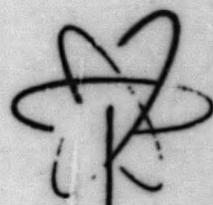


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GEAP-4361
AEC RESEARCH AND
DEVELOPMENT REPORT
SEPTEMBER 1, 1963



WASHER

ACCURATE NUCLEAR FUEL BURNUP ANALYSES

SEVENTH QUARTERLY PROGRESS REPORT JUNE 1963 - AUGUST 1963

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BY
B.F. RIDER
C.P. RUIZ
J.P. PETERSON, JR.
P.S. LUKE, JR.

U.S. ATOMIC ENERGY COMMISSION
CONTRACT AT(04-3)-189
PROJECT AGREEMENT 25

VALLECITOS ATOMIC LABORATORY

GENERAL  ELECTRIC

ATOMIC POWER EQUIPMENT DEPARTMENT
SAN JOSE, CALIFORNIA

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Printed in U.S.A.
Price 

Available from

Office of Technical Services
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1403-T10-2
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INTRODUCTION

Accurate measurements of fuel burnup are required to aid in reactor design for high burnup fuels and to aid in evaluation of nuclear fuel costs. Current radiochemical methods are limited in usefulness by limited availability of long-lived radioactive isotopes among the fission products, the accuracy with which these can be measured due to uncertainties in their physical constants, and the volatile nature of the few available isotopes. Mass spectrometric techniques are being explored as a solution to accurate burnup analysis by seeking to measure the quantity of the nonradioactive, refractory fission products produced from a variety of fuels. Use of the accurate isotope dilution technique on stable fission products eliminates the errors that limit the final accuracy of radiochemical measurements such as those contained in counting efficiencies, decay schemes, decay constants, and in-pile and out-of-pile decay corrections.

SUMMARY

The development of accurate methods for determination of nuclear fuel burnup began in December 1961. The report of the first year's progress, in cooperation with Phillips Petroleum Company, appeared in TID-17385, "Burnup Determination of Nuclear Fuel", January 1963. The first six quarterly reports appeared as GEAP-4053-1, GEAP-4053-2, GEAP-4082, GEAP-4137, GEAP-4201, and GEAP-4278. The work performed during the seventh quarter is summarized as follows:

1. Scope of Work

On August 26 and 27, 1963, the Burnup Task Force of the ASTM (E-10, Sub V) met at Vallecitos Atomic Laboratory, with representatives of 11 organizations present. They commented on the scope of the AEC-sponsored burnup program. The majority of task force members felt that increased emphasis should be put on fast plutonium reactor and thorium breeder reactor types of fuels. The most useful work would be in the measurement of fission yields and parasitic capture cross sections under fast reactor spectrum conditions in addition to the similar measurements under thermal neutron spectrum conditions already planned.

2. Variation of U-235 and Pu-239 Fission Yields With Neutron Energy

A total fission indicator for use in fast reactor fuel should have, among other characteristics, a nearly constant fission yield for all neutron energies and for all fissioning isotopes. A plot of the estimated Pu-239 and U-235 fast and thermal chain yields relative to the U-235 thermal chain yields indicates that such nuclides are to be found in the molybdenum-technecium region near mass 99 and in the neodymium region near mass 148. The only other region of near constancy (+ 10-30%) is in the region of the gaseous isotopes xenon-132 to -139, some of which decay to cesium, barium, and lanthanum.

3. Heavy Element Behavior on a Di(2-ethylhexyl) Phosphoric Acid Column

Fission product neodymium is separated on a Di(2-ethylhexyl) phosphoric acid (D2EHP) column for mass spectrometry. The behavior on the column of several other elements present in irradiated nuclear fuel and their possible influence on the separation of neodymium from irradiated fuel is reported.

4. The Behavior of Neodymium in High Heat Flux Fuels

Fission product neodymium behaved similarly to cerium, strontium, and zirconium in the UO_2 fuels observed. No migration of these elements has been observed in unmelted fuel. However, from a molten mass of UO_2 fuel, the uranium crystallizes from the cooler periphery inward and refractory fission products were observed to be concentrated among the last materials to solidify. For burnup analysis of melted fuel, a total dissolution of the fuel is the best way to assure correct fission product to uranium ratios.

5. Calculation of Nd-148 From Mass Spectrometric Ratios

When Nd-150 is used as an isotopic diluent for measurement of Nd-148 from fission, the 142 mass position is also scanned to check for natural neodymium contamination. Equations for calculation of Nd-148 and Nd-142 by use of 142/150 and 148/150 ratios have been reported in GEAP-4201. The ratio of mass 142/143 has been found to be more easily and accurately measured than the 142/150 ratio and to be less subject to mass discrimination effects. The equations for calculation of Nd-148 and Nd-142 from measured 142/143 and 148/150 ratios is presented.

PROGRAM PROGRESS

During the quarter, consideration was given to the scope of work which will best fill the needs for measurement of burnup in future reactor types; the variation of fission yields with neutron energy; and further work on stable fission product neodymium as a total fission indicator, its separation from irradiated fuels and its calculation from mass spectrometer ratios.

I. SCOPE OF WORK

On August 26 and 27, 1963, The American Society for Testing and Materials Committee E-10, Sub V, Burnup Task Force, met at Vallecitos Atomic Laboratory with a good attendance representing the Atomic Energy Commission, Atomics International, General Atomics, Pratt and Whitney, Argonne National Laboratory, Stanford Research Institute, Hanford Laboratories, Oak Ridge National Laboratory, NUMEC, Phillips Petroleum Company, and the General Electric Vallecitos Atomic Laboratory. Delegates commented on the scope of the AEC-sponsored burnup work. The meeting was classified to permit discussion of classified fuels.

