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***Mound Laboratory Chemistry
and Physics Progress Report:
April-June 1970***

AEC Research and Development Report

Monsanto

MOUND LABORATORY

operated by

MONSANTO RESEARCH CORPORATION

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for the

U. S. ATOMIC ENERGY COMMISSION

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MONSANTO RESEARCH CORPORATION

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Miamisburg, Ohio 45342

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Foreword

The Mound Laboratory Chemistry and Physics Progress Report, issued quarterly, is intended to be a means of reporting items of current technical interest in research and development programs. Since this is an informal progress report, the results and data presented are preliminary and subject to change.

These reports are not intended to constitute publication in any sense of the word. Final results either will be submitted for publication in regular professional journals or will be published in the form of MLM topical reports. Questions concerning the material compiled here should be directed to W. T. Cave, Director, Nuclear Operations.

Previous reports in this series are:

MLM-1726	MLM-1603
MLM-1612	MLM-1531
MLM-1609	MLM-1526
MLM-1606	MLM-1520

Summary

SEPARATION CHEMISTRY

Uranium-234 Recovery Previous failures of the final anion exchange process to remove plutonium-238 contamination from uranium-234 products were attributed to the presence of hexavalent plutonium. A more vigorous reduction of the feed solution and another final anion exchange run produced an acceptable uranium-234 product. The uranium was converted to oxide and shipped. (Page 6)

Thorium-229 Recovery One uranium-thorium separation run was made during this period; final purification of the product thorium-229 from this and the previous run is being done before shipment. A new batch of 62 g of uranium-233 has been dissolved for the next uranium-thorium separation run. (Page 7)

INORGANIC CHEMISTRY

Addition Compound of Uranium Tetrachloride with Tetramethylurea A new addition compound of uranium tetrachloride and tetramethylurea was prepared and characterized. The formula of the compound is $\text{UCl}_4 \cdot 2(\text{C}_5\text{H}_{12}\text{N}_2\text{O})$. (Page 8)

Distribution of Plutonium Valence States from the Disproportionation of Pentavalent Plutonium The distribution of plutonium valence states which derive from the disproportionation of pentavalent plutonium is presented graphically. Hexavalent and tetravalent plutonium are the principal species produced in strong acid. (Page 10)

Further Studies on New Rare Earth Hydrides A compound of yttrium was prepared by dissolving yttrium metal in a warm aqueous solution of monochloroacetic acid. The compound appears to be similar to rare earth hydrides prepared under similar conditions. (Page 11)

Studies on Metal Oxide Microparticles Thorium dioxide microparticles were prepared by a freeze-dry process. The dried particles were ignited at various temperatures, and properties such as crush strength, density and particle size were measured. (Page 12)

ANALYTICAL CHEMISTRY

Determination of Traces of Mercury in Oxygen-Free High-Conductivity Copper The concentration of mercury in oxygen-free high-conductivity copper is being determined by neutron activation. The 279-keV photopeak of mercury-203 is used in the analysis. Parts-per-billion quantities of mercury in copper can be determined by this non-destructive method. (Page 14)

Determination of Precision of Plutonium-238 Data on Various Recovery Solutions The analytical precision of plutonium-238 data was compared with sampling precision for tanks of recovery solutions. The investigation revealed that the major problem is sampling. (Page 14)

In-Line Gamma Scanning of High-Level Plutonium-238 Waste Actual values for plutonium-238 concentrations determined by in-line gamma scanning were compared with the known values. The maximum concentration limits were extended to 29 g with no significant decrease in accuracy. (Page 16)

Preparation of Plutonium-Containing Solutions for Impurity Analysis by Carrier Distillation A new procedure is being developed for the preparation of plutonium samples for analysis of the impurities. The procedure involves conversion of the nitrated species to their respective oxides. It was found to give results comparable to those obtained with ion exchange for all impurities except silicon, boron and nickel. The best comparative results were obtained when a Vycor crucible was used for the ignition. (Page 16)

