[redacted]

HW-24956

DECLASSIFIED

717106

BIOPHYSICS SECTION

RADIOLOGICAL SCIENCES DEPARTMENT

INTERIM REPORT ON THE RADIOCHEMICAL ANALYSIS OF HANFORD REACTOR EFFLUENT WATER

REPOSITORY PNL
COLLECTION General
BOX No. N/A
FOLDER N/A

Classification Cancelled and Changed To
DECLASSIFIED
By Authority of RLO-CG-4
S.E. Gydeson 11-11-88
By JJ Kearn 8-30-94
Verified By PM Eick 8-3

April 20, 1953

GENERAL ELECTRIC

HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

DECLASSIFIED

[redacted]

~~SECRET~~
~~Security Information~~

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

1107397

THIS DOCUMENT IS PUBLICLY
AVAILABLE

DECLASSIFIED

~~SECRET~~

[redacted]

E E Carey

HW--24956

DE90 006834

HW-24956

Health and Safety

This document consists of 39 pages. Copy No. ____ of 137 copies. Series A

INTERIM REPORT ON THE RADIOCHEMICAL ANALYSIS OF HANFORD REACTOR EFFLUENT WATER

0: INV 89

33520

April 20, 1953

by

R. L. Watters
E. E. Larkin

CLASSIFICATION CANCELLED
[redacted]
BY AUTHORITY OF S.E. Gydesen, RLO
BY T. Stein DATE 1-24-90
T. Davis 2-1-90

HANFORD WORKS
RICHLAND, WASHINGTON

Operated for the Atomic Energy Commission
by the
General Electric Company
under
Contract #W-31-109-eng-52

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

Route To	Read By	Date	Route To	Read By	Date

Photostat Charge $ 6.64 for
Access Permittees

Available from
Technical Information Service Extension
P. O. Box 1001, Oak Ridge, Tennessee

1-3

[redacted]

DECLASSIFIED

1107398

DECLASSIFIED

~~SECRET~~
~~Security Information~~

Health and Safety

INTERNAL DISTRIBUTION

COPY NUMBER	
1	H. M. Parker
2	C. N. Gross
3	W. K. MacCready - J. E. Maider
4	A. B. Greninger
5	R. S. Bell - E. P. Lee - W. M. Mathis
6	W. D. Norwood - P. A. Fuqua
7	W. A. McAdams
8	H. A. Kornberg - R. F. Foster - A. C. Case
9	D. W. Pearce - L. C. Schwendiman
10	P. C. Jerman
11	R. M. Fryar - M. Lewis - J. M. Atwood
12	H. R. Schmidt
13	J. M. Smith - J. W. Healy
14	Z. E. Carey
15	D. L. Reid
16	H. J. Paas
17	R. L. Watters
18	700 File
19	300 File
20	Yellow Copy

EXTERNAL DISTRIBUTION

21-28	Argonne National Laboratory
29	Armed Forces Special Weapons Project (Washington)
30	Army Chemical Center
31-33	Atomic Energy Commission, Washington
34	Battelle Memorial Institute
35-37	Brookhaven National Laboratory
38	Brush Beryllium Company
39	Bureau of Medicine and Surgery
40-41	California Research and Development Company
42-43	Carbide and Carbon Chemicals Company (C-31 Plant)
44	Carbide and Carbon Chemicals Company (K-25 Plant)
45-52	Carbide and Carbon Chemicals Company (ORNL)
53-54	Carbide and Carbon Chemicals Company (Y-12 Plant)
55	Chicago Patent Group
56	Chief of Naval Research
57	Columbia University (Failla)
58	Dow Chemical Company (Rocky Flats)
59-63	duPont Company
64-65	General Electric Company (ANPP)
66	Hanford Operations Office
67-72	Idaho Operations Office
73	Iowa State College
74-75	Knolls Atomic Power Laboratory
76-77	Los Alamos Scientific Laboratory
78	Mallinckrodt Chemical Works
79	Massachusetts Institute of Technology (Kaufmann)
80-82	Mound Laboratory
83	National Advisory Committee for Aeronautics, Cleveland

~~SECRET~~
~~Security Information~~

DECLASSIFIED

DECLASSIFIED

[redacted]

COPY NUMBER	EXTERNAL DISTRIBUTION
84	National Lead Company of Ohio
85	Naval Medical Research Institute
86	Naval Research Laboratory
87	New Brunswick Laboratory
88-90	New York Operations Office
91-92	North American Aviation, Inc.
93	Nuclear Development Associates, Inc.
94	Patent Branch, Washington
95	Public Health Service
96	RAND Corporation
97	Savannah River Operations Office (Augusta)
98	Sylvania Electric Products, Inc.
99	The Surgeon General
100	USAF Radiation Laboratory
101	U.S. Naval Radiological Defense Laboratory
102	UCLA Medical Research Laboratory (Warren)
103-106	University of California Radiation Laboratory
107-108	University of Rochester
109	University of Tennessee (Comar)
110	University of Washington
111-112	Vitro Corporation of America
113-116	Western Reserve University (Friedell)
117	Westinghouse Electric Corporation
118-122	Wright Air Development Center
123-137	Technical Information Service, Oak Ridge

[redacted]

DECLASSIFIED

[redacted] [redacted]

DECLASSIFIED

INTERIM REPORT ON THE RADIOCHEMICAL ANALYSIS OF HANFORD REACTOR EFFLUENT WATER

INTRODUCTION:

The Biophysics Control Laboratory, Radiological Sciences Department, has studied and analyzed reactor effluent water for the various beta particle emitting isotopes on a routine basis since late 1950. The objectives of this work were the identification of the radioactive isotopes present and the recognition and explanation, if possible, of any trends in the isotope spectrum throughout the year.

An earlier report (1) contains the routine sampling and analytical procedures with the presentation of earlier results. The present report gives a discussion of new isotopes found, corrections made in counting factors, and a detailed discussion of results obtained during the period from January, 1951, to June, 1952.

SUMMARY:

Several fission products have been identified and quantitatively determined in reactor effluent water. From the activity densities determined, an estimation of the uranium available for releasing fission products to the cooling water has been made assuming instantaneous release of the fission products to the water. The analytical data obtained indicate that approximately 85% of the total beta particle emitting isotopes have now been properly identified. Certain variations of total beta particle activity and isotopic spectrum between reactors and during different seasons are noted, but lack of sufficient data prevents actual definitions and correlations. Revised procedures for sodium and the rare earth elements are included in the appendix with those procedures adopted for the determination of iodine, strontium, and barium.

PROCEDURES:

The isotopes of strontium and barium were separated from the effluent water initially by precipitation of the carbonates in the presence of carrier. This pre-

[redacted] [redacted]

DECLASSIFIED

cipitate was dissolved and the strontium and barium separated from calcium by precipitation of strontium and barium nitrates in fuming nitric acid. The barium and strontium were finally precipitated as the chromate and oxalate, respectively. Decay curve analysis was used to identify the two isotopes Sr^{91} and Ba^{139}.

Separation of the total iodine isotopes was accomplished by a distillation method in which the iodide was oxidized to free iodine with nitrous acid and (3) collected in carbon tetrachloride. The iodine was reduced with sodium bisulfite, extracted into water and precipitated as palladous iodide. The decay curve of the total iodine isotopes was then analyzed to determine the amount of I^{131} present.

The analytical procedure for the rare earth isotopes (1) was revised to eliminate the zirconium iodate step and the consequent loss of part of the rare earth fraction to the iodate precipitate. This change was possible because the zirconium present was effectively removed in the fluoride precipitation and the low thorium concentrations did not significantly affect the rare earth determinations. This revision of procedure resulted in a significant increase in the rare earth concentrations reported.

The analytical procedure for sodium reported previously (1) has been shortened by limiting scavenging steps to one sulfide precipitation from an ammoniacal solution. A calcium oxalate precipitation could probably be made at the same time but this step has been carried out separately as a qualitative check on the activity from the alkaline earths.

Backscatter measurements were made by mounting the isotope on formvar film, which had previously been found to give negligible backscatter (4), and measuring the counting rate with and without stainless steel backing. The ratio of the counting rate with the stainless steel backing to that with formvar backing only, gave the backscatter correction factor.

[redacted]

The effluent analysis data were compiled for the period from January, 1951, to June, 1952, and a statistical study carried out. These data were analyzed for variation of total beta activity density and of the isotopic spectrum with different reactors and different seasons of the year. All data were adjusted for variations in power level and flow rate of cooling water before correlations were attempted by assuming linear relationship between activity and power level and by comparing activity per unit time rather than activity density. The significance of the differences existing in effluents was determined by T-test.