The majority of task force members felt that increased emphasis should be given to above-thermal neutron spectra because of the trend toward fast breeder reactors operating on the U-238 to Pu-239 cycle and toward the Th-232 to U-233 cycle. One of the principal ways that the program can contribute to the accuracy of burnup measurements is in the measurement of fission yields and parasitic capture cross sections in thermal, epithermal, and fast neutron spectra. In addition to the main three fissionable isotopes, U-233, U-235, and Pu-239, the fission yields of Th-232, U-238, Pu-240, and Pu-241 should be measured in a fission flux because of their importance in fast breeder reactors. Parasitic capture cross sections for thermal and epithermal neutrons should be measured for any nuclide selected as a burnup monitor. These include the very long-lived and stable nuclides.

It was felt that measurements such as the heats of fission were outside the scope of this burnup task force, except to review their deficiencies and to make recommendations. Several other topics were discussed, including (a) use of a light element reference spike, (b) relation of α to some measurable spectrum-dependent parameter, (c) non-destructive burnup methods, (d) measurement of half-lives of radioactive fission products, such as Sr-90, (e) migration of fission products at high temperatures, and finally, (f) dissolution problems for fuels of varied composition.

The conclusions drawn on the last six topics are briefly summarized as follows:

(a) Light Element Spike

The use of a light element reference spike for normalizing the heavy element nuclides creates an additional chemical analysis and a uniformity problem for the manufacturer. The correction for a small loss of the heavy element reference (U-238 or Th-232) does not seem to be much over 1-5% in presently conceived designs, and correction may be preferable to introducing an additional reference.

(b) Variation of the Capture to Fission Ratio, α

The variation of α with both neutron energy and fissioning nuclide makes the relation of this value to a single parameter unlikely. Individual values of α for each nuclide would be the goal for such a correlation since it is not clear what an average α value would mean or how it could be used.

(c) Non-Destructive Methods

No generally applicable methods are known for actual fuels with long or varied irradiation history.

(d) Half Lives of Long Lived Fission Products

The half life of Sr-90 has the greatest uncertainty among useful nuclides. The redetermination of this value would not remove the other considerable problems to the use of Sr-90 as a total fission indicator. These include the problems of absolute beta counting and the appreciable variation in fission yield with neutron energy. For these reasons, the half-life measurement of Sr-90 may be of less immediate interest than other measurements.

(e) Migration of Fission Products

The best solution to eliminate the effect of migration of fission products in a fuel to be analyzed for burnup appears to be a total dissolution of the fuel prior to analysis.

(f) Dissolution Problems

Fuel processing flowsheets appear to require solution by each individual reactor project. The most useful burnup research that can be performed is the development of a sound, general method which can be adapted to individual situations by workers in the field.

The scope of work will continue to stress the use of stable fission products as fission indicators and the measurement of fission yields and parasitic absorption cross sections of the most promising fission product nuclides under thermal, epithermal, and fast neutron irradiation for the important fissioning nuclides. As was stated in the August 26-27th meeting, this obviously is beyond the scope of work as defined for the three year program, and an additional effort will be required. Rather than expand the third year's effort, the additional work will be proposed as a fourth year extension.

II. VARIATION OF U-235 AND Pu-239 FISSION YIELDS WITH NEUTRON ENERGY

It is well established that the yield of fission product nuclides in fission is a function of neutron energy as well as the fissioning nuclide. The most desirable total fission indicator is one whose fission yield is least variable for different neutron energies and different fissionable nuclides. Recently, a compilation of estimated total chain fission yields for several neutron-induced fission processes was published in USNRDL-TR-633.

It proved helpful to plot the estimated total chain yield for each mass for fission of U-235 and Pu-239 in thermal, as well as fission neutron spectra, relative to the U-235 thermal fission yield. These are shown in Figure 1, and, although it is recognized that the estimates are subject to some uncertainty, it is plainly seen that fission yields are extremely energy dependent in the valley region corresponding to masses 106 to 129, as well as in the region of the wings corresponding to masses less than 80 and greater than 153. There are only a few regions of reasonable constancy. In the light masses, this occurs in the molybdenum-technecium region near mass 99. The gaseous xenon nuclides from 132 to 139, some of which decay to cesium, barium, and lanthanum, show a 10 to 30% variation. In the heavy masses, the region of constancy occurs in the neodymium region near mass 148. It appears, therefore, that these are the regions most likely to contain good total fission indicators whose yield is independent of fissioning isotope and of neutron energy, at least for reactors on the uranium-plutonium fuel cycle. Work has been in progress on neodymium separation and mass spectrometry, and is reported in GEAP-4053-1, GEAP-4053-2, GEAP-4082, GEAP-4137, and GEAP-4201. The chemical separation scheme reported there involves reversed-phase chromatography, using a D2EHP column. The effect of several other elements in the fuel on this separation are reported below.