INSTRUMENTATION AND METHODS

Efficiency Determination of a 30-cm³ Ge(Li) Detector The intrinsic detector efficiency was determined for a coaxial Ge(Li) detector to be used for quantitative measurements of gamma rays. Standard sources from the National Bureau of Standards were used for calibration of the device in various energy regions. The absolute detector efficiency for a given source-to-detector housing distance at a given energy can be found using the intrinsic efficiency curve and a calculation function. The data points on the calibration curve were accurate to $\pm 5\%$. (Page 18)

Separation Chemistry

URANIUM-234 RECOVERY

The rare isotope uranium-234 is being recovered from two sources at Mound Laboratory: waste anion exchange raffinate solution, and aged plutonium-238 "cows" or heat sources. After the initial separation and concentration steps, material from both sources is treated by two cycles of anion exchange. An intermediate anion exchange step consists of loading uranium and plutonium onto an anion column from an aluminum nitrate-salted medium, washing the uranium off with 7M HNO_3 , and eluting the plutonium with dilute HNO_3 . The final anion exchange step also uses an aluminum nitrate-salted medium to load uranium (and the small amount of plutonium remaining) on the column, but then uses 9M HCl and a reducing agent (iodide ion) to reduce and wash off the remaining plutonium before the uranium is eluted with dilute HCl .

In the previous report,¹ problems in the final anion exchange processing of batch U-5 were described. The plutonium-238 contamination had been reduced from 980 ppm (by weight) to 580 ppm in the first purification attempt, and to 300 ppm in a second attempt. The poor separation of the plutonium was attributed to the presence of plutonium in the hexavalent oxidation state, rather than quadrivalent, and failure of the reducing agents to reduce the hexavalent plutonium on the column. Therefore, in preparing the uranium-234 material for another purification, the dilute HCl solution from previous processing was evaporated to dryness and taken up in 1M HNO_3 . Addition of a small volume of 30% H_2O_2 produced a white precipitate, which decomposed slowly with gentle heating. A second treatment with peroxide and the addition of hydroxylamine as a holding reductant preceded the usual salting with aluminum nitrate. This time the uranium-234 product assayed only 3 ppm (by weight) of plutonium-238 contamination. This experience tends to confirm the belief that the valence adjustment step used earlier (peroxide and hydroxylamine at room temperature in aluminum nitrate-salted solution) is not sufficient for the reduction of gross amounts of plutonium(VI).

The purified U-5 material was precipitated with hydroxide, and calcined to constant weight at 800°C. The oxide, weighing 2.10 g, was shipped to the Heavy Elements Research Pool at ORNL. A final mass analysis is not yet available to calculate the weight of uranium-234, but the mass purity should be approximately 95%. (P. E. Figgins)

THORIUM-229 RECOVERY

A second batch of 60 g of uranium-233 was extracted with DSBPP-DEB (di-sec-butyl phenyl phosphonate in diethyl benzene), using the semi-automatic extraction apparatus described in the previous report.² The organic phase recirculation rate was maintained approximately equal to the strip rate. No thorium appeared in the strip and the uranium-233 was completely extracted, stripped and concentrated in 31 hr of operation.

The first attempt to extract the thorium-229 product from the original aqueous solution into a TOPO-DEB (tri-n-octylphosphine oxide in diethyl benzene) organic phase for purification resulted in the recovery of only 1 mg of product. Several attempts were made to improve this, including addition of aluminum nitrate, repeated fumings with concentrated HNO_3 and, finally, boiling the aqueous phase with NaOH and filtering. The last treatment was used on the assumption that some of the thorium may have been precipitated as the insoluble phosphate by reacting with degradation products of the phosphorus-containing extractants. A total recovery, for this run, of 1.9 mg of thorium-229 was obtained. Together with the 1.5 mg of thorium-229 from the previous run, a total of approximately 3.4 mg is being prepared for shipment.

After concentration of both of the above products, an isopropyl ether extraction, to remove iron, will be attempted before the material is shipped.

