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TITLE

IRRADIATION PROCESSING DEPARTMENT SPONSORED
RESEARCH AND DEVELOPMENT AND
PROCESS TECHNOLOGY PROGRAMS - FY-1966

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IRRADIATION PROCESSING DEPARTMENT SPONSORED
RESEARCH AND DEVELOPMENT AND
PROCESS TECHNOLOGY PROGRAMS - FY-1966

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September 1, 1965

HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

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IRRADIATION PROCESSING DEPARTMENT SPONSORED
RESEARCH AND DEVELOPMENT AND
PROCESS TECHNOLOGY PROGRAMS - FY-1966

I. INTRODUCTION

An allocation of \$300,000 O-2 Reactor and \$115,000 O-2 Metallurgy research and development funds has been made to Battelle-Northwest for FY-1966.¹ Process Technology funds to the extent of \$430,000 for reactor related work and \$45,000 for fuels related work were also assigned to Battelle. The funding was further broken down to the section level.^{2,3} Subsequent to the section breakdown discussions between Battelle and Irradiation Processing Department personnel developed programs within the funding levels. This document briefly summarizes the results of the discussions and delineates the work to be accomplished.

Early in 1965 research and development proposals were prepared using the usual 189 Form format.⁴ The Irradiation Processing Department programs were identified as follows:

- R-1 Radioisotope Reduction Reactor Effluent
- R-2 Nuclear and Radiation Safety
- R-3 Production of Clean Uranium-233
- R-4 Product Development Studies
- M-1 Alternate Cladding Process
- M-2 Fuel Development

Following the submission of the 189 Forms, the integrated five-year research and development program was formulated by the Product Departments with the assistance of Battelle-Northwest.⁵ The program identified the following 15 missions each requiring research and development work:

1. Letter, A. B. Greninger to S. L. Fawcett, "FY-1966 Sponsored Programs - Research and Development and Process Technology," dated July 5, 1965.
2. Letter, R. Nilson to Distribution, "Reactor Research and Development and Process Technology Funds Allocated to Battelle-Northwest," dated August 4, 1965.
3. Letter, J. T. Stringer to Distribution, "Fuels Research and Development and Process Technology Funds Allocated to Battelle-Northwest," dated August 24, 1965.
4. T. W. Ambrose and J. T. Stringer. "Budget for FY-1967 and Revision of FY-1966 Irradiation Processing Department O2 Research and Development," April 12, 1965.
5. RL-GEN-22 REV1. "Richland Integrated Process Development Program," July 30, 1965 plus Supplements 1-15.

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<u>Mission</u>	<u>Title</u>
1	Defense Plutonium Mission
2	Coproduct Mission
3	Integrated Site Operation
4	Non-Defense Plutonium Mission
5	Plutonium Burning Program
6	Pu-238 Program
7	U-233 Program
8	Co-60 Program
9	Po-210 Program
10	High Power Density Fuel Program
11	Small Reactor Modernization
12	Fundamental and General Studies
13	Nuclear Safety Program
14	Columbia River Studies
15	Waste Management; Fission Product Recovery

Although the five-year program has not been officially approved at this time, approval is anticipated. Thus, each area of research and development work is keyed both to a 189 Form and to its appropriate five-year program mission.

Continuing process technology programs of a routine nature such as graphite monitoring (\$30,000), thermal hydraulic testing (\$80,000), corrosion testing (\$30,000), and radiometallurgy support (\$75,000) have not been specifically discussed.

II. RESEARCH AND DEVELOPMENT FUNDS - REACTOR

A. Reactor and Materials Technology

1. Engineering Development

a. Thermal Hydraulics

Funding Level: \$40,000

IPD 189 Form Designation: R-2

Battelle Cost Code: ND

Five-Year R&D Program Mission: 3 and 7

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(1) Self-supported Fuel Studies

A full-scale model of a K-Reactor self-supported fuel column will be used to study the heat transfer conditions at significantly higher specific power. Specifically, the study will include the effect of support height and rear hardware flow resistance on the shape of steady-state boiling demand curves. The effect of coolant mixing in the annulus, as influenced by eccentric positioning of the self-supported fuel elements, will also be investigated. It is anticipated that laboratory tube powers will approach 200 per cent of the base level.

