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GENERAL ELECTRIC

HANFORD ATOMIC PRODUCTS OPERATION – RICHLAND, WASHINGTON

DATE

May 6, 1957

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TITLE

MONTHLY REPORT
APRIL 1957
CHEMICAL RESEARCH & DEVELOPMENT OPERA

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CHEMICAL RESEARCH AND DEVELOPMENT
MONTHLY REPORT
APRIL 1957

Compiled by Members of

Chemical Research, Chemical Development, Analytical Laboratories,
Engineering Development Planning, Chemical Effluents Technology

Submitted by: V. R. Cooper

May 6, 1957

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1st REVIEW-DATE: 3/6/81
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PR Eick 4-25-00

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

DOC-1973/Re-Reviewed
J E Savely
10-6-99

Reviewed and Approved for
Public Release by the Hanford
Declassification Project
D Briggs PNNL ADD
9/8/99 Date

HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

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Chemical Research & Development Operation

ORGANIZATION AND PERSONNEL

The planned interchange of personnel between Research and Engineering (CPD) and Chemical Research and Development (HLO) was extended to include eleven additional pairs through the balance of 1957.

Daisy C. Smith transferred into Analytical Laboratories Operation as a Laboratory Assistant from Exposure Records and Evaluation.

Carolyn L. Leinberger transferred into Analytical Laboratories Operation as a Counting Room Assistant from Exposure Records and Control.

Jack L. Welch was reactivated as an Engineering Assistant in Chemical Development Operation.

TECHNICAL ACTIVITIES

2000 PROGRAM - PRODUCTION OF FISSIONABLE MATERIAL

IRRADIATION PROCESSES

Reactor Effluent Studies

It is currently recommended that no reactor purges be made directly to the river when its temperature is above 15 C. It tentatively appears that the control of phosphorus-32 may be adequate if a limited number of purges are employed. The uptake by whitefish is not expected to be significantly increased by such action. Final recommendations will require the concurrence of Biology and Radioprotection Operations.

Direct measurement of isotopic content of reactor cooling water by gamma spectrometry was delayed by instrumentation problems. Arsenic-76 is now determined directly. Determination of Scandium-46 and Copper-64 has been slowed for lack of a large (3 inch) scintillation crystal

REACTOR EFFLUENT STUDIES

Process Assistance

Increases in amounts of radioisotopes discharged to the river have caused some concern that a production-limiting concentration may be approached in the not too distant future. Months of highest concentration are November through March. Low river flow during the winter period has contributed directly because of lowered dilution factors. As^{76}, Np^{239}, Rare Earths + Y, and the fission products in reactor effluent water have also increased during this period indicating that lowered dilution factors were not solely responsible. Of significance is the higher river turbidity reached during later 1956 and early 1957. This required increases in alum and filter aid to maintain cooling water quality. Increases in reactor power level during the interval also contributed to the observed increase. During this period also cooling water pH was decreased. Some data are available which indicate increases in effluent concentrations with decreasing pH. Although data are not available for quantitative determination of increases in reactor effluent isotopes from changes in power, river flow, turbidity and pH. They may well be responsible. Further study of this problem is underway.

Laboratory experiments with 107 Basin water and sludge were completed to investigate the effect of variables such as pH and concentration of Turco 4306-B, a decontaminating agent, on the desorption of Cs^{137} and Sr^{89-90} from the sludge. Conclusions from this study will be formulated soon.

Automatic Analyzing Monitor

Widths of channels to be used for Mn^{56}, As^{76}, and Na^{24} appeared to be satisfactorily stable, however, some drifting of the channel width for the low energy Np^{239} peak caused some concern, since Compton contribution in this region from higher energy emitters could markedly influence the counting rate of the Np^{239} channel. Increasing the channel width will improve the situation, but variations in channel width may well be the limiting factor on accuracy of the Np^{239} measurement.

Improvements in electrical and electronic features were incorporated including provision for manually advancing stepping switches, more convenient calibration of the count rate meter, minimizing sparking of the toothed-wheel digitizer, and the incorporation of a new interval timer to establish the length of time allowed for the recorder to read data on the Streeter-Amet printer.

Necessary improvement in the continuous removal of interfering radioisotopes from the P^{32} stream of the monitor was not realized. An analysis of decay curves from previous runs showed that interference from As^{76}, W^{187}, and I^{133} had been as low as 10 per cent, indicating that under some conditions satisfactory performance in this respect should be attainable. High Si^{31} initially in the sample will contribute significantly to the P^{32} count made after 26 hours, and this was accounting for a portion of the earlier observed high values for P^{32}. The contribution from P^{32} can be reduced through a longer decay period, or a computed correction can be applied to the P^{32} count. A detailed chemical study of the mode of removal of arsenic will be necessary to prescribe exact conditions to insure efficient removal of As^{76} from the P^{32} stream. Investigations on the separation procedure will be continued by the Analytical Chemical Operation.

Analytical Chemical

Determination of plutonium in normal solid and I&E type fuel elements will be made by coulometric titration. Some problems have developed in maintaining plutonium valance state. The shielded equipment for gravimetric determination of Pu is scheduled to be in place by August

Measurement of properties of organic coolant is undergoing improvements which should improve the flow of information. It was observed that about one-half of the tars formed in MIPB have melting points above 240 C vs. the assumed 220 C on the assumption that tesphenyl would have the highest melting point.

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SEPARATIONS PROCESSES

PUREX

Phase II Flowsheet

The proposed Purex Phase II Flowsheet* for the HA Column was tested in a dual-purpose glass pulse column. The extraction section was 3-inch diameter by 9-foot high and contained a nozzle plate cartridge. The scrub section was 4-inch diameter by 9-foot high and contained a mixed plate cartridge (4 stainless steel - 2 plastic sieve plate configuration). Except for the omission of louver plates and the use of fluorothene rather than polythene as the plastic plate material, the cartridge were identical to those specified for the plant HA Column.

Extraction Section. Flooding frequencies for the extraction section with the organic phase continuous were not determined since the scrub section controlled column flooding. With the aqueous phase continuous, the extraction section controlled column flooding, and flooding frequencies of 105 ± 5 and 85 ± 5 cycles/minute at capacity factors of 1.8 and 3.6, respectively, were obtained. Increasing the column temperature to 45 ± 5 C increased the aqueous phase continuous flooding frequencies by about 15 per cent.

Uranium waste losses ranged from 0.01 to 0.04 per cent, which is approximately the same as obtained with Purex Plant Flowsheet IIA. The choice of continuous phase did not appear to influence column efficiency.

Scrub Section. Both flooding and efficiency runs were made under Phase II Flowsheet conditions. Scrub section efficiency runs were also made with:

(a) The secondary organic stream (HASO) off.
(b) The aqueous backcycle (3WB) off.
(c) Both HASO and 3WB off (equivalent to Purex Plant Flowsheet IIA conditions).

The results are summarized below:

1. With the organic phase continuous the scrub section controlled column flooding. Flooding frequencies of 105 ± 5 and 80 ± 5 cycles/minute at capacity factors of 2.4 and 3.5, respectively, were obtained. Increasing the temperature to 45 ± 5 C increased the flooding frequencies 15 per cent. Scrub section flooding frequencies were not determined for aqueous phase continuous operation since the extraction section controlled column flooding.

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* Nomenclature used in this report approximates that used in HW-47889, "Purex Phase II Proposed Flowsheet," W. H. Swift and E. R. Irish, 2/1/57. For example, 3WB refers to the backcycled waste stream introduced below the feed point in the HA Column. On the otherhand, the flowsheet calls for two organic streams to be introduced above the feed point. These are the IBSU stream (the organic effluent from the IBS Column containing 0.065 $\underline{M}$ UNH) and the HAO stream (fresh organic stream from the organic header). For the purposes of the study reported here, these two organic streams were considered as one stream, of HAO composition (no uranium), flow equivalent to the sum of the two streams, and called HASO in this report.

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2. A good "zebra" effect (alternate phase inversion) was obtained under Phase II Flowsheet conditions. With the HASO stream off, the zebra effect was apparent, but the dispersion - especially, in the middle three-foot section - was considerably coarser. With the 3WB stream off, only a slight and erratic tendency toward a zebra effect was noted. With both streams off (equivalent to Purex Plant Flowsheet No. IIA conditions), the zebra effect was limited to the upper two-thirds of the scrub section.

3. The scrubbing efficiencies (measured by chloride transfer) obtained under Phase II Flowsheet conditions were approximately the same as obtained under Purex Plant Flowsheet No. IIA conditions. The efficiency was reduced by approximately 25 per cent by eliminating the backcycle (3WB). No appreciable effect was noted by eliminating only the HASO stream.

Permanganate Addition to Phase II Flowsheet

Since permanganate oxidation has been considered as one potential scheme to achieve neptunium recovery in the Purex Process, two runs were made in the equipment described above to observe the effect of potassium permanganate addition to an organic phase continuous HA Column. Purex Phase II Flowsheet was used with the following modifications:

(a) The HAS contained 0.02 $\underline{M}$ $KMnO_4$.
(b) 0.02 $\underline{M}$ Fe^{++} followed by 0.03 $\underline{M}$ $KMnO_4$ was added to the 3WB.

