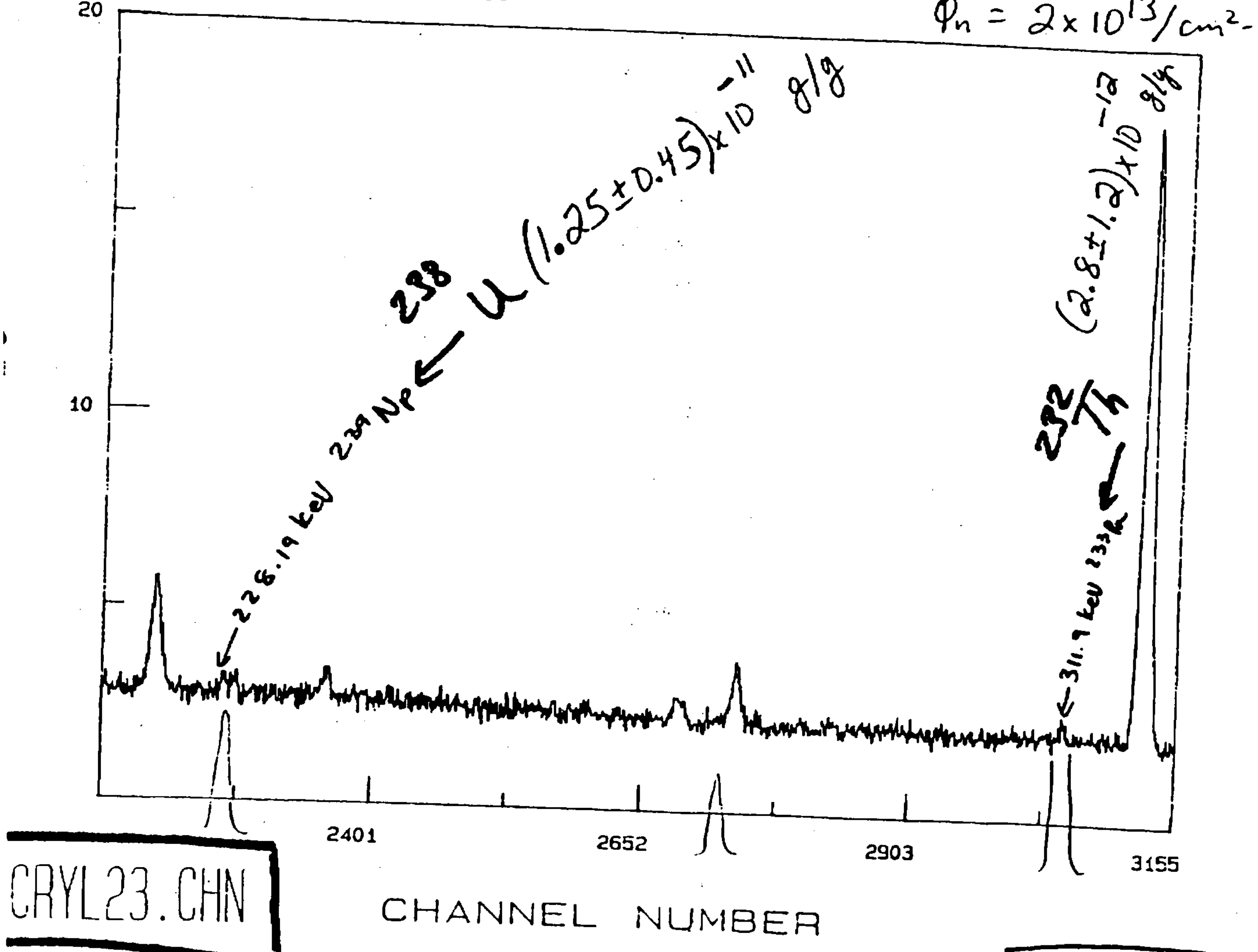


FIG. 3

with ϕ_n ~ 2x10^{13}/cm^2-sec

```
TYPE -   -1 MCA #  1 SEGMENT #   1
REALTIME =     1006.24 SECONDS, LIVETIME =     1000.00 SECONDS
DATA COLLECTED AT 10:19:00 ON 26-OCT-89
```

