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PACIFIC NORTHWEST LABORATORY

MONTHLY ACTIVITIES REPORT

OCTOBER 1971

Division of Production

and

Hanford Plant Assistance

Programs

by the

Staff of Battelle-Northwest

R. S. Paul, Director

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PACIFIC NORTHWEST LABORATORY

MONTHLY ACTIVITIES REPORT

DIVISION OF PRODUCTION AND HANFORD PLANT ASSISTANCE PROGRAMS

W. D. Richmond

ASSISTANCE TO ATLANTIC RICHFIELD HANFORD COMPANY

Summary

Program I

Modification and testing of a DuPont 400 Process Photometer for monitoring the sodium nitrite concentration of an organic process stream was completed.

Modification and testing of an electrolytic dissolver for dissolving plutonium button pieces in nitric acid are continuing. Tests with stainless steel as a stand-in for plutonium have yielded dissolution rates as high as 2.4 g/min at a current input of 350 amp.

Program III

The transmissivity iterative routine is being modified to account for the transient changes in the Hanford groundwater table. Pump test data are being reduced to transmissivity values utilizing Boulton's solution and the Theis modified non-equilibrium formula. Work is continuing on examining techniques to reduce computational time of the groundwater models.

The computer program for reducing gamma count data to moisture content data was tested and found satisfactory. An apparatus for increasing thermal diffusivities as a function of moisture content was nearly completed. The study on water yield production has been postponed due to lack of data. Six instrument cables for the lysimeters south of 200 East Area were tested and found in good condition.

A column study was made to continue the one-dimensional verification of the sorption model for predicting the movement of ^{85}Sr through soil systems as a function of throughput volume of a Na, K, and Ca solution.

A study to determine a depth series of soil chemical parameters for a group of 12 Hanford wells was continued.

Program IV

Fabrication of two low-level effluent monitors continued during the month.

A high capacity ion exchange gel for in situ polymerization in Hanford soil was prepared in the laboratory.

In the Cs-Sr packaging program, all originally scheduled compatibility tests have been completed. Supplemental tests scheduled will be complete in September, 1972.

No preferential corrosive attack at the vapor-liquid interface has occurred during eight months of exposure of 1020 mild steel coupons to boiling simulated alkaline high level radioactive waste. The tests were made in response to speculation that such preferential attack may occur in waste storage tanks.

Melting radioactively contaminated metal equipment under an inert atmosphere tends to fix radioactive constituents in the resulting ingot making subsequent leaching of them difficult.

Literature pertinent to purification of sodium metal by distillation is being reviewed in preparation for application of this technique in salt cake volume reduction studies.

Process Technology

An expanded version of the Hansen-Roach cross section library has been generated, which contains 130 different cross section data sets, for use with the KENO Monte Carlo code. A listing of the new cross section sets is available on request.

The criticality experiments on plutonium nitrate solutions with high burn up fuel (^{239}Pu - 41.4 wt%, ^{240}Pu - 42.9 wt%, ^{241}Pu - 10.8 wt%, and ^{242}Pu - 4.7 wt%) were analyzed by means of the KENO code with cross sections from the ENDF/B-II data library. The computed k_{eff} was found to be, on the average, about 2% above unity.

Assistance was provided the USGS in installing 8 new microseismic stations and relocating two existing stations. The Environmental Research Laboratory seismograph station was operated continuously during the report period. One magnitude 3.2 event occurred on the south side of the Saddle Mountains on October 25, 1971. The October 25 event was recorded by triaxial accelerometers located on the ground surface and the first floor level of the 221-U Building.

Tentative analytical results have been received for the remainder

of the experiments performed to determine the fractional release of active materials involved in shipping-type accidents. Approximately 11.4 percent of the uranium present in the source material was airborne from uranyl nitrate solution on a stainless steel surface at an air velocity of 23 mph during the 10 minutes required to burn a gallon of gasoline. This is the highest fractional release found among the 26 sets of conditions tested.

One additional resuspension experiment was performed during the month and is presently being analyzed.

Observations made during study of the relatively poor zirconium decontamination in the Purex HA column obtained during a recently completed processing campaign can be explained on there being in the HA column a solid which (1) slowly dissolves and releases a complexant that forms unscrubbable Zr species and (2) contains some Zr (sorbed or precipitated).

The inextractable plutonium in the Plutonium Reclamation Facility (PRF) CAW stream is due essentially entirely to solids coming into the process in the CAW rather than to solids which might form in the backcycle stream.

In mixer settler runs, Am extraction in the PRF E-1 column of 97.5 and 99.8% was achieved with total nitrate salting strengths of 4.5 and 5.5M respectively. Hydroxyacetic acid added to the E-1 column feed before pH adjustment helped prevent solids formation but hindered Am extraction in the E-1 column. In a series of S-1 column runs, the best Am stripping was achieved with a low acid flowsheet.

Studies were completed to develop basic extraction and stripping equilibrium distribution data for Pu(IV), Am and H^+ required for the Plutonium Reclamation Facility, which uses a 30% DBBP-CCl_4 organic extraction system.

Concentration of Am recovered in the Plutonium Reclamation Facility can be accomplished by ion exchange at faster flow rates than now used in the plant.

Three buttons of plutonium metal were dissolved during the month. Large amounts of brown solids were formed at low concentrations of HF, but redissolved when HF was increased to 0.05M. At lower nitric acid concentrations some solids were formed, but at 0.05M HF and HNO_3 concentrations above 9.7M no solids were formed.

Laboratory evaluation of oxidation-reduction potential measurements for possible application to Pu valence control in the Plutonium Reclamation Facility (PRF) indicates the technique is applicable. An electrodeless conductivity system for use in PRF CAF acidity control is under construction.

Design and development of circuitry necessary for selectable narrow and elevated span operation of the Purex HAW and HAS electrodeless conductivity monitors was accomplished. Revision of the HAS system was completed.

Testing and evaluation of two wire transmitters for application to plant instrumentation and control system has been conducted.