No adverse effect on phase dispersion was noted. The bulk of the manganese dioxide formed remained with the aqueous phase and was discharged from the bottom of the column. Some entrainment in the HAP was noted, however, causing a light brown color which disappeared after approximately 2-1/2 hours.

In the second run, the column was operated until steady state operation was reached. The addition of the HAS containing potassium permanganate was then stopped and regular HAS was started to determine if the manganese dioxide would scrub out of the organic and the length of contact time necessary. The organic phase in the cartridge section started clearing immediately, and the organic phase in the disengaging section was essentially free of manganese dioxide in approximately 15 minutes.

Organophosphorus Complexing Agents

The compounds ethyl dibutyl phosphonoacetic acid, dibutyl phosphonoacetic acid, butyl phosphonoacetic acid, and phosphonoacetic acid have been prepared and purified, and their properties as complexing agents for metal ions are currently under investigation. The physical properties of these compounds are summarized in Table I.

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TABLE I

PHYSICAL PROPERTIES OF PHOSPHONOACETIC ACID AND ITS ESTERS

	Boiling Point or Melting Point (deg C/mm Hg)	Density (g/cc)	Refractive Index (n_D^{25})	Solubility in H_2O (g/l)	Solubility in CCl_4 (g/l)	pK_a
1. Ethyl dibutyl phosphonoacetic acid $(BuO)_2P(O)CH_2C(O)(OEt)$ ("EDBPA")	121/0.85	1.0321	1.4340	-	-	-
2. Dibutyl phosphonoacetic acid $(BuO)_2P(O)CH_2COOH$ ("DBPA")	dec. > 50C	1.102	1.4444	17	-	3.4
3. Butyl phosphonoacetic acid $(BuO)(HO)P(O)CH_2COOH$ ("BPA")	dec. > 50 C	1.259	1.456	ca.250	0.2	2.1
4. Phosphonoacetic acid $(HO)_2P(O)CH_2COCH$ ("PA")	m.p. 143.4 C	--	--	984	>0.01	2.1

The completely esterified compound, 1, extracts uranyl nitrate in a manner similar to TBP. Compound 2, dibutyl phosphonoacetic acid forms salts which are organic soluble, the extraction reaction exhibiting the expected negative hydrogen ion dependence. Its behavior towards uranium and zirconium has been reported previously. Qualitative tests indicate that it forms organic soluble complexes with many ions, including cobalt, copper and uranium(IV). While cobalt and copper require a pH of about four for appreciable extraction (using 0.1 molar dibutyl phosphonoacetic acid in carbon tetrachloride), uranium(IV) extracts at a pH as low as 1. Table II shows typical extraction data for uranium(IV).

TABLE II

EXTRACTION OF 0.1 M UCl_4 WITH DIBUTYL PHOSPHONOACETIC ACID IN CCl_4

	E_a^0 U(IV)	
Conc. DBPA M	Aq. Phase pH = 1.0	Aq. Phase pH = 1.3
0.05	0.082	0.14
0.1	0.23	0.38
0.25	0.46	1.4
0.5	0.85	2.8

The extraction increases as the hydrogen ion concentration decreases as expected, and the extraction of uranium(IV) appears to be greater than that found for uranium(VI). However, the most interesting observation is that the uranium(IV)

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polymer is the species that is most highly extracted. The spectra of the organic phases obtained at high pH values (and high E_a^o values) lack the peaks characteristic of uranium(IV) and resembles that of the aqueous polymer. Further, the saturated organic phase is found to contain approximately a two to one mole ratio of uranium(IV) to DBPA.

The terminal hydrolysis product of the series, phosphonoacetic acid might be expected to form aqueous soluble complexes (or precipitates) as does phosphoric acid, with the added factor of possible chelate formation. Complexing of zirconium by this compound was studied by following its effect on partition of zirconium into TTA. A concentration of 0.1 molar TTA in benzene was used to extract 10^{-3} molar zirconium previously prepared by TTA extraction. The addition of phosphonoacetic acid strongly complexed the zirconium as the data in Table III show.

TABLE III

COMPLEXING OF ZIRCONIUM(IV) BY PHOSPHONOACETIC ACID
Organic phase 0.1 M TTA in benzene
Aqueous phase 5 M NO_3^-, H^+ as indicated

	E_a^o (Zr)	
M H⁺	No PA	0.1 M PA in Aq. Phase
0.5	1650	0.04
1.0	450	0.04
2.5	50	0.05
5.0	8	0.01

No complexing experiments have been performed with the intermediate ester, butyl phosphonoacetic acid.

NEW PROCESSES

Chemical Dejacketing of Zircaloy Clad Fuels

The effect of major variables, temperature concentrations, time, and pH as they effect the dissolution of zircaloy in NH_4F solutions have been investigated. The applicability of the process to separations operations appears favorable. An interim report has been issued summarizing the experimental studies, "Dejacketing of Zircaloy Clad Fuel Elements with Ammonium Fluoride Solutions," HW-49633 by J. L. Swanson. Further studies will provide quantitative information on the distribution of U and Pu into the cladding waste solution as well as possible steps for holding these quantities at desired levels.

Flurex

During the month a "trench-cathode" Flurex cell was completed and tested. Exposed cation and anion exchange membrane surfaces are 11-inches high and 2-inches wide with the feed chamber between about 1/2-inch thick. The anode is a platinum plate parallel to and about 1-1/4 inch from the anion membrane. The cathode consists of three mercury-filled trenches situated one above the other and parallel to the cation membrane. Mercury level in the bottom trench is about flush with the bottom of the cell. All three trenches are provided with plexiglass stirrers. This particular cathode arrangement was used in order to obtain a more uniform current density in the membranes than is possible with a single mercury-pool cathode in the bottom of the cell.

A special feed cell divided into five compartments was used in runs made to study the membrane current density distribution when using all or only one of the cathode trenches. Using solution compositions approximating those anticipated for a production Flurex cell, current density through the top feed compartment was 42 per cent of that through the bottom compartment when only the bottom trench was used as cathode. When all three trenches were used variation in the current density through the five feed compartments was generally less than 10 per cent. Membrane current densities observed agree well with calculated values based on solution and membrane conductivities reported previously. These observations lend confidence to calculations of cell performance vs. cell geometry based on measured conductivity of individual cell components.

Observed current efficiency (based on dried NH_4UF_5 product) for the trench-cathode cell was about 45 per cent when the mercury was not stirred compared to 93 per cent when stirred. These results demonstrate again the need for renewal of the mercury surface. Future runs in the cell will determine the degree of agitation required and will test several proposed methods of obtaining agitation.

Performance of the trench-cathode cell has been highly satisfactory and it is anticipated that some form of trench cathode will be included in design of the 321 Building pilot-plant-scale Flurex cell.

As part of a program aimed at determination of membrane useful life under operating conditions, the effect of time, temperature, and solution composition on pertinent properties of the membranes is under study. Initial experiments involved exposure of anion membranes (Nalfilm 2 and Permutit 3148) to nitric acid of various concentrations simulating possible anolyte compositions (0.1, 0.5, and 1.0 M). The solutions were maintained at 90 C and conductivity, physical dimensions, and permselectivity of the membranes were determined at various exposure times. Observations made after 80 hours exposure may be summarized as follows:

1. Resistance of the Permutit membrane decreased with exposure time to all three solutions, becoming about half its original value. Also the degree of permselectivity under a given set of conditions decreased with exposure to all three solutions, dropping from 0.9 to about 0.75. (Permselectivity is defined as the fraction of the current carried through the membrane by the anion, in the case of the anion membrane; and as the fraction of the current carried through the membrane by the cation, in the case of the cation membrane.) Change in area of the film was nil although an increase in thickness of about 10 per cent occurred.

2. Dimensional changes in the Nalfilm 2 membrane were absent. In 0.1 M HNO_3, resistance decreased to about 25 per cent of original while in 0.5 and 1.0 M HNO_3, resistance increased to nearly 10 times the original. Permselectivity dropped from 0.90 to about 0.55 in 0.1 M HNO_3 and to about 0.7 in 0.5 and 1.0 M HNO_3. Extended use of this film in HNO_3 at elevated temperature appears impractical due to increased resistance and/or decreased permselectivity.

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Attempted permselectivity studies with Ionics-CR-61 cation membrane revealed that the resistance of this membrane increases rapidly in use to a value many times the initial value under operating conditions approximating those for Flurex. Similar difficulties occurred in attempts to obtain current efficiency data for Nalfilm cation membrane. No explanation for this unexpected behavior of these membranes has been found. Similar resistance variations have not been observed with Permutit cation membrane.

Transfer of water through the anion membrane of a Flurex cell is dependent on the concentration of uranium in the feed compartment and ranges from 0.17 to 0.35 g H_2O per 1000 coulombs of current passed as uranium concentration is decreased from 1.1 to 0.1 molar.

CONTINUOUS METAL DISSOLUTION

Several runs were completed during the month in the two-inch-diameter continuous countercurrent tower dissolver using 60 per cent nitric acid as feed. Preliminary results are summarized in the following table.