RESULTS:

Six individual isotopes, as well as the rare earth group, all of which can be formed from fission, have been identified. The activity density of the effluent contributed by each of these isotopes is given in Table I along with similar figures for those isotopes formed by (n, γ) reaction. Measurements on individual samples are included in Table III. The summation of average contributions by the isotopes accounts for approximately eighty-five percent of the total beta activity.

TABLE I
BETA PARTICLE EMITTERS IN REACTOR WASTE EFFLUENT

Isotopes Formed By (n, γ) Reaction	Average Activity Density Units of 10^{-6} uc/cc	Ave. %	Isotopes From Fission	Average Activity Density Units of 10^{-6} uc/cc	Ave. %
Cu - 64	537	27	R.E. + Y(2)	159	6.6
Mn - 56	450	22	R.E. + Y(3)	66	3.3
Na - 24	302	15	Ba - 139(1)	29	1.6
Si - 31	175	7.3	Sr - 91	12	0.67
As - 76	50	2.2	Mo - 99	3.3	0.20
I (total)(1)	27	1.4	I - 131	0.33	0.014
Cr - 51	9.4	0.51	Ba - 140	0.37	0.020
P - 32	9.0	0.39	Sr - 89	0.12	0.0039
Fe - 59		0.01			
Ca - 41, 45, 47	< 1.1	< 0.05			

(1) This figure includes activity from both fission and (n, γ) reaction.
(2) Results of analysis for rare earth-yttrium fraction by revised procedure.
(3) Results of analysis for rare earth-yttrium fraction by original procedure(1).

It was found that there were no significant differences between Areas in the activity density measurements in 1951 but that the activity density of reactor effluent in 100-H Area was significantly lower than that in all other areas during the period in 1952. With the exception of 100-H Area, the measurements made in 1952 were significantly higher in all areas than they were during the same period of 1952. The increase could not be accounted for by increased power levels if the linear relationship is assumed. Some evidence of higher concentrations of total beta particle emitters in reactor effluent water during the spring season were noted during 1951, but it was neither pronounced nor definite. The marked increase of such measurements in 1952 may well be due to more pronounced effect of the possible seasonal variation.

During the period from January, 1951, to October, 1951, when water treatments in all areas were similar with use of ferric sulfate as coagulant, the only highly significant difference for those isotopes measured occurred in the presence of higher concentrations of Mn^{56} in the reactor effluent water at 100-DR than those in the effluent from other areas. This may possibly be explained by the limited chlorination of the water in this area before the coagulation step.

After October, 1951, when aluminum sulfate was substituted for ferric sulfate as the coagulating agent at 100-F, significantly lower concentrations of Cu^{64} were observed in the reactor effluent water from that area. These lower concentrations were significnatly different from previous values at 100-F and from concentrations in other areas during the same period.

Two further changes, the addition of silica as a coagulating agent at 100-F after January, 1952, and the omission of the sodium dichromate after April, 1952, at all areas cannot be fully evaluated from this data. Marked increases were noted during the early months of 1952 in the Mn^{56} content of the water at 100-F. This increased concentration, significantly different from that at all other areas, may be due to the presence of manganese in the lime which was added after the coagulation step in the water treatment at 100-F.

Concentrations of Si^{31} were higher in effluents from all areas during 1952 than during the same period in 1951 and the concentrations of this isotope in effluent from 100-B were significantly higher than in effluent from all other areas during 1952. Concentrations of As^{76} were higher in the effluents from all areas except 100-DR during 1952. Concentrations of Mn^{56} continued to be significantly higher at 100-DR than from all other areas except 100-F.

Counting corrections for certain isotopes have been revised and are given in Table III. All other factors are those used in previous calculations (1). These factors apply for measurements using the conventional mica-window counter.

TABLE III
REVISED COUNTER CORRECTIONS

Isotope	Window and Air Absorption Correction Factor	Backscatter Factor
Mn^{56}	1.05	1.04 *
Cu^{64}	1.2	1.2 [1]
R.E. + Y	1.3	1.3
Cr^{51}		
Shelf 1	1.5	
2	1.9	
3	2.0	

* Mounted on filter paper. All other isotopes are mounted on stainless steel, 0.006" thick.

[1] Recent measurements by the Radiochemical Standards forces of Radiological Sciences have indicated that the backscatter factor for Cu^{64} may be as much as 1.4 resulting in more conservative figures for Cu^{64}.

Earlier work has shown Ba^{140}, a fission product, to be present in reactor effluent water under normal operation conditions (1). However, concentrations were not accurately determined. Further work initiated in this group for the development of reliable analyses for fission product in the reactor cooling water after a slug rupture revealed that concentrations of I^{131} in normal reactor cooling water were of the same order of magnitude as those of Ba^{140} (5). This led to an intensive search for fission products in the cooling water during normal operation.

Two other isotopes with possible fission source that have been identified and determined quantitatively are Ba^{139} and Sr^{91}. The latter is apparently in a mixture of all the strontium isotopes which would be present four hours after release from the reactor and is the predominant isotope measured by the counting methods used. It should be noted that the Ba^{139} concentration may not all be produced by fission since this isotope may be formed from Ba^{138} by the neutron-gamma reaction. Indications of tellurium and zirconium isotopes have also been found but quantitative values await the development of specific and efficient separations.

The rare earth fraction is of special interest since it is possible that the bulk of this activity could come from fission isotopes. A comparison of the decay curve of this fraction with the decay curve of the rare earths formed by fission indicates that this is quite possible.

Calculations have been made to determine the uranium in the reactor available for release of fission products to the effluent water assuming immediate release of fission products to the water. The results of such calculations are shown in Table IV and are based on the assumption that the flow rate of reactor effluent was 130,000 liters/minute with neutron flux equal to 10^{13} n/cm^2/sec. These results indicate that some or all of the fission products may be retained in the film on tubes and slugs for an undetermined length of time.

TABLE IV
URANIUM NECESSARY FOR FISSION PRODUCTS IN REACTOR EFFLUENT

Isotope Used	Isotopic Activity in Effluent Leaving Reactor Units of 10^{-6} uc/cc	Conc. U g/Reactor
Ba^{139}	240	19
Ba^{140}	0.50	10
Sr^{89}	0.077	6.4
Sr^{91}	9.9	3.6
I^{131}	0.77	20

[redacted]

Possible sources of this uranium may be the natural uranium in the cooling water, uranium present on the outside of the slugs when they are inserted into the reactor, and that uranium possibly left in the tubes from previously ruptured slugs. Measurements in cooling water entering the reactor at 100-B indicate natural uranium concentrations in the cooling water equal to 0.25 ug/liter. This uranium in the cooling water may be a source of uranium in the film. Complete surface smears made on slugs just before they entered the reactor at 100-F on November 23, 1951, showed an average amount of uranium per slug removed by this means to be 7 ug with as much as 40 ug of uranium being removed from one slug. The order of magnitude of that uranium contributed by the third means has not been measured.

Further work will be carried out to determine the effect of seasonal changes on the effluent as well as the effect of changes in water treatment prescribed by the Water Quality Studies Group of the Technical Section, Engineering Department.

ACKNOWLEDGEMENTS:

The writers wish to acknowledge the assistance of the Radiological Sciences Department Regional Survey, and the Radiation Monitoring Unit (Reactor Section) personnel who collected the samples analyzed, and the Radiological Sciences Control Services personnel who carried out the statistical studies.

TABLE V
ACTIVITY OF MAJOR CONSTITUTENTS OF PILE EFFLUENT WATER

Sample Location	100-B	100-B	100-B	100-B	100-B	100-B
Sampling Date	2-15-51	2-23-51	3-13-51	3-20-51	4-6-51	6-12-51
Sampling Time	0940	0930	0930	0900	0840	0920
Total Beta Activity Density, units of 10^{-3} uc/cc	1.7	1.6	1.9	2.6	1.4	1.9
% Total Beta Activity						
Manganese - 56	-	-	17	23	-	22
Sodium - 24	-	13	21	22	-	12
Copper - 64	-	28	33	61	-	20
Arsenic - 76	-	1.3	3.5	1.5	-	1.3
Rare Earths + Y	5.3	-	-	4.8	3.2	-
Silicon - 31	-	-	-	-	-	-
Phosphorus - 32	-	-	0.19	0.66	-	0.45

TABLE V (Cont.)
ACTIVITY OF MAJOR CONSTITUENTS OF REACTOR EFFLUENT WATER

Sample Location	100-B	100-B	100-B	100-B	100-B	100-B	100-B	100-B
Sampling Date	6-27-51	7-24-51	8-2-51	9-11-51	9-24-51	10-30-51	11-15-51	12-5-51
Sampling Time	0930	0900	0915	0920	1005	0955	0930	0920
Total Beta Activity Density, units of 10^{-3} uc/cc	1.9	1.7	2.3	2.1	2 3	2.0	4.3	1.9
% Total Beta Activity								
Manganese - 56	21	31	19	15	15	17	7.6	12
Sodium - 24	12	12	8	17	13	10	6.2	17
Copper - 64	36	34	23	32	56	29	55	27
Arsenic - 76	3.9	1.0	0.7	0.5	0.9	0.7	0.4	4.5
Rare Earths + Y	-	-	3.0	-	-	-	-	14 *
Silicon - 31	6.4	5.8	7.0	8.2	5.8	7.8	3.2	5.5
Phosphorus - 32	0.08	0.02	-	-	0.22	0.78	-	0.48

* Results obtained by revised method of analysis.