ASSISTANCE TO ATLANTIC RICHFIELD HANFORD COMPANY

Program I - Basic Production

Nitrite Monitor for Organic Process Solutions
(R. D. Dierks, Chemical Development Section)

A DuPont 400 process photometer, originally acquired to monitor the Purex plant stack gases for NO_2 concentrations, was modified to monitor the nitrous acid concentration of the organic feed stream (1BFX) to the Purex process partitioning column. The modifications included replacing the 3/4-in. sample cell with a 20-in. cell, replacing the calibration filter, and installing a pneumatic operator for the calibration filter. The stability and the ambient temperature sensitivity of the instrument was established and previously reported. The system responded well to simulated 1BXF solutions with nitrous acid concentrations as low as $1x10^{-3}$M.

Evaluation of the instrument system has been completed and the monitor has been disassembled in preparation for installation in the Purex plant 1BXF sampler system.

Electrolytic Dissolution
(R. D. Dierks, Chemical Development Section)

An electrolytic dissolver for dissolution of metallic plutonium pieces in nitric acid is being developed. Stainless steel is being used as a stand-in for plutonium in dissolver design evaluation studies.

The original dissolver concept was for a contact-type dissolver where the metal piece (a 4-in.-diameter stainless steel disc) rests on a heavy gauge platinum wire grid. Testing at high power inputs showed substantial arcing between the stainless piece and the platinum grid; pitting of the platinum and burning of the stainless steel occurred. A sustained dissolution rate of 2.4 g of stainless steel per min was achieved with an input current of 350 amp.

To avoid arcing and the possibility of ignition of the plutonium pieces, modification of the anode to provide a solution-type contact was investigated. Methods of providing the minimum insulation necessary to prevent arcing are currently being evaluated. Automatic control systems have been incorporated in the test facility to maintain the solution temperature and acidity constant during the dissolution periods.

Program III - Environmental and Regulatory Technology

Radionuclide Transport Model
(D. B. Cearlock, Water Resources Systems Section)

Additional refinements to the transmissivity iterative sequence

computer program were made. A control program was written and implemented into the sequence to allow easier transfer between major programs, i.e., TRAVEL, STREAM AND FLOTUB. These programs and their associated data file have also been transferred to permanent magnetic tape storage in a new manner using a recently developed SAVE program. This program eliminates much time which was previously used in handling tapes. It also allows an operator unfamiliar with the computer system to operate the groundwater programs with a minimum of confusion. Many of the program controls have been turned over to the display keyboard and light-pen to facilitate ease of operation and faster response.

In analyzing the historical records of well hydrographs, it was observed that groundwater storage was important relative to groundwater movement in portions of the Hanford system. It appears that ignoring storage can cause errors in the calculated transmissivity distribution. Therefore, the iterative sequence is being modified to handle transient systems, i.e., account for storage. The modifications have been coded but not inserted into the program because of current disc memory unit problems.

Simultaneous with this work, pump test data, that were previously collected, are being reduced to transmissivity values. Two theoretical methods are being used: (1) Boulton's solution, and (2) the Theis modified non-equilibrium formula. The Boulton's solution is more exact than the Theis method since it satisfies the linearized free boundary condition at the water table surface. These data along with groundwater potentials will be used to calculate an improved transmissivity distribution.

The Partially Saturated Transport (PST) program was converted to handle an unequal grid so that the tank leak problem could be continued. Problems with library subroutines have been eliminated. Work also continued on reducing the computational time of the Variable Thickness Transport (VTT) and PST models by incorporating new solution techniques. Several problems related to boundary conditions on the Hanford Project have been discovered which prevent their present use. Work is under way to study the alternatives.

The data collected from the physical models being used for verification of the PST model are in the form of gamma ray counts. The computer program for reducing these data to moisture content by volume was tested and found satisfactory. This program will take care of the background radiation, the dead time of the counting equipment, etc. The weighted average of the data will be plotted by computer to provide curves of water content versus soil depth in the model at various time intervals.

The apparatus to measure the rate of water flow (liquid and vapor) induced by a thermal gradient is almost ready for testing. The unique feature of this design is its ability to eliminate the back flow of water which is not caused directly by thermal gradients. This apparatus

will be used to measure thermal diffusivities as a function of moisture content.

The study of a water yield production equation for arid regions based on plant type, water use profile and historic precipitation record has been postponed because information on the water use profile of Hanford local plants is not available. The purpose of the above study is to provide additional information to assist in determining if natural precipitation reaches the groundwater table at Hanford.

The six instrument cables assembled for the lysimeters south of 200 East Area were checked for electrical continuity to insure that the connections were good and that the psychrometers were not broken. The tests indicated that all the cables are in good order.

Soil-Waste Interactions

(R. C. Routson and R. J. Serne, Water and Waste Management Section)

A column study was made to continue the one-dimensional verification of the sorption model for predicting the movement of ^{85}Sr through soil systems as a function of the throughput volume of a sodium, potassium and calcium solution. The study was made with a TF sandy loam soil. Agreement between the predicted and the measured normalized effluent ^{85}Sr concentrations as a function of column volumes of effluent was generally within ±10% for C/C_o values up to about 0.8. The agreement was less good at higher C/C_o values. The disagreement at the high C/C_o values is probably due to slow chemical reactions which are not accounted for by the equilibrium sorption model. In the high C/C_o region of the curve, the prediction is conservative and primarily of academic interest since ground disposal has generally ceased long before a C/C_o of 0.8 is reached.

The TF sandy loam is finer soil which has considerably different sorption parameters than the Burbank sand on which the initial model verification was made. These two soils probably represent much of the range of soil parameter variation which can be expected from the soils of the unsaturated zone of the Hanford project.

Hanford Soil Properties Versus Depth

(R. C. Routson and R. J. Serne, Water and Waste Management Section)

A study to determine a depth series of soil chemical parameters for a group of 12 Hanford wells was continued. Parameters measured include: (1) moisture content-moisture potential relationships; (2) pH in a saturated paste and 0.01M $CaCl_2$; (3) cation exchange capacity (CEC); (4) mechanical analysis; (5) $CaCO_3$ content; (6) field moisture content; and, (7) porosity.

The porosity of the soil in Well 9 varies from 32 to 45% over the depth range of from 5 to 140 ft. There is no apparent correlation of porosity with depth.

Program IV - Waste Management

Gamma Cell Development

(R. D. Dierks, Chemical Development Section)

Progress was made during the month in the fabrication of two low-level effluent monitors. Headplates for the flow cells were fabricated and electropolished and are currently being finish-machined to receive the tapered, plastic scintillation crystals. One stainless steel flow cell has been electropolished and the other has been lined with Kynar plastic. The final assembly step, to fit the headplates to the flow cells, will be accomplished early next month. The monitors will then be installed in the Purex plant for field evaluation.

