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SEMIANNUAL RESEARCH AND DEVELOPMENT REPORT

DOUGLAS UNITED NUCLEAR, INC.

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AUTHOR

T. W. Ambrose

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SEMIANNUAL RESEARCH AND DEVELOPMENT REPORT

DOUGLAS UNITED NUCLEAR, INC.

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9-21-94.

T. W. Ambrose

Research and Engineering

November 30, 1965

DOUGLAS UNITED NUCLEAR, INC.

RICHLAND, WASHINGTON

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SEMIANNUAL RESEARCH AND DEVELOPMENT REPORT
DOUGLAS UNITED NUCLEAR, INC.

INTRODUCTION

Initially the O2 Research and Development Program for Douglas United Nuclear, Inc., for FY 1966 was contained in six 189-forms R-1 through R-4 and M-1 and M-2¹. Although we are currently in a period of transition to the Five Year Research and Development Program², this document reports on the programs as originally set forth.

The subprograms designated by R pertain to reactor research and development, and those designated by M pertain to metallurgy or fuels research and development. The report covers the progress in each of the subprograms on an individual basis for the period July 1, 1965 through October 31, 1965. Although the reports are brief, they are complete. A few of the items are listed in the reports on the subprograms that have been performed on product cost funds but were made possible by earlier research and development effort. The reports listed under Section VI, "Topical Reports Issued,"³ of each subprogram include only those issued since the last semiannual report³.

There are two changes in the estimated expenditure pattern for the balance of the fiscal year. These are as follows:

<u>Program</u>	<u>Previous FY Estimate</u>	<u>Present FY Estimate</u>
R-3 Production of Clean U-233	\$202,000	\$102,000
R-4 Product Development	175,000	275,000

1. Ambrose, TW and JT Stringer, Budget for FY-1967 and Revision of FY-1966 Irradiation Processing Department O2 Research and Development, Undocumented April 12, 1965 (CONFIDENTIAL)
2. Dickeman, RL Summary-Richland Integrated Process Development Program, RL-GEN-00022 REV1 July 30, 1965 (SECRET)
3. Ambrose, TW Semiannual Research and Development Report - Irradiation Processing Department, RL-REA-2156 June 1, 1965 (CONFIDENTIAL)

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Program: 02 Reactor (DUN)
Project: 02 50 10 10
Title: Radioisotope Reduction Reactor Effluent (R-1)
Responsible Manager: T. W. Ambrose
Responsible Investigator: R. G. Geier
Reporting Period: July 1, 1965 through October 31, 1965

	<u>Prior Year</u>	<u>FY to Date</u>	<u>Period</u>	<u>FY Estimate</u>	<u>Previous FY Estimate</u>
Costs (Thousands)	185	82	4 months	216	216
Scientific Manpower (Man-years)	3	1	4 months	4	4

I. PROJECT SCOPE AND OBJECTIVES

The program is designed to develop processes for reducing the amount of radioisotopes present in the coolant leaving the reactors. Areas of study include finding methods for: (1) removing the parent materials responsible for radioisotope formation from the process water, (2) eliminating or reducing the time of adherence of parent materials to the in-reactor films by changes in the chemical constitution of the coolant, chemical form of the parent materials, or changes in the reactor film itself, and (3) treating the effluent to remove the radioisotopes after formation.

II. MAJOR ACCOMPLISHMENTS TO DATE

During the course of the program, several methods have been devised which are capable of reducing effluent radioactivity by a factor of about two for the isotopes of most concern. The methods are:

1. Water treatment using high aluminum sulfate feed rates. This process has been in use since July, 1961.
2. Water treatment using high aluminum nitrate feed rates with nitric acid for pH adjustment.
3. The addition of sodium silicate to process water.
4. The passage of reactor effluent through aluminum beds.

In addition, schemes for effluent disposal by an inland lake and by trenching were conceived.

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III. PROGRESS DURING REPORT PERIOD

1. Laboratory and single-tube in-reactor tests had indicated that the addition of sodium silicate to process water at a concentration of 20 ppm SiO_2 would reduce the amount of radioactivity in reactor effluent. In order to determine the magnitude of the effluent activity reduction as well as the filming and corrosion effects, a half-reactor test was initiated at D Reactor on January 15, 1965. The silicate concentration was increased gradually, and test conditions were established on January 25. Other water treatment parameters such as 18 ppm alum, 1.0 ppm sodium dichromate, and a coolant pH of 6.6 have remained constant since the start of the test.

After an equilibration period of from 4 to 6 months the P-32, As-76, Cr-51, and Np-239 concentrations in the effluent are being reduced by factors of 2-3, 5, 5, and 4, respectively. Isotopes such as Cu-64, Mn-56, Zn-65 and Co-60 are not being affected, and the concentration of Na-24 has been doubled.

The length of the equilibrium period is well in excess of that normally associated with water treatment changes. While the long time may be necessary, it also appears that there might be a seasonal effect involved. Therefore, the test will be continued into the cold water season to evaluate this possibility.

The film from a downstream dummy on the test side of the reactor was found to exhibit a negative zeta potential as compared to the zero zeta potential of the film from a control side dummy. This may be at least a partial explanation of the fact that the isotopes whose concentrations are being reduced are those which exist in the anionic state.

2. Analysis of the river water from B Area continued and has now encompassed one year. Seasonal peaks were observed for iron and phosphate while sulfate remained relatively constant. The analysis for silica was included in September, 1965. Filtered water analyses were terminated because the phosphate results were consistently less than the analytical limit (5ppb) and the river sulfate plus added sulfate were within 1 ppm of the filtered water sulfate.
3. The addition of activated silica to process water showed reductions in single-tube tests equivalent to those obtained when sodium silicate is added.
4. Fuel coated with lithium silicate ($\text{Li}_2\text{O} \cdot 5\text{SiO}_2$) were charged to two single tubes during September. Analytical data on the effluent radioactivity are not available yet.