The "milked" uranium-233 from the current run has been calcined to U_3O_8 and returned to storage.

A batch of UF_4 containing 62 g of uranium-233 has been dissolved for the next uranium-thorium separation which will be started as soon as the current thorium-229 product has been shipped. (M. R. Hertz)

Inorganic Chemistry

ADDITION COMPOUND OF URANIUM TETRACHLORIDE WITH TETRAMETHYLUREA

The study of coordination compounds of actinide elements is of interest for several reasons, one of them being the preparation of new primary standard compounds for actinide elements. A compound of uranium tetrachloride and tetramethylurea has been prepared by dissolving uranium tetrachloride in hot tetramethylurea which had been boiled for a few minutes to expel moisture. When the solution of uranium tetrachloride in hot tetramethylurea was cooled, large green crystals separated. The crystals were separated from the mother liquor and washed with boiled and cooled tetramethylurea. They were then dried in vacuum. Analysis of the crystals indicated: C, 20.25%; H, 3.88%; N, 9.15%; U, 39.34%; Cl, 23.62%. The compound $\text{UCl}_4 \cdot 2\text{TMU}$ (TMU = tetramethylurea, $\text{C}_5\text{H}_{12}\text{N}_2\text{O}$) requires 19.6% C; 4.0% H; 9.15% N; 38.9% U; 23.2% Cl. In a melting point apparatus, visible decomposition of the green crystals occurred at 267°C . Differential thermal analysis showed the presence of only one thermal transition near 270°C . Thermogravimetric analysis showed a weight loss upon heating in air beginning near 190°C . The weight loss amounted to 55.3% of the original compound weight. Predicted weight loss for the reaction

is 55.9%. The x-ray powder diffraction pattern of the compound is shown in Table 1.

The density of the compound was about 1.9 g/cc. At room temperature one crystal of the material exhibited a magnetic moment of 2.2 Bohr magnetons (where 1 Bohr magneton is equal to 9.273×10^{-21} erg/gauss), while a powdered sample showed a magnetic moment of 1.9 Bohr magnetons, indicating magnetic anisotropy or, more likely, atmospheric oxidation of the powdered sample. Using the spin only formula, the magnetic moment of the U^{4+} ion with two unpaired electrons is predicted to be 2.4 Bohr magnetons. The infrared absorption of the compound in a Nujol mull is shown in Table 2.

Analogous compounds of thorium and plutonium were prepared at this time, but their x-ray powder patterns differed significantly from the powder pattern of the uranium compound. In the case of thorium tetrachloride, more than one tetramethylurea adduct may be possible, but none of these compounds were investigated further.

Table 1

X-RAY POWDER DIFFRACTION PATTERN OF
TETRAMETHYLUREA ADDUCT OF URANIUM TETRACHLORIDE

<u>Interplanar Spacing</u> <u>(Å)</u>	<u>Observed Intensity</u> <u>(Arbitrary Units)^a</u>
7.75	M
6.85	S
5.15	M
4.68	W
4.37	W
3.95	M
3.40	W
3.24	M
2.95	W
2.78	W
2.70	W
2.58	M
2.34	W
2.27	W
2.23	W

^aS = Strong; M = Medium; W = Weak.

Table 2

INFRARED ABSORPTION OF $\text{UCl}_4 \cdot 2(\text{C}_5\text{H}_{12}\text{N}_2\text{O})$ IN A NUJOL MULL

<u>Absorption Position</u> <u>(cm⁻¹)</u>	<u>Relative Intensity</u> <u>(Arbitrary Units)^a</u>
361	W
419	W
565	W
585	M
730	W
750	S
763	S
900	M
918	W
921	W
1050	W
1062	S
1140	W
1168	M
1238	M
1315	M
1405	W
1414	W
1532	M
1590	M
1618	W

^aS = Strong; M = Medium; W = Weak.