Fixation of Radionuclides in Soils and Salt Cakes with Chemical Grout

(B. W. Mercer, Water and Waste Management Section)

A high capacity ion exchange gel, which appears suitable for in situ polymerization in Hanford soil, was prepared in the laboratory from a sodium acrylate solution with ethylene glycol dimethacrylate as the crosslinking agent. The ion exchange capacity of the polyacrylate gel with 20% crosslinking agent was 5.5 milliequivalents per gram on a dry weight basis. The acrylate and ethylene glycol dimethacrylate grout can be prepared and polymerized in a manner similar to AM-9 grout except that about 20-30% ethanol must be added. The ethanol serves to increase the solubility of the crosslinking agent which has a low solubility in water. Substitution of N, N'-methylenebisacrylamide, which has a relatively high solubility in water, for ethylene glycol dimethacrylate may be a possible alternative to eliminate the use of ethanol. This approach will be investigated further.

Leaching experiments are in progress to evaluate the effect of the ion exchange sites in the polyacrylate gel structure on the diffusion of trace strontium ions from the gel.

Cesium Purification Flowsheet Studies

(J. A. Partridge, Chemical Development Section)

Two Zeolon-900 ion exchange beds have been conditioned and cesium purification tests with simulated feed solution will be performed with them. The alkali metal-ion ratios in the simulated feed prepared for the initial tests are Na/Cs = 6.5, K/Cs = 4.5 and Rb/Cs = 0.016. These ratios correspond to those expected in the B-plant 37-1 tank solution.

Approximately 8 gallons of diluted B-plant 37-1 tank solution (corresponding to about 1 1/2 gallons of undiluted 37-1 solution) was received during the month. This material, which contains large amounts of solids, will be used for tests to characterize the solids and for ion exchange flowsheet tests.

Cesium-Strontium Packaging
(H. T. Fullam, Chemical Development Section)

The 36-month compatibility tests on non-radioactive cesium and strontium have been completed. The test capsules have been sectioned and sent to metallography for evaluation. None of the capsules showed excessive attack.

The originally scheduled compatibility tests have now been completed. The only tests remaining are the supplementary tests of non-radioactive SrF_2 with 316L stainless steel which will be finished in September, 1972, after 36 months at temperature. Preparation of the final report on the compatibility studies is now under way.

Waste Management - Corrosion Studies
(R. F. Maness, Corrosion Research and Engineering)

Laboratory studies have shown that reproducible stress corrosion cracking (SCC) data can be obtained in nitrate and in caustic solutions with notched and precracked (pop-in crack) 1020 mild steel C-rings as test specimens. A study was initiated to evaluate the variables affecting SCC in the two environments. Variables in the nitrate systems include temperature, nitrate concentration, sulfate concentration, pH and the effect of oxidants. Variables in the caustic system include temperature, caustic concentration and the effect of oxidants. Only limited data from this study are now available.

There is some speculation that existing storage tanks containing alkaline high-level waste solution have been preferentially attacked at the liquid-vapor interface. A laboratory study in which 1020 mild steel specimens were exposed to two boiling simulated alkaline high-level waste solutions (fluoride-free and fluoride-bearing) revealed no preferential attack at the liquid-vapor interface after eight months exposure. Solvent (0.1 vol percent of 30% TBP-NPH) was added to the test solutions on a weekly basis for the first ten weeks of the test and on a monthly basis thereafter.

Equipment Melt-Down Studies
(H. T. Fullam, Chemical Development Section)

Tests continue to show that highly contaminated metal equipment can be melted down and cast to give metal ingots in which the contamination is dispersed and fixed in the metal phase. The contamination in the ingot is non-smearable and difficult to leach. Leaching a typical ingot (prepared from 304L stainless steel contaminated with Purex acid waste) with 8M nitric acid for one hour at 25 °C gave a leach solution having a total beta activity of < 200 d/min/ml. The total beta activity of the metal ingot, determined on an aqua regia solution used to completely dissolve the ingot, was about 10^6 d/min/g. Water leaching for 11 days

of a similarly contaminated ingot gave a solution having a total beta activity of 7 d/min/ml.

It was demonstrated that contaminated metal can be decontaminated using a fused nitrate salt at high temperature. The nitrate decomposes liberating oxygen which reacts and corrodes the metal surface. A decontamination factor of about 10^3 was obtained when contaminated 304L stainless steel was contacted with sodium nitrate at 800 °C for 2 hours. A second contact with fresh sodium nitrate at 800 °C gave an added decontamination factor of 10^2-10^3. The dissolution rate of the metal in the sodium nitrate at 800 °C was about 5 mils per hour. The preliminary data indicate a fused salt to metal weight ratio of about 10 is required to obtain an overall decontamination factor of 10^5-10^6.

Salt Waste Volume Reduction

(J. L. Swanson, Chemical Development Section)

The feasibility of purifying sodium from fission products by fractional distillation of the metal is being evaluated. Initial efforts involve becoming familiar with the literature on fractional distillation as it may be applied to the separation of sodium and fission products.

For the separation of sodium and cesium, no advantage is seen to isotopic dilution with inactive cesium (in the absence of any complicating impurity effects, at least). This conclusion is different from that reached in an earlier preliminary study.

Several papers concerning the vaporization of fission products from sodium were reviewed. These were concerned with the entry of fission products into the vapor space of a sodium-cooled reactor but should be applicable to the fractional distillation problem.

The only experimental results found to date are for cesium and iodine (as sodium iodide). Calculated values of the partial pressures of the less volatile fission products strontium, barium, tellerium, and antimony over sodium have been reported. Of these, strontium should be hardest to separate from sodium.

Process Technology

are currently being made, and are designed to speed up the data retrieval process by directly addressing the data rather than using the previous procedure of tape searching. A listing of the new cross section data sets is now available.