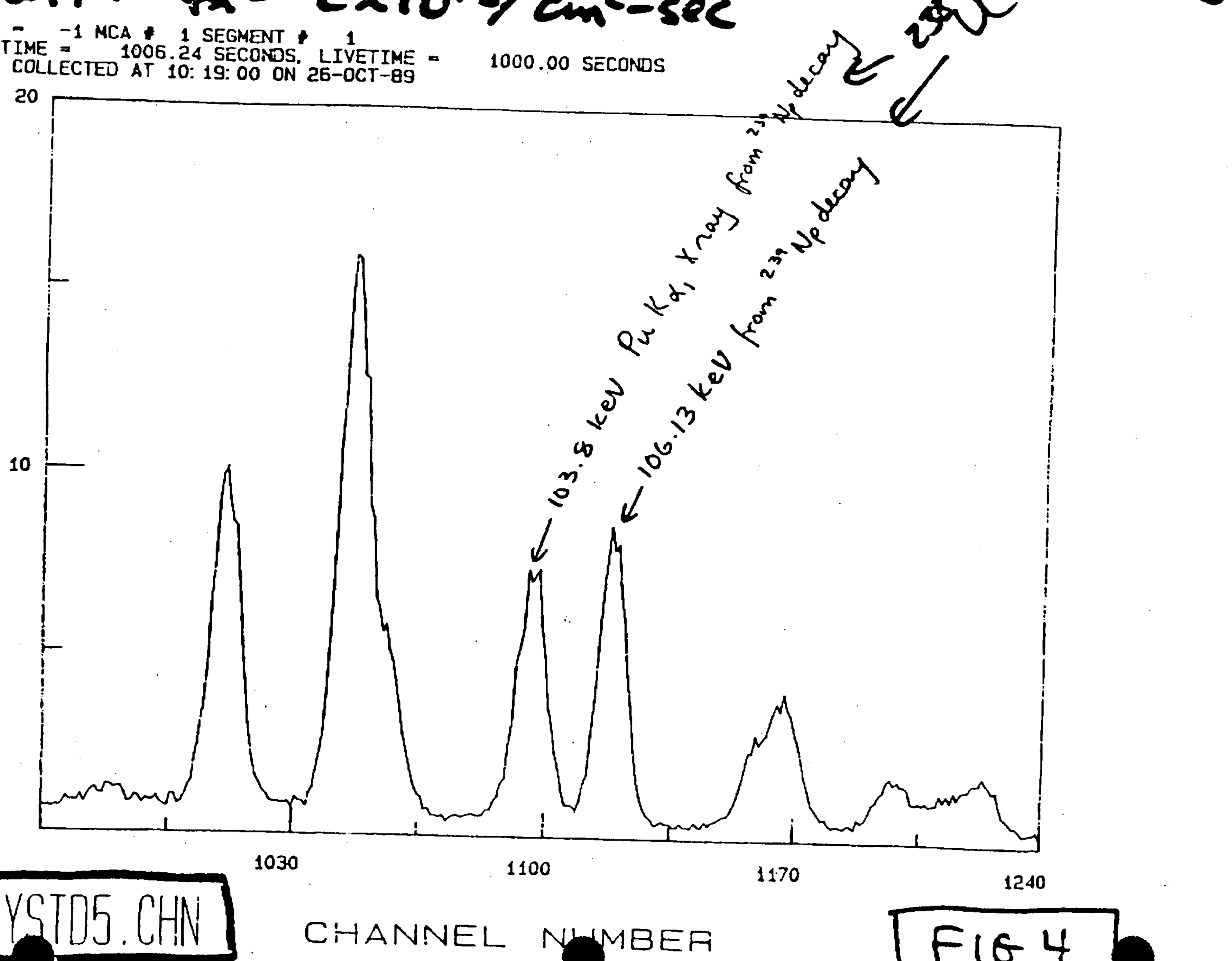


FIG.4

```
TYPE =    -1 MCA #  1 SEGMENT #    1
REALTIME =     1006.24 SECONDS. LIVETIME =     1000.00 SECONDS
DATA COLLECTED AT 10:19:00 ON 26-OCT-89
```