(G. L. Silver)

DISTRIBUTION OF PLUTONIUM VALENCE STATES FROM THE DISPROPORTIONATION OF PENTAVALENT PLUTONIUM

A new concept in the presentation of plutonium disproportionation stoichiometries has been proposed in recent times.⁸ The merits of this new concept are at present largely a matter of speculation. However, the concept allows an uncomplicated calculation of the equilibrium concentrations of all plutonium species which derive from tetravalent or pentavalent plutonium ions. A plot of the distribution of plutonium valence states which may derive from the disproportionation of pentavalent plutonium is shown in Figure 1. Complicating phenomena such as hydrolysis have been neglected in Figure 1. This figure is of interest because a diagram of this type does not appear to have been prepared before. The diagram shows that hexavalent and tetravalent plutonium are the principal species produced by the disproportionation of pentavalent plutonium in strong acid, and that pentavalent plutonium shows a decreasing tendency to disproportionate in solutions of decreasing equilibrium acidity. Both of these observations are in agreement with experiment.

(G. L. Silver)

FIGURE 1 - Equilibrium distribution of species produced by disproportionation of pentavalent plutonium (hydrolysis neglected).

FURTHER STUDIES ON NEW RARE EARTH HYDRIDES

In recent times, new hydride compounds of the lanthanide elements have been discovered.⁴ At the present time, results of research on these compounds indicate that they contain oxide anions, hydride anions and a few organic anions distributed throughout a fluorite lattice structure in an apparently random manner. The apparently random nature of the distribution allows a wide range of elemental composition for these compounds since hydride, oxide and appropriately sized organic anions may substitute for one another. The degree of substitution seems to depend upon the conditions of preparation since materials of differing compositions but similar x-ray powder patterns may be obtained by dissolving rare earth metals in a dilute organic acid under differing conditions of temperature and acid concentration.

A compound of yttrium was prepared by dissolving yttrium in warm, 5M monochloroacetic acid. The gray-green precipitate which resulted was washed by decantation three times with deionized water, and then three times with acetone. The material was dried in vacuum over phosphorus pentoxide, then analyzed by Galbraith Laboratories for the following elements: analysis indicated 66.60% Y, 2.19% Cl, 2.08% H and 2.82% C. It is difficult to formulate the yttrium compound from these data since the amounts of chlorine and carbon found by analysis do not agree with the formula of monochloroacetic acid. The x-ray powder diffraction pattern of the compound, shown in Table 3, is similar to the powder patterns of analogous rare earth compounds.

Table 3

X-RAY POWDER DIFFRACTION PATTERN OF COMPOUND PREPARED
BY DISSOLVING YTTRIUM METAL IN MONOCHLOROACETIC ACID

<u>d-Spacing</u>	<u>Observed Intensity</u>	<u>d-Spacing</u>	<u>Observed Intensity</u>
5.4153	3	1.7400	2
3.8575	2	1.6614	9
3.5132	1	1.6369	4
3.1704	10	1.5929	5
3.0538	6	1.5516	3
2.8631	1	1.5104	4
2.7601	5	1.4675	3
2.6645	4	1.4382	1
2.4695	3	1.4028	4
2.3603	8	1.3792	3
2.2261	5	1.3514	1
2.1926	1	1.3233	2
2.1405	1	1.3122	4
1.9525	5	1.2966	3
1.9291	9	1.2786	3
1.8695	5	1.2656	3
1.8138	1	1.2542	3

(G. L. Silver)

STUDIES ON METAL OXIDE MICROPARTICLES

Radioelement microparticles are of interest for medical and nuclear power generating purposes. Many methods are available for preparing metal oxide microparticles, including the sol-gel process, an ion exchange process,⁵ and utilization of a plasma torch.