DECLASSIFIED

TABLE V
ACTIVITY OF MAJOR CONSTITUENTS OF REACTOR EFFLUENT WATER

Sampling Location	100-B	100-B	100-B	100-B	100-B	100-B	100-B	100-B	100-B
Sampling Date	1-22-52	2-11-52	2-18-52	3-18-52	4-14-52	5-7-52	6-4-52	6-18-52	6-30-52
Sampling Time	0925	0900	0925	0930	0920	0910	0945	0926	0940
Total Beta Activity Density, units of 10^{-3} uc/cc	1.9	2.4	2.4	2.8	3.5	2.6	2.9	3.2	2.4
% Total Beta Activity									
Manganese - 56	12	6.5	13	25	22	17	16	-	16
Sodium - 24	17	18	15	12	13	14	12	15	15
Copper - 64	27	26	28	28	28	34	18	17	16
Arsenic - 76	4.4	3.3	0.47	1.5	5.3	3.0	1.5	0.90	-
Rare Earths + Y	-	7.7	8.3	5.4	6.3	5.9	5.2	9.4	5.1
Silicon - 31	9.3	7.6	6.3	6.5	9.7	13	12	14	16
Phosphorus - 32	0.77	1.3	0.37	0.58	0.26	0.42	0.50	0.51	0.38

DECLASSIFIED

TABLE V (Cont.)
ACTIVITY OF MAJOR CONSTITUENTS OF REACTOR EFFLUENT WATER

Sample Location	100-D	100-D	100-D	100-D	100-D	100-D	100-D	100-D
Sampling Date	2-27-51	5-8-51	5-10-51	5-18-51	7-3-51	8-1-51	8-28-51	9-17-51
Sampling Time	0930	1100	0910	0920	1000	0915	0915	0900
Total Beta Activity Density, units of 10^{-3} uc/cc	1.8	2.2	2.3	2.6	1.9	1.6	1.9	2.3
% Total Beta Activity								
Manganese - 56	20	21	31	20	23	20	15	14
Sodium - 24	15	14	9	13	-	19	16	11
Copper - 64	22	24	11	21	31	40	35	19
Arsenic - 76	3.0	3.1	3.1	0.3	0.1	0.9	2.6	0.2
Rare Earths + Y	-	-	-	-	-	3.4	-	-
Silicon - 31	-	-	-	9.6	9.0	7.7	6.6	6.1
Phosphorus - 32	0.37	0.11	-	-	-	-	-	-

TABLE V (Cont.)
ACTIVITY OF MAJOR CONSTITUENTS OF REACTOR EFFLUENT WATER

Sample Location	100-D	100-D	100-D	100-D	100-D	100-D	100-D	100-D
Sampling Date	11-13-51	11-26-51	12-11-51	12-26-51	2-4-52	2-12-52	3-3-52	4-3-52
Sampling Time	0915	1005	0935	0920	0915	0930	0945	0915
Total Beta Activity Density units of 10^{-3} uc/cc	1.5	2.3	2.2	2.4	2.5	2.1	1.8	3.0
% Total Beta Activity								
Manganese - 56	6.9	23	14	13	10	15	25	16
Sodium - 24	13	21	11	13	15	13	16	13
Copper - 64	42	18	20	32	38	30	42	19
Arsenic - 76	0.6	0.3	-	-	4.1	2.4	-	4.1
Rare Earths + Y	-	-	-	-	5.8	6.4	7.0	6.7
Silicon - 3.	3.8	6.0	6.2	5.8	7.4	7.5	9.2	6.2
Phosphorus - 32	0.8	0.28	0.53	0.59	0.42	0.43	0.46	0.52

TABLE V (Cont.)
ACTIVITY OF MAJOR CONSTITUENTS OF REACTOR EFFLUENT WATER

Sample Location	100-D	100-D	100-D	100-D
Sampling Date	4-30-52	5-20-52	6-9-52	6-25-52
Sampling Time	0925	0920	0920	1025
Total Beta Activity Density units of 10^{-3} uc/cc	2.7	2.9	2.5	2.5
% Total Beta Activity				
Manganese - 56	18	12	14	10
Sodium - 24	15	14	9.4	15
Copper - 64	23	20	18	17
Arsenic - 76	2.7	1.2	1.2	1.3
Rare Earths + Y	4.3	5.2	3.7	6.8
Silicon - 31	11	10	12	16
Phosphorus - 32	0.45	0.32	0.28	0.32

[redacted]

WATER

100-D	100-DR	100-DR	100-DR
4-16-5	4-24-51	5-1-51	5-29-51
0905	0905	0910	0940
1.8	2.7	2.3	1.7
7	34	29	45
0	8.3	18	14
0	19.2	19	35
2.8	2.4	4.0	-
-	2.2	-	-
-	-	-	10
0.06	0.18	0.03	0.03

DECLASSIFIED

TABLE V (Cont.)
ACTIVITY OF MAJOR CONSTITUENTS OF REACTOR EFFLUENT WATER

Sample Location	100-DR	100-DR	100-DR	100-DR	100-DR	100-DR	100-DR	100-DR
Sampling Date	6-4-51	7-5-51	7-11-51	8-6-51	8-13-51	8-30-51	11-8-51	12-4-51
Sampling Time	0945	0950	0920	0910	0925	0910	0945	0925
Total Beta Activity Density units of 10^{-3} uc/cc	2.3	2.5	2.6	2.0	2.3	2.4	2.3	2.5
% Total Beta Activity								
Manganese - 56	35	25	38	32	26	28	23	21
Sodium - 24	16	12	8	12	14	16	11	9
Copper - 64	27	25	29	31	25	33	25	30
Arsenic - 76	5.5	0.22	0.4	0.7	7.1	1.0	0.8	4.5
Rare Earths + Y	-	-	-	4.4	4.8	4.4	-	12 *
Silicon - 31	-	6.3	8.2	6.6	5.0	6.4	7.3	6.7
Phosphorus - 32	0.34	0.03	-	0.27	0.27	0.35	0.002	0.54

DECLASSIFIED

TABLE V (Cont.)
ACTIVITY OF MAJOR CONSTITUENTS OF REACTOR EFFLUENT WATER

Sample Location	100-DR	100-DR	100-DR	100-DR	100-DR	100-DR	100-DR	100-DR	100-DR	100-DR
Sampling Date	1-2-52	1-30-52	2-25-52	3-4-52	3-13-52	4-1-52	4-29-52	5-19-52	6-2-52	6-23-52
Sampling Time	0930	0940	0930	0915	0915	0905	0945	1005	0925	1040
Total Beta Activity Density, units of 10^{-3} uc/cc	2.3	2.0	2.9	3.0	3.0	3.0	2.5	2.4	2.4	2.4
% Total Beta Activity										
Manganese - 56	16	21	18	20	41	33	24	22	27	22
Sodium - 24	12	11	10	11	14	14	9.8	16	8.4	12
Copper - 64	30	25	25	23	18	20	19	22	11	25
Arsenic - 76	1.3	4.3	3.4	1.0	4.1	0.28	2.1	0.19	1.4	0.76
Rare Earths + Y	-	-	6.9	4.8	4.5	4.4	3.2	3.9	3.8	5.4
Silicon - 31	7.6	6.3	5.5	7.4	5.1	6.7	6.6	12	10	14
Phosphorus - 32	-	0.62	1.3	0.42	0.61	-	0.48	0.35	0.29	0.32