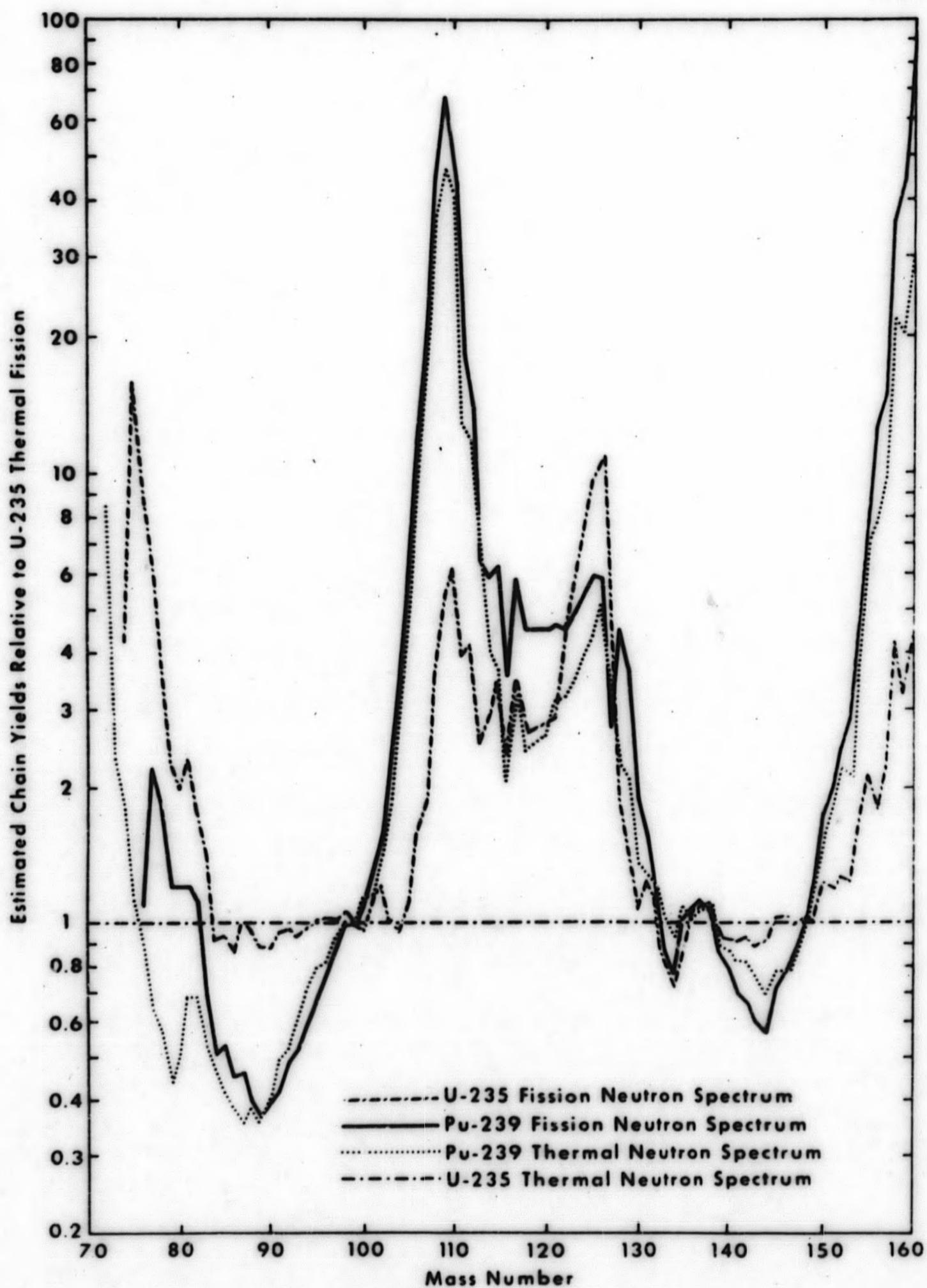


Figure 1. RELATIVE FISSION YIELDS FOR U-235 AND Pu-239

III. HEAVY ELEMENT BEHAVIOR ON A D2EHP COLUMN

The behavior of the rare earth elements on a D2EHP column has been fairly well studied, and this column has been incorporated into a chemical procedure for the isolation of neodymium.^[1] It was seen that the D2EHP column can separate the neodymium from group 1 (e.g., Cs-137) and group 2 (e.g., Ba) elements and also from adjacent rare earths. However, the behavior of the heavy elements which are present in fuel has not been studied.

Now the procedure which has been devised (previously reported in GEAP-4278) will separate the uranium and plutonium from the rare earths by the use of a Dowex-1 column which precedes the D2EHP column. However, this will not separate other heavy elements from the rare earth fraction. These elements include thorium, protactinium if fluoride ion is present, and the transplutonium elements. It is also of interest to study the uranium and plutonium behavior since it may be possible to simplify the neodymium separation scheme by eliminating the initial Dowex-1 column. Therefore, a study of the behavior of some of the heavy elements on D2EHP columns has been initiated.

The preparation of the D2EHP columns was identical to that described in Section II-B of GEAP-4278. The elements studied were Ra-226, Ac-227, Th-230, Pa-233, U-233, Pu-239, Am-241, and Cm-244 in various HCl concentrations at 87°C.

In order to obtain a general understanding of the behavior of ions having oxidation states ranging from +2 to +5, each of the elements except the americium and curium (+3) was studied on separate columns. In all cases, the eluate samples were obtained as a function of the elution volume (in ml). They were alpha-counted in all cases except Pa-233 which was gamma-counted. The results are summarized in Figure 2.