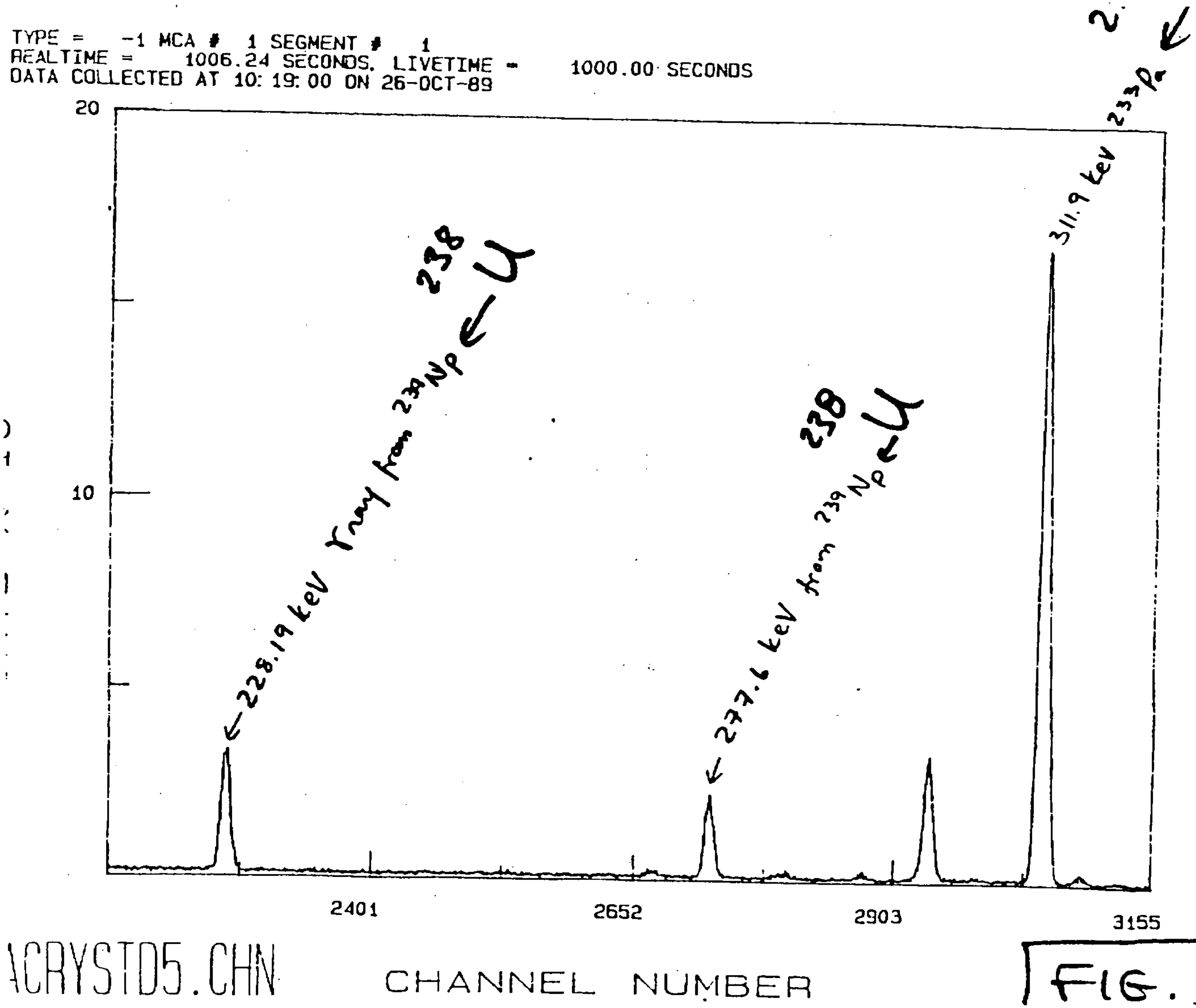
