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**Pacific Northwest Laboratory
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38 Pages

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RICHLAND

02 R&D PROGRAM

ANNUAL REPORT
SUMMARY

June 30, 1969

RICHLAND OPERATIONS OFFICE

ATLANTIC RICHFIELD HANFORD COMPANY

BATTELLE-NORTHWEST

DOUGLAS UNITED NUCLEAR, INC.

PREPARED BY RICHLAND STAFF

coordinated by

CHEMICAL PROCESSING DIVISION

and

PRODUCTION REACTOR DIVISION

Classification Cancelled and Changed To

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By Authority of RM-1ten
CG-NMP-1, 8-9-94
By J E Savely 8-18-94
Verified By Jessie Mally,
9-28-94

MASTER

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RICHLAND 02 R&D PROGRAMANNUAL REPORT SUMMARYINTRODUCTION

This document provides, by program, consolidated abstracts of two detailed annual R&D reports which individually cover separations (Atlantic Richfield Hanford Company), and reactors (Douglas United Nuclear, Inc.). Work performed by the Battelle Northwest Laboratory is included with and forms part of the basis for work described in the reports for separations or reactors. The complete annual report consists of this document and the two detailed reports. It is based on the objectives and milestones as listed in the Summary Document for FY 1968 (HAN-98341), in this summary, and the two contractor reports.

The pattern of missions shown on previous semiannual and annual reports have been consolidated into four programs. Under these headings are the subitems recognizable as titles of the former missions:

Basic Production

Product Flexibility

- 2 Tritium (Coproduct)
- 3 Transplutonium
- 4 Plutonium-238
- 5 Other Isotopes
- 7 Enriched Fuels (Oralloy)
- 7 Target Space Enhancement

Waste Management

Environmental, Safety and Regulatory Technology

- 8 Nuclear Safety
- 10 Columbia River
- 9 Radionuclides in the Soil

Estimated expenditures for fiscal years 1969 - 1971 are shown on page 4. The 25 percent reduction in funding has been effected principally in

Note: Numbers denote previous mission designation

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the fields of Basic Production and Product Flexibility, with increased emphasis programmed for Waste Management, Safety and Environmental Studies.

All work under Product Flexibility has been terminated, with the exception of minimal programs in Pu-238, Orallo, Other Isotopes, and Transplutonium. The portion of Columbia River Environmental Studies carried on by Battelle Northwest has progressed to process technology where it will be funded in FY 1970 and FY 1971 from funds programmed to the reactors operating program.

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Page 5

02 R&D PROGRAM COSTS
(thousands of dollars)

	<u>FY 1969(Est. Costs)</u>			<u>FY 1970(Est. Costs)</u>			<u>FY 1971(Est. Costs)</u>		
	<u>DUN</u>	<u>ARHCO</u>	<u>TOTAL</u>	<u>DUN</u>	<u>ARHCO</u>	<u>TOTAL</u>	<u>DUN</u>	<u>ARHCO</u>	<u>TOTAL</u>
BASIC PRODUCTION	<u>983.0</u>	<u>355.0</u>	<u>1,338.0</u>	<u>480.0</u>	<u>255.0</u>	<u>735.0</u>	<u>480.0</u>	<u>250.0</u>	<u>730.0</u>
PRODUCT FLEXIBILITY	<u>1,036.0</u>	<u>175.0</u>	<u>1,211.0</u>	<u>200.0</u>	<u>100.0</u>	<u>300.0</u>	<u>200.0</u>	<u>100.0</u>	<u>300.0</u>
Tritium(Coproduct)	12.0								
Transplutonium		3.4							
Pu-238	75.0	100.2		100.0	95.0		50.0	100.0	
Other Isotopes		50.0			5.0				
Enriched Fuels (Oralloy)	267.0	21.4		100.0			150.0		
Target Space Enhancement	682.0								
WASTE MANAGEMENT	<u>0</u>	<u>1,060.0</u>	<u>1,060.0</u>	<u>125.0</u>	<u>1,300.0</u>	<u>1,425.0</u>	<u>125.0</u>	<u>1,425.0</u>	<u>1,550.0</u>
ENVIRONMENTAL, SAFETY & REGULATORY TECHNOLOGY	<u>906.0</u>	<u>185.0</u>	<u>1,091.0</u>	<u>720.0</u>	<u>320.0</u>	<u>1,040.0</u>	<u>720.0</u>	<u>400.0</u>	<u>1,220.0</u>
Nuclear Safety & Regulatory	606.0	70.1		670.0	70.0		670.0	100.0	1,070.0
Columbia River	#300.0			50.0			50.0		50.0
Radionuclides in the Soil		114.9			250.0			300.0	
TOTAL	<u>2,925.0</u>	<u>1,775.0</u>	<u>4,700.0</u>	<u>1,525.0</u>	<u>1,975.0</u>	<u>3,500.0</u>	<u>1,525.0</u>	<u>2,175.0</u>	<u>3,700.0</u>

200 assigned to BNW. This work will
be process tech funded by DUN or
Work Order to BNW in 1970 & 1971

BASIC PRODUCTION PROGRAM

This program is aimed at providing advanced production reactor and separations technology, applicable to improved plutonium production efficiency and capability. The work involves activity in a wide variety of disciplines, such as chemistry, physics, thermal hydraulics, materials technology and fuels technology. The program spans the range from basic research to inplant development.

OBJECTIVES AND INCENTIVES

Efforts have been and in the future will be directed toward development of new reactor and separations production technology to:

Improve product quality

Reduce costs

Improve maintenance

Increase plant utilization

Extend plant life

Improve plant performance

Reduce product losses

In this way, the Richland complex can continue to perform consistent with or in excess of its commitments.

PROGRESS DURING THE YEAR

Reactors

C and K Reactors

Experimental and analytical programs are underway to assess the fracture potential for the K reactor inlet piping. Fracture toughness tests are essentially complete, and crack growth under conditions of both cyclic fatigue and steady stress is being determined.

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Zircaloy process tube hydriding studies have shown that complete removal of the case layer inhibits further hydriding. The grit blasting process previously developed does not always result in complete case layer removal. A new technique, employing electrochemical anodization, is being developed, for which laboratory data appear promising.

Graphite samples, irradiated in the Engineering Test Reactor (ETR) at K reactor operating temperature, continue to contract, but at a decreasing rate, while higher temperature samples have experienced "turnaround." Extrapolation suggests those operating at the normal K reactor temperature will begin to expand at an exposure equivalent to an additional four to five years of K reactor operation.

A laboratory technique to study localized fuel and tube erosion-corrosion phenomena has been developed and studies under varying simulated process water conditions initiated. Early results tend to confirm the efficiency of 1.0 parts per million (ppm) dichromate as an inhibitor.