^[1]GEAP-4278

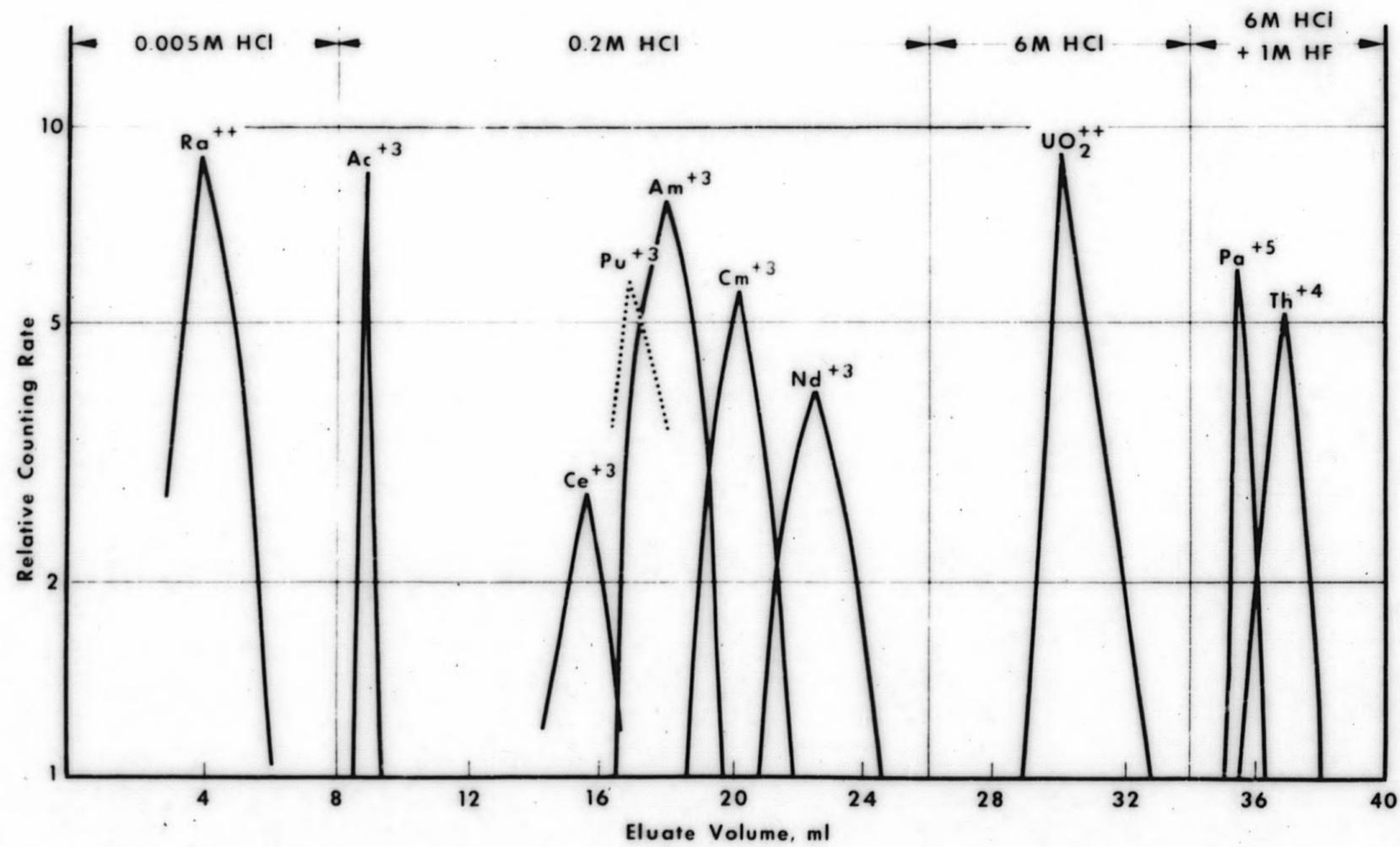


Figure 2. HEAVY ELEMENT CHROMATOGRAM, 0.1 gm D2EHP ON CHROMOSORB

It was found that the group 2 element radium was weakly adsorbed, as expected. The Ac^{+3} was more strongly adsorbed than Ra^{+2} but slightly less than La^{+3} . The UO_2^{++} was strongly adsorbed up to concentrations of 4M HCl . At 6M HCl , where the UO_2^{++} ion begins forming strong chloride complexes, the uranium was eluted. An attempt was made to elute the UO_2^{++} with 6M HNO_3 without success. In this case, only a weak complex is formed with the nitrate ion, which tends to indicate the complexing action of the chloride ion is responsible for the UO_2^{++} elution.

The Th^{+4} and Pa^{+5} were found to be strongly adsorbed at all HCl concentrations. These were eluted upon the addition of strong complexing agents, such as oxalate ion or fluoride ion. The elution curves in 1M $\text{H}_2\text{C}_2\text{O}_4$ and 6M $\text{HCl} + 1\text{M}$ HF for both the Th^{+4} and Pa^{+5} are shown in Figure 3. Attempts to elute the Th^{+4} by complexing with the nitrate ion in 8M HNO_3 were not successful.

The behavior of the Pu^{+3} , Am^{+3} , and Cm^{+3} was studied initially as a group, together with Ce^{+3} and Nd^{+3} which were used to correlate the position of these heavy element elution peaks relative to the rare earth peaks previously obtained. The Am and Cm were found to elute between Ce and Nd (Figure 2) but the Pu which had been reduced supposedly to the $+3$ state by treatment with Fe^{++} and NH_2OH remained tightly adsorbed. Attempts to reduce the Pu to the $+3$ state on the column (assuming it had been oxidized initially) were not successful.

In order to observe the oxidation-reduction behavior of the plutonium on the D2EHP column visually, use was made of the color differences of macro amounts of Pu associated with the different oxidation states. Dissolution of plutonium metal in HCl resulted in Pu^{+3} (blue) which was eluted in 3M HCl with very little hold-up. The Pu^{+4} (green) was obtained by the addition of BrO_3^- to Pu^{+3} solution with gentle heating.