Feed = 60% HNO_3 + 0.005 $\underline{M}$ $Hg(NO_3)_2$

Product Composition			Type of Slugs	Diss. Rate Lb. U/Day/Lb. Heel
UNH, $\underline{M}$	HNO_3, $\underline{M}$	HNO_3/UNH		
2.3	4.0	1.7	Unjacketed	1.6
3.0	1.7	0.6	Unjacketed	1.6
3.5	1.1	0.3	Unjacketed	1.2
2.3	3.6	1.6	Jacketed	1.8
2.4	2.5	1.1	Jacketed	1.1
2.5	2.0	0.8	Jacketed	0.8

Reduction of the residual acidity in the product appears to have a larger adverse effect on the dissolving rate of jacketed slugs than of unjacketed slugs. The results are encouraging and indicate high dissolution rates if some acid can be tolerated in the dissolver product. Further tests at lower residual acidity are planned.

CAUSTIC SCRUBBER STUDIES

The six-inch-diameter bubble cap caustic scrubber with six trays on two-foot spacing was operated to measure efficiencies of iodine removal. A concentration of 500 ppm iodine in air was used as the feed vapor. Efficiencies greater than 99.85 per cent were obtained consistently with 25, 15, and 5 per cent caustic scrub solutions. No decrease in efficiency occurred when synthetic coating waste solution was substituted for sodium hydroxide solution.

After 168 hours of continuous operation at nominal plant rates, no plugging of equipment or lines was detected. Analytical results, though incomplete, indicate almost complete conversion of the sodium hydroxide to sodium bicarbonate and carbonate.

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Improved Reactivity - UO_3

Among the various methods investigated for increasing the reactivity (in the fluorination process) of UO_3, the most effective treatment developed so far involves fluidized-bed reduction of the trioxide with hydrogen to produce UO_2 which is then reoxidized with air in another fluidized-bed to produce U_3O_8. This material made in this manner has satisfactory reactivity regardless of the quality of the starting UO_3 material. A study was initiated to establish the magnitude of the economic incentive for carrying out pilot-scale development work for such processing. Capital expenditure at Hanford would amount to about $450,000 after being credited with construction expenditures not required at Paducah. Annual operating cost savings of $40,000 (Paducah savings less Hanford operating costs) were shown. In the report of this study, HW-50081*, it was concluded that a full-scale process could not be justified at this time but that consideration should be given to pilot-scale development without priority over present programs.

Anion Exchange Process for Plutonium Recovery

An informal report, HW-49524, entitled, "Tentative Flowsheet for Recovery and Concentration of Plutonium by Continuous Ion Exchange," by J. L. Ryan, was issued to guide 234-5 Development Operation in demonstration of this process in the laboratory-scale continuous ion exchange unit. The flowsheet uses a feed solution containing 7.2 molar nitric acid and operating at a flow rate of 28 mg Pu/cm^2/min. The elutant is 0.35 molar nitric acid at a flow rate designed to yield a product concentration of 50 g Pu/l. The operating temperature of both extraction and stripping columns is 50 C.

Dissolution of Uranium by Bismuth Bromide

Preliminary experiments were conducted to determine the reaction characteristics of uranium and bismuth bromide, a reaction proposed by R. H. Moore in HW-48503 for the pyrochemical dissolution of uranium. The details of this brief study may be found in HW-49731, "The Reaction of Uranium and Bismuth Bromide - A Preliminary Investigation" by H. T. Hahn. The following conclusions were drawn: (1) the reaction occurs rapidly in the vicinity of 400 C with a dissolution rate greater than 0.32 grams per minute per square centimeter, (2) the product is a solid at the reaction temperature when the ratio of bismuth bromide to uranium is 1.6 to 1 or less; the conditions for product solubility are unknown, (3) the reaction apparently produces uranium tetrabromide rather than the tribromide, and (4) the reaction proceeds smoothly without violence and could undoubtedly be adapted to the large scale dissolution of uranium metal.

WASTE TREATMENT AND BY-PRODUCT RECOVERY

Neptunium Recovery

Data have been obtained on plant samples which define the path of neptunium in the Purex plant. They show that the fraction of Np appearing in the uranium product

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*HW-50081, "Uranium Oxide Activation Cost Study" by R. W. McKee, April 23, 1957.

[redacted]

since the first of the year is sensitive to solvent characteristics. Correlations of plant operation with laboratory experiments support a hypothesis that the extractibility of neptunium in the plant operation is due to degradation of the solvent to form compounds which preferentially extract neptunium.

Analyses made of the major Purex streams, and the results for samples taken on three different dates are shown in Table V.

TABLE V

DISTRIBUTION OF NEPTUNIUM IN THE HANFORD PUREX PLANT

Stream	Percent of Feed Neptunium		
	March 22	April 5	April 12
1AF	64	82	NA*
1AW	11	4	4
2DF	--	[illegible]1	39
2DW	--	41	19
2EUC	19	10	25
1BP	--	Negligible	NA*

* Data not currently available

It is noted that a major fraction gets past the HA-HC cycle. It then splits into essentially all of the aqueous streams. However, only negligible amounts are found in the 1BP or in the plutonium product, as would be expected based on the extractability of neptunium(IV) and the high nitric acid concentration in the 1B column.

Variation of the concentration of neptunium in the uranium product with time is shown in Table VI. The dates given are equivalent Purex dates, i.e., in the case of the car lot samples these are the date of oxide sampling less five days. On the basis of an average exposure of 650 MWD/T and a value of 0.0036 for the Np/Pu ratio, the calculated feed neptunium is equivalent to 1.5 grams per ton of UO_3. The percentages of feed neptunium in the product are based on this value and are only approximate. The three samples of 2EUC are essentially "grab samples" and would be expected to show more variation than the oxide samples.

TABLE VI

NEPTUNIUM IN PUREX URANIUM PRODUCT AS A FUNCTION OF TIME

Date	Car No.	gm Np^{237}/T UO_3	Percent Feed Neptunium
1/31	16	0.18	12
2/4	17	0.05	3
2/21	509	0.51	34
2/28	510	0.84	56
3/5	511	0.82	55
3/11	512	0.70	47
3/17	22	0.87	58
3/18	513	0.75	50
3/22	2EUC	0.3	20
4/5	2EUC	0.2	13
4/2	2EUC	0.4	27

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It will be noted that the concentration of neptunium in the uranium product increased markedly about February 21, remained high until March 18, and then declined. This behavior coincided with a change in the solvent wash procedure. During the period February 7 to March 17, the "front end" (IOO) solvent received a batch carbonate wash followed by a two per cent nitric acid scrub in the IO column. Prior to this time, a double carbonate wash was employed. This was reinstituted on March 18 and is still being used. A trend toward higher plutonium losses and poorer ruthenium decontamination was also noted during use of the nitric acid treatment, and was one reason for its abandonment. These results strongly suggest that some solvent degradation product is responsible for all of these phenomena, and the following laboratory experiments support this view.

A 30 per cent TBP-Shell Spray Base solvent was degraded by digestion for 48 hours with 6 M nitric acid containing 0.1 M nitrite, and the extraction coefficients of neptunium (IV), (V), and (VI) into this solvent from 2 M nitric acid were then determined both at room temperature and at 45 C, the approximate current temperature of Purex plant operation. The results are shown in Table VII with the corresponding values for untreated solvent in parenthesis. The extractability of both the (IV) and (V) states is increased by factors of 6 to 30, whereas the (VI) is not appreciably effected. In other experiments, carbonate washed plant IOO solvent gave neptunium(V) extraction coefficients from 2 M nitric acid of about 0.1 and 0.45 at room temperature and 45 C, respectively, and 2OO plant solvent gave 0.1 and 0.3. These numbers can be compared to values of about 0.02 with laboratory solvent and are adequately large to account for the observed plant behavior of neptunium. Dibutyl phosphate, on the other hand, when present to the extent of 10^{-4} M did not appreciably effect the extraction of either neptunium(IV) or (V), suggesting that degradation of the diluent is responsible for the observed effects.

TABLE VII
EFFECT OF DEGRADED SOLVENT ON NEPTUNIUM EXTRACTION

30% TBP-Shell Spray Base degraded by 48 hours digestion at 70 C with 6 M HNO_3-0.1 M nitrite. Extraction coefficients from 2 M HNO_3.

Neptunium Oxidation State	E_a^o	
	R.T.	45 C
IV	13 (1.7)*	7 (1.1)
V	0.4 (0.02)	0.6 (0.02)
VI	8 (12)	6 (9)

* Parenthetic numbers are controls with untreated solvent.

Workers at the Savannah River Plant have attributed the extraction of neptunium to a nitrite catalyzed nitric acid oxidation of neptunium(V) to (VI). In an experiment to check this idea, a 2 M nitric acid solution containing 0.1 M nitrite was agitated with 30 per cent TBP-Shell 2342 and the distribution coefficients of neptunium (initially V) followed as a function of time. At 45 C, the gross neptunium extraction coefficient increased from a value of 0.34 at ten minutes to 0.83 at one hour versus a constant value of about 0.02 in the absence of nitrite. The effect at room temperature was less marked. While these results taken by themselves, would seem to indicate oxidation to the extractable (VI), an explanation more consistent with the other work would be that the nitrite is bringing about a degradation of the organic phase.

Thus, increasing the temperature from 25 to 45 C was found to decrease the extraction coefficients of neptunium(IV), (V), and (VI) by about one-third when employing laboratory solvent. The presence of sulfate in the aqueous was found to markedly decrease

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the extractability of neptunium(IV) and would seriously interfere with any solvent extraction process based on extracting neptunium(IV). The concentration of sulfate should be kept as low as possible.