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5. Preliminary arrangements have been to send fuel elements to ORGDP to determine if metallic coatings can be applied by using plasma techniques. If so, several fuel charges will be coated so that the effect of fuel surfaces on effluent radioactivity can be studied.
6. Contacts have been made and water samples have been made available to ORGDP personnel so that the ultracentrifuge can be evaluated as a research tool.
7. A previous half-plant test at H Reactor demonstrated that if both the bauxite conversion acid and the pH adjustment acid are added through the bauxite conversion box, decreases in effluent radioactivity result. The modification was put into effect at C Reactor on a full plant basis in June. Although data analysis is difficult, it appears that some benefits have been obtained.
8. A program of film analysis to determine the inventory of radioisotopes and parent materials in fuel, tube, and spacer film as a function of fuel exposure, tube age, and location has been initiated. The emphasis to date has been on developing the appropriate analytical techniques such that a large number of samples can be processed efficiently.

IV. EVALUATION OF EFFORT TO DATE

The research and development program to date has encompassed both basic studies and the more practical aspects of the problem, such as the possibilities of making process changes to alleviate the situation. By far the major emphasis was placed on the latter in an effort to obtain results and hopefully reduction in effluent releases as soon as possible. Although some success has been achieved, and the answers to many specific questions obtained, it appears that the more readily applicable changes have been explored. Thus, the program is being redirected toward more basic studies.

V. NEXT REPORT PERIOD ACTIVITIES

1. The determination of the inventory of radionuclides and parent materials in the in-reactor film will continue.
2. In-reactor tests using deionized water and chemical additions thereto will be resumed.
3. The feasibility of applying metallic coatings to fuel elements using plasma techniques will be determined.
4. The usefulness of the ultracentrifuge as a research tool in the effluent activity program will be demonstrated.
5. The role of filtration and filtering techniques in parent isotope removal will be explored.

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6. Additional tests with silicate fuel coatings will be carried out.
7. Tests to demonstrate the effects of zeta potential and flocculation pH will be conducted in the water treatment pilot plant.
8. The half-plant sodium silicate test will be concluded.

VI. TOPICAL REPORTS ISSUED

1. Frymier, J. W. and R. G. Geier, Production Test IP-745 Half-Plant Validity Test, RL-REA-416, January 7, 1965
2. Frymier, J. W. and R. G. Geier, Production Test IP-745, Supplement 1, Half-Plant Validity Test, RL-REA-416SUPP1, March 30, 1965
3. Frymier, J. W. and R. N. Pitman, Final Report - Production Test IP-740 - Phosphate Capture Subsequent to Filtering, RL-REA-2008, May 5, 1965
4. Frymier, J. W., Final Report Production Test IP-745 - Half-Plant Validity Test, RL-REA-2108, May 20, 1965
5. Geier, R. G., Richland Integrated Process Development Program - Columbia River Studies, RL-GEN-00022SUPP14 REV1, July 30, 1965 (SECRET)
6. Geier, R. G., Interim Report I - Production Test IP-728 Half-Plant Sodium Silicate Test, RL-REA-2546, September 16, 1965
7. Silker, W. B., The Contribution of 1706 KE Process Piping to Effluent Radioactivity, BNWL-CC-264, September 22, 1965
8. Silker, W. B., Cross Mixing of Reactor Coolant, BNWL-CC-275, September 30, 1965

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Program: 02 Reactor (DUN)
Project: 02 50 10 10
Title: Nuclear and Radiation Safety (R-2))
Responsible Manager: T. W. Ambrose
Responsible Investigator: R. Nilson
Reporting Period: July 1, 1965 through October 31, 1965

	<u>Prior Year</u>	<u>FY to Date</u>	<u>Period</u>	<u>FY Estimate</u>	<u>Previous FY Estimate</u>
Costs (Thousands)	247	102	4 months	207	207
Scientific Manpower (Man-years)	6	2	4 months	4	4

I. PROJECT SCOPE AND OBJECTIVES

The continuing nuclear safety program has the prime objectives of upgrading the safety status of the reactors and of providing development as necessary to assure that deterioration of the reactor components and proposed operating techniques and conditions do not reduce the safety of the reactors.

The radiation safety program objectives are to establish methods leading to a reduction in the radiation exposure received by personnel and to improve the overall radiation safety aspects of the reactors.

II. MAJOR ACCOMPLISHMENTS TO DATE

1. Computer codes have been acquired or developed, codes have been grouped into chains, and codes have been modified to provide a useful and highly efficient library of codes applicable to nuclear safety studies.
2. Comprehensive theoretical and experimental work has been carried out over a number of years to enable evaluation of the consequences of the loss of coolant accident in the reactors.
3. Comprehensive exponential pile measurements of the multiplicative properties of enriched uranium - water lattices have provided basic critical mass data of importance to safe fuel storage.
4. Hazard Summary Reports for the production reactors have been prepared.