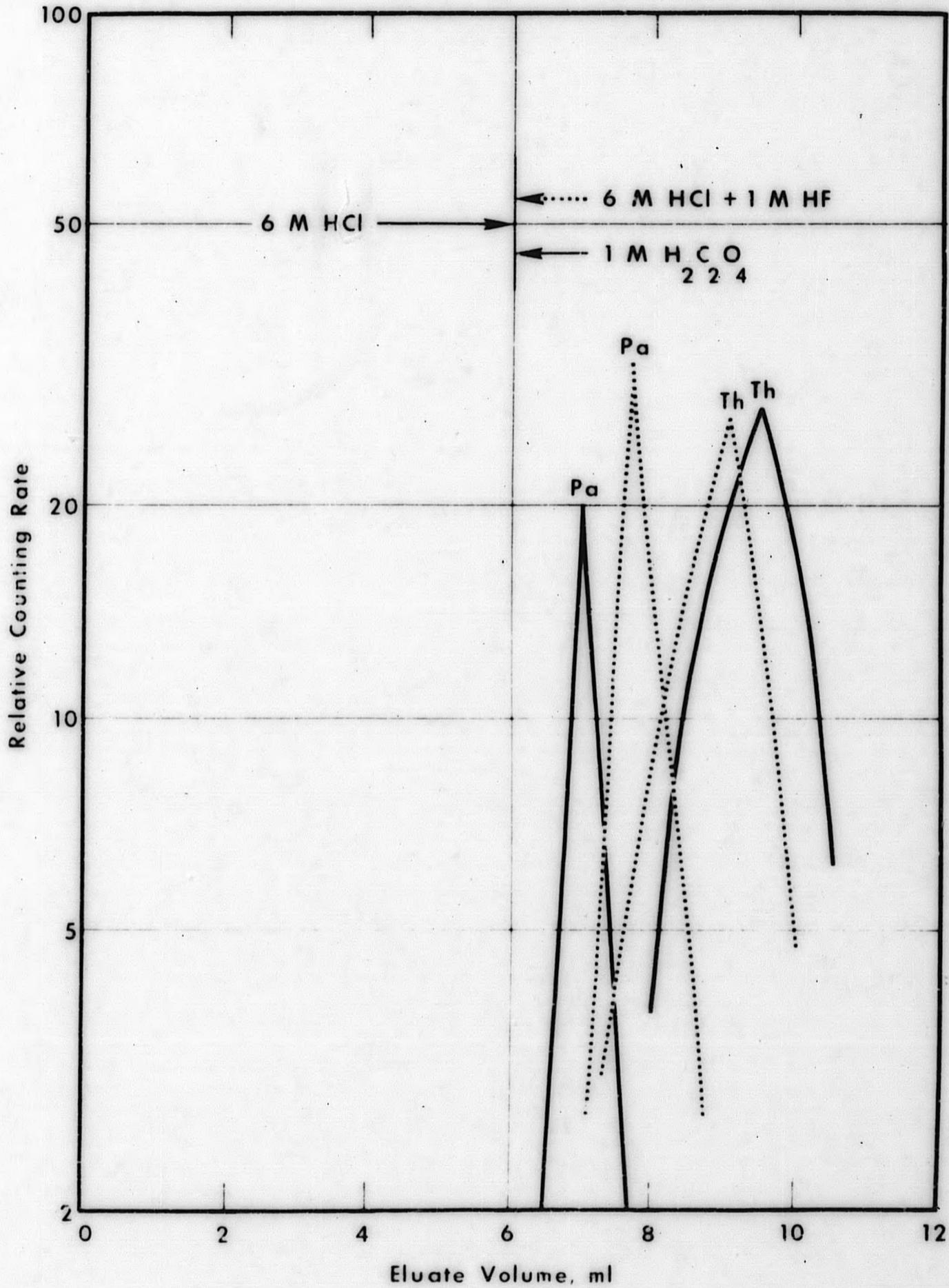


Figure 3. THORIUM AND PROTACTINIUM BEHAVIOR FOR 0.1 gm D2EHP ON CHROMOSORB

Upon transferring this solution to a D2EHP column, a green band adsorbed at the top of the column. A series of washes with 0.2M HCl to 6M HCl failed to move the band. Of the reducing agents tried, I^- , NH_2OH , Fe^{++} , $NH_2OH + Fe^{++}$, and Sn^{++} , only the latter two reduced the Pu^{+4} to Pu^{+3} . Various concentrations of HNO_3 from 1-6M were tried without moving the band. Apparently the anionic NO_3^- complex was too weakly formed in this system for elution. The Pu^{+4} band did appear to elute upon the addition of 6M HCl + 1M HF.

The preliminary results with PuO_2^{++} (light tan) are inconclusive. The light tan solution appeared to adsorb on the D2EHP column as a green band which had the same behavior as described previously for the Pu^{+4} . The reduction of the Pu^{+4} did not readily occur on the column with either Sn^{++} or $NH_2OH + Fe^{++}$.

From the results of the studies with macro amounts of plutonium, the following conclusions can be drawn: (a) reduction of Pu^{+4} to Pu^{+3} occurs with difficulty on a D2EHP column, (b) oxidation of Pu^{+3} to Pu^{+4} does not occur on a D2EHP column for macro amounts of plutonium, and (c) preliminary data with the PuO_2^{++} suggests reduction of this species to Pu^{+4} on a D2EHP column.

Preliminary results have been obtained with micro amounts of Pu^{+3} on a D2EHP column. In this case, Sn^{++} was present in the system at all times. The plutonium elution peak was found between the Ac^{+3} and the Am^{+3} peaks as expected (dotted Curve in Figure 2).