On the basis of successful half-plant testing, C and KE reactors have been operated at 0.5 ppm dichromate. Although generally acceptable results have been obtained, some erosion-corrosion has been observed, and further testing will be needed to define conditions under which such corrosion will occur.

K Tube Limits

Extensive physics analysis of reactor kinetics, related primarily to dual riser loss with the E-Q loading (uranium-233 production loading), has been conducted. Calculations, using analytical heat transfer models and measured two-phase flow heat transfer coefficients, of fuel temperature transients following pipe failure have been made. The models have been coupled with others for predicting coolant void-reactivity feedback to analyze associated reactivity transients.

Computerization

The following new codes have been developed:

A program for tube temperature instrumentation calibration.

A program for handling of Equipment Maintenance Standards.

A one-group, three-dimensional diffusion theory code.

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Thoria Sampling Results - U-232 and U-233

Radiochemistry analyses from two redundant sets of thoria samples indicate the yield of uranium-233 per unit of exposure to be less than previously anticipated and the uranium-232 buildup to be higher. The data indicate that an exposure cut of 10 to 15 percent will be required to meet the 8 parts per million U-232 specification, and that an additional five to seven percent loss in U-233 buildup-unit exposure will be experienced.

Reactor Fuels

Studies of uniform corrosion behavior of potential replacement aluminum cladding alloys indicate no significant differences between alloys. Erosion testing is in progress. A program to develop an electroplated nickel corrosion barrier is underway.

An in-reactor test to obtain high heat flux corrosion data using oralloy fuels has been completed. Data are being obtained to validate the corrosion equation.

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N Reactor

Reactor Materials

Small scale testing has shown that it is feasible, using organo-silanes, to apply SiC coatings to reactor graphite which will improve oxidation resistance. Measurements have indicated that the graphite stack is experiencing an oxidation rate of 0.02 percent per year.

Measured vertical contraction of the N graphite stack has averaged less than 0.1 percent for the year.

Zircaloy process tube monitoring continues to show that no loss of mechanical strength has occurred due to reactor operation, but that hydrogen content in the upstream spacer section has increased slightly.

Efforts are underway to improve understanding of the crud deposition and release process, as well as to develop methods for control. Ethylene-diaminetetracetic acid-citric acid-hydrazine solutions appear to have potential as additives in laboratory studies. Screening tests continue on improved single-pass decontaminants.

Ultrasonic surveillance of primary piping has been initiated; no indications of flaw growth have been observed in testing conducted to date.

Industry-wide experience indicates that stress corrosion cracking of Inconel-600 tubing can occur under certain conditions. A program has been initiated to evaluate the situation in the N steam generators for this type of problem. One tube from steam generator 4A with over five years of service has been removed and examined. - Less than one mil of corrosion penetration has occurred on this tube.

Reactor Physics

Physics parameters of the Mark IV loading were determined to be sufficiently close to those of the Mark I, so no new hazards are introduced. MOFDA computer code calculations indicate a slight reactivity loss with the Mark IV load. Incentives for increasing the base level enrichment are under study.

Thermal Hydraulics

Improved methods were developed for generation of technical bases for Zone Temperature Monitor (ZTM) limits, permitting more accurate determination of boiling burnout safety factors. These models have been applied to obtain ZTM limits for the Mark IV fuel.

Thermal hydraulic characteristics of the Mark IV fuel were evaluated in support of the transition to this fuel loading. Imbalance of enthalpy rise among fuel flow subchannels were within four percent of predicted values.

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Instrumentation and Control Systems

An incore flux monitoring system description was completed and functional criteria developed and documented.

Operational data were evaluated to determine the effectiveness of helical sample chambers installed on one of the rupture monitoring system turrets and a production test undertaken.

The ISI-609 computer was programmed to support the V-4 valve maintenance program. Programming for automatic tube power calculation is in process. Two codes were developed for application to analysis of the reactor coolant control systems. An evaluation of the effects of the N Reactor control system on-plant performance was completed. A systems study to determine benefits and costs of a new process computer system is 80 percent complete.

Reactor Fuels

Conversion of the central zone to the new Mark IV fuel was begun in January. Irradiation of test fuel to 3000 megawatt days per ton exposure has indicated a lower swelling rate than for the Mark I.

Irradiation testing of Mark IC fuel fabricated from upset forged and direct cast billets has begun.

Fuel assemblies were fabricated to permit reactor evaluation of Mark IVAA spike fuel thermal hydraulic behavior and the stability of flat and chevron brazed end closures. Unbonded chevron end closures are being prepared for mechanical and thermal cycling tests. Hydraulic testing of the end spider support design has shown promising results, and a relatively low cost stamping technique has been developed to reduce spider fabrication costs.

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PROGRESS DURING THE YEAR

Chemical Separations

Solvent Studies

Prior studies have shown that tributyl phosphate (TBP) is degraded to a greater extent when in dilute solutions, and thus dilute solutions of TBP are being used in the continuing search for causes and mechanisms of solvent degradation. Various aromatic organic compounds have been found successful in reducing the decomposition rate. A three percent TBP solution in diisopropyl benzene, demonstrated severe degradation by concentrated nitric acid. A solution of 10 percent biphenyl and three percent TBP in normal paraffin hydrocarbon (NPH) appears to be stable to chemical attack in fuel separation processes.

Studies were implemented to determine the impurities in NPH and their effect on the extraction process in an effort to improve Purex process performance. Gas chromatographic examination of impurities removed from NPH diluent by activated silica gel indicates at least 15 compounds are present, including aromatics with carbonyl, alcohols and unsaturated groups.

Zirflex Tonic Effect

Efforts were continued to determine the reason for the significant improvement in zirconium and niobium decontamination performance (over aluminum-clad feed extraction) in the first cycle extraction column (HA) when aluminum-clad feed solution is processed with zirconium-clad dissolver solution.

Investigation of the "Zirflex Tonic Effect" indicates reasonably consistent zirconium behavior but erratic niobium decontamination. No improvement in niobium decontamination was achieved by digesting with various complexants (fluoride, sulfate, phosphate, and oxalate), with stainless steel corrosion products, or with a solution obtained from nonirradiated N Reactor fuel.

N Reactor Fuel Processing Studies

Laboratory studies have not defined the reason for the high temperature reactions observed at NFS during the handling and processing zircaloy-clad N Reactor fuels. Literature review leads to the hypothesis that hydride products may be produced (UH_3 and ZrH_2) in ruptured or defected fuel elements.

The addition of small amounts of copper to normal Zirflex decladding solution was found to be effective in suppressing the evolution of hydrogen during Zirflex dissolution without affecting the reaction rate.

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Neptunium Chemistry

In low acid aluminum nitrate solutions, iron (III) was found to oxidize neptunium (IV) to neptunium (V) at rates fast enough to be of use in the Purex neptunium second cycle stripping column. These conditions should provide a procedure which can be used on a campaign basis to separate neptunium from accumulated impurity thorium.