Effect of ^{241}Pu on Criticality Safety Limits for ^{239}Pu

(R. C. Lloyd, Criticality Research and Analysis Section)

The critical experiments with high burn up fuel have been analyzed by means of the KENO Monte Carlo code utilizing ENDF/B-II cross section data and the GAMTEC code. The experiments involved criticality measurements on plutonium nitrate solutions in a 24-in. diameter, water-reflected, cylindrical vessel; the isotopic composition of the plutonium was: ^{238}Pu - 0.2 wt%, ^{239}Pu - 41.4 wt%, ^{240}Pu - 42.9 wt%, ^{241}Pu - 10.8 wt%, and ^{242}Pu - 4.7 wt%. The computed criticality factors are presented in the following table.

The results obtained by the KENO code, with cross sections from the ENDF/B data file, display a somewhat positive bias with k_{eff} being, on the average, about 2% above unity. Further efforts will be made to determine the reason for the bias, and to improve the correlation between experiment and theory.

Criticality of High Burn Up Plutonium Solutions

Pu Conc., g/ℓ	Nitric Acid Molarity	Critical Height in 24-in. Dia. Cylinder, cm	Critical Volume, ℓ	Computed k_{eff} with ENDF/B-II Date & KENO Code
140.0	5.02	54.10	158.3	0.998 ± 0.007
116.0	4.14	50.55	147.9	1.032 ± 0.006
99.3	3.67	48.26	141.2	1.022 ± 0.007
85.5	3.12	47.00	137.5	1.029 ± 0.005
75.6	2.74	47.29	138.3	1.016 ± 0.007
65.1	2.41	49.12	143.7	1.023 ± 0.007
56.3	2.05	52.83	154.5	1.031 ± 0.007
46.8	1.70	63.47	185.7	1.022 ± 0.005
40.6	1.46	80.92	236.7	1.015 ± 0.005

Field Monitoring

(D. D. Tillson, Water Resources Systems Section)

Water level recorders were installed on project well numbers 45-42, 53-35, 54-35 and 54-37 near Gable Mountain.

Work was started on establishing new well designation for the existing and proposed dry wells around the waste tanks in the 200 areas. The purpose of the change is to provide a system that more readily identifies any given well in relation to the waste tank it monitors.

Seismic Studies

(D. D. Tillson, Water Resources Systems Section)

Assistance was provided the USGS in installing 8 new microseismic stations and in relocation of two existing stations. For the second time in two months, 1100 feet of signal cable had to be replaced at the Midway station when it was found missing.

The hydroseismic system was maintained in continuous operation during the month. Seventeen teleseismic events were detected during the report period. A local event that occurred on October 25, 1971, caused a very small response that would not have been seen under normal circumstances.

The Environmental Research Laboratory (formerly National Ocean Survey) seismograph station on Gable Mountain was maintained in continuous operation during the last thirty days. Arrival times for 28 teleseismic events, 33 regional events and one nuclear test were reported to ERS National Earthquake Information Center, Rockville, Maryland. The frequency of occurrence of local seismic events was below normal for the month. One magnitude 3.2 event occurred on the south side of the Saddle Mountains on October 25, 1971, at approximately 1153 hours. Preliminary epicenter location was 46° 42.4' N, 119° 32.8'E (USGS) at a depth of 3 km. Reports of surface motion felt in the vicinity of the epicenter are being verified.

The U-Plant coupling experiment is proceeding according to schedule. The impedance matching system and amplifiers for 12 channels were completed and placed in service. Continuous recording on magnetic tape was initiated. The local event on October 25, 1971, was recorded on magnetic tape by two separate triaxial accelerometers located on the ground surface and first floor level of 221-U Building. Reduction of the tape data is now in progress. Latest information from the vendor indicates that the equipment necessary for remote location of a fourth accelerometer will be on project about the middle of November.

Atmospheric Release of Radioactive Particles

(L. C. Schwendiman and J. Mishima, Atmospheric Analysis Section)

Tentative analytical results of the samples taken to determine the fractional release under selected conditions from uranium as dioxide powder or nitrate solution deposited on stainless steel or macadam surface have been received. Not all results have been finalized due to unresolved procedural instrument problems with the fluorometric procedure. Only the last few results are involved--a single impactor stage on experiment SA-20b and the filter samples from experiment SA-21a. Replicate analyses indicate that incomplete dissolution (the presence of particles) may be contributing to the uncertainty in the impactor stage result.

In these experiments, the uranium compound was deposited on the surface to be tested and placed in the furnace section of the wind tunnel.

A gallon of gasoline was poured onto the surface and ignited. Air at the preselected velocity was drawn over the burning mass and sampled downstream. Three samplers were used--a filter sampler at the top of the duct near the centerline, a filter sampler at the center of the duct and a cascade impactor sampler at the bottom of the duct near the centerline. Sampling rates varied in the filter samples during the burning cycle due to high loadings. Airflow and sampling were stopped when the flames were extinguished. The samplers were replaced and the experiment was continued.

In these five experiments, 22 grams of UO_2 powder, and 50 cc of uranyl nitrate solution deposited on stainless steel surfaces were used in one series. In another series, 50 cc of uranyl nitrate solution and UO_2 powder deposited on a 15-in. diameter macadam (asphalt-gravel mixture) surface were used. From the 26 sets of conditions selected for evaluation, it is concluded that:

- The highest fractional release was during a gasoline fire involving uranyl nitrate solution on a stainless steel surface into air at 23 mph, during the 0.17 hr burning period. Around 35 percent of the particles were in the nominal inhalable size range and thus 4.3 percent of the total source would be an inhalation hazard.

- The next largest fractional release was for UO_2 sprinkled on vegetation-covered soil during and after a gasoline fire into air at 23 mph. A value of 2.9 percent was calculated for the amount made airborne during the fire. Only 19 percent of the material airborne was less than 10 microns in size.

- The degree of hazard of a particular set of circumstances appears to depend upon the interaction of effects. Liquids penetrating into soil are immobilized on drying and less material is available for release. Spreading the liquid in a thin film prior to drying (on vegetation or a hard surface) can make significant quantities of particles available upon film breakup. Preformed particles (powder) can be entrained from surfaces if the physical conditions can provide a sufficient lifting force but the presence of cover (vegetation) or hard surfaces appears to decrease the release under the conditions used in this study.

Background experiments on the fractional release of active particles during the combustion of flammable waste/active materials mixtures were performed and the fractional release experiment using UO_2 incorporated in flammable waste was carried out.