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5. Measurements were made on the control worth of splines and how the worth varied with boron depletion and spline corrosion.
6. A comprehensive analytical review of instrumentation protection has been completed. The review established quantitatively and realistically technical bases for upgraded reactor instrumentation.
7. Unstable boiling and burnout conditions for Hanford process tubes under a large variety of possible operating situations have been identified.
8. The behavior of the fuel columns during flow-loss and power-surge accidents has been established
9. The effect of local disturbances in the coolant flow passage, such as self-supports or bumpers on the fuel surface, on the conditions resulting in local boiling have been evaluated.
10. The effort to improve the understanding of and compensate for moderator contraction has resulted in the following achievements:
 - a. Efficient channel broaching techniques have been developed.
 - b. Projections of future stack distortion have been obtained from experimental irradiations.
 - c. A probe was developed which provides accurate measurement of channel dimensions and distortion.
 - d. A flexible safety rod which can follow channel curvature has been developed.
11. In the area of fixed radiation monitoring instrumentation, a log picoammeter was developed which appears to far surpass anything in the line of area radiation monitoring equipment produced to date.
12. A dose rate telemetering has been developed to a good state of refinement. The unit has the capability of integrating dose rate information to dose.

III. PROGRESS DURING REPORT PERIOD

1. Measurements of the wet and dry k_{∞} of the K Reactor enriched fuel (0.947 w/o per cent U-235) were performed in the Physical Constants Test Reactor. The data when analyzed will be used to normalize calculational models for evaluating reactor safety under current and proposed operating conditions.
2. The mathematical formulation was completed and the computer programming initiated of the transient thermal hydraulics in a fuel tube resulting from a power ramp. The completed code will be used to

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evaluate in detail the fuel temperature, reactivity effects, and tube pressure gauge response to a power excursion incident.

3. A test program is evaluating the deterioration and response characteristics of resistance temperature detectors. In particular, strap-on and immersion type units have been compared and evaluated with respect to meeting nuclear safety monitoring requirements.
4. A study has been performed and limits established for using existing and authorized instrumentation for providing automatic full range power protection for the small reactors such that reactor safety is assured should all safety procedures fail.
5. A two-dimension, diffusion theory, multi-group computer code (EXTERMINATOR) has been procured and modified for the reactors. The code can be used for calculating the reactivity effects of fuel melting, water loss, poison splines, and reactor loading and control arrangement. Use of the code will significantly extend the capability for performing nuclear safety calculations.
6. Thermal hydraulic studies were directed toward determining unstable boiling and fuel burnout conditions for self-supported fuel elements in smooth-bore process tubes at specific powers up to 100% above current operating conditions. Steady-state tests using rods with normal tube-to-fuel centering were completed with both standard and more open tube outlet hardware.
7. The confinement studies have redefined the accidents which would be required to cause fission product release from 1, 50 and 100 per cent of the fuel in the production reactors. The studies have also determined general methods for limiting fission product release to the environs.
8. Studies were begun to develop safe handling procedures for Np-237 materials.
9. Tracerlab, Inc. delivered the six-channel dose rate telemetering prototype system it was contracted to fabricate. The accuracy in reading the true dose above 500 mR/hr is the only thing lacking in the ability of the system to meet specifications. A development test was written for its use for demonstration purposes at KE Reactor.
10. Tests were run at the F Reactor, during the period it was prepared for abandonment, to determine the efficiency of the confinement system. Although all the tests did not produce conclusive results, they did show: 1) excellent effectiveness on the part of fog spray for halogen removal (reduction factor of 500); 2) the fine reliability of the condensation nuclei counter for in-line measurement of the particle penetration of the confinement filters when compared with other measuring methods; 3) that foam generated within the

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exhaust system to encapsulate radioactive gases and radioaerosols has much potential as a confinement aid; and 4) that as much as 86% of iodine released plates out on reactor surfaces and in the exhaust duct.

11. The development of an articulated uncooled horizontal control rod for use in distorted channels was completed.

IV. EVALUATION OF EFFORT TO DATE

The research and development effort in support of nuclear safety has always been of primary importance, and a continuous program has been in progress. The fact that the reactors are operating at many times their design basis without nuclear incident is conclusive proof of the value of the program.

The success of the radiation safety program is evidenced by:

1. Reactor decontamination procedures have reduced dose rates received by personnel working in the discharge area during outages by up to a factor of four.
2. Satisfactory equipment has been made available for replacing outmoded fixed radiation monitoring equipment.
3. Three instruments have been developed for warning personnel of high dose and dose rate conditions. The use of the instruments should help alleviate the need for continuous radiation monitor coverage during radiation zone work performed by operating and maintenance personnel.

V. NEXT REPORT PERIOD ACTIVITIES

1. The measured wet and dry k_{00} of the K Reactor enriched fuel will be included in total and speed-of-control calculations and be factored into the establishment of appropriate limits.
2. The single-tube thermal hydraulics computer code will be completed and used to evaluate the safety protection provided by the individual tube pressure monitor in limiting power excursion.
3. Limits will be established for full range power protection at the K Reactors utilizing the newly installed high speed scanner.
4. The thermal hydraulic studies will be extended to obtain steady-state data at specific powers up to 100 per cent above current values using heater-rods placed 50 per cent eccentric in relation to perfect centering in the process tube. The coolant flow, coolant pressure,

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and fuel temperature transients following simulated flow reduction accidents at higher powers will also be investigated.

5. Confinement studies will be continued and experimental work will be conducted to define the rate of fission product release for the fuel heating cycles expected in loss-of-coolant accidents. Further effort will be applied toward defining the methods for limiting the accidental release of fission products.
6. The field testing of the dose rate telemetering prototype and the pocket dose alarm devices will be completed.