Transient tests are planned to investigate the relationship between fuel temperatures and Panellit pressures following power surges and flow decreases. The present procedures available for calculating the minimum Panellit pressure will be reviewed and adjusted for higher specific powers. The results of this effort will be used to establish tube power limits for self-supported fuel geometry.

The test section for this work is in place, and the program is expected to be completed by January, 1966.

(2) Burnout Heat Flux Program

A program to more closely define burnout heat flux conditions in the present reactor fuel geometry under present and possible future operating conditions is planned. The results of the tests will be used to establish a better burnout correlation which will be used in hazards analysis for the present reactors operating at current and higher specific powers.

It is expected to complete the evaluation of the burnout heat flux for the inner surface of the I&E fuel geometries by the end of the fiscal year. In addition, experimental data will be compiled to obtain preliminary data with respect to the outer or annular surface.

2. Ceramics Research and Development

a. Fuels Design and Evaluation

Funding Level: \$50,000

IPD 189 Form Designation: R-4 and R-3

Battelle Cost Code: NB and NC

Five-Year R&D Program Mission: 3 and 7

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(1) UO₂ Studies

Three columns of aluminum clad UO₂ fuel elements will be fabricated for testing in a K Reactor to demonstrate the feasibility of irradiating oxide fuel elements in an Irradiation Processing Department reactor. The effects of fuel density and specific power will be investigated.

(2) Thoria Studies

Thoria development work will be performed to identify the significant parameters affecting the production of acceptable thoria feed material and to provide information to improve fuel material specifications. Particular emphasis will be placed on gas and moisture content, particle density and particle size.

B. Chemistry

1. Chemical Research

Funding Level: \$70,000

IPD 189 Form Designation: R-1

Battelle Cost Code: NA

Five-Year R&D Program Mission: 14

a. Determination of the Influence of In-reactor Surfaces on Radioisotope Production

Laboratory tests and the experience at the K Reactors have indicated that the type of in-reactor surfaces involved play a significant role in the extent of film formation and the production of radioisotopes which are in the reactor effluent. The studies would include determining the differences in radioisotope production in process channels fueled with nonautoclaved fuel, fuel coated with lithium silicate, and metals such as Zircaloy, nickel, and tungsten. Other coatings considered attractive from an experimental standpoint include the ceramics, e.g., alumina, zirconia, and glass. A detailed inventory of both the fission products and the parent isotopes adsorbed on the fuel element film will be made. Similar inventory determinations will be carried out on representative process tubes. Out-of-reactor studies will demonstrate the effect of coolant velocity and temperature on the amount of film formed per unit area of exposed surface as well as the concentration of parent isotopes in the film.

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b. Determination of the Influence of Ionic Impurities in Reactor Coolant on Radioisotope Production

The relative concentrations of the various radioisotopes in the reactor effluent are found to vary as the concentration of the parent materials in the influent water vary. In some cases it has been demonstrated that it is possible to reduce the concentration of a given radioisotope by increasing the concentration of a seemingly nonrelated isotope. The demineralizing facilities will be operated using the present carbon steel piping. The piping will be changed to stainless steel and base conditions redefined. Following this, chemical additions to the deionized water will be initiated. The first effort will be to define the fraction of P^{32} which results from the irradiation of S^{32} as opposed to that originating from P^{31} . This will be followed by the addition of other chemicals to determine their relationship to the radioisotopes produced as well as the kinetics of parent isotope adsorption and radioisotope release.

c. Determination of the Effect of Water Treatment Process Variables on Radioisotope Production

Studies in the area of water treatment process variables will include the variation of the Zeta potential of the floc to determine the degree of parent isotope removal using activation analysis, the determination of the effectiveness of the nitrate-permanganate flocculation process, and the addition of oxygen-scavenging agents.

d. Determination of the Physical Form of Effluent Radioisotopes and the Effects of Influent Water Composition

An example of the work to be done in this area is making a material balance for the particulate and soluble iron entering and leaving a process tube. The characterization of the physical form of the radioisotopes in the reactor effluent and the characterization of parent material associated with the soluble and insoluble portions of the influent will be investigated. These efforts necessitate the use of activation analyses, membrane filtration techniques, and possibly the use of an ultracentrifuge.