The turbine flowmeter to measure large volume sampling equipment flow was placed in service. The meter was coupled to a LUDLUM Scaler; a holding stand and transition pieces to connect the meter to sampling equipment were fabricated. Various other necessary pieces of equipment were designed and are under construction. The four high-volume ROTRON

pumps to be used for sampling are being fitted with a power cord switch and an elapsed time meter.

Particulate Resuspension Studies

(C. E. Elderkin, G. A. Sehmel and J. V. Ramsdell, Atmospheric Physics Section)

Particle resuspension caused by passage of a 3/4-ton truck was measured on the same day that the tracer was placed on the asphalt road. The data are being analyzed. Results from this series of tests will be compared with previously obtained data for resuspension after four days weathering.

Purex Studies - Zirconium Decontamination in HA Column

(J. L. Swanson, Chemical Development Section)

Poor decontamination from zirconium in the Purex plant extraction column (HA) continued to the end of the recently completed processing campaign. At the end of this campaign the column was operated for several hours with feed containing no plutonium or fission products. Analysis of the organic product stream (HAP) after five hours of such operation showed that the zirconium content had decreased only two-fold while the plutonium content had decreased seven-fold. This behavior suggests the presence of a source of zirconium within the column.

After plant shutdown, the HA column was flushed with nitric acid, then with a caustic-tartarate mixture, and then with a nitric acid-oxalic acid mixture. All of these flushes were found to contain considerable zirconium; the ratios of activity in the flushes to that in the HAP during operation with cold feed were about 6, 20, and 5 respectively. Extraction-scrub tests showed that the first two flushes contained materials that form unscrubbable zirconium species. Very little zirconium could be extracted from the third flush, doubtless because of oxalate complexing.

The observations described here (and in the report for last month) can be explained by hypothesizing the presence within the HA column of a solid material that (1) slowly dissolves during plant operation to yield a complexant that forms unscrubbable zirconium species and (2) contains some zirconium (sorbed or precipitated). Testing of this hypothesis may not be possible, however.

Plutonium on Solids in the Plutonium Reclamation Facility (PRF)

(J. L. Ryan, Applied Chemistry Section)

Work was continued on identifying the cause for poor extraction of Pu from the PRF extraction column waste (CAW). It was concluded on the basis of centrifugation studies that all of the Pu in the E1 column waste

(E1W) was present as solids. Centrifugation of a CAW sample indicated that about 60% of the Pu present was in the form of solids. From this work, it was concluded that all soluble Pu is being efficiently extracted in the E1 column and that the losses are due essentially entirely to Pu on solids coming into the process in the CAW (rather than solids which might form in the backcycle Pu stream). Although the solids were not identified, there is some reason to believe (color and solubility in HNO_3-HF) that they may be largely PuO_2.

Solvent Extraction in the Plutonium Reclamation Facility (PRF)

(D. G. Bouse, Applied Chemistry Section)

Mixer-settler runs were continued to test flowsheets for the PRF waste treatment system E-1, S-1 and S-2 columns. The evaluation of the effect of total nitrate salting strength on Am extraction in the E-1 column was extended to include feed nitrate salting strengths of 4.5 and 5.5M. Americium extraction for these runs was 97.5 and 99.8%, respectively.

During the E-1 column feed pH adjustment step, considerable amounts of iron and aluminum precipitates can be formed and are slow to redissolve. One E-1 column feed was prepared to contain 0.25M hydroxyacetic acid to complex the iron and aluminum. Neutralizing this feed with 9.5M NaOH produced no iron precipitate and only a small amount of aluminum precipitate which redissolved within 3-4 minutes; however, Am waste loss with hydroxyacetic acid present was fairly high with 17.5% being in the E-1-W.

Various combinations of S-1-X compositions and aqueous to organic flow ratios have been tested in a series of S-1 column demonstration runs. Very poor analytical data were obtained from these runs. Americium losses to the S-1-W ranged from 16% using 1.0M hydroxylamine nitrate for the S-1-X and an aqueous to organic ratio (L/V) of 0.2, to 37% using HNO_3 at 0.8 to 1.2M and L/V's at .05 and .33. One run was made with low acid (S-1-X = 0.1M HNO_3) and an L/V of 0.27. Ninety-eight percent of the Am was stripped into the S-1-P in this run.

Solvent Extraction of Americium and Plutonium(IV) into Dibutyl Butylphosphonate (DBBP)

(L. A. Bray, Applied Chemistry Section)

Extraction distribution ratios for Pu(IV), Am(III) and H^+ were determined for the 30% DBBP-CCl_4 extraction system, as the equilibrated aqueous H^+ concentration varied from 0.1 to 10M. Maximum Pu, Am, and H^+ ratios were determined as 300, 0.2, and 0.3 respectively, at 2M H^+.

Also, Pu, Am and H^+ stripping distribution ratios were determined as both the H^+ and $NaNO_3$ concentrations varied from 0.1 to 2M. Distribution ratios (E_d) for both Pu and Am increased with increasing $NaNO_3$ at constant H^+, and increased with H^+ with constant $NaNO_3$.

Further, Pu, Am, and H^+ distribution ratios were also determined for 0.2M ferrous sulfamate-0.25M HF - HNO_3 and for 0.1M HF - HNO_3 stripping systems. The ferrous sulfamate lowered the E_a^o for Pu by reducing the plutonium to (III), and lowered the E_a^o for Am by forming a sulfate complex.

Several experiments were completed to test methods for adding aluminum salts, such as $Al(NO_3)_2 \cdot OH$ and $NaAlO_2$, to the extraction column waste (CAW) solutions (feed to the E-1 column) to increase the Am extraction. Combinations of carbonate or hydroxide were investigated both as mixed caustic-aluminum systems and as individual additives for adjusting the pH to 0.5-0.7. Salting of CAW solutions with aluminum appears to be very difficult because solids form locally and will not redissolve in the plant-imposed 2 minute time period. Complexants such as sodium gluconate and hydroxyacetic acid provide excellent solids control when combined with additional aluminum salting, but they also lower the distribution of Am 2- to 3-fold which then defeats the benefits of aluminum salting.