VI. TOPICAL REPORTS ISSUED

1. Scott, H. H. and J. D. Hamric. Kinetics Characteristics of the Ball-3X System at B-C-D Reactors, RL-REA-930. February 10, 1965 (SECRET)
2. Smith, M. L. Development Test IP-739 F Reactor Confinement System, RL-REA-1093. May 25, 1965 (UNCLASSIFIED)
3. Vaughn, A. D. Effective Strength of the Safety System of Hanford Reactor, RL-REA-2326. July 1, 1965. (SECRET)
4. Geier, R. G. Richland Integrated Process Development Program - Defense Plutonium Mission, RL-GEN-22SUP 1, REV 1, July 1, 1965 (SECRET)
5. Leverett, M. C. and R. E. Trumble. Richland Integrated Process Development Program - Nuclear Safety Program, RL-GEN-22 SUP13 REV1 July 30, 1965 (SECRET)
6. Smith, M. L. Development Test IP-739 Supplement 2 F Reactor Confinement System, RL-REA-1093 SUP2. August 5, 1965 (UNCLASSIFIED)
7. Copeland, H. C. Reactor Plant Deactivation Test, 105 F and H Storage Basin Layaway, RL-REA-2536 September 14, 1965
8. Smith, M. L. Confinement Test Results for Development Test RL-REA-1093, RL-REA-2548, September 30, 1965 (UNCLASSIFIED)
9. Windsheimer, W. W., H. C. Copeland, D. C. Greenhalgh, and A. W. Hervin. Reactor Plant Deactivation History, 100 DR, H and F Areas, RL-REA-2686. October 25, 1965
10. Smith, M. L. and G. L. Erickson. Development Test DT-IP-782 Radiation Dose Rate Telemetry and Integration Systems Test at KE Reactor, RL-REA-2093. October 15, 1965 (UNCLASSIFIED)

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Program: 02 Reactor (DUN)
 Project: 02 50 10 10
 Title: Production of Clean U-233 (R-3)
 Responsible Manager: T. W. Ambrose
 Responsible Investigator: W. K. Woods
 Reporting Period: July 1, 1965 through October 31, 1965

	<u>Prior Year</u>	<u>FY to Date</u>	<u>Period</u>	<u>FY Estimate</u>	<u>Previous FY Estimate</u>
Costs (Thousands)	400*	31	4 months	102	202
Scientific Manpower (Man-years)	5	1	4 months	2	4

*Includes \$191,000 0-2 miscellaneous funds.

I. PROJECT SCOPE AND OBJECTIVES

The program is designed to develop and demonstrate the technology required to optimize a process for the large-scale production of uranium-233 containing low amounts of uranium-232. It includes studies to establish thorium powder specifications, investigations to define low cost methods of fabricating thorium target elements for irradiation and to intensively examine the reactor physics and reactor engineering related to thorium loadings in the reactors.

The research and development required to define a thorium dissolution process, a chemical reprocessing scheme, and the conversion of uranyl-233 nitrate and thorium nitrate to other forms is covered in a separate program, of a different Hanford operation.

II. MAJOR ACCOMPLISHMENTS TO DATE

1. The rate of formation of U-232 and U-233 in the reactors has been established by small scale irradiations, and the feasibility of producing U-233 containing only lppm of U-232 has been demonstrated.
2. The effect on U-232 production by thorium-230 contamination of thorium-232 was established. Photoneutron and fast neutron effects contributing to U-232 formation were observed and evaluated.
3. An economical method of fabricating thorium target elements was demonstrated and interim thorium specifications were developed.

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4. The ratios of enriched uranium fuel to thorium targets required to provide proper reactivity balance were demonstrated on a small scale and confirmed for both fringe and core loads during the irradiations to produce 140 kg of U-233.

III. PROGRESS DURING THE REPORT PERIOD

1. The examination of thorium target element failures which occurred during irradiation confirmed that the mode of failure was water entry through imperfect closures. An investigation was initiated to determine the relative integrity of target element closures produced by AC and DC tungsten inert gas welding, inert friction welding, and by a cold forming point-closure technique.
2. A program was begun to obtain data with which to establish optimum product specifications. Variables to be investigated include the correlation of thorium surface area with moisture and gas absorption, particle density, and dissolution rate.
3. Approximately 300 pounds of thorium produced by the GE-NMPO process were received for evaluation. In the process, stream denitrated thorium is mixed with an organic binder, extruded as rod and cut to 1/8-inch diameter pellets, fired at 300 - 1000 C, sintered at 1600 C and crushed to desired particle density. Except for a low compaction density (7.2 g/cc) attributed to particle porosity, the quality of the material was as good as the best Sol Gel material received to date.

IV. EVALUATION OF EFFORT TO DATE

A complete evaluation of the program must await the reprocessing of the 140 kg production effort which will not be completed until early FY 1967. However, as evidenced by the ability to fabricate target elements on a production scale and the results from the irradiation of 6 tons of thorium, which confirm that U-233 containing small quantities of U-232 can be produced economically in the reactors, the effort has been eminently successful.

V. NEXT REPORT PERIOD ACTIVITIES

1. The investigation of alternate end closure and welding techniques will be completed.
2. The evaluation of GE-NMPO crushed thorium will be extended to a five ton lot for ex-reactor and in-reactor testing.
3. A small amount of GE-NMPO thorium extruded to target element size will be obtained for ex-reactor tests.
4. Studies directed toward increasing the compressive strength of target elements will continue.

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5. The correlation of thoria surface area with moisture and gas absorption will be completed. If strong correlations are observed, surface area specifications will be used instead of loss on ignition specifications.
6. Target elements containing thoria having various properties will be fabricated and their dissolution rate determined. The irradiation behavior of target elements which appear promising will be characterized.
7. A physics manual for the production of U-233 in the reactors will be prepared.
8. Since minimum cost U-233, regardless of U-232 content, may be desired, studies will be initiated to determine the irradiation effects in thoria targets subjected to higher accumulated exposures than have yet been encountered.