2. Analytical Laboratories

Funding Level: \$15,000

IPD 189 Form Designation: R-4

Battelle Cost Code: NB

Five-Year R&D Program Mission: 5, 6, 7, 8, and 9

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It is anticipated that as further progress is made in the production of materials other than plutonium in the Richland reactors, the need for special analysis and the development of analytical techniques will arise. The funds are assigned to handle such an eventuality, although specific products or programs cannot be defined at this time.

C. Physics and Instruments

1. Experimental Physics Research

Funding Level: \$10,000

IPD 189 Form Designation: R-4

Battelle Cost Code: NB

Five-Year R&D Program Mission: 12

A basic neutron cross-section measurement program is conducted by Battelle-Northwest. Inasmuch as the data generated are used in reactor calculations, these funds are provided in support of the continuing program.

2. Reactor Physics

Funding Level: \$90,000

IPD 189 Form Designation: R-4

Battelle Cost Code: NB

Five-Year R&D Program Mission: 5, 7, and 10

The principal objectives of the physics studies to be performed will be to obtain experimental data which can be used for normalizing mathematical calculations of key safety and conversion rate parameters for new reactor loading proposals.

- a. A series of reactivity measurements of Pu-fueled-thoria supercells will be performed in the PCTR. The measurements will include the wet and dry reactivities, thermal utilization, and flux distribution in the lattice. The data will be used to determine water loss effect of plutonium fuel and as a basis for normalizing computational models for calculating reactivity characteristics and production rate of the plutonium fueled supercell lattice. Dependent on the results of these tests and the priority attached to this program, plans will be formulated for extending the tests in future years to higher plutonium concentrations, and investigations of the effects of melting temperature changes on reactivity.

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- b. A series of reactivity measurements of slightly enriched uranium (~ 1.25 w/o U-235) fueled-thoria supercells will be performed in the PCTR. The measurements will include the determination of the wet and dry reactivities, thermal utilization, and flux distribution in the lattice. The data will be used to extend the calculational reliability for higher enriched supercells and most specifically for determining the reactivity effect of water voiding.
- c. As more specific priorities are assigned to the five-year research and development program mission, plans will be made for measuring temperature effects on reactivity, use of even higher enrichment levels and other target materials, and possibly the use of oralloy fuel.

3. Fuel Cycle Analysis

Funding Level: \$25,000

IPD 189 Form Designation: R-4

Battelle Cost Code: NB

Five-Year R&D Program Mission: 12

These funds are to be used for the generation and development of new concepts which may enhance the flexibility of the Richland reactor complex.

III. RESEARCH AND DEVELOPMENT FUNDS - METALLURGY

A. Reactor and Materials Technology

1. Metallurgy Research

Funding Level: \$10,000

IPD 189 Form Designation: M-1 and M-2

Battelle Cost Code: NK and NL

Five-Year R&D Program Mission: 1 and 10

Laboratory studies will be made to develop new aluminum cladding alloys or coatings for fuel elements which offer potentially improved corrosion resistance under present or more severe irradiation conditions. Both chemical and metallic coatings will be investigated.

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2. Metallurgy Development

Funding Level: \$25,000

IPD 189 Form Designation: M-1, M-2, and R-3

Battelle Cost Code: NK and NL

Five-Year R&D Program Mission: 1, 7, and 10

This work will consist of basic studies: 1) to improve hot-die-sized fuel element and thorium target element closure integrity, and 2) to characterize and define the nickel-diffusion bonds produced by the hot-die-sizing process. Fuel closure development will include studies of optimum end cap design for TIG welding, and the evaluation of alternate end closure welding or brazing methods. Diffusion bond characterization will involve identification and measurement of the intermetallic compounds in the bond layer to increase understanding and ability to specify optimum process parameters.

3. Ceramic Research and Development

Funding Level: \$10,000

IPD 189 Form Designation: R-3

Battelle Cost Code: NM

Five-Year R&D Program Mission: 7

Data will be developed for a thorium surface area off-gas specification as an alternate for the present loss-on-ignition specification, which is unsatisfactory. Samples of thorium powder representing a wide range of surface area will be off-gassed and then exposed to various atmospheres. The amount of gas and moisture adsorbed will be measured to develop the relationship between surface area and gas content. Use of a well-defined thorium surface area specification is expected to prevent excessive pressure buildup from gas evolution during irradiation.