PRF Flowsheet Studies - Am Concentration by Cation Exchange
(J. L. Ryan, Applied Chemistry Section)

To test the feasibility of diluting the entire S-1P stream (Am solvent extraction product) to 0.25M HNO_3 and feeding it to a cation exchange resin column instead of backcycling two-thirds of it, a cation exchange column loading was made simulating the proposed conditions. The effect of such a flowsheet modification will be to decrease the Am concentration in the column feed by a factor of three and to increase the volume flow rate by a factor of three over the present flowsheet, thus giving no change in the Am mass flow rate (assuming complete Am recovery by solvent extraction in either case). It can be reasonably expected, however, that the impurity ions concentrations in the feed (Fe, Al, etc.) will be the same in either case. Hence, feeding the entire stream will have the net effect of lowering the Am concentration and decreasing the ratios Am^{3+}/H^+ and Am^{3+}/impurity ions, and thus will be expected to decrease the resin equilibrium capacity for Am. The higher volume flow rate in itself should have a negligible effect.

A simulated feed of the composition shown below was fed to a two-foot column of Dowex 50W, X-8 (50-100 mesh) at 10 ml/min/cm^2 (present plant flow is 3.2 ml/min/cm^2 on a two-foot column). The Am concentration shown corresponds to that expected with no Am backcycle, and the impurity metals concentrations are in all cases a factor of three higher than those that would be present in this stream based on an analysis of a sample of S-1P from a recent mini-mixer-settler run and are much higher than observed in one analysis of a plant S-1P solution.

Feed Composition

Fe(III)	0.0007M
Al	0.0002M
Cr(III)	0.00004M
Mg	0.0006M
Pb	0.00001 4M
HNO_3	0.25M
Am	0.40 mg/ℓ

Sixteen hundred column volumes were fed to the column in 155 hours; Am breakthrough at termination was 10.9%. At this point total loading was 0.635 mg Am/ml resin and total Am loss was 0.77%. At 1% breakthrough (1340 column volumes) total loading was 0.535 mg Am/ml resin and total Am loss was only 0.014%. The breakthrough curve was steep and there would be no significant gain in loading beyond about 5-10 percent breakthrough. Iron breakthrough occurred at about 410 column volumes indicating that Am is held stronger by the resin than Fe(III). This also indicates that, from this feed composition, the resin loads to at least 40% of its exchange capacity with Fe(III) and presumably to a considerably higher percentage with total trivalent ions including Al and Cr. Any decrease in the concentration of these competing impurity ions over that used in this run will increase Am capacity to breakthrough. Also, since the Am distribution onto H^+ form resin is inversely proportional to the third power of the H^+ concentration, any decrease in acidity below 0.25M will increase Am capacity, particularly if trivalent impurity ion concentrations prove to be actually lower than these used here.

The column was eluted with 7M HNO_3 at 0.44 ml/min/cm^2 (equal to 4.8 ℓ/hr for the plant column). The Am was almost completely removed in three column volumes and the eluate concentration was below the feed concentration at the end of four column volumes. The peak Am product concentration was about 0.7 g/ℓ.

Electrolytic Dissolution of Plutonium Metal

(E. J. Wheelwright, Applied Chemistry Section)

Three additional buttons of plutonium metal were dissolved during the month in experiments designed to determine operating conditions which would avoid the generation of solids in the dissolver solution.

In each of the three experiments, dissolving was interrupted after each hour, the button was removed and weighed, one liter of dissolver solution was removed from the dissolver system and replaced by one liter of fresh dissolvent. The addition of fresh dissolvent was made to hold the acid concentration above 9M throughout the operation. In the first run, the initial dissolvent was 12M HNO_3 - 0.05M HF, and periodic replacement was with the same dissolvent composition. The rate of dissolution was low because the button geometry (2 1/4 in. diameter x 2 in. high) did not utilize the full cross-sectional area of the electrodes. Solids

could not be seen in the dissolver solution. However, at the end of the 5th hour, filtration of the solution slowed down as the filters became partially blinded by a very small amount of solids. Starting at the 9th hour, 15.6M HNO_3 - 0.05M HF was added each hour rather than the weaker acid and filtration improved as the dissolver acid concentration increased above 10M.

In the second run the initial acid contained no HF and the replacement acid was 14M HNO_3 - 0.05M HF. After only 10 minutes, large amounts of brown solids were present in the system so HF was added. In a short period of time, the solids were dissolved and were not thereafter observed. Solids were not found on the filters. The incremental addition of 14M HNO_3 - 0.05M HF correctly compensated for the loss by electrolysis and held the acid concentration constant.

A flat polyethylene washer, designed to fit around the Pu button and blind the uncovered area between the electrodes, was used in some dissolving periods. This had no effect on the rate of dissolution until the button height was decreased to about 3/4 in. At that point, with the electrodes increasingly closer together, the gasket was effective in increasing the rate of dissolution compared with no gasket. Even at full power, 150 amps could not be approached with the gasket in place.

The third run involved a plutonium button nearly four inches in diameter. It had a much more favorable geometry in the dissolver basket and the overall dissolution rate was much higher. The button had been quartered before delivery to BNW. The problem of manipulating the four sections and the deterioration of the bottom of the plastic basket caused problems near the end of the operation which prolonged dissolving of the last 150 grams of metal. Dissolver current was maintained at 150 amp and, except for the last 100 g of the button, the dissolution rate averaged 323 g/hr which is excellent.

The initial dissolvent concentration in this run was 12M HNO_3 - 0.005M HF. Incremental dissolvent additions were 14M in HNO_3.

Copious amounts of brown solids were generated during the first 10 minutes, and the HF concentration was increased to 0.025M. This change seemed to eliminate the solids for about 2 hours. When an increasing amount of solids was observed, the HF concentration was increased to 0.05M. Even this did not completely eliminate the solids problem. The decreasing acid concentration (below 10M) probably contributed to the solids problem. Since all of the solids were soluble in 14M HNO_3 - 0.05M HF, the presence of solids in the dissolver solution of this experiment, compared with the absence of solids in the previous experiment, must have been due to the higher Pu concentration in the dissolver solution or to the more rapid dissolution rate. The concentration effect is more suspect. In a production line, the decrease in acid concentration and the high Pu concentration could be corrected by a more rapid addition of fresh dissolver solution.

Samples were taken of the dissolver off-gas during the 1st, 3rd and 6th hours of operation. Mass spectrometric analytical results are given below.