TABLE V (Cont.)
ACTIVITY OF MAJOR CONSTITUENTS OF REACTOR EFFLUENT WATER

Sample Location	100-F	100-F	100-F	100-F	100-F	100-F	100-F	100-F	100-F	100-F
Sampling Date	1-2-51	2-14-51	2-20-51	2-28-51	3-12-51	3-27-51	4-2-51	4-9-51	4-18-51	5-14-51
Sampling Time	0855	1040	0900	0900	0920	0900	0945	0855	1005	1000
Total Beta Activity Density, units of 10^{-3} uc/cc	1.4	2.0	1.5	1.9	2.3	2.5	2.4	2.6	1.9	1.9
% Total Beta Activity										
Manganese - 56	24	-	-	26	24	16	30	22	30	37
Sodium - 24	-	-	29	22	-	14	21	24	5	9
Copper - 64	-	-	-	28	21	22	20	16	24	40
Arsenic - 76	-	-	-	12	2.6	7.3	3.3	2.7	3.9	4
Rare Earths + Y	-	6.2	-	-	-	1.6	-	3.8	-	-
Silicon - 31	-	-	-	-	-	-	-	-	-	-
Phosphorus - 32	0.9	-	-	0.52	0.33	0.04	0.14	0.24	0.24	0.3

TABLE V (Cont.)
ACTIVITY OF MAJOR CONSTITUENTS OF REACTOR EFFLUENT WATER

Sample Location	100-F	100-F	100-F	100-F	100-F	100-F	100-F	100-F	100-F	100-F
Sampling Date	6-20-51	7-17-51	7-19-51	8-15-51	9-4-51	9-12-51	10-15-51	10-24-51	11-19-51	12-10-51
Sampling Time	0910	0940	0930	0910	0925	0930	1030	0940	0930	0910
Total Beta Activity Density, units of 10^{-3} uc/cc	2.2	1.6	2.0	2.2	0.9	1.8	1.6	1.8	1.7	1.8
% Total Beta Activity										
Manganese - 56	17	22	18	27	20	27	26	27	36	38
Sodium - 24	15	22	15	17	13	-	20	12	13	15
Copper - 64	17	32	12	30	23	31	23	20	20	17
Arsenic - 76	0.3	1.1	1.4	0.8	-	1.1	0.7	1.5	1.0	1.2
Rare Earths + Y	-	-	-	-	-	-	-	-	8.0 *	12 *
Silicon - 31	9.4	7.9	7.5	8.3	6.6	6.9	7.1	6.4	8.2	5.5
Phosphorus - 32	-	0.06	0.013	0.37	-	-	0.56	0.63	0.3	0.37

TABLE V (Cont.)
ACTIVITY OF MAJOR CONSTITUENTS OF REACTOR EFFLUENT WATER

Sample Location	100-F	100-F	100-F	100-F	100-F	100-F	100-F	100-F
Sampling Date	1-3-52	2-6-52	2-27-52	3-27-52	4-16-52	5-6-52	5-26-52	6-12-52
Sampling Time	0930	0930	0905	0905	0925	0925	0935	0930
Total Beta Activity Density units of 10^{-3} uc/cc	1.9	2.5	2.3	3.0	4.4	2.7	3.5	2.9
% Total Beta Activity								
Manganese - 56	27	26	28	47	52	55	37	44
Sodium - 24	14	16	16	13	9	12	6.6	10
Copper - 64	18	14	18	7.4	7.7	12	9.7	9.1
Arsenic - 76	0.46	4.2	3.5	5.5	2.3	3.9	2.2	3.5
Rare Earths + Y	-	7.3	7.5	7.1	-	6.7	6.9	5.6
Silicon - 31	6.8	7.3	5.6	3.3	4.2	6.6	8.2	11
Phosphorus - 32	0.16	0.41	0.51	-	0.087	0.51	0.19	0.36

TABLE V (Cont.)
ACTIVITY OF MAJOR CONSTITUENTS OF REACTOR EFFLUENT WATER

Sample Location	100-H	100-H	100-H	100-H	100-H	100-H	100-H	100-H	100-H	100-H
Sampling Date	1-10-51	1-16-51	1-29-51	2-15-51	3-19-51	3-28-51	4-3-51	4-10-51	4-23-51	5-16-51
Sampling Time	0840	0945	0850	0910	0915	0915	0935	0845	1000	0940
Total Beta Activity Density, units of 10^{-3} uc/cc	1.9	1.8	1.8	1.6	2.3	2.5	2.8	2.1	2.0	2.5
% Total Beta Activity										
Manganese - 56	27	21	15	-	28	26	26	32	25	18
Sodium - 24	23	17	17	-	42	21	7.4	18	13	8.9
Copper - 64	27	27	9.3	-	32	16	23	28	-	22
Arsenic - 76	4.1	-	3.8	-	2.5	4.4	2.1	5.2	1.1	0.5
Rare Earths + Y	-	-	-	5.4	2.6	1.5	-	3.4	3.3	-
Silicon - 31	-	-	-	-	-	-	-	-	-	-
Phosphorus - 32	-	-	0.87	-	0.42	0.09	-	0.05	-	0.16

TABLE V (Cont.)
ACTIVITY OF MAJOR CONSTITUENTS OF REACTOR EFFLUENT WATER

Sample Location	100-H	100-H	100-H	100-H	100-H	100-H	100-H	100-H	100-H	100-H
Sampling Date	6-5-51	6-25-51	7-9-51	7-25-51	9-5-51	9-20-51	10-2-51	10-22-51	11-28-51	12-18-51
Sampling Time	0935	0920	0935	0905	0920	0930	0915	0950	0925	0930
Total Beta Activity Density, units of 10^{-3} uc/cc	2.1	1.9	1.8	1.6	1.8	2.0	1.3	1.6	2.2	1.9
% Total Beta Activity										
Manganese - 56	21	27	21	24	16	20	16	15	16	21
Sodium - 24	17	16	15	19	17	15	12	14	12	16
Copper - 64	15	29	31	21	33	34	25	34	28	11
Arsenic - 76	1.4	0.2	0.2	0.5	0.4	0.8	0.7	1.0	0.9	0.7
Rare Earths + Y	-	-	-	-	-	-	-	-	13 *	-
Silicon - 31	-	9.0	6.5	5.4	5.4	5.9	5.6	5.0	6.2	8.5
Phosphorus - 32	0.25	-	0.05	-	-	-	0.4	-	0.90	0.73

TABLE V (Cont.)
ACTIVITY OF MAJOR CONSTITUENTS OF REACTOR EFFLUENT WATER

Sample Location	100-H	100-H	100-H	100-H	100-H	100-H	100-H	100-H
Sampling Date	1-7-52	1-14-52	2-20-52	3-10-52	4-18-52	4-22-52	5-28-52	6-16-52
Sampling Time	0940	0930	0915	0915	0915	0930	0925	0915
Total Beta Activity Density, units of 10^{-3} uc/cc	1.2	1.9	2.2	2.0	2.7	2.5	2.0	1.8
% Total Beta Activity								
Manganese - 56	23	14	9.7	11	14	21	22	19
Sodium - 24	19	13	16	16	13	12	13	15
Copper - 64	42	24	27	37	26	29	29	23
Arsenic - 76	7.3	2.7	3.7	-	2.5	2.6	0.11	1.4
Rare Earths + Y	-	-	11	8.3	5.0	4.6	5.5	4.8
Silicon - 31	7.5	6.5	6.2	4.9	5.1	7.9	11	15
Phosphorus - 32	0.087	-	0.55	0.51	0.39	0.44	0.23	0.35

DECLASSIFIED

LITERATURE CITED:

1. Larkin, E. E., Watters, R. L., Carey, Z. E., and Singlevich, W., HW-21844, Preliminary Report on the Radiochemical Analysis of Hanford Pile Effluent Water, August 1, 1951.

2. Hillebrand, W. F., and Lundell, G. E. F., Applied Inorganic Analysis, 1929, pp. 491, 504-505.

3. Furman, N. H., Scott's Standard Methods of Chemical Analysis, Volume I, Fifth Edition, pp. 450-451.

4. Healy, J. W., Schwendiman, L. C., and Thorburn, R. C., HW-18258, Counter Calibrations in the Health Instrument Methods Group, July 1, 1950.