B. Chemistry

Chemical Research

Funding Level: \$35,000

IPD 189 Form Designation: M-1 and M-2

Battelle Cost Code: NK and NL

Five-Year R&D Program Mission: 1 and 10

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Uranium plating studies will be continued to increase the resistance of the electrodeposited nickel plate in hot-die-sized fuel to reactor coolant penetration in the event of a cladding failure. This work will include: 1) an evaluation of techniques for reducing nickel plate porosity, 2) correlation of nickel plate continuity and uranium-aluminum-diffusion, and 3) the development of alternate metallic plates which offer greater resistance to hydrogen and water penetration. Salt-bath plating techniques will also be investigated, including the feasibility of applying corrosion resistant zirconium coatings to the fuel cladding by this method.

C. Physics and Instruments

Instrument Research and Development

Funding Level: \$20,000

IPD 189 Form Designation: M-1

Battelle Cost Code: NK

Five-Year R&D Program Mission: 1 and 10

Studies underway will be continued to develop reliable nondestructive testing methods to measure nickel plate integrity and thickness in finished hot-die-sized fuel elements. A magnetic sensing device is being evaluated for this application. High and low frequency ultrasound techniques will also be investigated as a means of differentiating between mechanical and metallurgical diffusion bonds.

D. Applied Mathematics

Statistics

Funding Level: \$15,000

IPD 189 Form Designation: M-1, M-2, and R-3

Battelle Cost Code: NK, NL, and NM

Five-Year R&D Program Mission: 1, 7, and 10

These funds will be used for consultation and assistance on the statistical design and analysis of complicated fuel and target element evaluation and production tests. Specialized services are required in this area.

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IV. PROCESS TECHNOLOGY - REACTOR

A. Reactor and Materials Technology

1. Metallurgy Research

Funding Level: \$90,000

Battelle Cost Code: NT

a. Process Tube Hydriding Studies

The program begun in FY-1965 will continue to better define the extent of hydriding in the reactors, to determine methods for surveillance of the rate of hydriding, to determine process tube hydriding mechanisms, and to develop remedial actions.

Sets of zirconium alloy coupons of several compositions with varying surface conditions will be charged in the rear gunbarrel of selected process tubes. These coupons will be inserted in both aluminum and stainless steel sample holders. Periodic evaluation of hydrogen pickup will be made. Sequential withdrawal and hydrogen analyses of coupons is expected to become an important factor in estimating hydriding rates and approximate date of initiation of tube hydriding.

Present information indicates strongly that the hydrogen is introduced into the Zircaloy by galvanic coupling of the process tube to the aluminum fuel clad and to the aluminum dummy elements used to center the fuel columns in the reactor. In addition, other mechanisms are being studied. It is extremely important to establish the mechanism responsible for process tube hydriding. Experiments in progress are expected to provide for such a decision.

If galvanic coupling of the Zircaloy to aluminum proves to be the source of hydrogen, remedial action could involve replacement of the downstream aluminum (6063 alloy) dummies with dummies made from some other material or aluminum alloy. Alternate dummy materials will be evaluated, including stainless steel, aluminum alloys, and coated aluminum alloys. A possible solution to the tube hydriding problem may be to rejuvenate existing tubes by removing the hydride by mechanical or chemical methods possibly followed by oxidizing to form an abrasion resistant barrier to further hydrogen absorption. Studies will be made to determine the thickness of oxide film required to confer abrasion resistance to the sliding column of self-supported fuel elements and methods of developing this thickness in-reactor.

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Accomplishments expected in this program for the fiscal year are the definition of the basic mechanism of process tube hydriding and in-reactor demonstration of remedial measures.

b. Corrosion Resistant Coatings

The corrosion resistance of aluminum process tubes may be considerably improved by the addition of corrosion resistant coatings. A minimal program is planned to further develop the diffusion heat treatment of nickel plates and chromic acid autoclaved coatings. Nickel plating techniques to provide a plate which can be subsequently diffusion bonded will be developed and evaluated in reactor tests. Commercial alloys will be investigated to determine those best suited to plating and heat treatment. Chromic acid autoclaving techniques are well established and will be further evaluated by in-reactor testing.