	Mole %		
	Sample No. 1	Sample No. 2	Sample No. 3
Co_2	<0.01	<0.01	<0.01
Ar	0.57	0.05	<0.01
CO_2	19.1	17.8	57.0
N_2	48.0	25.9	22.9
CO	3.0	2.5	<0.10
H_2	1.36	6.19	1.42
CH_4	<0.01	<0.01	<0.01
N_2O	1.49	7.2	4.5
NO	26.2	40.4	14.2

Air contains about 0.9% Ar; contamination of the sample by air can be judged by the Ar content. Reasons for the fluctuations in the other values are not known. The hydrogen content appears to be safely below explosive concentrations.

Instrumentation for the Plutonium Reclamation Facility
(O. H. Koski, Chemical Development Section)

The measurement and control of Pu valence state is desired in the control of the Plutonium Reclamation Facility CC column (strip column) and in the CC column product oxidation for recycle to the CA column (extraction column).

Preliminary tests using the Fe(II)-Fe(III) system has shown that oxidation-reduction potential techniques appear applicable. Of most pertinence to column control was a demonstration of a differential oxidation-reduction potential measurement which gave an output which was related to the difference in Fe(II)/Fe(III) ratios in two streams. This system used two low impedance platinum electrodes and a salt bridge. Results suggest that platinum electrodes inserted at selected levels in the column would indicate the comparative Pu valence ratios and hence provide for better CC reductant control. In the system the column contents would act as the salt bridge.

An electrodeless conductivity system for use in the CA column feed acidity control is being constructed. A flanged Kynar cell with temperature compensation and the associated electronics will be supplied. Delivery is scheduled for the middle of November.

Electrodeless Conductivity for Purex Plant HAS
(O. H. Koski, Chemical Development Section)

The electrodeless conductivity system which was provided for the

Pure plant HAS stream malfunctioned in the compensated mode when used with a recorder output. A new unit was provided and the original was examined for defects. Except for a long-term drift of the power supply no defects were found. However, the new unit provided, combined with a cabling installation check by plant personnel, has provided satisfactory service.

A revision of two electrodeless conductivity systems is under way to provide a selectable narrow elevated span operation so as to provide a large output for small changes in conductivity and therefore allow closer control of Purex HAS and HAW stream acidities.

Miscellaneous Instrumentation Studies

(O. H. Koski, Chemical Development Section)

The multichannel analyzer, which was a replacement for a system which became contaminated during in-plant testing, was found defective and returned to the manufacturer for repair. Upon return the system was found to give satisfactory operation.

A selection of newly developed two-wire transmitters for Emf and resistance transmission have become commercially available. They are designed for field installation near the signal source and to operate as signal variable impedance on a two-wire DC transmission line. They are adaptable to most conventional DC current type control systems. Of the transmitters tested, all gave a stabilities on the order of 1/2 percent during a four-month test period. For plant applications the choice of a transmitter for general plant use would require variable span and zero with input-output isolation and operable from a wide range of signal sources impedances. One of the transmitters tested meets all of the above requirements and gives less than 1 percent error for a change in source impedance from zero to 4000 ohms.

Equipment Development - Plutonium Reclamation Facility (PRF)

(J. Dunn, Chemical Development Section)

Centrifugation has been selected as the most promising method for the removal of undissolved solids from the dissolver effluent in the Plutonium Recovery Facility. Initial testing with a special Clarifuge manufactured by L. J. Barrett Co. and modified at Hanford to provide a critically safe geometry has been done with a wide variety of solutions and under different flow conditions. To determine the ability of the centrifuge to handle high flows (2.5 gpm) of the dissolver heel during clean-out, the solids were jetted directly into the bowl instead of being transferred to the bank-tanks. Solids recovery on the first fast transfer was 93.74%. Recycle of the centrifuge effluent at a reduced flow (0.9 gpm) resulted in recovery of an additional 6.07% of the solids for a total recovery of 99.81%. The Clarifuge has been transferred to the PRF for plant use.

Investigations using a second Clarifuge unit (a stock Model 125) employed simulated interface crud composed of Mistron, calcium oxide, silica powder, dissolver fines, nitric acid, aluminum nitrate and carbon tetrachloride. Runs were also made using trichlorethylene as a stand-in for carbon tetrachloride. In each case feed rates were set at 2.5 gpm. The unrecovered solids amounted to about 11% (measured in the centrifuge overflow) and recovery of the organic phase averaged 75%. The only plausible explanation for loss of solvent is evaporation due to the turbulence and high vapor pressure. No organic material was detected in the overflow; all was retained in the bowl.

The Sihi vacuum pump system tested for suitability in PRF transfer applications was shipped to the plant for installation.

Operation of the glass mock up of the PRF plant dissolver has been resumed primarily to generate solids for use in extraction column tests and for centrifuge tests.

TECHNICAL ASSISTANCE TO THE HANFORD PLANT

Summary

Environmental Evaluations

Several radioactive particles were found near switchyards during the annual plant railroad survey. A comparison of aerial survey capabilities between EG&G survey (ARMS) aircraft and the aerial monitoring equipment routinely flown by the Environmental Evaluations section indicated comparable sensitivity.

Dosimetry Technology

A new contract for providing inserts for the Multipurpose TL Dosimeter has been negotiated with Mil-Lor Company. The completed order should be received by November 15, 1971.

The automatic reader for TL dosimeters was successfully demonstrated for U. S. Testing and AEC representatives.

Radiation Protection

A contract was negotiated to re-package used basic TL dosimeters for reuse at Hanford.

The internal and/or external personnel exposures resulting from several incidents were evaluated.

Radiation Standards and Engineering

The mobile tube stand for the new x-ray machine was received and installed. A computer code for determining the radiological consequences of chronic radionuclide releases was completed.

Columbia River Studies

A resurvey of coastal population groups was completed and analyses of the data are in progress. Body burdens of ^{65}Zn measured in the coastal population during the fall of 1971 were about 60% of those measured during the spring of 1970. Estimates of the ratios of measured-to-computed ^{65}Zn body burdens were close to unity. Preparations were completed for a resurvey of Riverview residents beginning November 3, 1971.