Batch extraction schemes, with a dibutyl butyl phosphonate (DBBP) solvent, for recovery of neptunium from Purex concentrated waste (LWW) and Purex Acid Sludge (PAS) have been developed and tested in bench-scale tests. The feasibility of neptunium recovery from LWW has been successfully demonstrated on a plant-scale with wastes from both aluminum- and zircaloy-clad fuels.

Reusable Plutonium Reduction Vessel Studies

Full-scale reductions in a tantalum crucible have been made. Performance was promising, but further improvements in the crucible are required.

Direct Plutonium Oxide Reduction

About one kilogram of plutonium has been produced by the direct reduction of plutonium dioxide using a magnesium-zinc alloy in a salt matrix process. A tungsten-coated graphite crucible will withstand repetitive heating and exposure to a molten salt mixture (calcium-calcium fluoride-calcium chloride) at 800-1000°C. Batch sizes up to 400 grams of plutonium dioxide have been reduced. The production scale process equipment for denitration and pyroreduction demonstration is being designed.

Plutonium-Uranium Partitioning, Plutonium Scrap Processing

Laboratory and Reclamation plant testing show promise for successful partition of relatively small amounts of uranium from plutonium by flowsheets designed around coextraction-partitioning and partitioning-extraction. Removal of trace plutonium from uranium by use of a silica gel bed appears possible.

Reprocessing of Plutonium-Uranium

Metallurgical Scrap

Studies on oxides produced by burning zero power production reactor alloy fuel scrap (68% UO_2 - 28.6% PuO_2 - 3.4% MoO_3) indicate low plutonium and uranium content in residues remaining after dissolution in nitric-hydrofluoric acid solutions. Complete dissolution was accomplished in some cases.

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Process Materials

Compatibility with the Reclamation process was determined for a number of oils and greases which are under consideration as protective films or cleaning agents for hood panels. Silicone (trade name - General Electric) R-407 and R-855, greases A-07, A-09, oils A-4, A-6, A-18 (Hannford designations for oils and greases), and other were not detrimental to the solvent extraction process at contamination levels up to one percent.

Laminations of Kynar (trade name - PennWalt Corporation) and poly-trifluorochloroethylene film provided improved acid corrosion protection to hood plexiglass windows.

Kynar valves are suitable replacements for stainless steel valves in contact with hydrofluoric-nitric acid solutions.

An aluminum solution made from monobasic aluminum nitrate shows promise as a solvent extraction salting agent.

Process Equipment

Work with the glass replica of the Reclamation Facility Tank 08 slag and crucible dissolver with synthetic ash feeds resulted in revisions to the production equipment. Although major improvements were realized, complete ash dissolution has not yet been achieved.

Control Instrumentation

The variable area flowmeter was operated over the range from 5 to 200 milliliters per minute. However, it is anticipated that alignment and hang-up problems would cause difficulties in remote operation and maintenance of the flowmeters.

The thermal flowmeter gave good sensitivity to extremely low flows. However, the calibration was found to change drastically between flowing organic and water.

A preliminary scope of the mechanical structure, sensor housing and procedural methods for measuring the annulus of a Purex dissolver has been written.

Several methods of solvent extraction column pulse measurement have been tested successfully.

A corrosion probe, which is not cast in plastic, shows promise for use in high radiation fields in existing plant equipment. Three probes are on order.

Analytical Instrumentation

Analytical method development was completed for laboratory determinations of a number of trace elements based on techniques of selective coprecipitation or extraction, with measurement by x-ray fluorescence. A sensitive and readily automated spectrophotometric method for in-line determination of plutonium shows promise for plant application. Considerable progress has been made with preparations to automate either x-ray or gamma-ray spectrometry using a high resolution silicon (lithium-drifted) detector.

A colorimetric ferrous ion monitor has been evaluated and tested in the laboratory on a twenty-four hour schedule. The instrument is specific for ferrous ions in the Purex partition IBX stream.

Tests have been initiated to determine if nitrite concentration can be monitored in the feed to the second plutonium cycle (2AF) by measuring the nitrogen dioxide concentration in the sampler airlift gas.

Tests with samples of cerium-activated Vycor glass suggest that a prescribed depth of grinding could be used to produce a sensor optimized with respect to alpha to beta sensitivity ratio.

The use of lithium-drifted germanium detectors permits gamma energy analysis for plutonium-239 in packages.

Inaccuracy of solution conductivity correlations tend to eliminate this method as a possible measurement of nitric acid in the slag and crucible dissolvers.

An alpha energy analysis resolution of 14.5 keV full width at half maximum (FWHM) has been attained with outstanding stability using the surface barrier-type silicon detector. Analysis of fractional parts per million plutonium-236 in plutonium-238 with stable week long counts were possible.

Nonproduction Plutonium

Tests indicated that pressing and sintering procedures are sensitive in achieving high plutonium oxide densities. A literature search indicates that oxide produced by calcining oxalate at 750° C may have more desirable properties than that produced at lower temperatures.

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PRODUCT FLEXIBILITY PROGRAM

This program consolidates research and development work formerly carried on under the following missions and subitems of missions:

Tritium (Coproduct)
Transplutonium
Plutonium-238
Other Isotopes
Enriched Fuels (Oralloy)
Target Space Enhancement

With the exception of a small amount of work under Pu-238 and Oralloy, and a very minor amount on Other Isotopes, these programs are phased out in FY 1969. The scope and objectives are shown separately along with the progress during the past year under each of the subitems.

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PRODUCT FLEXIBILITY

TRITIUM (COPRODUCT)

SCOPE AND OBJECTIVES

The Richland reactor complex has a wide range of potential production capability to meet varying needs for tritium by using slightly enriched uranium. The objective of the program is to establish an economic and timely productive capacity on which defense planning can rely.

Long-Term Potential and Incentives

The technology developed under this program should provide the capability to produce large quantities of tritium. The production of tritium in conjunction with plutonium in the Richland reactors increases the neutron utilization of the lattice, providing up to 20 percent increase in production when compared on a gram-equivalent basis. Higher uranium enrichment permits a longer exposure for weapon grade plutonium, thus reducing the annual fuel throughput. The increased production and lower fuel throughput provide a substantial decrease in product cost compared to weapons grade plutonium only.

PROGRESS DURING THE YEAR

Reactors

A final report on the N Coproduct Demonstration was issued.

A shipment of the target elements selected for determining N Reactor coproduct conversion efficiency data was made to Savannah River at the end of CY 1968.

The lithium-aluminum (LiAl) elements from isotope control rod No. 27 were discharged with no apparent difficulties. The target elements are to be examined for physical effects prior to their inclusion with other material for normal separations processing.