VI. TOPICAL REPORTS ISSUED

1. Production Fuels Section, IPD. Engineering Specifications, Thorium Oxide Target Element, RL-REA-2177. June 15, 1965
2. Gross, P. D. and K. L. Hladek. PITA-31, Fringe-Blanket Irradiation of Thorium Oxide Supplement VI, HW 84021 SUP 6. July 1, 1965.
3. Woods, W. K. Richland Integrated Process Development Program U-233 Program, RL-GEN-22 SUP7 REV1, July 30, 1965

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Program: 02 Reactor (DUN)
 Project: 02 50 10 10
 Title: Product Development Studies (R-4)
 Responsible Manager: T. W. Ambrose
 Responsible Investigator: R. Nilson
 Reporting Period: July 1, 1965 through October 31, 1965

	<u>Prior Year</u>	<u>FY to Date</u>	<u>Period</u>	<u>FY Estimate</u>	<u>Previous FY Estimate</u>
Cost (Thousands)	227*	128	4 months	275	175
Scientific Manpower (Man-years)	6	1	4 months	6	4

*Includes \$188,000 of O-2 miscellaneous funds.

I. PROJECT SCOPE AND OBJECTIVE

The program is expected to develop the technology for the efficient production of specific isotopes, particularly exploiting the unique advantages of the reactors, and to develop the technological basis for producing a wide variety of products. Irradiation techniques receiving consideration are different fuel - higher enriched metallic uranium, enriched uranium - aluminum alloys, and plutonium-aluminum alloys; the production of plutonium of high Pu-240 content from the irradiation of depleted uranium; the creation of a high flux zone in a reactor for materials testing and rapid product formation; and the use of test hole facilities for special irradiations.

II. MAJOR ACCOMPLISHMENTS TO DATE

1. Studies of Np-237 production in the reactors have led to a quantitative understanding of its formation rates in nonrecycle fuel. Similar studies of Np-237 in recycle fuel have established the advantages of recycle.
2. Studies to determine the production capability of isotopes such as Co-60, Po-210, Tm-170, Cm-242, Cm-244 and Am-241 have been instrumental in critically assessing the flexibility of the reactors and have led to new irradiation techniques of significant importance.

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3. An optimum E-N lattice has been developed through successive irradiation cycles in H Reactor. The loading, which produces tritium and plutonium, has proved to be safe and significantly more productive than a normal tritium producing load (11 per cent).
4. A comprehensive study of an overbored lattice has been carried out. Nuclear safety advantages and improved product yields have been identified and demonstrated. Laboratory measurements verified that at 500 mils overbore the lattice is fail safe on coolant loss. In-reactor conversion ratio tests verified expected gains in plutonium conversion, Np-237 production and higher isotope yields.

III. PROGRESS DURING REPORT PERIOD

1. The irradiation of 22 tons depleted uranium (0.22 w/o U-235) to demonstrate a process for producing plutonium containing a high concentration of Pu-240 was successfully completed at both KE and B Reactors. It is estimated that 50 kg of plutonium containing 25 w/o Pu-240 was produced as compared to the initial program objective of producing 25 kg of plutonium containing 18 w/o Pu-240.
2. Sixty uranium oxide fuel elements (1.3 w/o U-235) were fabricated, and the production test authorizing their irradiation has been approved. The irradiation is expected to start mid-FY-1966.
3. Five columns of metallic uranium fuel (1.25 w/o U-235) were fabricated, and the production test authorizing their irradiation was approved. Uranium alloying requirements were developed for future testing programs.
4. A computational model was developed and used to evaluate recycle techniques for enhancing the production of Np-237 from U-236.
5. A study has been completed and a report issued outlining the Pu-238 and Np-237 production capability. Estimated capital and development costs for implementing the program are included in the study.
6. To confirm theoretical calculations, a small-scale irradiation test consisting of 380 gm of Np-237 has been designed. Target elements are being fabricated for irradiation in a K Reactor. Of the 12 elements to be fabricated, 4 will be irradiated for 3 $\frac{1}{2}$ months and 8 will be irradiated for 6 months.
7. Calorimetric measurements have been completed on overbore-size bismuth target elements. It is concluded from the data that irradiating bismuth in a hard flux spectrum will not increase polonium production rates.
8. Chemical and mass spectrograph measurements have been initiated to determine the Bi-207 and Bi-208 isotope buildup in irradiated bismuth.

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The feasibility of recycling bismuth will depend on the buildup of the two isotopes.

9. The measurements of the wet and dry reactivities of plutonium-aluminum fuel in a super-cell arrangement with thoria have been completed. The results which will be used to normalize computational models are being analyzed.
10. The irradiation of Pu-Al fuel elements for a scheduled 12-18 months was initiated. The irradiations will provide isotopic yield data.

IV. EVALUATION OF EFFORT TO DATE

The program has been successful in establishing incentives in many areas of diversification. Four programs - the tritium producing E-N load, a modernized (0.5 inch overbored) reactor, Np-237 production, and U-233 production are quantitatively understood. Others are in various stages of development. A substantial effort to define the physics, engineering and economic parameters of new processes is anticipated in future years.