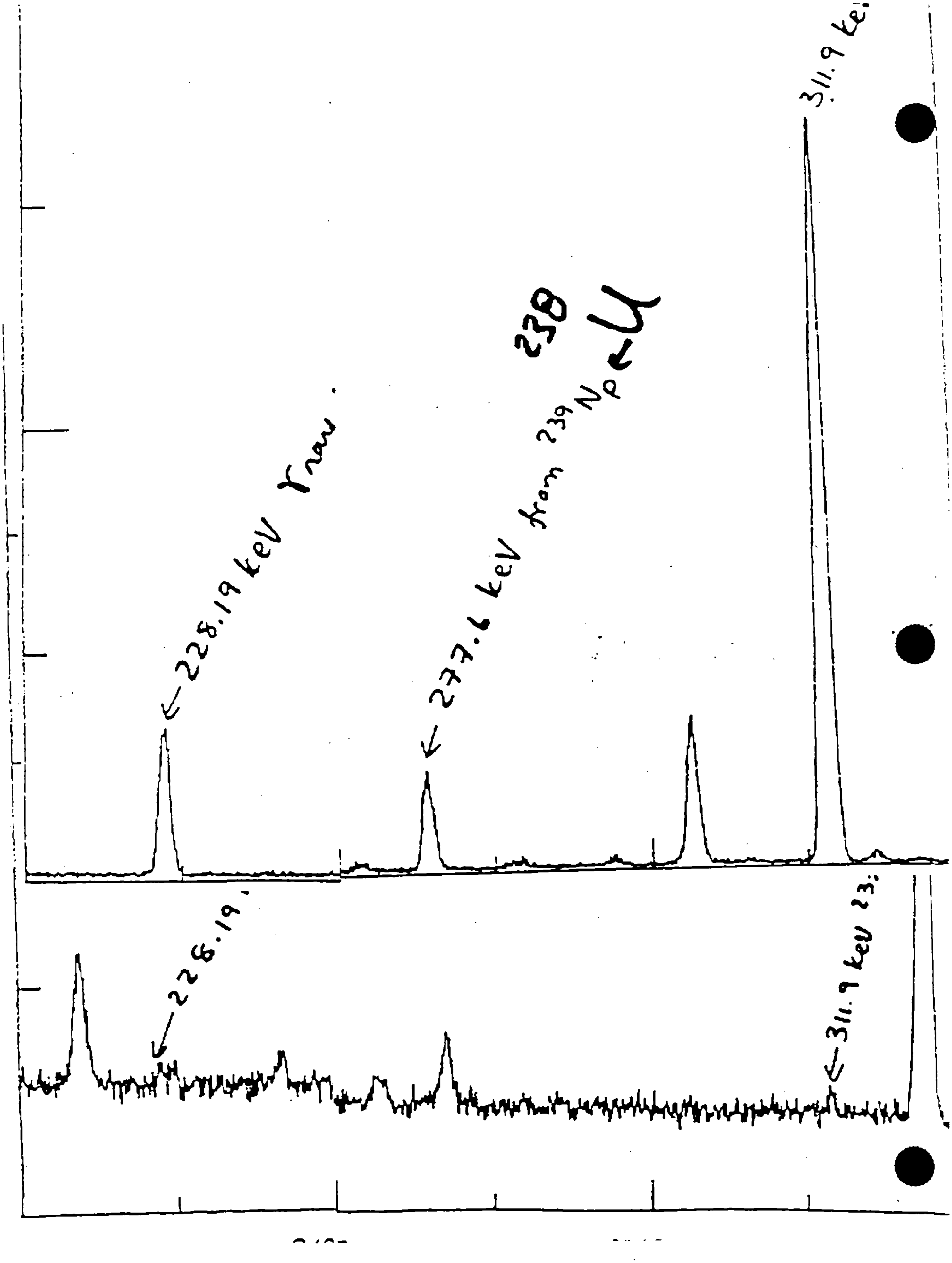