5. Quimby, G. R., Unpublished Work, Laboratory Notebook HW-4708-T, pp. 21-23, September, 1951.

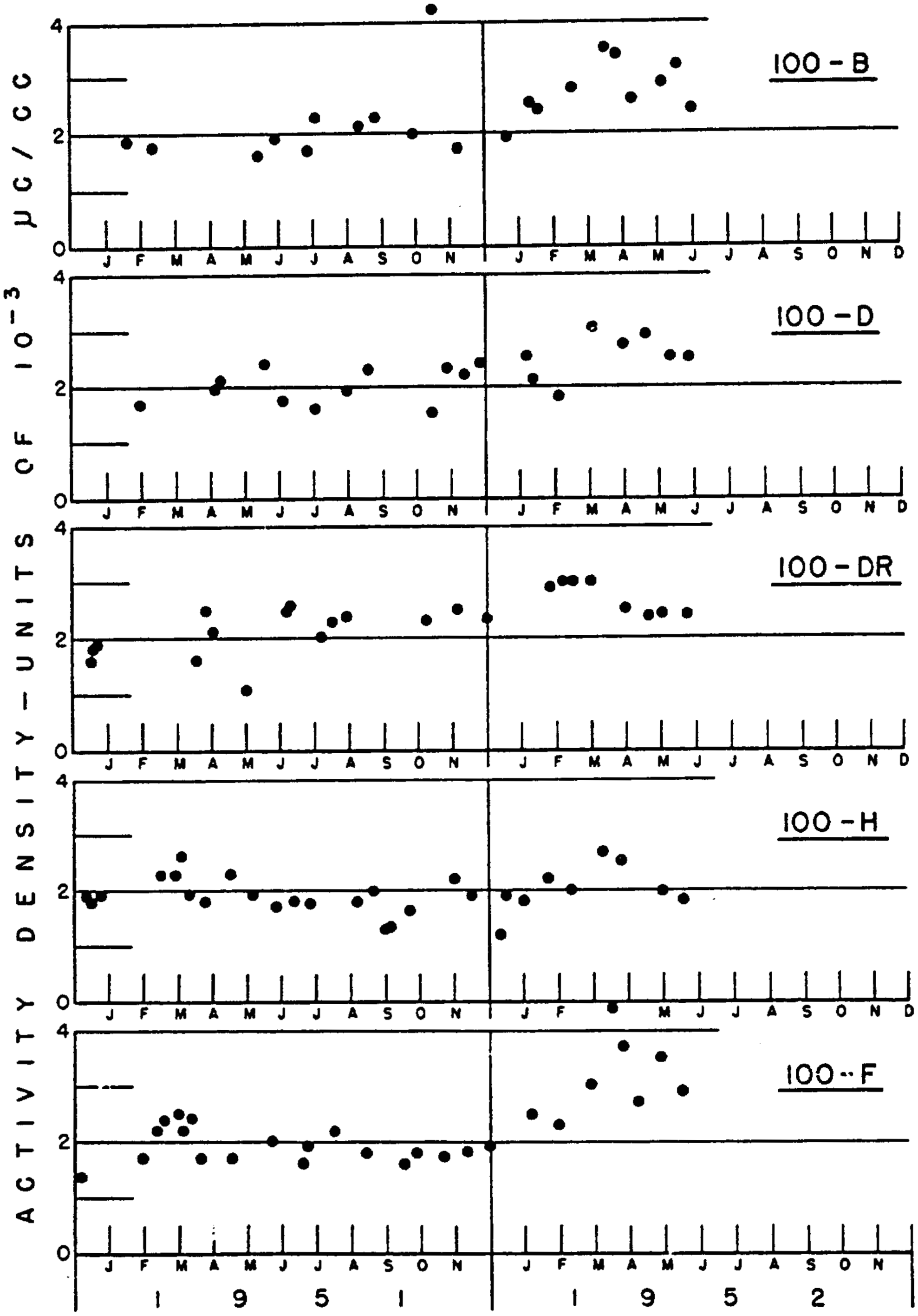
6. West, J. M., and O'Connor, J. J., 3-3398, Final Report on Production Test #305-6-P Activation of Process Water Additives, January 29, 1946.