Manganese dioxide, particularly co-formed, has been previously reported to be effective for oxidizing neptunium to the (VI) state, and column evaluations are in progress. Additional experiments have now shown that sodium bismuthate and lead dioxide are equally effective, as was sodium bromate (previously evaluated at ORNL and rejected for corrosion reasons). Persulfate was ineffective due to unfavorable kinetics.

Appraisal of the situation leads to the view that the use of an oxidizing agent in the first solvent extraction cycle is the most positive means for forcing the Np to accompany the Pu and U at this point. It is relatively well established that conditions in the partition column (1B) will drive the Np to the IV state and that it will follow uranium to the second solvent extraction cycle. Introduction of sulfate at this point is expected to force the Np with second cycle waste. This waste can be processed through ion exchange to separate Np. The balance of the waste will be back-cycled to the first solvent extraction cycle.

Ion exchange studies have shown that semicarbazide is a satisfactory reducing agent for neptunium in high acid (8 M) solutions and that it gives almost as high distribution ratios onto Dowex-1 as are obtained with ferrous sulfamate. Column runs are in progress but are limited to a very small scale inasmuch as only 100 milligrams of neptunium-237 are available. Reasonably good breakthrough and elution curves have been obtained in columns containing 0.1 ml of resin; however, there is evidence that channelling and kinetic factors in these small columns may cause erroneously low apparent capacities.

An interim status report on the neptunium recovery work is in preparation and will soon be released as HW-49825.

Concentration of Np-237 in Irradiated Uranium

Analyses of Purex feed (HAF) solution from 730 MWD/T metal yielded a Np-237/Pu ratio of 0.004. An uncertainty of as much as ± 20 per cent is inherent in the counting statistics. However, the value agrees well with 0.0036 computed from nuclear crosssections and a more rigorous calculation by the Theoretical Physics Operation.

WASTE TREATMENT STUDIES

Cesium Isolation and Packaging Facility

The first engineering flowsheet defining a batch-type calcination process to convert an aqueous slurry of zinc-cesium-ferrocyanide to a dry cesium chloride product was prepared. Laboratory experience has shown that the presence of nitrate in the influent slurry introduced a potential hazard to the calcination step. The flowsheet will be modified to eliminate nitrate before calcination.

Semiworks Waste Self-Concentrator

During the month, 378 liters of condensate from the boiling waste concentrator were collected and cribbed, reducing the liquid level in the tank to approximately 9-1/2 feet. This corresponds to 22 per cent of the original volume charged to the tank. (If tank contents were homogeneous, sodium ion concentration would be approximately 23 M).

No pressure "bumps" have been recorded since March 21. However, the condensate rate has evidenced a definite boiling cycle. Every 12 to 18 hours, the condensate rate peaks at about 24 ml/min. with the 1/2-inch vapor valve open and at about 50 ml/min. with the one-inch vapor valve open. Between peaks, the condensate rate ranges from 0 to 6 ml/min. The tank temperature profile also reflects the 12 to 18 hour cycle.

During the last month, tank temperatures below the liquid surface have steadily gained from 13 to 17 F, the largest gain occurring at the bottom. The actual temperature range from 259 F one foot beneath the liquid surface to 302 F six inches off the tank bottom.

An attempt was made to determine the liquid and sludge consistency by turning the manual agitator paddles which were installed in the tank before filling. Normal friction in the system made it impossible to detect any difference in the torque required to turn the top four paddles. Three of these paddles are immersed in liquid while the upper one is "free" in air. Evidence that the sludge has set up like "cement" was obtained when a fifth agitator in the sludge layer (approximately one foot off tank bottom) was "frozen" solid. The paddle did not break free when a force great enough to break the 9/16-inch stainless steel rod was applied. Calculations indicate that a force of greater than 500 lb./sq.in. was exerted on the sludge by the paddles. Although three of the agitator paddles were beneath the liquid surface, no significant changes in temperature, pressure, or boil-off rate were noted as a result of turning the paddles. Actual speed of the paddles was a jerky one to two revolutions per minute for two to five minutes.

Special Geological Studies

Drilling on the fixed-price portion of the CA-700 (FY-1957) Chemical Effluents Technology drilling program by the Hatch Drilling Company began April 8th. In approximately two weeks of drilling three wells were completed and two more under way, about two weeks ahead of the contract requirements. The Geological Survey drillers bettered their drilling progress and are currently less than 150 feet behind schedule. Probable completion of the CA-700 project for an estimated cost 28 per cent less than originally scheduled is attributed to competitive bidding, to improvements in drilling techniques as employed by fixed-price contractors, and to the successful efforts of the Geological Survey drillers to increase performance and decrease costs.

Data recently procured further indicate the desirability of testing the use of rotary drilling equipment at Hanford. Savings up to one-half the present costs appear possible, moreover drilling rates up to five-times the present speeds are indicated. Negotiations between the Atomic Energy Commission and Geophysical Service, Inc. of Dallas, Texas, for the proposed geophysical evaluation study continued. Evaluation of the rotary drilling techniques as used by that company will further determine the desirability of use of rotary equipment here.

Well 299-W15-5, drilled to basalt at a depth of about 530 feet, encountered subnormal, non-artesian water beneath the two clay horizons of the uppermost part of the lower Ringold formation for more than 100 feet of depth. The data, from the east side of a deep structural basin in upper Cold Creek Valley, confirm similar data obtained in 1948 in a well toward the west side of that basin nearly three miles away. The subnormal level indicates no appreciable recharge from deeper artesian aquifers in the basalt and no appreciable hydraulic continuity with shallower ground waters there. Storage of large volumes of wastes in this deep Ringold aquifer may be feasible.

Disposal to Ground

An investigation of the possibility of direct disposal of aluminum coating waste to the ground was started concurrently with the study of gelling of the material. The preliminary results of a soil column test with a sample of supernatant liquid (aluminum coating waste) from the 105 C tank were as follows: essentially none of the 7.5 $\mu c/ml$ of Ru^{106} was adsorbed by the soil, the concentration of Cs^{137} was reduced from 4.1 $\mu c/ml$ to $< 1.2 \times 10^{-4}$ $\mu c/ml$, and the concentration of Co^{60} was reduced from 1.2×10^{-2} $\mu c/ml$ to 2.1×10^{-3} $\mu c/ml$ at one column volume of effluent. Plutonium and uranium were adsorbed sufficiently to permit disposal to the ground, but the preliminary analytical results for Sr^{90} were not sensitive enough to evaluate the extent of adsorption of this radioisotope by soil. This test represented the first instance of Co^{60} adsorption by soil, however the adsorption was still insufficient to permit direct disposal to the ground based on present limits. Samples of aluminum coating waste from tanks 106-T and 109-U were obtained for additional experimental work.

A laboratory soil column test was begun with A-8 waste (Purex tank farm condensate) to estimate the "crib life" of the A-8 crib for the higher activity density wastes, which are being produced under present operating conditions. The activity density of current A-8 waste is approximately 100 times greater than that produced six months ago, however this is probably due, at least in part, to the reduced volumes of condenser cooling water.

Approximately one million gallons of TBP scavenged waste supernate having a Co^{60} concentration greater than 4×10^{-4} $\mu c/cc$ were discharged to the 216-BC trenchsite during the month. This brings the total volume of scavenged waste disposed on a specific retention basis at this site to 11.5 million gallons. Remaining unused trench space will accomodate 5 million gallons of non-cribbable supernate.

In-farm scavenging operations at C Farm were started on March 31. The first batch of settled supernate (9-112C-102C) was sampled and analyses should be available by the end of the month. Analytical and soil column testing results will determine the suitability of disposing of this waste to the 216-BC cribsite on a test basis. Chemical Processing Department was requested to delay disposals to the 216-BC cribsite until test wells can be drilled and hydrological testing performed before the wastes enter the regional ground water. An outline of proposed geological and hydrological tests and monitoring and analytical requirements concerned with the test disposal of high Co^{60} scavenged waste at the 216-BC cribsite was under preparation.

Recommendations were made to Chemical Processing Department for the disposal of 21,000 gallons of contaminated organic solvent stored at the Metal Recovery Plant. Disposal, on a specific retention basis, to a trench to be located west of U-Plant and immediately adjacent to a site which received organic waste several years ago was recommended.

Observation Wells

The addition of new monitoring wells in the vicinity of the BC cribs and trenches permitted a more detailed analysis of the ground water elevations in this area. This analysis revealed the presence of a small ground water mound beneath this site. This mound can only be caused by recharge of the ground water by liquids disposed to these facilities. The mound complicates the selection of monitoring well sites for the special disposal test proposed for these disposal structures.

Samples of ground water obtained from wells adjacent to the 216-S-1 and 2 cribs contained detectable concentrations of Sr^{90}. This is the first time Sr^{90} has been

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detected in the ground water at Hanford. These cribs received large volumes of D-1 and D-2 wastes from the Redox plant until they were replaced by the 216-S-7 crib over a year ago. The 216-S-1 and 2 cribs were abandoned when soil column tests of the wastes being discharged to the site demonstrated very poor strontium removal. The wastes were discharged at a low pH (2-4), conditions not suitable for strontium removal by soils; such waste streams are now neutralized before discharge to the ground. The ground water samples were found to have pH 7.5-8.0, at which strontium removal should be readily achieved. It is felt that the strontium contamination cannot move far in the ground water without being removed by the soil under these conditions. The highest Sr^{90} concentration found in the ground water was 1×10^{-6} μc/cc, or 2-1/2 times the Handbook 52 MPC for drinking water. The occurrence of radio strontium in the ground water at the measured pH indicates that the soil beneath the crib is still draining although no liquids have been discharged to the site for many months.