FIG. 5



311.9 ke
228.19 keV from
277.6 keV from 239Np ← 238U
228.19
311.9 keV 23

Summary of Neutron Activation Results in 1989

Material	Th ppt	U ppt	Comments
Cyro	5	21	large error H2O leak
Cyro	13	32	large error
Cyro	16	15	large error laser cut

at this point Edwards introduced chemical separation after irradiation.

Material	Th ppt	U ppt	Comments
Poly	48	8	after chem. sep.
Poly	20	4	" " "
Poly	3	12	Counted at LBL
no ID	34	19	after chem. sep.
Cyro	143	60	before chem. sep.
	149	76	after chem. sep.
	182	<105	at Queen's aft. sep.
Poly	174	80	at Queen's bef. sep. after chem. sep.

Ge counter backgrounds at 300 keV

	counts/hour/keV
CRNL 15% with 4" Pb	21
LBL 25% with 2" Pb	7.4
with Comp. Sup.	? (about 2)
Queen's 18% in Comp. Sh.	7.8
with shield on	1.8
Guelph 48% with 6" special Pb, 2" Hg and cosmic veto.	1

Considerations:

1) Is compton suppression sufficient so that chemical separation is not required?

2) Do we need a well type Ge detector for chemically separated samples?

0w66t4i

Alpha Spectrometric Measurements of Th and U in Acrylic

In order to check on possible disequilibrium in the thorium and uranium families (i.e. the presence of short-lived daughters, unsupported by the parent isotopes ^{232}Th and ^{238}U), it is necessary to measure the last long-lived member of each chain, prior to the gamma emitter of interest. Alpha spectrometry is the only technique capable of detecting ^{228}Th and ^{226}Ra at the very low activities anticipated.

By adopting current ultra trace element analytical techniques (Class 100 acids, use of Teflon ware throughout, etc.) it has been possible to lower backgrounds in the relevant regions of interest of the spectra to 2 to 4 counts in a 20,000 minute period. Even so, in order to detect these radioisotopes at < 1pg.g^{-1} Th or U equivalent, it has been necessary to ash 10 kg of acrylic and to count samples and blanks in excess of 10 days each to obtain statistically significant results.

Initially an electroplated source was prepared directly from the dissolved ash without prior chemical separation, however the sources ~~so~~ obtained were thick and the spectra difficult to analyze. A step for thorium purification was first introduced, and subsequent to Jan.'88 a minimum number of analytical procedures have been added to purify and electroplate U, Th and Ra separately. Average recoveries by this method, measured on spiked 200g samples of acrylic, are as follows:

5/ng Ca

U	40±2 %
Th	70± %
Ra	30± %

The lower than usual recoveries for U and Ra have been shown to result from our inability to volatilize trace residues prior to plating, when working in Teflon. In order to check for possible losses during the volatilization, chemical separation has been omitted for some of these spiked samples. Close to quantitative recoveries were observed utilizing methods such as counting dried sources, thermal ionization mass spectrometry, and in the case of Ra, liquid scintillation counting. It must be recognized, however, that such checks, while encouraging, do not rule out the possibility of increased losses at this stage if the radioisotopes have been bound into the organic matrix at the time of polymerization.

Accumulated Analytical Data

Date	Weight	U(pg.g^{-1})	Th(pg.g^{-1})
Mar'87	10.96 10kg [illegible]	^{230}Th 1.0+0.22	^{228}Th 5.1+1.0
Dec'87	11.6kg	^{230}Th 0.8+0.20	^{232}Th 4.3+0.80 ^{228}Th 3.7+0.60
April'88	10.6kg polycast	^{235}U 0.3+0.20 ^{230}Th 0.8+0.30	^{232}Th 2.0+1.0 adj to 1.7 ^{228}Th 2.2+1.0 " to 0.66
July'88	9.8kg	^{235}U 2.1=0.80 ^{230}Th 0.8+0.20 ^{226}Ra 1.2+0.80	^{232}Th 2.6+0.60 ^{228}Th 3.9+0.80
July'89	8.72 10kg Rohm?	^{235}U 0.3+0.13 ^{230}Th 0.08+0.07 ^{226}Ra below LLD	^{232}Th 0.41+0.29 ^{228}Th 0.60+0.35
Dec'89	15.5kg Cyro		

Preliminary measurements in monomer, crosslinker and polymer, prior to application of separation techniques, have provided useful information on the sources of U and Th contamination, and identified other radio and stable elements present in the samples (e.g. ^{210}Po, ^{239,240}Pu, Fe and Si)