V. NEXT REPORT PERIOD ACTIVITIES

1. The irradiation of the uranium oxide fuel elements will be completed and their irradiation behavior evaluated.
2. An evaluation of the depleted uranium irradiation programs for the production of plutonium containing a high Pu-240 content will be completed.
3. The irradiation of five columns of 1.25 w/o U-235 will be completed and tests to evaluate uranium alloys and cladding alloys at 150 per cent of current power will be underway.
4. The fabrication of the Np-237 target elements will be completed and their irradiation initiated.
5. Studies on methods of producing Np-237 target elements on a large scale will begin.
6. The feasibility and problems involved in handling recycle bismuth will be evaluated.
7. Safety evaluations will be directed toward operation with composite loadings of enriched fuel and target elements.
8. The development of lithium bearing splines for producing tritium will be started.
9. The reactivity and flux distribution in a 1.25 w/o U-235 - thoria super-cell will be measured in the Physical Constants Test Reactor.

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10. The capability to shape and control axial flux to increase power density will be investigated.
11. The economic, engineering and physics parameters associated with the production of other isotopes and other modes of operation will continue.

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3. Gross, P. D. and K. L. Hladek. Supplement B to Production Test IP-669-AC Depleted Uranium Irradiation Smaller Reactors, HW-81379 G. June 14, 1965. (SECRET)
4. Gross, P. D. and K. L. Hladek. Supplement 1 to Production Test IP-694-A, Depleted Uranium Irradiation - K Reactors, HW 82744SUP1. June 14, 1965 (SECRET)
5. Fullmer, G. C. and E. G. Pierick. Richland Integrated Process Development Program - Coproduct Mission, RL-GEN-22SUP2REV1. July 30, 1965 (SECRET)
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17. Hladek, K. L. Production Test IP-817, Irradiation of 1.25% Enriched Uranium Fuel Columns, DUN-191. November 1, 1965 (SECRET)
18. Carlson, P. A. and K. L. Hladek. Production Test IP-803, Irradiation of Enriched Uranium Oxide Fuel Elements, RL-REA-2578. November 5, 1965. (SECRET)

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Program: 02 Metallurgy (DUN)
 Project: 02 50 10 05
 Title: Alternate Cladding Process (M-1)
 Responsible Manager: J. T. Stringer
 Responsible Investigator: H. F. Tew
 Reporting Period: July 1, 1965 through October 31, 1965

	<u>Prior Year</u>	<u>FY to Date</u>	<u>Period</u>	<u>FY Estimate</u>	<u>Previous FY Estimate</u>
Cost (Thousands)	245*	71	4 months	302	302
Scientific Manpower (Man-years)	4	1	4 months	4	4

*Includes \$18,000 0-2 miscellaneous funds.

I. PROJECT SCOPE AND OBJECTIVE

Attractive fuel manufacturing incentives and payout exist for replacing the present ALSi process with an alternate hot-die-sizing process. Other advantages of the hot-die-sizing process are: 1) increased flexibility in fuel geometry and length, 2) amenability to mechanization, 3) ease of control, and 4) a more uniform and higher quality fuel to satisfy current or more severe irradiation conditions warranted by product diversification programs.

The program will advance the technology of the hot-die-sizing process. Major areas of investigation include: 1) the effects of uranium and process variables on fuel quality and irradiation behavior, 2) process refinement, 3) establishing the technology to use tubular and potentially more corrosion resistant high-silicon aluminum cladding alloys, and 4) the development of techniques to produce self-supported fuel with integral ribs.

II. MAJOR ACCOMPLISHMENTS TO DATE

1. Equipment has been installed for the development and pilot-scale demonstration of the hot-die-sizing process. Production capability for up to 5,000 fuel elements per month for irradiation was established in February, 1965.
2. Improvements made in uranium core surface pretreatment and plating

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rack design have provided an electro-deposited nickel plate with excellent adhesion properties and uniform thickness, which is relatively free of defects and low in porosity.

3. Process parameters have been developed to produce fuel elements with high integrity uranium-nickel diffusion bonds, having at least twice the tensile strength (26,000 psi) of the best AlSi fuel bonds. End closure quality has been enhanced by increasing cladding component upset in the die sizing step to form a tight friction weld between the end caps and lateral cladding. End closure rejects have been reduced to less than one per cent.
4. A fuel diameter growth problem, experienced during the initial irradiation testing of hot-die-sized fuel, was resolved through the use of uranium cores that are heat treated in hollow blank form and oil-quenched rather than in solid core blank form and water-quenched. Average diameter growth at the mid-point of hot-die-sized fuel elements was reduced from 12 mils to less than 4 mils, as compared to about 5 mils for standard water-quenched AlSi-bonded fuel elements.
5. Because of inherent process advantages in terms of heat conduction, control, and cycle time, induction heating was developed as a replacement for resistance heating in the end bonding operation.

III. PROGRESS DURING REPORT PERIOD

1. Pre-canning examination and testing of natural uranium cores received for a hot-die-sized fuel core optimization test were completed. This test is designed to evaluate the effects of uranium alloying (Al, Fe, and Si at 50, 175, and 300 ppm) and five different heat treatments on irradiation behavior. Uranium cores were also received for a test to compare chloride salt and induction heat treating.
2. Nickel plating studies were directed toward reducing plate porosity. Incomplete laboratory tests indicate that nickel-plated uranium having improved resistance to water (steam) penetration can be produced using a H_2SO_4 -HCl electrolytic etch rather than the conventional H_3PO_4 -HCl electrolytic etch.
3. An investigation of methods to nondestructively measure the thickness of the nickel layer in finished hot-die-sized fuel led to the selection of a magnetic sensing (Hall Effect) device for pilot plant evaluation. This device has an estimated accuracy of ± 0.1 mil for a nickel thickness range of 0.3 to 1.0 mil through 30 mils of aluminum cladding. It is also capable of measuring nickel plate thickness on as-plated uranium cores.