Shipment of the LiAl target samples from the 1967 small-scale 2.1-spiked K block has been accomplished. Productivity measurements on the samples at Savannah River are expected to be completed by the end of the fiscal year.

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Further analysis and testing of the three splines which were irradiated 43 days indicated they were embrittled to the point where they could not be handled without breaking. Since irradiation periods this long, or longer, would be required in normal usage, it has been concluded that the 3.2 percent LiAl alloy is not an acceptable spline material.

TRANSPLUTONIUM

SCOPE AND OBJECTIVES

The program is directed toward the production of higher weight plutonium isotopes and transplutonium isotopes through the irradiation of transuranium isotopes and plutonium. Specific products which are of prime interest are curium-244 and plutonium containing a higher concentration of either the Pu-240 isotope or the Pu-242 isotope. Because of the potential interest in Cm-244 as a heat source in the late 1970's, primary emphasis will be placed on that isotope. It is proposed to explore and develop the capability for producing these materials in the Richland reactors.

Plutonium can be irradiated in the reactors while they remain basically uranium fueled. However, the plutonium inventory will shift the reactivity and safety characteristics such that safety, operability, and economic evaluations must be performed prior to an assessment of capability. In addition, optimization of fuel design and fabrication is required.

Long-Term Potential and Incentives

The incentives for irradiating plutonium are directly dependent upon the market for the ashes of the irradiation. However, since the plutonium would be utilized as an enrichment for other irradiations, the reactor processing costs will be relatively small. At present the following potential markets are foreseen: Cm-244, californium-252, plutonium fuel of specific isotopic content for the fast reactor development program, and a high Pu-240 content plutonium for exploiting the Phoenix fuel concept. In a Cf-252 program the Richland reactors would be principally useful in producing a satisfactory feed for a high flux or resonance reactor.

The Richland reactors are ideally suited for producing Pu-242, which is an intermediate isotope in the production of Cm-244. The advantages are as follows: First, the higher temperature thermal neutron flux

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of the graphite moderated reactors should maximize the quantity of feed Pu-239 converted to Pu-242; second, the high heat removal capability of the Richland reactors permits burning of the fissionable isotopes, Pu-239 and Pu-241, in a single irradiation; and third, the reactors have a versatile capability for utilizing neutrons produced in the plutonium fuel at the start of the irradiation in producing a variety of products and for adjusting the loading as the plutonium burns out without compromising the overall neutron utilization efficiency. In addition, a substantial portion of the Pu-242 can be converted to americium-243 and Cm-244 directly without an intermediate separation and fabrication step.

PROGRESS DURING THE YEAR

Reactors

The production test of the plutonium-aluminum fuel elements was approved and the elements loaded into KE reactor in February, 1969. The Pu-Al fuel elements were loaded over splines into 13 process tubes as a slab in the central zone of the reactor. These were surrounded by 56 process tubes containing thoria and around these were 156 process tubes containing 94 metal.

Process column power levels measured after startup proved the design calculations to be safe. Splines were removed, and at a later time, thoria was removed to increase operating power levels of the plutonium-aluminum elements.

PLUTONIUM-238

SCOPE AND OBJECTIVES

The objective of this program is to increase the long-term national capability for production of Pu-238 in as efficient and economical a manner as practical while giving full attention to satisfying the short-term market requirements for Pu-238.

This program consists of three principal parts. The first is to maximize the production of the intermediate isotope, neptunium-237, through careful management of the uranium fuel cycle. The second area is the conversion of Np-237 to Pu-238 utilizing the fabrication, irradiation, and reprocessing capability at Richland; and the third area is the production of Pu-238 from the irradiation and reprocessing of americium-241.

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To maximize the production of neptunium, principal effort is placed on better quantification of the conversion rates of Np-237 from U-236, fuel management techniques for maximizing the U-236 in fuel streams within economic limitations, and optimum utilization of uranium fuel in the reactors consistent with production requirements.

In producing Pu-238 from the irradiation of Np-237, several factors must be considered in accelerating the conversion rate while maintaining efficiency. Emphasis will be placed on optimizing the neutron flux level, the related target and reactor load design, and the exposure per cycle. Further, reprocessing steps will be implemented which achieve a minimum out-of-reactor time between successive recycles of the Np-237 and reduce the neptunium and plutonium losses. Optimizing the process will also consider utilizing existing facilities for separations and fabrication in order to minimize costs associated with a production-scale program. Principal future work will be directed to improving the various neptunium and Pu-238 production steps, and reducing the Pu-236 contaminant level to 0.3 parts per million (ppm) or less.

Long-Term Potential and Incentives

The operation of the Richland production reactors results in the production of substantial quantities of Np-237 as a by-product. Since the production of Pu-238 is limited by the national availability of Np-237, there is considerable incentive for adapting feasible and economically attractive methods for enhancing the production of Np-237.

Advantages at Richland for converting Np-237 to Pu-238 include a favorable thermal neutron energy spectrum (which results in a low Pu-236 contaminant level and in a significantly lower ratio of Pu-238 destruction neutron cross section to Pu-238 formation neutron cross section compared to thermal reactors with lower moderator temperatures); the availability of a closely integrated fabrication, irradiation, and separation process with relatively low operating cost (which permits minimum out-of-reactor time for recycled neptunium); and the reactor capability to vary the flux level to optimum values.

Production of Pu-238 from Am-241 provides a potential for substantially increased Pu-238 production capability which will be significant in the middle and late 1970's when quantities of Am-241 from spent power reactor fuels become significant. However, it is expedient that the technology be developed now to provide well-integrated expansion into this production area.

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PROGRESS DURING THE YEARReactors

Irradiation of the 94 metal having high (1100 ppm) U-236 content for Np-237 buildup determination has been completed. Elements are available for separations analysis having calculated Pu-240 buildup of 6, 12, and 13 percent.

Neptunium target elements were charged in a 2 x 2 column cluster during the last quarter of FY 1969 to verify calculated reductions in Pu-236 content. The Pu-236 content is expected to be about a 0.2 ppm level. Thirty target elements of about 40 grams neptunium each were made from the 1200 grams of neptunium authorized for this test. Twelve of these elements were neptunium-aluminum alloy, and the remaining eighteen elements were a homogeneous distribution of neptunium oxide in a graphite matrix. Six of the latter elements were encapsulated in aluminum and the remaining twelve in Zircaloy-2.

The twelve neptunium-aluminum and twelve of the neptunium oxide-graphite elements will be loaded as a 2 x 2 column cluster in the central zone of KW reactor. The remaining six neptunium oxide-graphite elements will be loaded into an outer fringe column to determine the minimum Pu-236 contamination level attainable in the low fast flux portion of the Hanford reactors.