~~SECRET~~

DECLASSIFIED

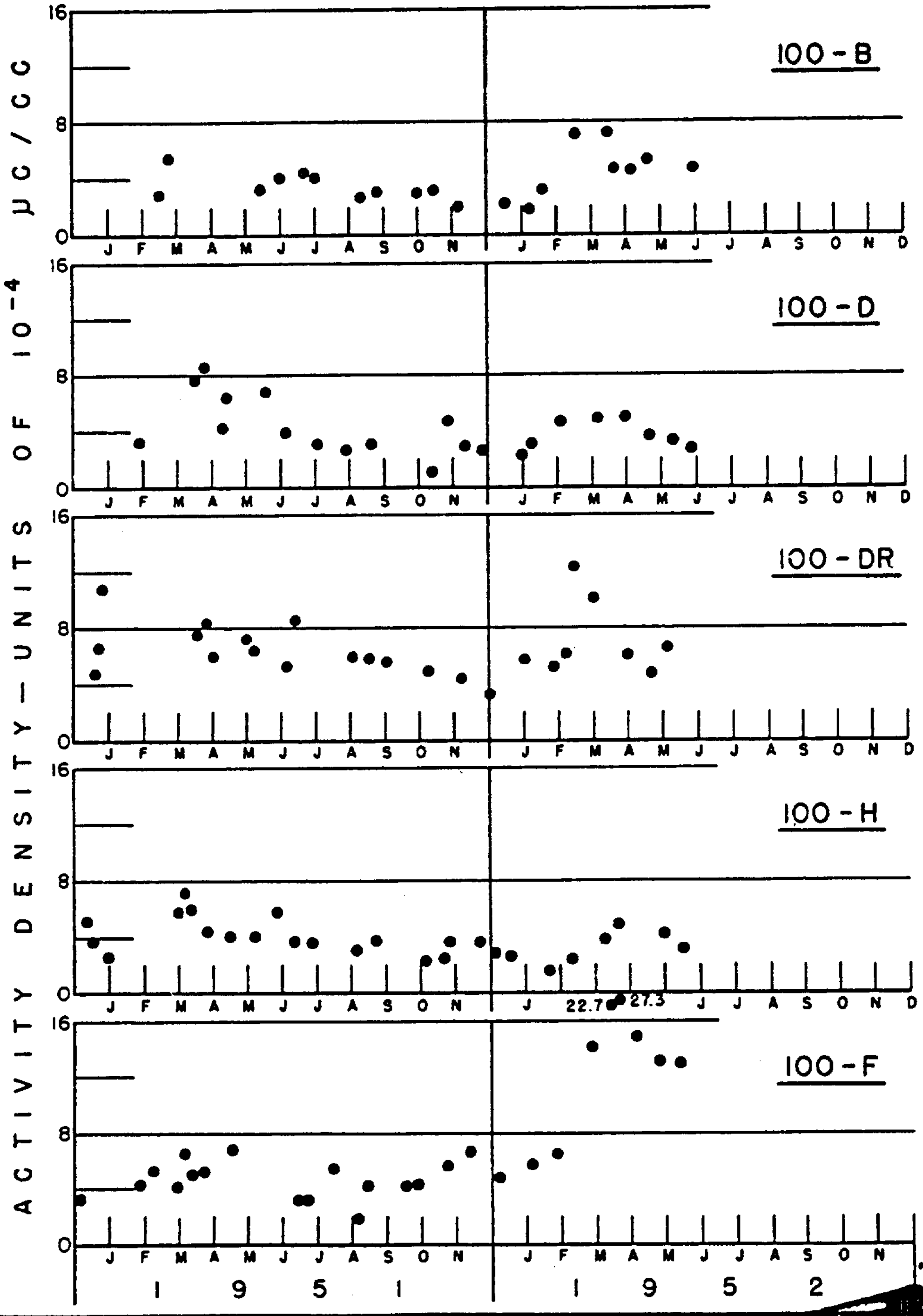
GROSS BETA ACTIVITY IN REACTOR EFFLUENT WATER

FIGURE - 1



MANGANESE^{56} ACTIVITY IN REACTOR EFFLUENT WATER

FIGURE - 2

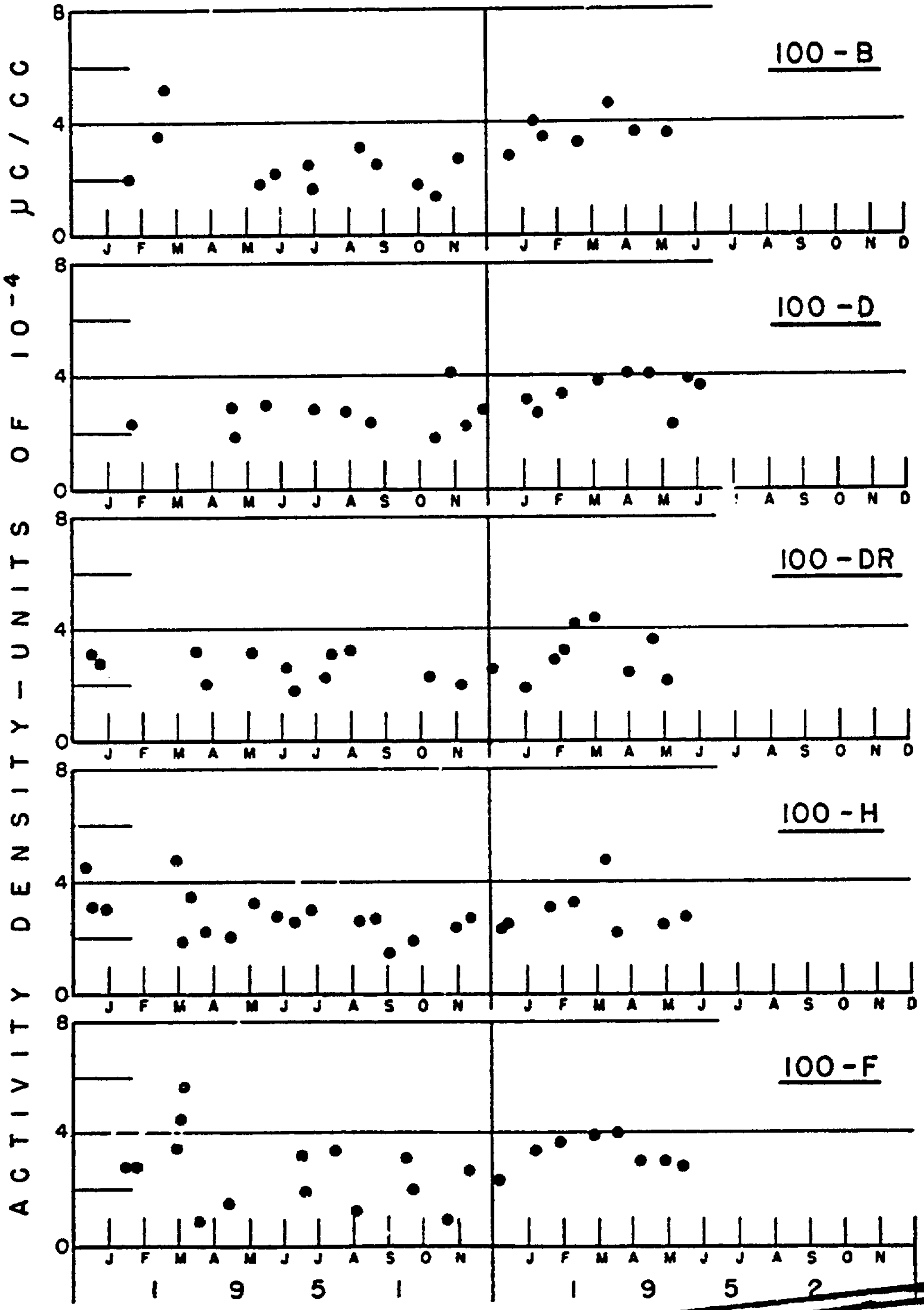


DECLASSIFIED

[redacted]

SODIUM^{24} ACTIVITY IN REACTOR EFFLUENT WATER

FIGURE - 3

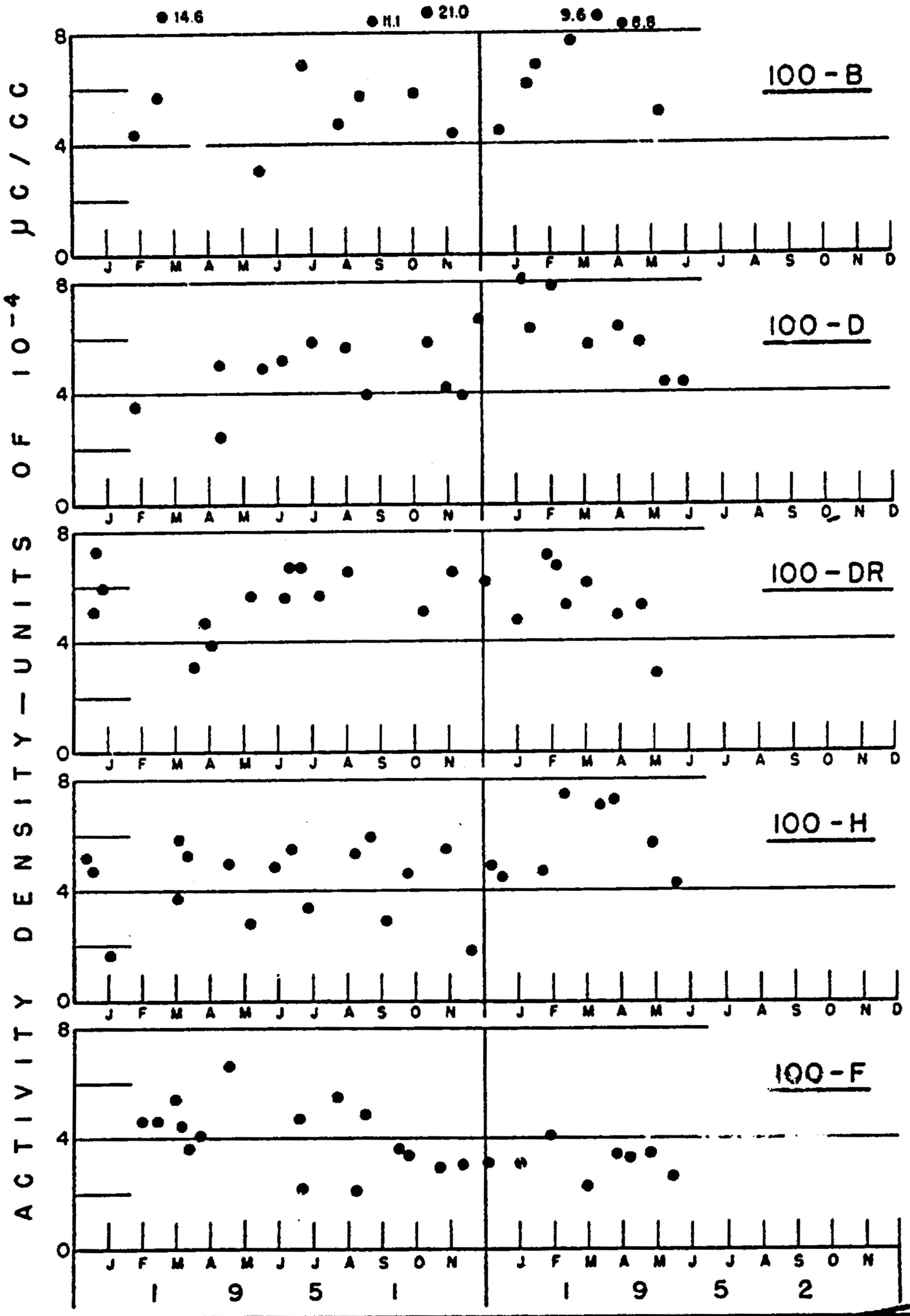


[redacted]