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4. Formal engineering specifications for hot-die-sized fuel were prepared for review and approval.
5. The out-of-reactor corrosion evaluation of high-silicon and aluminum cladding alloys was completed. Based on the results, the alloy containing 10 per cent silicon, 1.0 per cent nickel, and 0.6 per cent iron is being procured for in-reactor testing.
6. A production test (IP-781) to compare the side "hot spot" corrosion resistance of 0.947 per cent enriched hot-die-sized and AlSi-bonded fuel elements was discharged from D Reactor after sustaining three hot-die-sized fuel failures, and one confirmed and one unconfirmed AlSi failure. The hot-die-sized failures occurred at exposures ranging from 420 to 870 MWD/T, and the AlSi failure at about 870 MWD/T. Since all of the failures were attributed to severe ledge and groove corrosion, no statistical conclusion could be reached from the test results on resistance to side failures described by the rupture model.
7. The irradiation of self-supported hot-die-sized fuel elements in the K Reactors indicated that hot-die-sized fuel elements are more susceptible to ledge and groove corrosion of the external cladding in the downstream high temperature positions than AlSi bonded fuel. Examination of hot-die-sized pieces exposed to 50 per cent and 100 per cent of goal in these reactors during September showed groove corrosion on the outer cladding up to 20 mils in depth or about twice as severe and at a higher frequency than observed on AlSi-bonded fuel irradiated under comparable conditions.
8. As a result of the groove corrosion problem, an extensive program was initiated to determine the cause of the observed differences in groove corrosion resistance between 8001 alloy clad hot-die-sized and AlSi-bonded fuel. Investigations include:
 - a. Analysis of the physical, metallurgical, and chemical properties of the cladding.
 - b. Effect of hot-die-size process parameters on cladding grain growth, surface hardness, and possible carbon retention from the die lubricant.
 - c. Effect of fuel design (clad thickness and core diameters), finished dimensions, and irradiation stability.
 - d. Effect of cladding component fabrication and alternate alloys.
 - e. Comparative out-of-reactor corrosion tests on hot-die-sized and AlSi-bonded fuel in a device designed to duplicate groove corrosion by impingement of high velocity (80 ft./sec.) tap water at above 100 C over a small area of the cladding surface.

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Studies thus far have reconfirmed that hot-die-sized fuel cladding has large grains (up to 50 mils diameter) in varying patterns extending from the cap and base ends, as compared to relatively uniform small grains in AlSi fuel cladding. Surface hardness also varies considerably on hot-die-sized fuel. Second phase dispersion in the cladding appears to be uniform, and does not differ between the two types of fuel elements. Analytical samples show some evidence of carbon retention on the surfaces of hot-die-sized fuel from the die lubricant after chemical cleaning.

9. A feasibility study is being conducted by Aeroprojects on possible applications of ultrasonic vibration to diffusion bonding under contract AT(30-1)-2910. This work is directed toward beneficial application of ultrasonic energy in combining the die-sizing and end-bonding operations or to accelerate diffusion bonding in the separate steps.

IV. EVALUATION OF EFFORT TO DATE

Engineering and development studies encompassed by the effort to date have demonstrated the feasibility and versatility of the hot-die-sizing process for fuel production. Definition of process parameters required to support the conversion of fuel manufacturing facilities to the hot-die-sizing process has progressed essentially on schedule. No serious difficulty is apparent in resolving existing or foreseeable problem areas. Although continuing research and development will be required to facilitate smooth production transition, present technology is ample to warrant project authorization.

V. NEXT REPORT PERIOD ACTIVITIES

1. Basic studies of nickel plating will be continued to minimize porosity and reduce plate thickness.
2. Formal engineering specifications for the hot-die-sizing fuel will be issued.
3. Irradiation tests designed to optimize hot-die-sized fuel cores from the standpoint of alloying and heat treatment will approximate goal exposure.
4. Resolution of the 8001 aluminum alloy cladding groove corrosion problem will receive high priority.
5. Preliminary in-reactor testing of high-silicon aluminum alloy cladding will be initiated.
6. Process optimization will be further enhanced through pilot-scale fabrication, irradiation, and evaluation of both natural and enriched fuel models clad in tubular components.

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7. Laboratory studies will be conducted of alternate closure and welding techniques to further optimize end closure quality.
8. Feasibility studies on the beneficial application of ultrasonic vibration in the diffusion bonding of hot-die-size fuel will be completed under an off-site contract.

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2. Strand, C. A. The Effect of Holding Time Between Nickel Plating and Hot Die Sizing, RL-REA-497. January 27, 1965. (UNCLASSIFIED)
3. Strand, C. A. Status of Development Work on the Hot-Die-Size Fuel Element Growth Problem, RL-REA-59REV. February 18, 1965 (CONFIDENTIAL)
4. Baars, R. E. PITA-IP-32, Use of Semi-Production Quantities of Hot-Die-Sized Fuel Elements, RL-REA-42. March 12, 1965. (SECRET)
5. Schweikhardt, G. M. Evaluation of the AlSi Wafer in HDS Fuel Element Production, RL-REA-839. March 15, 1965. (CONFIDENTIAL)
6. Stringer, J. T. Summary of Technical Programs and Progress on Hot-Die-Sizing, RL-REA-928. March 30, 1965. (SECRET)
7. Peterson, D. W. Evaluation of Vacuum Drying and Argon Backfilling of Fuel Elements Manufactured by the HDS Process, RL-REA-2107. May 18, 1965. (UNCLASSIFIED)
8. Strand, C. A. Increased Sizing Speed for HDS Fuel Elements, RL-REA-2086. May 20, 1965. (CONFIDENTIAL)
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11. Bowen, H. C. Corrosion Evaluation of High-Silicon Aluminum Alloys, BNWL-125. September, 1965. (UNCLASSIFIED)
12. Jacky, G. F. Interim Report No. 1, Investigation of Hot-Die-Size Fuel Behavior with Respect to Ledge and Groove Corrosion, Undocumented. September 17, 1965. (UNCLASSIFIED)