Design efforts have been started on a thin annulus target element with a graphite core. Calculations show this design will produce about the same quality Pu-238 as the homogeneous graphite matrix now in use, but will simplify the separation operations.

Approximately 150 grams of americium oxide was encapsulated by the Livermore Radiation Laboratory. A secondary encapsulation of the target capsules was performed by DUN personnel in the Transuranium Pilot Plant. The eighteen capsules were incorporated with six aluminum dummy capsules into twelve target elements of suitable geometry for K reactors. Each target element also contained a 0.020-inch diameter flux wire of aluminum 0.1 percent cobalt to measure integrated neutron flux. The targets were charged into KE reactor in May, 1969.

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Separations

Twelve hundred grams of neptunium nitrate were converted to oxide for use in the clean plutonium-238 program. The denitration was carried out in a continuous tube calciner. The process was not entirely satisfactory, and further work is necessary.

Limited testing indicates feasibility of a tributyl phosphate extraction scheme which employs dual first cycle extraction columns for recovery and separation of neptunium-237 and plutonium-238. A high acid first cycle flowsheet for coextraction of Np-237 and Pu-238 has been demonstrated in mixer-settler runs with actual dissolver solution. Batch contact data indicate a low acid co-extraction flowsheet can also be devised.

OTHER ISOTOPES

SCOPE AND OBJECTIVES

The purpose of this program is to develop the technology for producing and recovering certain radioisotopes which are not covered elsewhere. Emphasis will be placed on production capability and definition, on minimum cost, and on characterization of the radiation product. Isotopes covered are: thulium-170, thulium-171, polonium-210, cobalt-60, uranium-233 and special fission products. Other activities supported are isotope separation and encapsulation studies.

Long-Term Potential and Incentives

The balanced, long-range interest requires definition of the production capability and product characteristics of reactor products produced in its production complex. This data will help establish firm bases for planning for new production programs, consistent and compatible with market and/or use development.

PROGRESS DURING THE YEAR

Reactors

Uranium-233 Production

Target elements from the current E-Q (uranium-233 production) loadings in the K reactors were analyzed for U-233 yield and U-232 contaminant buildup. Inasmuch as this is now considered a production program, the results of the analyses are included under basic production.

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Physical Constants Test Reactor (PCTR) results on the 1.25 E-Q loading were reviewed and evaluated with respect to more efficient loading patterns.

Thoria wafering technology advanced to the stage where it is now being used on a production basis.

Cobalt-60

A Haynes Alloy 25 (nominally 51 percent cobalt) tube was installed in KW reactor to evaluate cobalt-60 production and to determine if such tubes can be operated without contributing radioactive cobalt to the effluent stream. Irradiations of short sections of the cobalt-plated aluminum process tubes, overlaid with aluminum plating, were initiated in a KE test channel. The Co-59 under irradiation to produce high, specific reactivity Co-60 was discharged from C reactor prior to its shutdown. Calorimetric measurements have been made, and a report is in preparation.

Separations

Isotopic Composition of Rhodium in Current Purex Wastes

Laboratory studies were undertaken to evaluate the rhodium in current Purex waste for feasibility of recovery. The first fission-product rhodium has been isolated from (one-year old) current Purex process acid waste, decontaminated from other fission products and the radioactive composition determined to be significantly less than expected from fission-yield calculations. The Rh-102m (1050 day half-life) is 7 ± 2 microcuries per gram and the Rh-102 (205 day half-life) is 27 ± 10 microcuries per gram.

ENRICHED FUELS - TARGET SPACE ENHANCEMENT

SCOPE AND OBJECTIVES

The prime objective of this effort was the development of fuel and reactor technology to establish an improved irradiation capability to produce special nuclear materials in the Hanford reactors. The

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program did include many activities leading to improved production capabilities, reduced unit cost of non-defense plutonium and overall increased product flexibility. These were concentrated into three areas: 1) the channel enlargement program, 2) the development of highly enriched (Oralloy) fuels, and 3) the development of high power density slightly enriched fuels. A separate discussion for Oralloy is given at the end of this section.

PROGRESS DURING THE YEAR

Reactors

Channel Enlargement

Extensive equipment development and testing have been accomplished in support of the overbore program. The continuous feed graphite boring head and the graphite cutter drive and universal have been fabricated and are undergoing development testing. Designs have been completed for the crossheader dimpler and gas sleeve remover. Feasibility testing was conducted of a vacuum system for moving hot flared sleeves and tube sleeve tips from the work platform to shielded storage.

Thermal hydraulics analysis was completed to determine individual channel protection limits for C overbore fuel (replacement loading for the 44-tube block). The basic technology for a full overbore load in the K reactors was developed from analysis of experimentally determined boiling pressure drop characteristics of a candidate fuel geometry.

Reactivity and control calculations for the C reactor overbore block utilizing a combination of natural uranium and 94-metal were completed.

Summary of reactivity and control parameters influencing channel size and number selection was carried out for the K reactor, and documentation was scheduled for completion at the end of FY 1969.

Recently received Physical Constants Test Reactor (PCTR) test results on the flooded overbore lattice showed good correlation with FLEX and MOFDA computer code calculation predictions.

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High Power Density Fuels

The high power density fuel work has been postponed to a large degree due to the tube power limits at the K reactors because of the brittle fracture situation. Some work was done in the laboratory to determine the subcooled burnout heat fluxes. In addition, criticality data for 150 and 170 metal were developed.

Separations

A rate expression has been derived for calculating dissolution rates of 10 to 40 percent uranium-zirconium alloys using a modified Alniflex process.

Oralloy

Scope and Objectives

The program is designed to develop highly enriched (Oralloy) fuel to be used as the sole fuel or in conjunction with slightly enriched uranium fuel elements (spike concept) to produce a desired product mix in the reactor. Initial work is to be directed toward the spike concept in the single-pass reactors with the objective of developing a loading using 94 or 72 metal with highly enriched fuel to achieve the equivalent of a 210 metal loading. The research and development program to be explored includes:

Reactor life effects from increased heat generation in the graphite moderator.

Nuclear safety studies to define fuel behavior following a loss of coolant.

Fuel design and fabrication development.

Development of both the supercell and the integral fuel-target irradiation modes.

Long-Term Potential and Incentives

The Oralloy program is based upon providing the capability to supply the long-range needs for production of special nuclear materials with the Richland reactors continuing to make a significant contribution.

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The successful development of highly enriched fuel for the K reactors will permit the diversion of a greater amount of reactor production to isotopes such as tritium, Pu-238, or U-233 other than those of plutonium. An additional incentive for the use of highly enriched fuels is the possibility that an inventory charge may be applied to slightly enriched uranium.

Progress During the Year

A study was undertaken to determine the relative total (gamma plus neutron) heating from all sources in an operating reactor. Two types of fueling were considered: K5N and Oralloy. The total heating rate (integrated over volume) in the graphite for Oralloy fuel was calculated to be twice that of K5N Fuel.