DECLASSIFIED

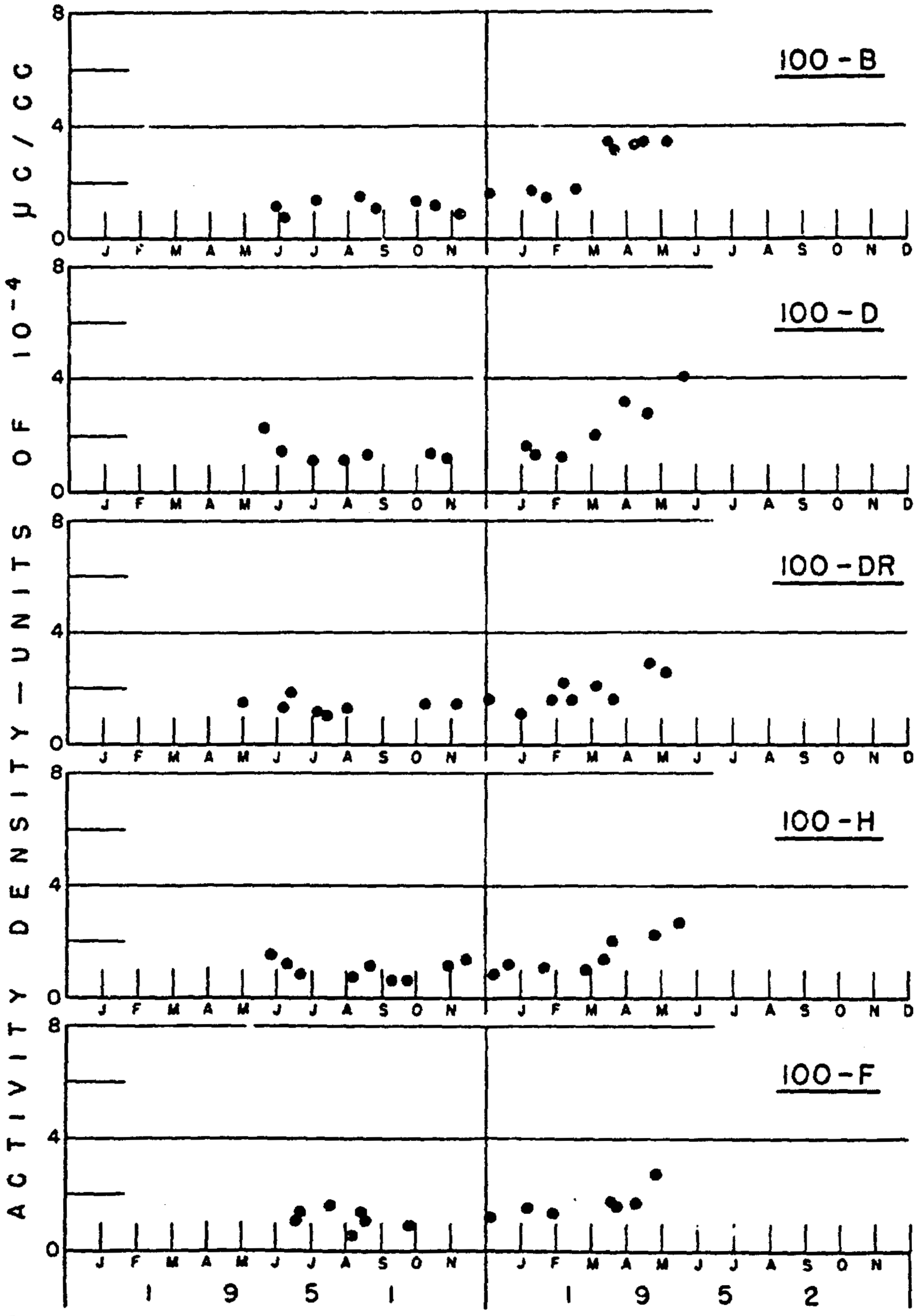
COPPER^{64} ACTIVITY IN REACTOR EFFLUENT WATER

FIGURE - 4



SILICON^{31} ACTIVITY
IN REACTOR EFFLUENT WATER

FIGURE - 5

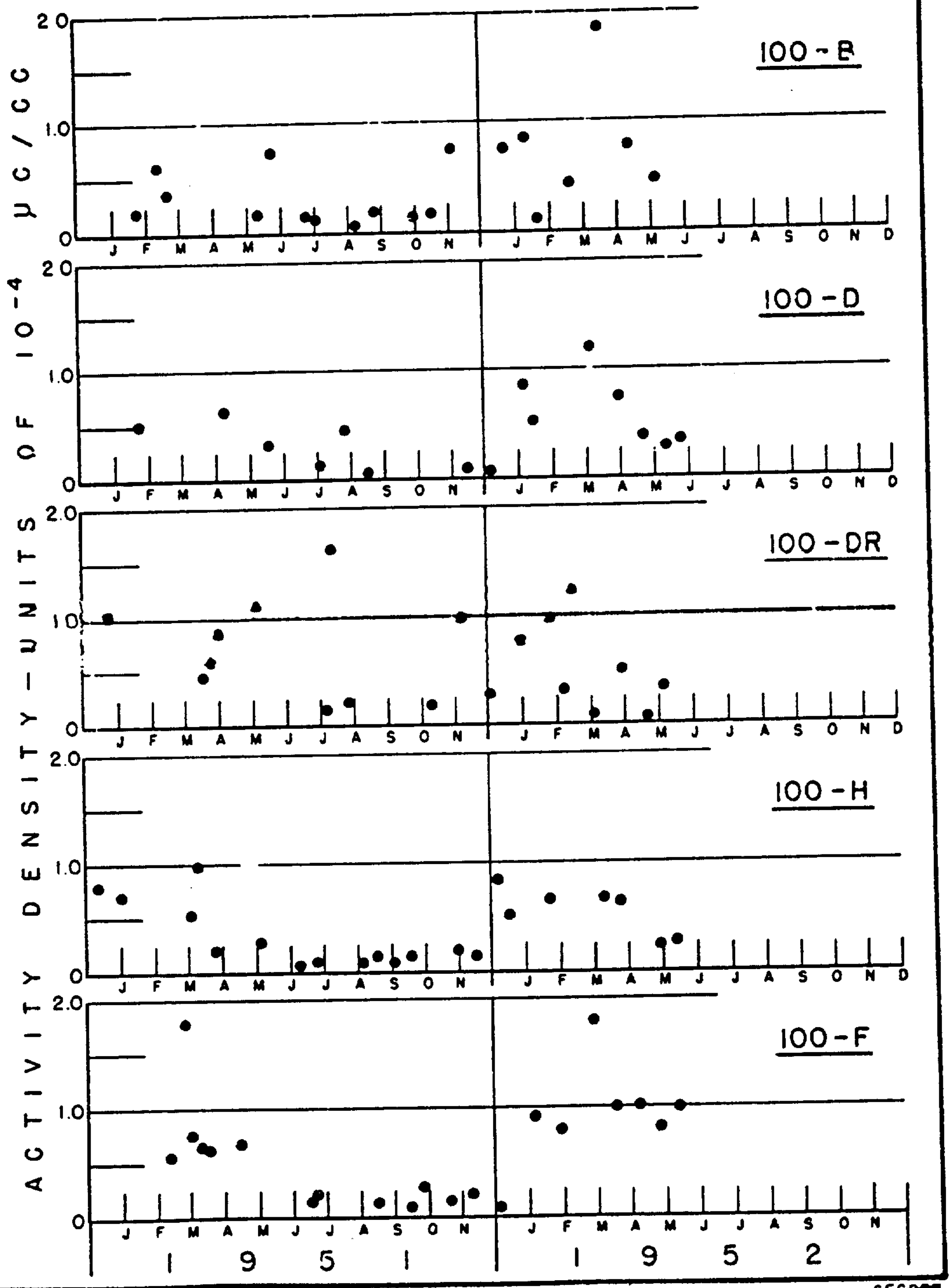


DECLASSIFIED

~~SECRET~~

ARSENIC^{76} ACTIVITY IN REACTOR EFFLUENT WATER

FIGURE - 6



~~SECRET~~

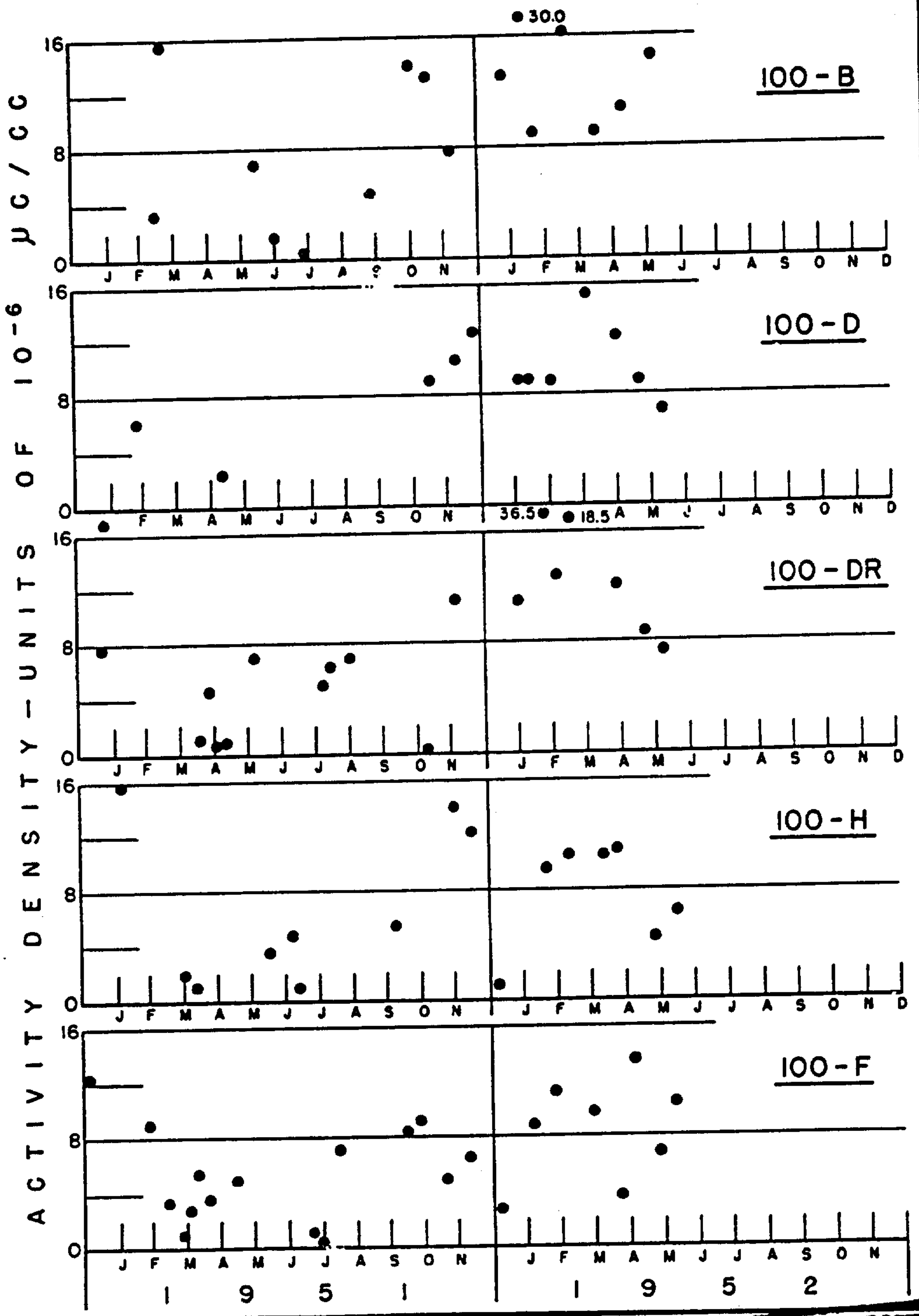
DECLASSIFIED

DECLASSIFIED

~~SECRET~~

PHOSPHORUS^{32} ACTIVITY IN REACTOR EFFLUENT WATER

FIGURE - 7



A E C G E RICHLAND WASH

~~SECRET~~

1107430

DECLASSIFIED

[redacted]

DECLASSIFIED

APPENDIX

DETERMINATION OF STRONTIUM AND BARIUM
Fuming Nitric Acid Method

PRINCIPLES AND LIMITATIONS OF METHOD:

This procedure is based upon the solubility of calcium nitrate in concentrated nitric acid while barium and strontium nitrates are not appreciably soluble. This procedure gives an excellent separation if the calcium present is not more than 0.1 gram. Chemical yield is approximately 80%. The time required is about two hours.

REAGENTS.

1. Carriers (Ag, Cu, Fe, La, Sr, Ba, Ca, and Na), 10 mg/ml standardized.
2. Nitric Acid, fuming.
3. Ammonium Hydroxide, c.p.
4. Hydrogen Sulfide, gas.
5. Sodium Carbonate, saturated solution.
6. Ammonium Bichromate, 100 g/l solution.
7. Ammonium Oxalate, saturated solution.
8. Ammonium Acetate, 3N solution.

PROCEDURE:

1. Transfer 100 ml of reactor effluent water to a 250 ml beaker.
2. Add 2.00 ml of Sr and Ba and about 2 ml of Ag, Cu, Fe, La, Ca, and Na.
3. Make ammoniacal and precipitate with hydrogen sulfide.
4. Centrifuge and save supernate. Discard precipitate.
5. Bring solution to boil and acidify. Boil until hydrogen sulfide is gone.
6. Add saturated sodium carbonate solution until basic and cool.
7. Filter through #42 filter paper, wash with 1% sodium carbonate solution, and discard filtrate.

[redacted]

DECLASSIFIED

8. Cautiously dissolve precipitate through the paper with dilute nitric acid and wash thoroughly with water.
9. Evaporate to 5 ml.
10. Add 30 ml of fuming nitric acid. Cool and centrifuge.
11. Decant solution and discard.
12. Dissolve precipitate in 2 ml of water and add 15 ml of fuming nitric acid. Cool and centrifuge.
13. Dissolve precipitate in 20 ml of water.
14. Add 5 ml of 3N ammonium acetate solution and bring to boil.
15. Add 5 ml of ammonium bichromate dropwise and cool.
16. Centrifuge. Save supernate and wash precipitate until washings are colorless. Add washings to supernate.
17. Dissolve precipitate in 2N nitric acid and re-precipitate with ammonium hydroxide.
18. Centrifuge. Discard supernate and wash once with water.
19. Transfer the barium chromate precipitate to a stainless steel plate and dry. Count at hourly intervals.
20. Add 10 ml of ammonium oxalate solution to the supernate in step (16).
21. Bring to boil and make ammoniacal.
22. Cool, centrifuge, and discard supernate.
23. Wash three times with water.
24. Count at hourly intervals. Transfer the strontium oxalate precipitate to a stainless steel plate and dry.

[redacted]

DETERMINATION OF SODIUM
Group Separations Method

PRINCIPLE AND LIMITATIONS OF METHOD:

Sodium^{24}, which has a half-life of 14.9 hours, constitutes approximately twenty percent of the total activity of reactor effluent water.

In this procedure, the analytical Groups I, II, and III are removed as sulfides from a basic solution and Group IV is removed as the oxalate. The solution is then evaporated to dryness and ignited to remove ammonium salts. The residue is taken up with water, filtered, acidified and plated.