Schnettler, Monforte and Rhodes⁶ have proposed a method whereby microparticles of a metal compound are prepared by drying droplets of a solution of the metal frozen in hexane at dry ice temperature. Several batches of ThO_2 microparticles were prepared from ThO_2 sols of varying thorium content. The dried particles were ignited, and properties such as crush strength and density were measured. Three sols were used in these experiments: Sol #1 contained 220 mg/ml thorium, Sol #2 contained 629 mg/ml, Sol #3 contained 1194 mg/ml. Table 4 summarizes some properties of microparticles prepared from these sols. Only Sol #2 and Sol #3 produced unbroken microspheres. Sol #2 seemed to give the better microspheres. (G. L. Silver)

Table 4

PROPERTIES OF MICROPARTICLES PREPARED BY A FREEZE-DRY PROCESS

<u>Particles from Sol #</u>	<u>Ignition Temperature (°C)</u>	<u>Crush Strength (g)</u>		<u>Size (μm)</u>		<u>Tap Density (g/cc)</u>
		<u>Average</u>	<u>Range</u>	<u>Average</u>	<u>Range</u>	
1	Not ignited	0	0	486	297-676	0.17
2	Not ignited	0	0	716	524-890	0.43
2	Not ignited	0	0	714	437-917	0.47
3	Not ignited	0	0	894	666-1110	0.81
1	800	0	0	453	283-600	0.19
2	800	3.6	1.2- 9.2	559	366-748	0.51
2	800	4.3	1.2- 32.2	713	441-993	0.54
3	800	12.5	1.2- 38.2	713	455-921	0.97
1	1000	0	0	416	293-560	0.26
2	1000	8.6	1.2- 21.9	472	324-593	0.72
2	1000	6.2	1.2- 17.3	451	255-689	0.79
3	1000	30.1	9.2- 58.7	680	468-876	1.44
1	1200	0	0	370	228-559	0.28
2	1200	25.2	4.8- 57.6	501	317-776	0.82
2	1200	24.1	3.5- 86.3	527	262-800	0.85
3	1200	59.0	5.8-161.0	603	379-821	1.62

Analytical Chemistry

DETERMINATION OF TRACES OF MERCURY IN OXYGEN-FREE HIGH-CONDUCTIVITY COPPER

Neutron activation analysis is being used to determine the concentration of mercury in oxygen-free high-conductivity (OFHC) copper. Four standards containing 0-100 ng of mercury and six 2-g copper samples containing no added mercury were sealed in quartz vials and irradiated for 100 hr at a thermal neutron flux of about 5×10^{12} neutrons/cm²/sec. Two weeks were allowed for the decay of the copper isotopes before the standards and the samples were counted with a Ge(Li) detector. No chemical separations were performed. The standards were counted in the quartz vials because of the possibility of a small amount of mercury being adsorbed on the quartz. The copper samples were removed from the quartz vials for counting.

A 400-channel analyzer was calibrated so that there was approximately 0.5 keV per channel. The number of counts in the 279-keV photopeak of mercury-203 was determined with a five-point spectrum smoothing computer program and corrected for decay. Linear regression analysis and a statistical evaluation of the standards were performed with a computer program, S-473. Analysis of these samples and standards indicated that the OFHC copper contained 38 ± 12 ppb mercury.

This non-destructive method for the determination of parts-per-billion quantities of mercury in OFHC copper appears to give satisfactory results. (R. K. Gillette)

DETERMINATION OF PRECISION OF PLUTONIUM-238 DATA ON VARIOUS RECOVERY SOLUTIONS

Investigations are complete on the remaining eight tanks of recovery solutions to compare analytical precision of the plutonium-238 data with the sampling precision. The data are shown in Table 5. The result of these investigations is that the major problem is sampling.