Experiments were carried out in the PCTR using highly enriched uranium in a three-to-one supercell lattice with lithium-aluminum targets.

Experiments were also carried out in the PCTR using highly enriched fuel and 94 metal plus lithium-aluminum targets. The data from both of the experiments has been received, and analysis with respect to a water loss incident is in progress.

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WASTE MANAGEMENT PROGRAM

Objectives

Assure the isolation of hazardous radionuclides from life forms for centuries, minimize the long-term cost of storage and surveillance operations, and reduce significantly the amount of radioactivity released to the environment.

Only separations waste management is covered for this report period. Fiscal year 1970 and 1971 will contain a minimal reactors waste management program.

PROGRESS DURING REPORT PERIOD - SEPARATIONS

Low and Intermediate Radioactive Level Waste

A prototype monitoring system for low-level radioactive waste streams was installed on the Purex Plant cooling water line, and evaluation of a polyethylene sample cell is in progress.

Two prototype sample cells fabricated by the single step Kynar (trade name - PennWalt Corporation) process have been obtained for evaluation of applicability to gross gamma monitoring of low- and intermediate-level radioactive waste streams.

Preliminary results indicate that good decontamination of Purex process acid condensate can be achieved in a single pass through an experimental reverse osmosis unit. An unfavorably high concentrate volume to product volume ratio (15:1) was obtained. No plugging or membrane deterioration was detected.

Laboratory studies were conducted to evaluate the flocculation-sedimentation-scavenging characteristics of combinations of hydrous ferric and manganous oxides. A combination (10 milligrams/liter Mn^{2+} , 40 milligrams/liter Fe^{3+} and 2.5 milligrams/liter poly-electrolyte) was found to give excellent flocculation and sedimentation for Purex tank farm condensate. A total beta decontamination factor (DF) of 140 was obtained by successive treatment with flocculant in a tube settler (a mixed media filter), a mixed bed cation exchange column of Amberlite IR-120 (trade name - Rohm & Haas) and Linde AW-500 (trade name - Union Carbide) and an anion exchange column containing hydroxide form Dowex 1X8 (trade name - Dow Chemical).

Chromatographic analyses of gas samples from the Purex vessel vent system obtained for radioiodine trapping studies indicate that the silver reactor is ineffective in trapping halogenated hydrocarbons.

Salt Waste Processing

Techniques have been demonstrated for monitoring and maintaining the strontium extraction capacity of plant solvent. Procedures have

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been devised for fractionating degraded B Plant solvent into a reusable normal paraffin hydrocarbon (NPH) phase and a di-(2-ethylhexyl) phosphoric acid (HDEHP)-tributyl phosphate (TBP) fraction for discard. Recent laboratory data have confirmed that strontium extraction capacity of an irradiated HDEHP extractant is strongly dependent on the presence and composition of an aqueous phase during the radiolysis. Particularly severe effects are observed when the aqueous phase is 1M nitric acid.

Laboratory-scale leaching studies have defined conditions of acidity, time, and temperature to minimize gelation of stored Purex Acid Sludge (PAS) solutions. Beneficial effects of commercially available flocculating agents in enhancing settling rates of water and acid leached sludge have also been demonstrated.

Greater than 98 percent removal of cesium from Purex Current Acid Waste (CAW) by phosphotungstate precipitation was demonstrated in a second plant test. The compatibility of the resulting dissolved cesium product with the Linde Molecular Sieve Type AW-500 (trade name - Union Carbide Corporation) ion exchange recovery process was confirmed both in laboratory and plant tests.

A low priority effort was continued in the laboratory testing of ABEDEM Ti-A (trade name - Seria) resin for the recovery of cesium from current acid wastes (CAW). The process is being investigated as a possible lower cost alternative for the phosphotungstate precipitation process.

Investigation of Polyantimonic Acid ion exchange (trade name - Applied Research Inc.) as an alternate means for strontium recovery from current acid waste (CAW) was started in the laboratory with synthetic CAW solutions adjusted to pH 7 with nitrilotriacetic acid (NTA) and hydroxyacetic acid (HA). Both batch and continuous methods have tested with excellent recovery (95 to 99 percent) of strontium being demonstrated.

Corrosion rates of 304L, Uranus S, and 310L were determined in simulated low-acid waste solutions. Variables included nitric acid and ferric ion content, presence or absence of fluoride ion and solution temperature. Corrosion of all materials was slight at 90° C. However, only Uranus S was entirely satisfactory for containing boiling solution.

Waste Solidification in Tanks

Laboratory studies of air-dried tank 108-SX Redox process sludge were continued. All of the thermal data acquired to date confirm the practicality and safety of storing air dried and air cooled sludge in tank 108-SX.

Correlation of temperature data from underground waste storage

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tanks containing solidified wastes resulted in better estimates of solidified waste thermal properties, allowable heat duties, and the effect of air cooling by force-fitting the computer models to the data.

Waste Packaging

The flowsheets for strontium fluoride (SrF_2) and cesium chloride (CsCl) preparation have been demonstrated and improved process techniques developed where warranted. The three-month duration nonradioactive capsule compatibility tests have been completed. The capsules have been sectioned and are being evaluated by metallographic techniques. Preliminary results indicate that 316L stainless steel and Hastelloy C are satisfactory encapsulation materials for CsCl and SrF_2 , respectively.

Impact consolidation has been chosen as the method for strontium fluoride compaction, and tests have started with material produced in pilot plant-scale equipment.

Vendor pan drying tests on cesium chloride have been completed, and preliminary melt-cast studies are complete. Although both process methods appear acceptable for plant-scale use, the melt-cast is favored and will be developed through the prototype stage.

The thermal expansion of strontium fluoride and cesium chloride relative to the cladding materials was calculated in order to determine the allowable compaction densities. The allowable density was between 70 and 80 percent of theoretical for CsCl and was 96 percent of theoretical for SrF_2 .

Several remote rotary welding concepts have been studied and design of the unit has been started.

Ultrasonic inspections of weld samples prepared in the welding study were performed and defects were observed. Acoustic emission is being investigated as a means for weld testing.

Salt Waste Volume Reduction

Hydrazine-stabilized uranium (IV) nitrate is presently the leading alternate reductant candidate for the Purex process to reduce the salt content of high-level wastes. A satisfactory method of preparing a uranium (IV) solution for Purex Plant partition column (LBX) test was developed. A pilot plant test to demonstrate the path of uranium (IV) in the LBX Column was completed. Uranium (IV) was introduced six feet above the bottom of the LBX cartridge and 78 to 90 percent of it was found in the exit streams, mostly in the uranium-bearing organic stream (LBU). Only 0.4 to 1.4 percent of the uranium (IV) was found in the aqueous plutonium stream (LBXP).