THIS PROCEDURE HAS BEEN TESTED ONLY ON REACTOR EFFLUENT WATER.

REAGENTS:

1. Ammonium Hydroxide, c.p. concentrated.
2. Ammonium Oxalate Solution, saturated aqueous.
3. Carriers (silver, copper, iron, lanthanum, calcium, and sodium), 10.
4. Hydrogen Sulfide, gas.
5. Nitric Acid, c.p. concentrated.

PROCEDURE:

1. Transfer 100 ml of reactor effluent water to a 250 ml beaker. Add 2.00 ml of sodium carrier and 2 ml of silver, copper, iron, lanthanum, and calcium carriers.
2. Add 1 ml of ammonium hydroxide and pass in hydrogen sulfide for two minutes.
3. Centrifuge and decant the supernate through a #40 Whatman filter paper.
4. Add 5 ml of ammonium oxalate solution to the filtrate and heat to boiling.
5. Centrifuge, decant the supernate through a #40 Whatman filter paper, and evaporate the filtrate to dryness.
6. Ignite the beaker and contents over a Meker burner until all of the ammonium salts are removed.

7. Add 20 ml of distilled water and heat to boiling.
8. Filter, wash the paper with hot water and collect the filtrate and washings in a 100 ml beaker.
9. Add 2 ml of nitric acid, evaporate to a volume of 3 to 4 ml, and transfer to a one inch stainless steel plate.
10. Dry under an infra-red lamp. Count at hourly intervals.

DECLASSIFIED

[redacted]

DETERMINATION OF TOTAL RARE EARTHS AND YTTRIUM

Ceric Fluoride Method

PRINCIPLE AND LIMITATIONS OF METHOD:

The rare earths are precipitated as fluoride using a cerium carrier. The zirconium is removed by complexing with the hydrofluoric acid. Barium and strontium are removed by precipitation of the rare earths as hydroxides in the presence of a strontium holdback carrier. The hydroxide precipitate is then plated and counted.

This method was adapted from a fission product procedure and was not designed primarily for the analysis of reactor effluent water.

REAGENTS:

1. Ammonium Hydroxide, concentrated.
2. Boric Acid Solution, saturated, aqueous.
3. Carriers (cerium, strontium), 10 mg/ml standardized.
4. Hydrochloric Acid, 6N.
5. Hydrofluoric Acid, 27N.
6. Nitric Acid, c.p. concentrated.
7. Nitric Acid, 4N.
8. Sodium Hydroxide, 24%

   Dissolve 240 g c.p. sodium hydroxide in 1000 ml of water.

PROCEDURE:

1. Place a 50 ml sample of reactor effluent water (concentrate if necessary) in a 100 ml lusteroid test tube.
2. Add 2.00 ml of cerium carrier, 4 ml of zirconium carrier and 15 ml of concentrated nitric acid.
3. Add 2 ml concentrated hydrofluoric acid, stir and centrifuge.
4. Decant supernate and dissolve the precipitate in 2 ml saturated boric

~~SECRET~~

DECLASSIFIED

acid solution and 1 ml of concentrated nitric acid. Add 2 ml zirconium carrier.

5. Dilute to 10 ml, add 2 ml concentrated hydrofluoric acid, stir and centrifuge.
6. Decant supernate, dissolve the precipitate in 2 ml of saturated boric acid solution and 1 ml of concentrated nitric acid.
7. Make the solution basic with sodium hydroxide, centrifuge, and wash the precipitate twice with water.
8. Dissolve the precipitate in 1 to 2 ml 6N hydrochloric acid and add 2 ml strontium carrier.
9. Dilute to 15 ml, make the solution basic with ammonium hydroxide, and centrifuge.
10. Decant the supernate, dissolve the precipitate in 1 to 2 ml 6N hydrochloric acid. Dilute to 15 ml.
11. Make the solution basic with ammonium hydroxide, centrifuge, wash the precipitate twice with water and mount on a stainless steel plate. Ignite. Measure the counting rate hourly.