Further work will include a study of the Pu^{+3} to determine its exact elution peak position relative to Ac^{+3} and Am^{+3} . The Pu^{+4} system will also be observed such that the relation between Pu^{+4} , Ib^{+4} , and Pa^{+5} is understood.

Since many fuels also contain aluminum, a preliminary investigation of the behavior of aluminum on D2EHP columns was made. Aluminum was found to behave somewhat similar to trivalent americium. The most effective separation of both aluminum and americium from neodymium tried to date has been a Dowex-50Wx8 column (200-400 mesh) on which neodymium adheres strongly, but americium and aluminum pass through with 20% ethanolic 12-13M HCl. When aluminum is a major constituent, an initial Dowex-50 separation may be desirable to avoid saturation of the D2EHP column.

IV. BEHAVIOR OF NEODYMIUM IN HIGH HEAT FLUX FUELS

In order to observe the behavior of neodymium, the radial samples taken in Specimens A and B (reported in GEAP-4137 and GEAP-4278) were analyzed for neodymium. These radial samples were very small (< 15 mgs) and burnup was also low (0.0008 to 0.0030 atom percent fission). A typical analysis showed 6 nanograms of fission product Nd-148 and 3 nanograms of natural Nd-148. The analysis of such a small quantity of fission product above background is difficult and can be made with only limited accuracy.

Table I and Table II show the behavior of stable neodymium compared to radioactive nuclides for Specimens A and B, respectively. It is seen that there is a similar distribution for both stable neodymium and refractory radioactive fission products. The distribution of neodymium and radioactive fission products observed in Specimen B may well be a result of the crystallization process during the freezing of the melt. Since the neodymium values are not sufficiently accurate to permit a valid point-to-point comparison, the most significant observation is that the neodymium does not behave differently from other refractory fission products. In fuel which has melted, total dissolution of the fuel is the best way to obtain a representative sample for burnup analysis.

TABLE I
NORMALIZED FISSION PRODUCT TO URANIUM RATIOS FOR SPECIMEN A

Distance From Center (mils)	Atom Ratios of Fission Product to Uranium				
	Zr-95	Sr-89	Sr-90	Ce-144	Nd-148
246	2.52	2.54	2.03	2.57	4.25
254	1.99	1.61	1.46	2.14	4.63
270	.99	1.10	1.01	.83	1.08
278	--	1.14	1.10	--	--
286	.85	1.03	.95	.80	1.88
306	1.18	1.10	1.01	1.08	1.72
318	.94	1.05	.92	.84	--
346	.98	1.11	1.06	.76	--
350	--	1.11	1.03	.78	1.09
370	.79	1.05	1.11	.79	--
386	.84	1.09	1.13	.93	1.39
406	--	--	--	--	--
410	.85	1.12	1.15	.80	1.76
426	.63	1.06	1.05	.84	1.63
475	.76	1.14	1.13	.77	1.39
571*	1.00	1.00	1.00	1.00	1.00

* Normalizing Point

TABLE II

NORMALIZED FISSION PRODUCT TO URANIUM RATIOS FOR SPECIMEN B

Distance From Center (mils)	Atom Ratio of Fission Product to Uranium				
	Zr-95	Sr-89	Sr-90	Ce-144	Nd-148
66	1.42	1.36	1.48	1.07	1.16
103	1.07	1.12	1.27	1.07	.76
116	.95	1.06	1.16	1.08	--
124	1.10	.90	.97	1.09	2.08
140	1.10	1.20	1.30	1.05	.92
173	1.26	--	1.12	1.06	1.17
268	1.15	1.12	1.16	.99	--
371	.97	.89	.91	.90	1.07
475	.60	.42	.45	.74	.77
495	.92	.79	.84	.93	.97
499	.78	.56	.61	.88	--
512	1.00	.98	1.09	1.02	1.00
520	1.01	1.04	1.09	1.25	--
524	1.05	.98	1.08	.98	--
528	1.08	1.02	1.12	1.09	--
545	1.21	.95	.96	1.03	1.14
553	1.18	.99	1.16	1.06	--
574	1.07	1.02	1.09	.95	--
590*	1.00	1.00	1.00	1.00	1.00

*Normalizing Point.

V. CALCULATION OF Nd-148 FROM MASS SPECTROMETRIC RATIOS

When Nd-150 is used as an isotopic diluent for measurement of Nd-148 from fission, the 142 mass position is scanned for natural neodymium contamination. Equations for calculation of Nd-148 and Nd-142 by use of 142/150 and 148/150 ratios have been reported in GEAP-4201. The ratio of mass 142/143 has been found to be more easily and accurately measured than the 142/150 ratio and to be less subject to mass discrimination effects. The equations for calculation of Nd-148 and Nd-142 from measured 142/143 and 148/150 ratios have been formulated. These equations, which can be programmed on a computer in preference to hand calculations, are derived and listed in Appendix I of this report. Experience has shown that such equations are useful to individuals performing the analyses and that those who make use of such equations find their derivation simpler to follow than to reconstruct.

APPENDIX IDERIVATION OF EQUATIONS FOR CALCULATION Nd-148 AND Nd-142

The equations below were formulated to determine fission product Nd-148 and natural Nd-142 in samples spiked with Nd-150. Only the 142/143 and 150/148 ratios need be measured on each spiked sample. All other ratios for pure Nd-150 spike and natural neodymium are known from prior measurements.