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The platinum-catalyzed hydrazine reduction of plutonium (IV) in nitric acid solution was studied more fully. Results indicate that plutonium (IV) and uranium (VI) were reduced simultaneously by the catalyzed hydrazine reactions. Also, the uranium (IV) which is formed on reduction of uranium (VI) reacts rapidly to reduce more plutonium (IV).

The feasibility of crushing N Reactor type fuel elements has been demonstrated. Because of handling and containment advantages, mechanical crushing has been selected over detonation methods for future refinement. Crushing will permit a reduction in salt waste volumes by eliminating the dissolution operation. However, a removal system must be developed.

A conceptual waste treatment flowsheet for slightly acidic Z Plant aqueous wastes, containing varying amounts of organics, was tested. It was shown that the wastes could be evaporated and calcined to dry, granular material suitable for burial in 55-gallon steel drums.

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ENVIRONMENTAL, SAFETY AND REGULATORY TECHNOLOGY PROGRAM

This program consolidates research and development work for laboratories, separations and reactors on the following general areas:

Safety
Columbia River Studies
Atmospheric Dispersion of Radioactive Particles
Radionuclides in Soils
Seismic Investigations

Nuclear Safety studies continued to provide safeguard guidelines against nuclear accidents. Columbia River studies are aimed at a reduction of radioactivity in reactor effluent, determining mechanisms of environmental exposure and effect of the reactor effluent on the quality of Columbia River water. The portion of Columbia River Environmental studies carried on by Battelle Northwest has progressed to process technology where it will be funded in FY 1970 and FY 1971 from funds programmed to the reactors operating program. Burning studies were continued to determine the Atmospheric Dispersion of Radioactive Particles from contaminated combustible material. Environmental studies of Radionuclides in Soils incorporated a variety of detailed work on movement and distribution for an assortment of radionuclides. Seismic Investigations determined information needed for the safety of separation and reactor plants.

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ENVIRONMENTAL, SAFETY AND REGULATORY TECHNOLOGY PROGRAM

SAFETY - Reactors

SCOPE AND OBJECTIVES

Nuclear Safety studies comprise the broad experimental and analytical effort directed toward assurance against nuclear accidents. A formal objective is the development of technical bases for defining and controlling the consequences of postulated incidents.

Long-Term Potential and Incentives

Previously, hazards evaluations have been based largely on pessimistically conservative estimates of maximum severity and consequences of postulated accidents. This program is designed to substitute facts for those conservative estimates and is expected to demonstrate that the reactors are actually safer than previously indicated. Expected benefits from such a demonstration include greater protection for the public against nuclear accident consequences, enhanced economy in waste management, increased utilization of surrounding land and water resources, and formal compliance to the intent of AEC commercial reactor regulatory standards.

PROGRESS DURING THE YEAR

Reactors

Further insight into the nuclear safety characteristics of the Richland facilities was developed through extensive experimental and analytical studies. Significant program results were as follows:

Fuel Performance in a Loss-of-Coolant Accident

Calculations of the N Reactor lattice heating characteristics with the Mark IV fuel have been completed. The results were used as a basis for recalculating fuel temperature transients after loss of cooling and for recalculating process tube temperatures during normal operation.

The occurrence of an exothermic reaction between uranium and molten aluminum has been confirmed and characterized.

Experimental apparatus to measure the overall heat transfer from a heated full size fuel piece to the N Reactor process tube and graphite has been designed and installed in the BNW Thermal Hydraulics Laboratory.

The flow stoppage event of tube 3560 at the K West reactor was analyzed and evaluated, resulting in significant progress toward our nuclear safety objectives:

1. The temperature excursion following loss of coolant has been characterized based on metallurgical examination.

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2. The postulated interaction of molten aluminum with uranium fuel was confirmed.

The 292-T irradiated fuel testing facility was used to conduct metal-water reaction tests with full sized N Reactor fuel elements.

Construction of the 324-D facility for fuel meltdown experiments has been completed under Project BCP-018. The first metal-water reaction test with high-exposure full-size N fuel was successfully conducted in the facility on June 5, 1969.

Fission Product Transport Studies

Analysis of data from the N Reactor crib indicates that ground water travel times from the crib to the river bank are about 5 days, as compared to previous estimates of 70-100 days. This conclusion has allowed significant revision to routine waste cleanup practices and has stimulated new studies of accident waste handling.

A meteorology data acquisition system for N Reactor has been installed and satisfactorily demonstrated. Analytical codes and associated file processing software have been prepared, and data covering a five months' period have been accumulated.

Three shallow wells have been dug to aid in determining permeabilities as input to a virtually complete ground water flow computer model.

Calculational models were developed for determining the condensation and washout capacity for the non-clogging fog spray nozzles installed in the N Reactor confinement system.

The MELT computer program was refined for evaluation of assumed accident cases involving loss of coolant, and apparatus for experimental confirmation was completed.

In-place testing of the charcoal filter banks in the confinement system has been successfully completed at all reactors, and results are being integrated into Process Standards and routine procedures.

Systems Reliability Studies

Seismic studies and evaluations of regional seismicity by outside consultants are being incorporated into a seismic criterion for the assessment of existing and modified (or new generation) reactors.

A review of the degree of compliance of the K plants to the intent of the 70 General Design Criteria (of 10CFR50) was completed, and a similar study for N plant is nearing completion. Required modifications have been scoped. Periodic reviews have been held with Division of Reactor Licensing, HQ. Key pivotal items have been identified and

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efforts are underway to demonstrate compliance with the intent of applicable regulations.

The quality of the K Reactor coolant supply piping was reviewed, and the limited amount of currently available data indicated a potentially low resistance to brittle fracture. An autonomous program has been initiated to more accurately characterize the piping properties and to design system modifications to alleviate the problem.

Selected critical components (e.g., solenoid valves, control rods) have been tested under simulated conditions of earthquake acceleration.

COLUMBIA RIVER STUDIES

Scope and Objectives

The two Richland K production reactors use treated Columbia River water as a coolant on a once-through basis. For this reason, radionuclides formed largely from activation of river salts and chemicals are released to the river. Heat is released from all reactors. These releases form the basis for a program having three distinct areas of study, namely (1) methods of reducing the radioactivity released, (2) determination of the means by which people might become exposed to the radioactivity, and (3) the effects of heat and chemicals on the river water and possible methods of reducing the effects.

Long-Term Potential and Incentives

Environmental surveillance and other measurements have shown that the effects of the radioactive and chemical discharges on downstream water users are well within AEC limits. Similarly, the discharge of heat has not been shown to have any appreciable harmful effect on fish. However, the increasing emphasis on reducing all types of pollution necessitates an aggressive program in that area. In addition, an understanding of the behavior of the releases and the possible effects in the river is considered vital not only for the Hanford plant but for the nation as a whole.