A statistical evaluation is being performed to enable estimates of precision on future samples from the same tanks. (M. J. Garrod, R. D. Terrell, C. R. Brown and W. R. Amos)

Table 5

COMPARISON OF ANALYTICAL RESULTS OF VARIOUS PLUTONIUM-238 SAMPLES

Sample Description	Sample Number	Analytical Dilution #1 (mg ²³⁸ Pu/ml)	Maximum Deviation among Samples (%)	Analytical Dilution #2 (mg ²³⁸ Pu/ml)	Maximum Deviation among Samples (%)	Deviation between Dilutions (%)
MS Lot	1	7.94		8.24		3.8
	2	8.26	4.0	7.97	4.4	3.5
	3	8.01		7.89		1.5
Ultrasonic Cleaner	1	0.072		0.063		12.5
	2	0.412		0.408		1.0
	3	0.055	749.0	0.058		5.5
	4	0.073		0.071		2.7
	5	0.063		0.082		30.2
Evaporator Concentrate	1	1.55		1.32		14.9
	2	1.47		1.48		0.7
	3	1.41	9.9	1.50	20.4	6.4
	4	1.49		1.59		6.7
	5	1.48		1.59		7.4
Bubbler	1	0.00113		0.00115		1.8
	2	0.00110		0.00104		5.5
	3	0.00120	234.0	0.00150	260.0	25.0
	4	0.00107		0.00110		2.8
	5	0.02500		0.02700		8.0
Filter	1	0.028		0.025		10.7
	2	0.022		0.022		0
	3	0.032	45.4	0.022	36.4	31.2
	4	0.028		0.025		10.7
	5	0.022		0.030		36.4
Vacuum B	1	2.082		2.355		13.1
	2	2.330		2.170		6.9
	3	2.300	16.2	2.340	8.5	1.7
	4	2.310		2.320		0.4
	5	2.4200		2.340		3.3
Raffinate	1	0.00086		0.00082		4.7
	2	0.00080		0.00080		0
	3	0.00090	12.5	0.00090	25.0	0
	4	0.00080		0.00100		25.0
	5	0.00090		0.00100		11.1
Filtrate	1	0.675		0.650		3.7
	2	0.600		0.537		11.4
	3	0.643	26.2	0.607	22.2	5.6
	4	0.535		0.532		0.6
	5	0.590		0.566		2.4

IN-LINE GAMMA SCANNING OF HIGH-LEVEL PLUTONIUM-238 WASTE

Work was completed on the modification of the in-line gamma scan system with an automatic shielding device to provide more accurate plutonium-238 data in the higher concentration ranges. Data obtained with known quantities of plutonium-238 are shown in Table 6.

Table 6

COMPARISON OF PLUTONIUM-238 RESULTS BY IN-LINE GAMMA SCANNING

<u>Sample Number</u>	<u>^{238}Pu Added (g)</u>	<u>^{238}Pu Found (g)</u>	<u>Difference (%)</u>
1	4.6	5.3	15
2	10.1	10.1	0
3	15.3	15.5	1.3
4	20.0	21.4	7.0
5	24.3	24.1	0.8
6	29.3	29.4	0.3

Comparison of actual values with known values shows a maximum difference of 15%. The maximum concentration limits are therefore extended to 29 g without significant decrease of accuracy. (J. Y. Jarvis and K. M. Muldoon)

PREPARATION OF PLUTONIUM-CONTAINING SOLUTIONS FOR IMPURITY ANALYSIS BY CARRIER DISTILLATION

A new procedure for preparing samples for impurity analysis which involves conversion of the nitrated species to their respective oxides is being investigated. The present emission spectrographic impurity analysis of plutonium-containing solutions requires a somewhat involved separation step to remove the interfering plutonium. The separation is performed by passing the solution through nitrated anion exchange resin and collecting the impurity-containing effluent for analysis. The plutonium remains on the column as a nitrate complex.

Six different plutonium nitrate solutions have been prepared for analysis by the new procedure. An aliquot of the sample solution is evaporated to dryness and then ignited at 900°C for 1 hr. The oxide residue is then transferred to the emission spectroscopy laboratory for impurity analysis by carrier distillation. Five of these samples compare between the two procedures for all impurities except silicon, boron and nickel. These samples were ignited in porcelain crucibles. The sixth sample was ignited in a Vycor crucible and compared in both respects with the exception of boron, as shown in Table 7. It is apparent that silicon and

boron (and nickel to a smaller extent) contamination is occurring from the porcelain crucible. Switching to Vycor indicates the possibility of eliminating most contamination with the exception of boron. It has been noted that the sample can be removed from the Vycor with absolute ease where much difficulty is experienced in removing samples from the porcelain.