A. Notations Used in Equations

- a, b, etc. = the last digit of the mass number, i.e., 2, 5, 6, 8, 0 for 142, 145, 146, 148 and 150 masses.
- $M_{a/b}$ = atom ratio as measured in mass spectrometer of (mass a/mass b) in the mixtures.
- $C_{a/b}$ = atom ratio measured in mass spectrometer of (mass a/mass b) in natural contamination.
- $S_{a/b}$ = atom ratio measured in mass spectrometer of (mass a/mass b) in the spike.
- $F_{a/b}$ = atom ratio as measured in mass spectrometer of (mass a/mass b) in fission product Nd.
- c_a = atoms or neodymium mass a from natural contamination.
- f_a = atoms of neodymium mass a from fission products.
- s_a = atoms of neodymium mass a from spike.

B. Calculation of Fission Product Atoms

It was assumed that no Ce or Sm isotopes are present; and that only $M_{0/8}$ and $M_{2/3}$ mixture ratios are required; and that Nd-150 was added for spike.

Defining the ratio $M_{2/3}$:

$$M_{2/3} = \frac{c_2 + s_2}{c_3 + s_3 + f_3} \quad (1)$$

Solving for c_2 and c_3

$$c_3 M_{2/3} - c_2 = s_2 - s_3 M_{2/3} - f_3 M_{2/3} \quad (2)$$

$C_{3/2}$ previously measured

$$C_{3/2} = \frac{c_3}{c_2} \quad c_2 C_{3/2} = c_3 \quad (3)$$

Substituting (3) into (2) and rearranging

$$c_2 C_{3/2} M_{2/3} - c_2 = s_2 - s_3 M_{2/3} - f_3 M_{2/3} \quad (4)$$

$$c_2 = \frac{s_2 - s_3 M_{2/3} - f_3 M_{2/3}}{C_{3/2} M_{2/3} - 1} \quad (5)$$

Defining $S_{2/0}$, $S_{3/0}$ and $F_{3/8}$

$$S_{2/0} = \frac{s_2}{s_0} \quad s_2 = s_0 S_{2/0} \quad (6)$$

$$S_{3/0} = \frac{s_3}{s_0} \quad s_3 = s_0 S_{3/0} \quad (7)$$

$$F_{3/8} = \frac{f_3}{f_8} \quad f_3 = f_8 F_{3/8} \quad (8)$$

Substituting (6), (7), (8) after rearrangement into (5)

$$c_2 = \frac{S_{2/0} s_0 - S_{3/0} M_{2/3} s_0 - f_8 F_{3/8} M_{2/3}}{C_{3/2} M_{2/3} - 1} \quad (9)$$

The equation of measured $M_{0/8}$ is found to be

$$M_{0/8} = \frac{s_0 + f_0 + c_0}{s_8 + f_8 + c_8} \quad (10)$$

Rearranging

$$M_{0/8} (s_8 + f_8 + c_8) = s_0 + f_0 + c_0 \quad (11)$$

Solving for f_8 and f_0

$$f_8 M_{0/8} - f_0 = s_0 + c_0 - s_8 M_{0/8} - c_8 M_{0/8} \quad (12)$$

Equation for $F_{0/8}$

$$F_{0/8} = \frac{f_0}{f_8} \quad f_0 = f_8 F_{0/8} \quad (13)$$

Rearranging (13) substituting into (12)

$$f_8 M_{0/8} - f_8 F_{0/8} = s_0 + c_0 - s_8 M_{0/8} - c_8 M_{0/8} \quad (14)$$

Previously measured equation for $S_{8/0}$, $C_{0/2}$ and $C_{8/2}$

$$S_{8/0} = \frac{s_8}{s_0} \quad s_8 = s_0 S_{8/0} \quad (15)$$

$$C_{0/2} = \frac{c_0}{c_2} \quad c_0 = c_2 C_{0/2} \quad (16)$$

$$C_{8/2} = \frac{c_8}{c_2} \quad c_8 = c_2 C_{8/2} \quad (17)$$

Substitution of equation (15), (16), and (17) into equation (14)

$$f_8 M_{0/8} - f_8 F_{0/8} = s_0 - s_0 S_{8/0} M_{0/8} + c_2 C_{0/2} - c_2 C_{8/2} M_{0/8} \quad (18)$$

Substituting equation (9) into equation (18) we obtain for the right side:

$$\begin{aligned}
 &= s_0 - s_0 S_{8/0} M_{0/8} + \frac{C_{0/2} S_{2/0} s_0}{C_{3/2} M_{2/3} - 1} - \frac{C_{0/2} S_{3/0} M_{2/3} s_0}{C_{3/2} M_{2/3} - 1} \\
 &- \frac{C_{8/2} M_{0/8} S_{2/0} s_0}{C_{3/2} M_{2/3} - 1} + \frac{M_{0/8} C_{8/2} S_{3/0} M_{2/3} s_0}{C_{3/2} M_{2/3} - 1} + \frac{C_{8/2} f_8 F_{3/8} M_{2/3} M_{0/8}}{C_{3/2} M_{2/3} - 1} \\
 &- \frac{C_{0/2} f_8 F_{3/8} M_{2/3}}{C_{3/2} M_{2/3} - 1} \tag{19}
 \end{aligned}$$