Process Development

The investigation of the use of ion exchange resins for the disposal of condensate wastes was continued. The extremely high affinity of the resins and the relatively low concentration (on a weight basis) of radioisotopes continued to interfere with attainment of reliable breakthrough data. Although such efficient removal of radioisotopes appeared at first glance to assure the successful application of ion exchange resins, preliminary estimates indicated that an extremely high resin column efficiency will be required to compete with disposal to the ground on an economic basis because of the deep soil profiles of the Separations Areas plateau. Samples of A-8 waste undiluted by condenser cooling water were obtained and additional Sr^{90} added. Experiments were begun to obtain breakthrough data for Sr^{90} using this waste. Preliminary attempts to elute Sr^{90} from Duolite C-3 resin indicated sulfuric acid or sodium chloride solutions as possible eluting agents. The sodium salt would be preferable since the resin then would be regenerated to the original sodium form during the elution step. Elution data will be necessary to determine whether it is more economical to discard or regenerate resin which has been loaded with radioisotopes from A-8 waste.

Gelling of Wastes - Field Work

It was demonstrated in the laboratory that a Vanton pump will provide sufficient mixing of a sodium silicate synthetic coating waste solution to form an aluminosilicate gel. The 1/3 gpm positive displacement pump provided suction on two containers, one containing 20 per cent sodium silicate, the other synthetic-coating waste. The solutions were blended at the pump inlet, mixed in the pump chamber, and discharged as a milky liquid which set to a gel in 2 - 3 minutes. Although there was no evidence of plugging in the lines or pumps, some difficulty was experienced in attempting to maintain flow control so that the solutions blended in a 2:5 silicate to waste volume ration.

Preliminary planning for field tests indicated that the most satisfactory method for mixing the streams and disposing of the gel will be realized by pumping each stream into a common mixing nozzle or chamber located near the end of the discharge line.

A four foot long, 6-inch diameter lucite column partially filled with soil and topped with a one foot thick layer of aluminosilicate gel is being set up to evaluate

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moisture loss to the soil due to compaction of the gel and to blotter effects exerted by the soil. Data obtained from this evaluation will be used to determine the length of column(s) necessary to evaluate loss of moisture from greater gel bed depths which would be encountered in plant scale disposals.

The economics of gelling vs. other methods of obtaining tank space was studied. Using actual costs of the waste scavenging process as a basis and applying the necessary adjustments it was estimated that coating waste may be gelled, discharged into an excavation, and covered at a cost of eight to nine cents per gallon. While being about three times as expensive as direct disposal to the ground, this process will be about two cents per gallon less expensive than evaporation and about eleven cents per gallon less expensive than tank storage.

ANALYTICAL AND INSTRUMENTAL DEVELOPMENT

Mass Spectrometry

Work was continued on the thermal emission mass spectrometer with most of the effort concentrated on the scanning method instead of the pulse counting method. Precision of the U-235 to U-238 ratios obtained by the scanning method was improved nearly ten-fold to about ± 1.5 per cent. The improved performance is attributed to the careful repositioning of the beam deflecting magnet which resulted in a narrower beam width of 10.5 mils. Additional improvements in the scanning method were made by modifying the magnet power supply to scan in both forward and reverse directions. An averaging of the results from both the forward and reverse scans eliminates the corrections for the gradual but noticeable change in the ion beam current with time. Further improvements in results will be realized when modifications to the micro-microammeter are made to remove the transients between range changes which tend to mask the peaks.

Controlled Potential Coulometry

It has been demonstrated that the plutonium concentration of a solution can be determined by titration from the three to the four valence state with a controlled potential coulometer. Preliminary precision measurements indicate that the method will be capable of obtaining the desired precision of ± 0.2 per cent (95 per cent confidence level). The range of 1 to 40 mg of plutonium has been investigated, and about four mg was chosen as the optimum sample size. Repeat titrations of a four mg sample (re-oxidation coulometrically in situ between titrations) resulted in a precision of ± 0.1 per cent, but titrations of separate samples showed a precision of ± 0.4 per cent. The optimum oxidation potential has been found to be ± 0.1 to 0.08 V with respect to a standard calomel electrode while the optimum reduction potential is about - 0.2 volts. These are the minimum potentials which will completely and reproducibly oxidize and reduce the plutonium in the citrate medium being used. Any substance which will reversibly change valence state between these two potentials will act as an interference to the estimation of plutonium by this technique.

Analysis by X-ray Fluorescence

The x-ray fluorescence method is useful for quantitative analysis of individual elements. The sample is irradiated by monochromatic 50-KV x-rays which are absorbed by each element and re-emitted as fluorescent x-rays having wave lengths characteristic of the atomic number. The wave length can be accurately measured with aid of a diffracting crystal and goniometer, and the intensity, which is a function of the concentration of the element, can be readily determined. With the present detector, an argon-filled Geiger-Mueller tube, and a three inch sodium chloride analyzing crystal, it was possible to determine elements in the range from iron (at. No. 26) to silver (at. No. 47) at a concentration of 50 mg/l with a precision of about ± 5 per cent. The elements from Hg (at. No. 80) to the end of the periodic table could be determined at a concentration of 100 mg/l with the same precision. Ten milliliter samples were used in these direct measurements; however, by evaporating the samples to dryness on a small disc of filter paper, a ten-fold increase in sensitivity was obtained, thus lowering the minimum determinable amount to 5 and 10 mg/l, respectively, for the light and heavy groups.

Contact Alpha Counter

The life test of a number of phosphors of different configuration is continuing. After five weeks, no major attack appears to have taken place, although some very small blisters have appeared on the Teflon surface of several of the phosphors. The small blisters only appear on the Teflon that is directly covering the zinc sulfide. The jig for making phosphors was modified to prevent the somewhat excessive flow of fluorothene previously experienced and to give a somewhat better dimensional stability to the phosphor material. Some 50 phosphors recently made with the modified equipment will be tested for uniformity of response to a fixed alpha source to evaluate the present method of phosphor production.

The proposed Purex Phase II Flowsheet calls for an 8 gram per liter plutonium concentration in the 2BP stream, which is higher than originally anticipated by a factor of four. Additional testing with the alpha counter covering this concentration range will be made since completed tests only include concentrations up to 6 grams per liter.

Interface Detector

A liquid interface detector-controller using a float-driven armature in a stationary coil has been fabricated for Facilities Engineering for prototype testing in the Purex 1A Column (Dwg. H-2-56729). The float and controller-transmitter unit were calibrated in the Purex Instrument Shop. The float has a 5-1/2 inch travel from zero to full scale and responds well to a 0.12 density difference between phases. Consideration is being given to prototyping a second unit in the IO column.

Plutonium Resin Column Controls

Reservoir Level Indicator. The resin level indicator has operated satisfactorily during plant cold runs using a cation exchange resin. Tests are underway to determine if the unit can be modified to operate using an anion exchange resin in the column. The conductivity difference between resin and solution with the two types of resins requires conductivity measurements of different orders of magnitude. The anion resin in the reservoir will probably be in equilibrium with 7 to 8 molar nitric acid, whereas the cation resin is in equilibrium with demineralized water.

"C" Column Acid-Water Interface Detector. The acid-water interface detector operated satisfactorily during plant cold runs with cation exchange resin. This instrumentation will also have to be reevaluated for use with an anion exchange resin. With cation resin, the instrument detects the difference between water and dilute acid, whereas with an anion resin it must detect the difference between two levels of rather concentrated acid.

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Absorptometer for Plutonium Loading on Resin. The absorptometer unit for detecting the concentration of plutonium on the resin was tested (with uranium) on the prototype column in the 321 Building. Preliminary tests revealed that the calibration data based on equivalent lead absorbers were in error by as much as 200 to 300 per cent. The unit was returned to the Laboratory for recalibration using uranium-loaded resin. The apparent discrepancy using lead absorbers will also be studied in greater detail but will require more time.

Purex AAA (F-3) Sampler Mock-Up

A design proposed by Facilities Engineering for the F-3 gamma monitor at Purex was tested for operability in the 321 Building. The design incorporates an "in-cell" degasser and jet. It does not require valves in the process lines and allows the sample cell to drain by shutting off the air to the jet. The design, as prototyped, was not operable. However, by changing line sizes and minimizing sample holdup volumes, the system could be made to operate satisfactorily. At the completion of these tests, the changes incorporated in the design will be recommended to Facilities Engineering.

Redox 1AFS (F-1) pH Prototype

As a result of piping changes, the buffer and rinse solutions now flow by gravity through the pH cell. A problem still remains with sampling the process stream which is thought to be due to restrictions in the valves in the sample manifold. Valves are being modified to improve their flow characteristics and will be used if their performance is acceptable for this service; otherwise, a valve-free manifold will be installed.

During periods of satisfactory sampling, the monitor gives pH data which compares favorably with the limited laboratory results.