Table 7

COMPARISON OF IMPURITIES FOR TWO METHODS OF SAMPLE PREPARATION

Sample Number	Boron		Silicon		Nickel	
	IE ^a	CD ^b	IE	CD	IE	CD
1	-	0.005	-	0.018	-	0.005
2	<0.002	0.007	-	0.035	<0.001	0.005
3	<0.001	0.007	-	0.015	-	-
4	<0.001	0.005	-	0.01	<0.003	0.03
5	<0.001	0.005	-	0.02	0.005	0.02
6 (Vycor)	<0.005	0.02	-	<0.005	0.004	<0.005

^a Ion exchange

^b Carrier distillation

The new procedure eliminates handling and disposing of the potentially hazardous nitrated resin. It also eliminates possible contamination of the sample by excessive handling and decreases the amounts of equipment, reagents and time necessary for the preparation of samples. A colorimetric iron determination, previously required, is eliminated. Finally, it works to eliminate the excessive number of rerun analyses due to incomplete plutonium separation.

(D. C. Carter, W. R. Amos and M. J. Garrod)

Instrumentation and Methods

EFFICIENCY DETERMINATION OF A 30-cm³ Ge(Li) DETECTOR

In order to perform quantitative measurements on the intensities of the gamma rays emitted by heat source material, the intrinsic detector efficiency of a 30-cm³ coaxial Ge(Li) detector manufactured by Princeton Gamma-Tech, Inc., was measured.

Single spectra of calibrated gamma-ray sources were obtained with 700 V reverse bias across the detector diode. The electronic system consisted of a Tennelec TC-130 preamplifier, a Tennelec TC-203BLR linear amplifier, and a Nuclear Data 50/50 analyzer system with an 8192 analog-to-digital conversion gain. The resolution for this system was 4.2 keV at 1332 keV (FWHM).

The gain setting of the amplifier was adjusted to approximately 0.5 keV per channel to provide a peak width of 7-10 channels (FWHM) over the energy region 160-3500 keV. Since the peak width containing 99% of the peak area is approximately $2.4 \times (\text{FWHM})$,⁷ a peak area of 25 channels was used for the intensity measurements. The background contribution was assumed to be linear over the peak area. The average of the counts from five channels on each side of the peak was used for the average counts per channel for the background.

The following standard sources, obtained from and calibrated by the National Bureau of Standards, were used: 166 keV from ¹³⁹Ce, 662 keV from ¹³⁷Cs, 835 keV from ⁵⁴Mn, 511 keV (1.8 rays per 1274-keV gamma ray) and 1274 keV from ²²Na, and 1174 and 1332 keV from ⁶⁰Co.

Absolute efficiencies (ratio of detected gamma rays to the number of gamma rays emitted by the source) were obtained at five distances from the detector housing for each energy. By using different combinations of source-to-detector housing distances for a given gamma-ray energy, the source-to-detector distance was determined using inverse square law calculations. These calculations indicated that the detector was 3.14 cm behind the detector housing.

The shape of the efficiency curve was determined in the energy region from 80 to 400 keV using a ¹³³Ba source and from 800 to 3500 keV using a ⁵⁶Co source, based upon the relative intensities of the gamma rays emitted by these two sources as listed in Marion.⁸

The intrinsic efficiency curve determined in this way is shown in Figure 2. The absolute data points are those determined using the NBS-calibrated sources. The absolute detector efficiency for a given source-to-detector housing distance at a given energy can be found from the figure by multiplying the efficiency value taken from the graph at this energy by $(d + 3.14)^{-2}$, where d is the distance from source to housing in centimeters. The accuracy of all of the data points in Figure 2 is $\pm 5\%$. (F. X. Haas)

FIGURE 2 - Intrinsic efficiency curve for the 30-cm³ Ge(Li) detector.

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