In order to simplify equation (19), the following substitutions were made:

$$O = \frac{F_{3/8} M_{2/3}}{C_{3/2} M_{2/3} - 1} \tag{20}$$

$$Q = \frac{S_{2/0}}{C_{3/2} M_{2/3} - 1} \tag{21}$$

$$R = \frac{S_{3/0} M_{2/3}}{C_{3/2} M_{2/3} - 1} \tag{22}$$

By rearrangement of equation (19) and substitution of equations (20), (21), (22), we obtain

$$\begin{aligned}
 &f_8 \left[M_{0/8} - F_{0/8} + C_{0/2} O - C_{8/2} M_{0/8} O \right] = \\
 &s_0 \left[1 - S_{8/0} M_{0/8} + C_{0/2} Q - C_{0/2} R - C_{8/2} M_{0/8} Q + C_{8/2} M_{0/8} R \right] \tag{23}
 \end{aligned}$$

Solving for f_8

$$f_8 = s_0 \frac{1 - S_{8/0} M_{0/8} + C_{0/2} (Q-R) - C_{8/2} M_{0/8} (Q-R)}{M_{0/8} - F_{0/8} + C_{0/2} 0 - C_{8/2} M_{0/8} (0)} \quad (24)$$

C. Derivation of the Equation for Natural Nd-142, c_2 , Contaminating the Samples

Equation (5) of Part B was solved in terms of natural Nd-142, c_2 and is a logical starting point.

$$c_2 = \frac{S_{2/0} s_0 - S_{3/0} M_{2/3} s_0 - M_{2/3} f_3}{C_{3/2} M_{2/3} - 1} \quad (25)$$

Measure relationship $M_{8/0}$

$$M_{8/0} = \frac{s_8 + c_8 + f_8}{s_0 + c_0 + f_0} \quad (26)$$

Rearranging

$$s_0 M_{8/0} + c_0 M_{8/0} + f_0 M_{8/0} = s_8 + c_8 + f_8 \quad (27)$$

Solving for f_0 and f_8

$$f_0 M_{8/0} - f_8 = s_8 + c_8 - s_0 M_{8/0} - c_0 M_{8/0} \quad (28)$$

Since we know that

$$s_{8/0} = \frac{s_8}{s_0} \qquad s_8 = s_0 s_{8/0} \qquad (29)$$

$$F_{0/3} = \frac{f_0}{f_3} \qquad f_0 = f_3 F_{0/3} \qquad (30)$$

$$F_{8/3} = \frac{f_8}{f_3} \qquad f_8 = f_3 F_{8/3} \qquad (31)$$

and

$$C_{0/2} = \frac{c_0}{c_2} \qquad c_0 = c_2 C_{0/2} \qquad (32)$$

$$C_{8/2} = \frac{c_8}{c_2} \qquad c_8 = c_2 C_{8/2} \qquad (33)$$

Substituting equations (29), (30), (31), (32), and (33) into (28)

$$f_3 F_{0/3} M_{8/0} - f_3 F_{8/3} = s_0 s_{8/0} + c_2 C_{8/2} - s_0 M_{8/0} - c_2 C_{0/2} M_{8/0} \qquad (34)$$

Solving for f_3

$$f_3 = \left[\frac{s_0 s_{8/0} - s_0 M_{8/0} + c_2 C_{8/2} - c_2 C_{0/2} M_{8/0}}{F_{0/3} M_{8/0} - F_{8/3}} \right] \qquad (35)$$

Substituting into (25)

$$c_2 \left[C_{3/2} M_{2/3} - 1 + \frac{M_{2/3} C_{8/2}}{F_{0/3} M_{8/0} - F_{8/3}} - \frac{M_{2/3} M_{8/0} C_{0/2}}{F_{0/3} M_{8/0} - F_{8/3}} \right] =$$

$$s_0 \left[s_{2/0} - s_{3/0} M_{2/3} - \frac{s_{8/0} M_{2/3}}{F_{0/3} M_{8/0} - F_{8/3}} + \frac{M_{2/3} M_{8/0}}{F_{0/3} M_{8/0} - F_{8/3}} \right] \qquad (36)$$

Divide both sides by $M_{2/3}$

$$c_2 \left[C_{3/2} - \frac{1}{M_{2/3}} + \frac{C_{8/2}}{F_{0/3} M_{8/0} - F_{8/3}} - \frac{M_{8/0} C_{0/2}}{F_{0/3} M_{8/0} - F_{8/3}} \right] = s_0 \left[\frac{S_{2/0}}{M_{2/3}} - S_{3/0} - \frac{S_{8/0}}{F_{0/3} M_{8/0} - F_{8/3}} + \frac{M_{8/0}}{F_{0/3} M_{8/0} - F_{8/3}} \right] \quad (37)$$

In order to simplify, T is substituted in equation (37)

$$T = \frac{1}{F_{0/3} M_{8/0} - F_{8/3}}$$

One obtains

$$c_2 \left[C_{3/2} - \frac{1}{M_{2/3}} + C_{8/2} T - M_{8/0} C_{0/2} T \right] = s_0 \left[\frac{S_{2/0}}{M_{2/3}} - S_{3/0} - S_{8/0} T + M_{8/0} T \right]$$

Solving for c_2

$$c_2 = s_0 \left[\frac{\frac{S_{2/0}}{M_{2/3}} - S_{3/0} - S_{8/0} T + M_{8/0} T}{C_{3/2} - \frac{1}{M_{2/3}} + C_{8/2} T - M_{8/0} C_{0/2} T} \right] \quad (38)$$

ACKNOWLEDGMENTS

The Research and Development Program which is sponsored by the AEC is being conducted by the General Electric Company with the following personnel contributing to the project this quarter:

Project Scientist

B. F. Rider

Chemistry

C. P. Ruiz

J. P. Peterson, Jr.

P. S. Luke, Jr.

F. R. Smith

Mass Spectrometry

W. E. Duffy

S. E. Haugland

E. B. Clayton

END