EQUIPMENT DEVELOPMENT

Redox Spare Pump (Bingham Deepwell Turbine)

A spare deepwell turbine pump equipped with glass bearings operated about 200 hours pumping dilute simulated Redox 1AFS solution (Redox Flowsheet No. 5). The test was discontinued when the motor shaft seized. The glass bearings were in perfect condition. The ball bearings in the motor were frozen and the motor windings were burned. Failure was apparently caused by lack of lubrication in the motor bearings although grease was present in the housings above the bearings.

Valves

"Valvair" Solenoid Valve. This valve has operated 207,000 times (16 operations per minute) opening and closing against 83 psi air. This valve has given better service than other solenoid valves tested.

Hills-McCanna Saunders-type Valve. A spare Purex Plant Hills-McCanna Saunders-type valve was equipped with a Teflon coated neoprene diaphragm molded in the closed position. The Teflon and neoprene were badly cut and cracked after 230 forceful closures. Also, the threads on the small (8-32 threads) screw molded into the diaphragm were stripped. Super Dylan (Ziegler Polyethylene) diaphragms will be tested when they are available.

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Jamesbury Ball Valve. A pneumatically operated plug-type valve has operated for 120 hours (115,000 operations at 16 operations/minute). The valve was closing against 100 psig water requiring 20 psig air for operation. After 2800 operations closing against water containing ion exchange resin, the valve started sticking. The cylinder of the air cylinder was scored and neoprene "O"-ring" on the end of the piston was destroyed. The presence of ion exchange resin in the system did not necessarily cause the valve failure.

Materials of Construction

K-1047 Silicone Rubber, milled by Huntington Rubber Company from a raw gum manufactured by Dow Corning, was tested for chemical compatibility and the effects of irradiation on chemical compatibility.

This elastomer failed in 50 per cent caustic soda by hardening and spalling of the surface. It dissolved in Recuplex CAX and Purex HAX. Only minor shifts in these tendencies were caused by irradiation. Dosages of 10^7r and over caused some hardening. Immersion in 10 per cent nitric acid for 35 days seemed to make little change but immersion in distilled water for the same time caused a noticeable softening. There was no change in linear dimension noted on either of these two samples.

Immersion for 35 days in 60 per cent nitric acid caused a marked increase in hardness which decreased slightly with increased irradiation. There was little change in length noted, 12 per cent for the 10^8r sample, in 60 per cent nitric acid.

The samples tested in hexone and in carbon tetrachloride both swelled markedly, approximately 30 per cent in hexone and 40 per cent in carbon tetrachloride. This swelling was accompanied by a reduction in hardness. Both changes decreased upon increasing radiation dose.

LS 53 Silicone Rubber, milled by Huntington Rubber Company from a raw gum manufactured by Dow Corning, was tested for chemical compatibility and for the effects of irradiation on chemical compatibility.

LS 53 rubber failed in 50 per cent caustic soda by surface deterioration. The material dissolved in Purex HAX. It swelled severely and lost strength in Recuplex CAX and Hexone but there was no indication of dissolution. Changes in dimension and hardness in 10 per cent and 60 per cent nitric acid, carbon tetrachloride and distilled water were negligible. Radiation dosage up to 10^8r caused no noticeable changes in the chemical compatibility of this material but it did cause the material to lose tensile strength.

Seilon DP and Seilon Glare-Ez, transparent vinyl-type plastic sheet material manufactured by Seiberling Rubber Company was tested by static immersion at room temperature for 28 days. Both these materials successfully withstood immersion in 60 per cent nitric acid, 50 per cent caustic soda, carbon tetrachloride, Recuplex CAX, Purex HAX, and distilled water. The material delaminated after less than 24 hours in hexone.

Seilon DP is furnished in a standard green tint and Seilon Glare-Ez in a standard blue-grey tint. Crystal clear sheet is not available since tinting is required for light stability.

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Corrosion Studies

Corrosion Rates by Activation Analysis Techniques. Several stainless steel coupons irradiated for use in corrosion studies are now being exposed to various corroding media. Corrosion rates based on gross gamma determinations are two- to three-fold lower than rates determined from weight loss under similar conditions. This discrepancy is currently thought due to preferential dissolution of iron which has a lower specific activity than chromium in the irradiated samples. Gamma scan determinations now in progress may provide an explanation.

Redox Waste Storage. The first set of samples from the 107-SX tank was removed April 3, 1957. The operation went smoothly with the exception of the initial freeing of the lead radiation plugs. Apparently, corrosion products from the mild steel pipe had flaked off from above and deposited around the shoulder of the plug making removal of the plugs somewhat difficult. It is quite possible that future removals will be more difficult because of this. The samples were delivered to Radiometallurgy for examination.

Effect of Dichromate and Sulfate on the Corrosion of A55 Titanium in Nitric Acid. The corrosion rate of A55 titanium in boiling 65 weight per cent nitric acid was not affected by dichromate in concentrations from 0.01 to 5 g Cr/l. Sulfuric acid increased A55 titanium corrosion rates in boiling 65 weight per cent nitric acid by factors of 3 to 5 at 0.01 $\underline{M}\ SO_4^=$ and 10 to 20 at 1 $\underline{M}\ SO_4^=$.

Effect of Fe^{+3} on Corrosion of 304L Stainless Steel in HNO_3. Ferric ion had no effect on the rate of corrosion of 304L stainless steel in boiling 65 weight per cent nitric acid at concentrations from 0.01 to 1 g/l. At 5 g/l, the corrosion rate was increased by a factor of 7. The effect of ferric ion on corrosion of stainless steels by nitric acid is apparently similar to that of Cr(VI), i.e., there is a critical concentration below which corrosion acceleration is negligible.

Mock-up Heat Exchanger Studies. After 1.75 months exposure to boiling 60 weight per cent nitric acid at a steam temperature of 175 C, a 304L bayonet suffered a 14 per cent weight loss while an A-70 titanium bayonet lost only 0.035 per cent of its weight. Preferential attack near welds was very apparent on bayonets of 446, 310 Cb, and 16-2 manganese substitution stainless steels exposed to 60 weight per cent nitric acid at steam temperature of 145 C.

MISCELLANEOUS

Hot Semiworks Maintenance Activities

The maintenance program at the Semiworks is 85 to 90 per cent complete. Process piping work is 85 per cent complete.

The feed centrifuge, G-6, was dismantled, cleaned, assembled, and tested. Flush lines for improved decontamination were added. New thick walled (3/4-inch pipe) rings were placed in the No. 1 acid concentrator tower and revised packing supports were installed. The corroded rings were removed from the No. 2 acid concentrator tower and a layer of thick rings were added in the bottom section of the tower. New one-inch rings with a Huey test corrosion rate of 0.0015 ipm were used in the remainder of the tower. The wall cover skirt (secondary seal on the wall) was completed. This concludes the work on the stainless steel floor in A Cell except for

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final leak testing. Revisions on the remote connector pan for the A Cell pumps were completed. The unit was installed in the cell and repiped. The permanent lighting system for A Cell was placed in service. Improved visibility and safety has resulted.

Annular 2A Column. Fabrication of the annular column is proceeding on schedule. The contacting section will be ready for tie-in upon completion of the following items:

1. Fabrication and installation of feed distributors.
2. Fabrication and installation of samplers.
3. Fabrication and installation of "Plexiglas" windows for four observation ports.
4. Welding end flanges on 16-inch shell.
5. Final assembly.

Work is in progress on the first three of these items and all are expected to be finished on or before May 17. Tie-in is scheduled for completion on May 31.

Anion Exchange for 1WW Rework. The test unit is now complete and operation should be underway in May.

Purex 2A Column Capacity Tests. Erection of a 6-inch-diameter, 10-foot-long glass column is about 75 per cent complete. Processing capacities using the Purex Phase II flowsheet and possibly a new "ultra-low-acid" flowsheet are to be determined.

Operation of the test unit is expected to commence during the week ending May 5.

3000 PROGRAM - WEAPONS

In support of Plutonium Metallurgy studies, emission spectrographic methods are being adapted for measuring both zirconium (from crucibles) and titanium at the 1 per cent level in plutonium. Zirconium is being separated from plutonium by anion exchange while separation is unnecessary for titanium. Working curves are being prepared.

4000 PROGRAM - REACTOR DEVELOPMENT

Impregnated Graphite Studies

Another experiment was performed to determine the uranium distribution in a impregnated graphite cyclinder. Four equal-volume samples which were machined from a piece loaded to an average concentration of 46.2 mg U/ml of graphite analyzed 79.4, 44, 33.5, and 28.0 mg/ml, respectively, from the periphery to the core. The average of the four samples was 46.2 mg/ml which is in agreement with the over-all concentration and indicates the uniformity of distribution along the axis.

Three test pieces of graphite, 0.426-inch in diameter and 0.500-inch in length were impregnated simultaneously in a nitrate solution containing 312 mg U/l. An apparent inverse linear relationship between density of graphite and extent of loading was observed when the data were plotted. Thus, the possibility of predicting the extent of loading from the solution concentration and the graphite density exists.

A series of tests was made to determine the effect of pre-oxidation of the graphite on the volume uptake during the impregnation step. By oxidizing at 600 C in air for 50 hours, the external dimensions of a piece were not measurably altered but the apparent density fell from 1.65 grams/cm^3 to 1.47 grams/cm^3. The measured uptake indicated about 85 per cent available pore volume compared to a previous 70 per cent. This technique may be useful if more uniformly impregnated pieces and higher loadings are desirable.