2. Engineering Development

a. Coolant Systems Development

Funding Level: \$40,000

Battelle Cost Code: NT

Studies will be conducted to determine the role of fretting between the aluminum spacers and the Zircaloy tube on the hydriding of Zircaloy process tubes. The parameters of flow, mass of spacer, geometry of spacer, and spacer material will be investigated. Experiments using both coupons and specially prepared spacers will be carried out in laboratory mockups and in-reactor.

Studies initiated in FY-1965 to investigate the effectiveness of inhibitors other than sodium dichromate should continue. An evaluation of aluminum corrosion at high rates of heat transfer should be undertaken. Particular emphasis should be given to determining the causes of and corrective measures for the nonuniform corrosion of aluminum.

Half-plant tests are conducted frequently to evaluate changes in water treatment. The effects of the water treatment changes on the corrosion of reactor components will require evaluation as in the past.

3. Materials Research and Service

a. Graphite Research and Development

Funding Level: \$20,000

Battelle Cost Code: NT

REF ASSIFIED

Tests will be performed to measure the oxidation rates of large graphite blocks at various temperatures in a flowing atmosphere containing various quantities of water vapor. In particular, it is desirable to measure the oxidation gradient through the blocks under these conditions and the effect this gradient has on the block strength since the ultimate objective of the oxidation limit is to limit the reduction in block strength by oxidation. These data are necessary because of the uncertainty of the effects of water vapor in the gas atmosphere on stack oxidation, particularly if higher graphite temperature operation is anticipated.

B. Chemistry

1. Chemical Effluents Technology

Funding Level: \$40,000

Battelle Cost Code: NT

The studies conducted by Chemical Effluents Technology will be in two areas: (1) reactor effluent radioactivity and (2) support for confinement studies and/or hazards analyses.

a. Determination of the Influence of Filtration Variables on the Removal of Parent Isotopes from Reactor Coolant (\$20,000)

It has been shown that the coolant treatment process is quite efficient in removing many of the parent isotopes. However, there is still room for improvement and one of the key areas in the process is filtration. The proposed studies on filtration will include an investigation of floc loading, polyelectrolyte additions, flow surges, and backwash techniques. The chemical aspects of floc formation and adsorption as well as the colloidal characteristics of the floc will be undergoing study. The effect of irradiation on flocculation and settling will be determined.

b. Confinement Studies (\$20,000)

These funds are expected to cover specific experiments, e.g., gaseous release following jacket melting or the melting of Li-Al in a Zircaloy tube, as required in the course of confinement studies or hazards evaluation of alternate product loadings.

C. Physics and Instruments

1. Reactor Physics

Funding Level: \$10,000

Battelle Cost Code: NT

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The reactivity, wet and dry, of the K5E fuel will be measured in the PCTR.

2. Instrument Research and Development

Funding Level: \$15,000

Battelle Cost Code: NT

Nominal support is being given to continued analog computer program development. In addition, requests for nonroutine instrument development are anticipated.

V. PROCESS TECHNOLOGY - METALLURGY

A. Reactor and Materials Technology

1. Metallurgy Research

Funding Level: \$15,000

Battelle Cost Code: NU and NW

Funds will be used in support of out-of-reactor tests to aid in the selection of potentially improved fuel element aluminum cladding alloys or coatings developed for in-reactor evaluation. Physical metallurgy studies will also be made to obtain a better understanding of the hot-die-sized fuel core diameter growth problem during irradiation.

2. Metallurgy Development

Funding Level: \$10,000

Battelle Cost Code: NU and NW

Special uranium, cladding component, and fuel fabrication work will be performed in support of the over-all fuels program that cannot be accomplished in Production Fuels facilities.

B. Physics and Instruments

Instrument Research and Development

Funding Level: \$20,000

Battelle Cost Code: NU and NW

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These funds will be used to cover consultation and assistance in the application of new or improved testing methods developed in the laboratory. Specific applications will include: 1) a gas evolution sensing device to detect pinholes in fuel element end closures during welding, 2) magnetic principal for measuring nickel diffusion bond integrity and plate thickness in finished hot-die-sized fuel elements, and 3) the use of high and low-frequency ultrasound to distinguish between metallurgical and mechanical bonds.

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