TECHNICAL ASSISTANCE TO THE HANFORD PLANT

Environmental Evaluations

(J. P. Corley, Environmental Evaluations Section)

The annual plant railroad survey was completed, using the Environmental Evaluations gamma road monitor. Four particles ranging from 1000 to 50,000 counts/minute (GM) were found at two switchyard points plus one small area reading up to 600 counts/minute on the spurline outside the 300 Area. Gamma spectra of the particles showed cesium-137 and cobalt-60 to predominate. The detected contamination was reported to appropriate contractors for follow-up action. No particles were detected on the 1970 survey.

Detailed comparison of an aerial survey conducted in May for Environmental Evaluations by the EG&G survey (ARMS) aircraft and one performed by the Environmental Evaluations aerial monitor (EEAM) revealed general agreement. However, the EEAM was apparently more sensitive, showing larger meter responses for radiation anomalies detected by both systems. Gamma spectra taken by the EG&G equipment showed ^{40}K and uranium and thorium daughters.

Two mice collected at the U-Swamp in September showed 65 and 25 pCi/g cesium-137, an order of magnitude higher than has usually been observed in small mammals collected on the site. The data are not believed to indicate a significant dose to people.

Airborne particulate radioactivity was significantly lower in September than the previous month, both onsite and offsite. An exception was the 200 East Area North Center location, for which a gross beta activity on air filters continued an upward trend. The maximum value observed during the month was 4.1 pCi/m^3 gross beta, and is believed to be associated with waste-site operations.

Dosimetry Technology

(A. J. Haverfield, Radiological Physics Section, and
K. R. Heid, Personnel Dosimetry Section)

Development of the Multipurpose Dosimeter

A new contract to replace the former contract with PECO has been negotiated with Mil-Lor Company, which provides for new dosimeter cards using the Teflon sandwiches assembled by PECO. A total of 5937 Teflon-TLD sandwiches were sent to Mil-Lor for assembly in new dosimeter cards. The bodies for all the red cards have been molded and assembly has begun. Mil-Lor has also started molding the new gray cards. Target date for completion of the order is November 15.

The operation of the automatic reader was demonstrated for U. S. Testing and AEC personnel. The readout of several dosimeter cards was

accomplished satisfactorily, including readout of dose and payroll numbers. Several cards with calibrated 500 mR exposures were read out and ranges of readings at each position varied by less than ± 10 percent. Each of the five light pipes leading from the TL dosimeter to the photo multiplier tube has its own light transmission factor, so each position has its own calibration factor. The total system seems to be performing about as expected.

Studies of long-term decay of TL signal are continuing.

Radiation Protection

(K. R. Heid, Personnel Dosimetry Section)

A contract was negotiated with Mil-Lor to re-package ∿4200 used Basic TL dosimeter cards which were no longer usable.

Special chest area in-vivo measurements were made on a subject, for whom 4nCi ^{241}Am had been measured in the chest area, to determine if there is any detectable liver deposition. The results indicate the deposition is uniformly distributed in the chest area. There is some indication of possible concentration in the tracheal-bronchial lymph nodes.

Examination of a puncture wound using a NaI wound counter indicated 4.1 nCi ^{239}Pu. This was reduced to 3.7 nCi using epidermal excision techniques. Follow-up examination on Monday indicated 0.2 nCi at the wound site. The employee had been working with gasoline and paint thinner over the weekend resulting in removal of the contamination.

Assistance was provided in evaluating the skin dose received by an employee when the right shoulder of his protective coveralls became contaminated by radioactive material. Based on this evaluation, the employee received a dose of 5 rad to approximately 200 cm^2 of skin.

Problems encountered in the 4-crystal array ^{241}Am, natural uranium, chest counter were resolved by replacing the two back detectors. Recalibration and background subject counting indicate that the system is now stable and the routine programs may be resumed.

In-vivo counts were performed on 196 employees during August including 68 lung and incident-associated counts. Total in-vivo counts to date number 2420.

Radiation Standards and Engineering

(H. V. Larson, Radiation Standards and Engineering Section)

The mobile tube stand was received from the Picker Corporation and was placed on the new balcony platform in the south room of the 3745 Building. Permanent installation of the x-ray tube should be complete

within a couple weeks, pending installation of a raised platform for the transformers and oil cooler. Data gathered during the month indicate that the machine can be used for calibrating the high range instruments, with an effective beam energy of at least 250 keV.

A computer code for determining radiological consequences of chronic radionuclide releases (for Environmental Impact Statements) was completed, permitting the following calculations:

1. Sector average air concentrations based on joint probability meteorological data.

2. Whole body gamma dose from finite clouds for each sector as a function of downwind distance. The gamma dose to tissue may be calculated as a function of tissue depth.

The Emergency Control Center, Room G-52, is no longer being used as a conference room four days a week for the Racial Awareness Seminars. This returns the facility to the personnel responsible for the Emergency Preparedness Program and greatly facilitates its state of preparedness for use in emergencies.

Columbia River Studies

Mechanisms of Environmental Exposure

(T. H. Essig, Radiological Physics Section)

Whole-body counting of previously-surveyed populations in Washington and Oregon coastal communities was concluded on October 5. A total of 109 residents of the Ilwaco, Washington, area and 140 residents of the Rockaway, Oregon, area were counted in the survey. About 3/4 of the total population participating in the resurvey was composed of high school students. Of the 249 persons measured, about 40% represented recounts.

The initial analyses of the measured and computed total body burdens of ^{65}Zn indicate that the body burdens measured during September-October, 1971, were about 60% of those measured during April-May, 1970, and that the ratio of measured-to-computed body burdens was close to unity for everyone except the Rockaway students where it was 2. More detailed analyses of the data may improve the ratio of measured-to-computed body burdens and will permit estimation of the effective half-life (T_e) and the fractional absorption from ingestion (f_w) for ^{65}Zn. The estimation of T_e and f_w for these (or any other) population groups will be a complex iterative process, since these factors are normally separable (i.e., capable of independent assessment) only in a well controlled uptake/retention experiment.

Preparations have been completed for a resurvey of Riverview (Pasco) residents, scheduled to begin November 3. This effort will include fifth and sixth grade students at the Mark Twain School and many Riverview families.

Meteorological Services

(E. H. Phinney, Synoptic Meteorology Section)

Meteorological services, viz., weather forecasts and observations and climatological services were provided to plant operations and management on a routine basis.

OCTOBER, 1971

Type	No. Made	% Reliability
Production	62	84.1
General	62	87.9
Special	157	91.1

Number of calls processed by Code-A-Phone: 4,454