Aluminum-Plutonium Alloy Preparation

Five 100 to 200 gram batches of 1.65 weight per cent plutonium-aluminum were prepared by the reduction of plutonium dioxide with aluminum under cryolite Na_3AlF_6 as a flux. Although analyses have not been made to determine the extent of plutonium losses, the appearances of the metal reguli are indicative of the completeness of reaction.

In each of four runs, the entire charge was placed in the graphite crucible prior to heating and the furnace cycle completed under an argon atmosphere. When a temperature of 1100 C was reached and held for 20 minutes, a very clean metal button was obtained. On the other hand, when a temperature of 1050 C was used with no hold period, the metal was somewhat contaminated and the flux showed evidence of some unreacted plutonium dioxide. No mechanical stirring was used in these experiments although some stirring by convection was apparent, especially in the 200 gram batches. In a single run, the plutonium dioxide was added with the cryolite as an aluminum foil wrapped mixture. In this procedure, the crucible had access to air, and the flux appeared to have a greater oxide content, sticking so tightly that its release could be effected only by breaking the crucible.

A much smaller scale experiment was successfully completed to yield about 22 grams of aluminum - 15 weight per cent plutonium alloy. The button and flux appeared very clean, with very little evidence of unreacted plutonium oxide. This reduction method for preparing aluminum-plutonium alloys proceeds smoothly with no violence and is simple and direct.

Bismuth-Plutonium Dioxide Slurry Studies

An experimental study was started to investigate some of the chemical and physical properties of the system bismuth-plutonium oxide slurry as a "fluid fuel" for the PRP reactor. Several experiments were performed in which bismuth (or bismuth-lead eutectic), uranium dioxide, and magnesium in mole ratios of 7.3:1:0.5 were contacted in cold rolled steel capsules for three hours at 650-700 C. Uranium dioxide is being used as a "stand-in" for plutonium oxide. In each case (except one when the capsule failed), dispersion of the uranium dioxide in the bismuth occurred as determined by microscopic examination and X-ray diffraction analysis. These results are in substantial agreement with findings at KAPL. The dispersion appeared uniform except near the edges, a condition possibly brought about by the capsule material which was wetted, in each experiment, by the slurry. Better dispersion occurred in the bismuth-lead eutectic than in pure bismuth.

Plutonium Oxide Fuels

A series of experiments have been performed on the production of a UO_2-PuO_2 fuel material suitable for use in the PRP reactor. In agreement with the KAPL work, coprecipitation of an ammonium diuranate-plutonium hydroxide precipitate followed by hydrogen reduction results in mixed crystals provided that the material is heated from room temperature to the 900 C reduction temperature in an atmosphere of hydrogen (actually a nitrogen-1 to 2 per cent hydrogen mixture). When the reduction was preceded by air calcination (900 C), it was not possible to reduce the resultant material to mixed crystals of uranium dioxide and plutonium dioxide. The mixed crystals are desired because of their ease of dissolution for reprocessing.

A total of three reductions were carried out. In the first, the uranium/plutonium ratio was five. Precipitation was with an ammonia-air mixture. The precipitate filtered easily and was washed with ammonium hydroxide solution. It was air dried, then dried overnight at 100 to 120 C, ground to a powder, and reduced in the nitrogen-hydrogen mixture at 900 C for one hour (the sample was introduced into the furnace at room temperature and the furnace brought up to temperature while sweeping with the nitrogen-hydrogen mixture). The resulting powder was brownish-black in color, and X-ray diffraction showed it to be the desired mixed crystal material. An identical sample of precipitate was calcined at 900 C before reduction and did not yield mixed crystals.

A similar preparation was made with a uranium/plutonium ratio of 1.25. The precipitate was not as crystalline as the above, was difficult to filter, and peptized when washed with water but not when washed with ammonium hydroxide. Mixed crystals were not obtained; however, the powder had been calcined prior to reduction. Similarly, denitration at 500 C of a mixture of uranium and plutonium nitrates followed by calcination at 900 C and reduction did not yield mixed crystals. Studies of the rates of dissolution of these various oxides are being made, but results are not yet available.

In another approach, a scouting experiment indicated that uranium dioxide is one of the products of the thermal decomposition of ammonium diuranate in an inert atmosphere. If the diuranate can be decomposed so that the dioxide is the only product, it may be possible to eliminate the use of hydrogen in producing the mixed crystal material. Elimination of hydrogen would be desirable from a safety standpoint.

Crucible Treatment

The possibility of fluorine liberation from graphite crucibles treated with fluorine was investigated and found to be improbable. No fluorine found by mass spectrometer analysis upon heating sample of crucible to 1800 C in vacuum. It is concluded that use of these crucibles in a hydrogen atmosphere furnace is safe from the viewpoint of fluorine corrosion of the heating elements.

6000 PROGRAM - BIOLOGY AND MEDICINE

Geology and Hydrology

Discussions with Dr. Clifford Willis, chief geologist of the Harza Engineering Company, engineer architects for the Grant County P.U.D. for the Priest Rapids and Wanapum Dams, assured us of the desired geological data from the site.

Completion of several geological cross sections in the Wahluke Slope and Pasco Slope areas and correlation of well logs with each other and with the results of field work by others resolved numerous inconsistencies and anomalies and more precisely defined the Pasco Basin to the east. It also permitted the determination of a detailed Ringold formation stratigraphic sequence. The location of the main course of the Columbia River at each stage of deposition, hence of the nature of the sediments to be expected on the project, is a major result of the correlation; behavior of the ground waters can be more accurately predicted and monitoring wells more reliably and advantageously located.

A new well was drilled to basalt in the 200 West Area near the Z-Plant Area. When the blue clay zone of the Ringold formation was penetrated at a depth of 450 feet it was found that the water bearing sediments below the blue clay were at a significantly higher hydrostatic head than those above. These artesian conditions confirm the belief that the blue clay zone is a good aquaclude and must be extensive areally.

Work was initiated to evaluate the influence of dissolved solids and air in the water used for conducting model studies. It was found that as much as one cc of air per 1000 of water was removed from solution by a tension of 5 feet of water. Tensions of this order are frequently encountered in model studies and much higher tensions probably occur in the unsaturated flow zone beneath waste disposal sites. This released air undoubtedly affects the water transmitting properties of the soil and is particularly important in model studies where reproducible results are required. A design study was initiated to provide equipment to deaerate water for research models.

Geochemical and Geophysical Research

The U.S. Bureau of Reclamation Hydraulic Laboratories and the U.S. Geological Survey Hydrological Laboratory, Denver, Colorado, were visited to obtain information on laboratory soil permeability equipment and methods. As a result, equipment was chosen for a laboratory soil permeability apparatus to permit routine permeability tests on well drilling samples.

Use of shaped charges for well-perforating was discussed in a conference with B. M. Caldwell, president of Jet Research Center, a shaped charge application development company. It was indicated that shaped charges could be applied to the anticipated work here, and that the company, which holds important licensing rights, would cooperate fully. Only a modest investment would be needed for an initial application.

The gamma scintillation spectrometer well probe and associated components were studied to improve field performance. Calculations were made of the anticipated response to uniform cylindrical sources with the probe on the axis. It was determined that a uniform Cs^{137} source 12" in radius will give approximately 90 per cent of the response of the infinite radius source. A calibration source need not exceed about 15" radius if it consists of Cs^{137} deposited uniformly in soil surrounding a section of well-casing, or similar configuration.

The ground water flow direction indicator using conductance probes and gyro-compass orientation indication was made available from Radiation Protection Instrument Research and Development. First testing was unsuccessful because of corrosion in the electrolyte delivery unit of the indicator.

Soil Chemistry

Further research confirmed the unusual soil retention characteristics of Ce(III) previously observed. In the three variable system: per cent retention on soils, pH, and Ce-ion concentration, a region was found where the retention on soils was essentially nil. This region occurs between cerium concentrations of 1 x 10^{-6} to 2 x 10^{-6} M and in the pH range 9.5 to 11.5. Since the natural pH of the soil is near this range, it is conceivable that solutions in the critical concentration range could be added to a crib with a very low removal of Ce. The adsorbed Ce can be readily removed by HCl solutions but it is difficult to remove with solutions of salts.

The Gelling of Wastes

Batch equilibrium experiments in water with freshly formed aluminosilicate gel containing Sr^{90} and Cs^{137} indicated that the latter was removed from the gel rather readily by the water leach but that the former was largely retained by the gel. Equilibrium experiments in which dry gel residue was contacted with water and with synthetic aluminum coating waste containing these radioisotopes verified the observation that the aluminosilicate gel had a relatively high affinity for Sr^{90} but a low affinity for Cs^{137}; for example, distribution coefficients for the adsorption by gel from distilled water were $>$500 and $<$1, respectively. Although data from the batch experiments with aluminum coating waste were not complete, the same general trend was evident.

The experimental evidence suggested that liquid which is removed from freshly formed gel by compaction and the tension forces of adjacent dry soil probably will contain appreciable concentrations of cesium. If however, the apparent high affinity of soil for cesium in the original waste is also observed for cesium in liquid extracted from the gel, the radioisotope should be removed from solution rather effectively by the soil. Nevertheless, the apparent low affinity of the dried alumonosilicate gel for Cs^{137} would be a distinct disadvantage if the gel were calcined and an attempt made to use the calcined product to "clean up" additional radioactive waste.