PROGRESS DURING THE YEAR

Effluent Activity Reduction Studies

1. An apparent increase in the concentrations of several radionuclides prompted the initiation of a program to analyze Columbia River water for 21 elements by neutron activation techniques. In comparison with data obtained in 1962, the present values for Na, Co, and Zn are higher, Ca is lower, and As, U, and Sc are essentially the same.

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2. Ferrous sulfate-potassium permanganate was evaluated as a coagulant in the water treatment pilot plant. The test was terminated due to the inability to control manganese breakthrough.
3. The test to determine the effect of individual ionic impurities on effluent radioactivity by synthesizing process water with additives to dionized water was initiated.
4. The effect of increased coolant turbidity on effluent is being tested on a half-plant at KE Reactor. Initial results showed that increases of a factor of 2, 4 or 6 had little effect on reactor filming and effluent radioactivity.
5. A test is underway in single pass tubes to determine the effect of Zeta Potential on film formation characteristics.
6. Computer Simulation: Four differential equations were developed to describe the variations of both parent and daughter nuclides in the liquid and the film as a function of position and time. Additions of arsenic and bromine were made to the center of a fuel column to test the equation and develop the required constants.

Effect of Reactor Effluent on the Quality of Columbia River Water

1. Improved verification and operational flexibility were added to the basic COL HEAT computer program in order to account for short term transient effects on the order of an hour or less. Specifically, the current regime was better related to the stream tube concept so that exact duplication of dispersion can be achieved by assignment of transport and lag coefficients between and among sections and stream tubes in the processing of the heat and material budget.
2. Verification of the thermal trends using statistical procedures was achieved. The validity of thermal measurements at the Dalles and Bonneville Dams as a basis for drawing conclusions regarding Hanford effects (retention of 40 to 60 percent of Hanford heat in the river as far downstream as Bonneville Dam) was reexamined and found accurate.
3. A study of 1967 river temperatures was used to evaluate the incentives for relocating the temperature monitoring stations immediately above and below the reactors. It was found that locations closest to points of discharge (100°F) were most favorable and that weekly or seven day rolling mean averages were the least restrictive techniques of relating production to the Washington State standards.

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4. The thermal conditions in the mixing zone immediately beyond the discharge point (0-300 yards) were closely evaluated in order to develop a basis for a predictive model of the probability of juvenile fish being swept into high temperature regions of the discharge plume. High dilutions were found and the dangerous temperature zone was determined to be quite small and limited to the immediate vicinity of the discharge point (less than 50 yards downstream).

Mechanisms of Environmental Exposure

1. Measurements of radioactivity in school children and study of their diets were completed at three elementary schools and two secondary schools in the area. About 1500 children and teenagers were contacted in this way. The study also obtained data on the utilization of the Columbia River for recreation.
2. A program was completed in cooperation with home economics classes in local schools to survey the serving sizes of foods taken by members of the students' families. The data helped to fill a gap in the food consumption estimates used in environmental dose calculations.
3. A computer file of whole-body counting data and diet questionnaire results was utilized to calculate average environmental dose for Richland residents. The same file of data was used to identify the appearance of Hanford radionuclides in Richland residents whole-body counting results during the period when Columbia River pumps began supplying most of Richland's sanitary water supplies.
4. Calibration functions related to body size were developed for the mobile whole body counter, including scattering-coefficient functions to correct for Compton scatter from higher energy peaks.

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Separations

Objectives

Determine and explore the parameters for predicting the consequences from planned or accidental release of radionuclides to the environment.

PROGRESS DURING REPORT PERIOD

Atmospheric Dispersion of Radioactive Particles

Burning studies were continued to measure the airborne release of plutonium by using uranyl nitrate and uranium oxide (as stand-in compounds for plutonium) from contaminated combustible materials to postulate fire situations including: waste hutment fires, shipping accidents, metal fires, and explosions involving plutonium compounds in powder form and in solution. Simulation can be used in all cases except for the waste hutment incident. Finely divided uranium dioxide powder was more readily entrained by an air sweep than was uranium nitrate in tests where the flammable materials were either tissue paper or cotton cheesecloth. During burning of impregnated tissue paper, 36 to 55 percent of the uranium dioxide was airborne, and 33 to 38 percent was carried to the filter. In the case of uranium nitrate 14 percent was entrained, and six percent reached the filter.

Radionuclides in Soils

A more rapid, lower cost method for calculating streamlines, permeability and travel times for groundwater were developed based upon continuity of flow. This method replaces the elaborate method of fitting of functions based on energy dissipation.

The permeability distribution for the entire Hanford groundwater flow system was generated.

Contamination patterns and groundwater movement rates were determined for the confined aquifer beneath the Hanford project. Results include a plot of the groundwater potential of the confined aquifer, designation of possible recharge areas, tritium and nitrate contamination patterns, pump test results from selected wells, and recommendations for further study.

Equations describing the rates of reactions of selected nuclides with Hanford soils were incorporated in the transport equation resulting in a set of simultaneous partial differential equations. The set has the capability to handle both transient and steady flow, and sorption precipitation, and competing ion reactions for the important nuclides.

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Work continued on developing a sorption model which describes columns distribution and breakthrough of radioactive macro and microcomponents based on discontinuous, steady thermodynamics of soil-solution chemistry.

Sorption capacity of soil for radionuclides is not affected by residual moisture nor by previous disposals of uncontaminated solutions.

An intensive study of the mechanisms of the downward movement of moisture derived from incident precipitation was initiated during this report period. Results thus far are inconclusive.

Seismic Investigations

Evaluation of the U. S. Coast and Geodetic survey of benchmark changes with respect to the regional geologic structure was completed and documented during this report period. Although slow tectonic deformation appears to have occurred (average of 2 mm per year maximum), no unusual changes or rate of change were noted.

In support of the field study being made by the U. S. Geological Survey relative to possible young faulting on the Hanford site, exploratory excavations removed 50,000 to 75,000 cubic yards of glacial outwash sands and gravels from Gable Mountain and Gable Butte. Areas of concern on Gable Butte were unbroken and continuous through the apparent offset zone. Areas of concern on Gable Mountain appear to be due to differential erosion of folded basalts and interbeds caused by glacial melt water impinging on the toe of the anticlinal ridge. Thus far faulting per se has not been substantial.

Highly sensitive geophones were placed strategically in an array approximately fifty miles in diameter around the periphery of the Hanford Reservation by the USGS National Center for Earthquake Research as part of a program sponsored by the Division of Reactor Development and Testing. Findings are important to separations facilities and long-term radioactive waste storage programs. For the period March 23 through April 23, 1969, fifteen micro-earthquakes have been detected; all were outside of the network.

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