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13. Jacky, G. F. Interim Report No. 2, Investigation of Hot-Die-Size Fuel Behavior with Respect to Ledge and Groove Corrosion, Undocumented October 4, 1965. (UNCLASSIFIED)
14. Strand, C. A. Improve Weld Closure of HDS Fuel Elements, RL-REA-2623. October 12, 1965. (UNCLASSIFIED)
15. Strand, C. A. Hot-Die-Sizing Process Development Work-Ledge and Groove Corrosion, RL-REA-2690. October 10, 1965. (UNCLASSIFIED)

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Program: 02 Metallurgy (DUN)
 Project: 02 50 10 05
 Title: Fuel Development (M-2)
 Responsible Manager: J. T. Stringer
 Responsible Investigator: J. T. Stringer
 Reporting Period: July 1, 1965 through October 31, 1965

	<u>Prior Year</u>	<u>FY to Date</u>	<u>Period</u>	<u>FY Estimate</u>	<u>Previous FY Estimate</u>
Cost (Thousands)	141	34	4 months	198	198
Scientific Manpower (Man-years)	2	1	4 months	2	2

I. PROJECT SCOPE AND OBJECTIVE

About one-third of the funds for this project will be used for the "C" Basin examination of fuel irradiated under development programs. Other work under this project includes: 1) basic studies of uranium additives, heat treatment and fabrication to provide the technology to produce relatively stable fuel elements at minimum cost to satisfy present or more severe irradiation conditions under product diversification, 2) investigation of new cladding alloys or coatings offering potentially improved corrosion resistance, and 3) basic studies required to resolve fuel process or performance problems.

II. MAJOR ACCOMPLISHMENTS TO DATE

1. An evaluation of induction heat-treated, oil-quenched uranium cores showed no significant difference in the dimensional behavior of this material and standard salt heat-treated, water-quenched cores.
2. An initial test to compare the dimensional stability of water-quenched six-inch and ten-inch enriched fuel cores showed no significant difference in average warp (about 5 mils), and only minor differences in diameter change. Excessive warp was experienced in the canning of 12-inch, natural fuel cores, which was attributed to the need for mechanical straightening the core blanks after heat treating.
3. Advances in nondestructive testing technology have resulted in improved tester reproducibility and accuracy.

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4. A stronger arch-rail was developed and accepted as a replacement for the former collapsible-bridge rail on all self-supported fuel models.
5. Fuel evaluation studies have shown a strong correlation between the rupture resistance of non-projection fuel and fuel warp and total bond count.

III. PROGRESS DURING REPORT PERIOD

1. Post-irradiation measurements were continued in "C" Basin in support of fuel development programs.
2. An irradiation test was charged in D Reactor to evaluate the dimensional behavior of oil-quenched, 10-inch, enriched (0.947 w/o U-235) uranium fuel elements.
3. The economics for longer fuel elements were reviewed, taking into account revised cost estimates received from the uranium feed site, to update justification and establish the priority for future work on this program. Substantial Hanford economic savings will result from the use of longer fuel, and it is planned to carry out future development in conjunction with the hot-die-sized fuel program.
4. Initial test quantities of I&E fuel cores were hollow, alpha-extruded using the N-Fuels press, machined into core blanks, and heat treated. Pre- and post-heat treatment measurements indicated no difficulty in retaining dimensions within tolerable limits, and possibly extruding to or near finished fuel core dimensions. The inside diameter of the core blanks extruded over a mandrel increased eight mils during heat treatment, and those extruded by a filled (mild-steel graphite) billet technique increased two mils.
5. Pilot-scale evaluation of induction heat treated oil-quenched uranium was continued with favorable results.
6. A test to determine the effects of uranium additives (Al, Fe, and Si up to 300 ppm) on the dimensional behavior of AlSi-bonded fuel has been deferred pending the results of a similar irradiation test on hot-die-sized fuel cores. A test to determine the effect of increasing the as-heat treated solid core blank diameter 27 mils on fuel stability is ready for charging in a small reactor. Allowing for uranium warp during heat treatment, use of larger core blanks provides a more uniform texture in the machined cores.

IV. EVALUATION OF EFFORT

Advances in basic technology falling within the scope of this program have resulted in improved fuel quality and performance. Future development is oriented toward further improvements in fuel uniformity, irradiation stability, and lower fabrication costs.

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V. NEXT REPORT PERIOD ACTIVITIES

1. Sponsored research and development of new aluminum cladding alloys or coatings offering improved corrosion resistance will be continued.
2. Fuel examinations will continue in the "C" Basin examination facility in support of development programs.
3. An irradiation test to evaluate 10-inch, enriched, oil-quenched fuel cores will be completed.
4. Approximately 1,000 hollow, alpha-extruded, uranium cores will be fabricated for out-of-reactor evaluation and irradiation testing.
5. Test quantities of electro-reduced uranium will be obtained from Mallinckrodt Chemical Works for preliminary evaluation.
6. Irradiation of fuel cores fabricated from larger diameter solid core blanks will be completed.

VI. TOPICAL REPORTS ISSUED

1. Bowie, W. & G. L. Hammons, Interim Report, Quality Certification Program, RL-REA-2034. May 3, 1965. (CONFIDENTIAL)
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