Special Studies

The isotherms for the adsorption of ruthenium tetraoxide from carbon tetrachloride solutions by activated alumina were determined at 40 C and 25 C. The higher temperature appeared to favor greater adsorption, however it was planned to determine the isotherm at 15 C to verify this trend.

Studies on the adsorption and desorption of ruthenium tetraoxide vapor by activated alumina indicate a very appreciable hysteresis. This hysteresis effect suggested that a surface reaction had occurred resulting in a greater retention of adsorbed ruthenium tetraoxide molecules than would be predicted from capillary condensation theories. A logical interpretation of the isotherms suggests that several layers of ruthenium tetraoxide were adsorbed by a chemisorption mechanism followed by physical adsorption of the bulk of the tetraoxide molecules. The data have application to any procedure designed to remove or recovery ruthenium by adsorption processes.

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Environmental and Radiation Chemistry

A two month's study was completed of the application of gamma spectrometric and beta absorptiometric methods to the analysis of aquatic biology samples independently of chemical separations. The study included 143 samples representing nearly all types of aquatic biology specimens. The results show that as many as 17 radioisotopes may be identified and measured in certain types of samples without chemical separation. These isotopes are: Na^{24}, P^{32}, Sc^{46}, Cr^{51}, Mn^{54}, Mn^{56}, Fe^{59}, Co^{60}, Cu^{64}, Zn^{65}, Zn^{69}, As^{76}, Zr^{95}-Nb^{95}, Cs^{137}, Ba^{140}, La^{140}, and Np^{239}. The precision with which each isotope can be measured in a particular sample depends upon the relative amount present. For the majority of samples in this study, Na^{24}, P^{32}, Sc^{46}, Cr^{51}, Fe^{59}, Co^{60}, Cu^{64}, Zn^{65}, and As^{76} can be measured with a precision of about three per cent. Ba^{140}, La^{140}, and Np^{239} can usually be measured with a precision of ten per cent, while the precision of the Mn^{54}, Zr^{95}-Nb^{95}, and Cs^{137} measurements may be only 25 per cent. Mn^{56} and Zn^{69} determinations were not made on these samples, but could be if the samples are counted immediately after sampling. P^{32} could not be measured in the muscle tissue of any fish without chemical separation, and some degree of chemical separation would be required to improve the precision of measurement of the other isotopes. The small aquatic organisms such as algae, plankton, insects, and insect larvae contained a wide spectrum of the short-lived radioisotopes and relatively small amounts of longer-lived isotopes which were measured after some decay. Fish, except for the juveniles, contained mainly the long-lived isotopes, P^{32}, Zn^{65}, and Cs^{137}. The ratio of P^{32} and Zn^{65} in small organisms was about 10:1. In suckers, squawfish, sculpins, whitefish, and carp, the ratio varied from 1:1 to 1:10.

Gamma-gamma coincidence counting of the 1.17 and 1.33 Mev gamma rays can be used to determine Co^{60} in reactor effluent water without chemical separation. Because of the very small amount present, the residue from a one liter sample must be counted for an hour after a delay of one week (to allow the Na^{24} to decay) to obtain the accuracy required.

Fe^{59} and Ga^{72} radiochemical analyses on reactor effluent water were made over a period of several weeks with the separated isotopes placed on an automatic sample changer for beta decay curve determination. In the cases of both isotopes some of the decay curves showed the presence of an impurity of from one to four days half-life. Further studies are under way to identify and remove the interference from this isotope or isotopes.

The over-all concentration of gamma emitting radioisotopes on vegetation in the plant environs has doubled since December, 1956. In addition to the cerium, iodine, ruthenium, zirconium, and niobium isotopes previously measured, Ba^{140}-La^{140} is now present in measurable quantities. The relative amounts of these isotopes suggest that they are debris from atomic weapons tests. Measurements on off-site samples will be made to verify this conclusion.

Dosimetry

A comparison was made of the chemical protection afforded aqueous erioglaucine solutions by urea and thiourea. The protective capacities differ by a factor of one thousand despite the similarity in structure. The protective action measured as the ratio of the rate of the radical-protector reaction to the rate of the radical-dye reaction was found to be linearly related to the concentration for both urea and thiourea over a 50-fold change in concentration.

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Experimental determinations were repeated of the fraction of energy emitted by a spherical, homogeneous, aqueous solution source of P^{32} which is absorbed in the source itself. Two lucite-walled cavities, 2.222 cm diameter, were used and the absorbed dose was measured by means of the ferrous sulfate dosimeter after known periods of time. In each case 90-93 per cent of the energy liberated was absorbed by the source itself. A slight but steady decrease in energy absorbed at longer times was noted. This might be due to impurities in the P^{32}, high background radiation, or some chemical effect on the dosimeter. These factors will be investigated, and the experimentally determined absorbed dose values will be compared with values obtained using the various theoretical equations proposed for this calculation.

Nine off-site samples of underground water were analyzed for natural tritium content.

CUSTOMER WORK

Iodine-131 Analysis

Analysis of vegetation I-131 by chemical leaching-beta counting was found to be related significantly to the time lapse between deposition of I-131 on the vegetation and actual removal by leaching. Iodine was added by spiking with tracer solution, admittedly different from deposition of plant releases. Iodine-131 recovery any time after the day of deposition was about a factor of three low--the factor used for several weeks to increase I-131 observed by leaching-beta counting.

Day Analyzed	0	1	2	3	4	5	7
% I-131 Recovered	78	36	31	28	27	30	25

To measure vegetation I-131 more accurately a four-channel gamma counter is being assembled and tested. As soon as the instrument functions, Regional Monitoring anticipates reducing their requirements for I-131 analyses tenfold. One person within the Analytical Laboratories can be reassigned.

The work of one-fourth of a measurements clerk will be eliminated by a completed program for machine calculation of activity densities of radioisotopes in the river at Pasco. The Pasco densities are based upon measurements of radioisotopes in reactor effluent cooling water.

Redwing Program

The Redwing analytical program is 90 per cent complete on the basis of received and scheduled samples. Marine "life" received has included one octopus fragment and several sea snakes. The best monitor of bomb debris radioisotopes seems to be the clam. Recently Mn-54 was identified and is being measured for 44 selected specimens.

V R Cooper

Manager
Chemical Research & Development

VR Cooper:bp

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VISITS TO HANFORD WORKS

Name	Dates of Visits	Company or Organization Represented and Address	Reason for Visit	HW Personnel Contacted	Access to Restricted Data
Brandon Gunyou	4/15-16/	Koppers Company Pittsburgh, Penn.	Discuss experiences in operation and direct maintenance at the Hot Semiworks and in-line instrument development.	RJ Sloat	Yes
L. J. Barron	4/10/	Pigments Department duPont Company Wilmington, Del.	Applications of titanium to processing equipment	OF Hill AE Smith	Yes

VISITS TO OTHER INSTALLATIONS

Name	Dates of Visits	Company or Organization Represented and Address	Reason for Visit	HW Personnel Contacted	Access to Restricted Data
J. R. Raymond	4/1-2/	US Geological Survey Hydrologic Laboratory Denver, Colorado	Examine hydrological lab equipment for possible application at HAPO	AI Johnson	No
D. W. Pearce	4/4/	Washington State College Pullman, Washington	Present paper at 8th Annual Conference on Waste Disposal		No
D. W. Rhodes	4/5/	Phillips Petroleum Co. Idaho Falls, Idaho	Discuss possible work in waste disposal at the MRTS	CE Stevenson CM Slansky WH McVey	No
R. E. Burns	4/1-5/	Iowa State College Ames, Iowa	Recruiting trip		No
G. J. Alkire	4/8/	Savannah River Plant Augusta, Georgia	Discuss problems associated with instrument development activities.	CH Ice	Yes
	4/9-11/	National ACS Meeting Miami, Florida	Attend Meeting and present technical paper		No

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VISITS TO OTHER INSTALLATIONS

Name	Dates of Visits	Company or Organization Represented and Address	Reason for Visit	HW Personnel Contacted	Access to Restricted Data
R. F. Maness	4/9-11/	Corrosion Short Course Spokane, Washington	Attend Corrosion Short Course		No
R. L. Moore	4/7-12/	National ACS Meeting Miami, Florida	Present a paper and recruit technical personnel at Miami.		No
	4/15-16/	duPont Company Savannah River, Ga.	Inspect hot cell equipment and confer on process chemistry at duPont and ORNL	DG Karraker	Yes
	4/17/	Oak Ridge National Lab. Oak Ridge, Tennessee	Inspect hot cell equipment and confer on process chemistry at duPont and ORNL	FL Culler	Yes
V. R. Cooper	4/8-12/	National ACS Meeting Miami, Florida	Chairman of Committee		No
	4/9/	AEC Washington Office Washington, D.C.	Meeting on chemical reprocessing of power reactor fuel elements	WN Munster	Yes
	4/15/	ORNL Oak Ridge, Tennessee	Discuss new developments on separations processes and equipment.	FL Culler	Yes
	4/16-17/	Argonne National Lab. Lemont, Illinois	Discussion on separations technology, waste products disposal.	S. Lawroski	Yes

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