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REACTOR AND METALLURGY R&D PROGRAMS
IRRADIATION PROCESSING DEPARTMENT
FY-1963 - FY-1964

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IRRADIATION PROCESSING DEPARTMENT
FY-1963 - FY-1964

by

O.H. Greager

September 14, 1962

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I. INTRODUCTION

This document describes the objectives, targets and scope of the development programs to be sponsored and conducted by the Irradiation Processing Department in support of the Hanford production reactors during FY-1963 - 1964. It is anticipated that the basic funding level for the IPD Reactor and Metallurgy R&D programs for the next two years will total \$6,930,000 annually including overhead. Approximately 70 per cent of these funds will be directed in support of the N Reactor, and the remainder in support of the older reactors. The funding levels for the various reactor and metallurgy programs requiring technical support are reflected in HW-72790, "Budget Proposals for 02 Program Research and Development, FY-1963 and FY-1964, Irradiation Processing Department", O. H. Greager, April 23, 1962. This latter document also contains Supplementary Budget Proposals to the Commission for work associated with the Plant Improvement Program for the older reactors and alternate production concepts for the N Reactor, which require funds in addition to the basic program. For current planning purposes, however, the technical efforts described within this Program Document have been restricted to those within the basic funding levels, and if Supplementary Programs are authorized, adjustments will be made accordingly.

II. FUNDING ALLOCATIONS

It is anticipated that the following direct fund allocations will be made to carry out these R&D programs for FY-1963.

The Metallurgy allocation to HLO will be via FPD. In addition, HLO will be allocated \$360,000 Process Technology funds from IPD for those aspects of development activities which are in direct support of the production program.

<u>Component</u>	<u>Funds - Thousands</u>		
	<u>Reactor</u>	<u>Metallurgy</u>	<u>Total</u>
<u>IPD</u>			
R&E	\$ 801	\$ 585	\$1,386
FES	221	0	221
NRPSP	600	0	600
Total IPD	<u>\$1,622</u>	<u>\$ 585</u>	<u>\$2,207</u>
<u>FPD</u>	\$ 0	\$ 795	\$ 795
<u>HLO</u>	<u>\$1,325</u>	<u>\$1,300</u>	<u>\$2,625</u>
Total Direct	\$2,947	\$2,680	\$5,627
Overhead (IPD)	680	623	1,303
	<u>\$3,627</u>	<u>\$3,303</u>	<u>\$6,930</u>

III. SUMMARY OF DEVELOPMENT SCHEDULES FOR MAJOR R&D EFFORTS

Following is a summary of the major targeted R&D accomplishments for FY-1963 and FY-1964 necessary to support the basic IPD production program.

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A. Effluent Control

By December, 1962, establish the most effective methods for reduction of radioisotopes in water discharged from the reactors to the Columbia River, and by July, 1963, determine the most effective methods to obtain a factor of ten reduction in the radioisotopic content of the reactor effluent.

B. Nuclear Reactor Safety

Upgrading of the zone temperature monitors at the six older reactors will commence in FY-1963 to enable placing this system in the safety circuit. Project action will be initiated early in FY-1963 to place intermediate log N flux monitors and power-rate-of-rise meters in all eight reactors. Development and testing will continue through FY-1964 on in-core flux monitors. Installation of a high speed scanner for temperature monitoring at one K Reactor will begin in FY-1963 if the project for this action is approved.

C. Hazards Reviews

Rough drafts of updated hazards reports will be completed for the eight existing Hanford production reactors by mid-September, 1962. A rough draft of Volume II, Hazards Review for the N Reactor will be completed in July, 1962, and a final draft will be completed in August, 1962.

A Supplementary Hazards Review for the N Reactor will be available in outline form by July, 1963.

A draft of N Reactor Technical Specifications will be completed by December, 1962.

D. N Reactor Startup and Operation

All phases of the Reactor and Metallurgy development efforts which are necessary to provide technical support for the various phases of N Reactor startup and operation, including Acceptance Test Procedures, physics tests, low power operation, and power ascension, will continue with high priorities. N Reactor startup is anticipated in October, 1963.

E. K Reactor Zircaloy Retubing

Provide adequate technical support to initiate retubing the K Reactors with Zircaloy tubes in the period October - December, 1962.

F. C Reactor Overbore Test

Provide continuing technical support to the overbore test facility at C Reactor to obtain fuel performance data, conversion ratio data, and other data necessary to provide sound evaluations of the technical and mechanical component requirements for this production improvement concept.

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G. E-N Loading

Obtain and interpret additional data by December, 1962, to provide a complete technical evaluation of the E-N loading demonstration test which was conducted at H Reactor.

IV. PROCESS CONDITIONS AND OPERATING ASSUMPTIONS

The following illustrate maximum process conditions required to meet scheduled production rates for the existing reactors during FY-1963 - 1964.

A. Bulk Outlet Temperature

A maximum of 95°C bulk outlet temperature may be assumed for all reactors.

B. Reactor Flow Rates - Thousands gpm

<u>Reactor</u>	<u>Flow</u>
B	90
C	102
D	88
DR	88
F	90
H	88
K	208

C. Power Level Capability - MW (Based on 5°C Inlet)

<u>Reactor</u>	<u>Level</u>
B	2130
C	2420
D	2090
DR	2090
F	2130
H	2090
K	4930

$1 \text{ gal/min} = 63.1 \text{ cm}^3/\text{sec}$
 $102 \times 10^3 \times 63.1 \text{ cm}^3/\text{sec} \times 90 \text{ sec} \times 4.18 \text{ J/cal} = 2420 \text{ MW}$

Note that these power levels exceed the current administrative limits, and assumes relaxation of the limits after appropriate reviews by the various reactor safety groups.

D. Specific Power of Fuel - KW/ft.

Assuming achievement of the above process conditions, the following typical values for maximum fuel element specific power KW/ft., are to be anticipated.

<u>Reactor</u>	<u>Specific Powers</u>
B	90
C	95
D	90
DR	90
F	90
H	90
K	100

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- E. Goal discharge levels of fuel elements will be maintained at values consistent with the product specification of six per cent Pu-240.
- F. It is anticipated that the central zone process channel at the K Reactor will be refitted with Zircaloy-2 process tubes late in CY-1962, and will require the utilization of self-supported fuel elements.
- G. During FY-1961 - 1962, an overbore test facility was established at C Reactor in which large diameter, self-supported fuels were irradiated in smooth-bore process tubes. Maximum specific power of these fuel elements are on the order of 120 KW/ft. Fuel element testing in this facility will be continued during FY-1963 - 1964.

V. REACTOR PROGRAMS

A. Nuclear Physics

The broad objectives of the Nuclear Physics R&D studies are to provide sound and adequate experimental data with comprehensive interpretation of results and correlation with theory to enable a better understanding of the fundamental nuclear process in the Hanford production reactors.

1. Old Reactors - FY-1963

The bucklings of the C-pile mockup with present C-pile fuel will be measured with 0, 1, and 6 control rods for water cooling and then in the flooded pile condition with 0 and 6 rods. The bucklings with overbore fuel will be measured with 0 and 6 control rods for water cooling.

The thermal diffusion length and thermal neutron extrapolation distance into a control rod will be measured in the C-pile mockup.

The change in k_{∞} of the K-pile lattice upon loss of coolant will be measured with zirconium tubes and the latest fuel elements late in the year.

The change in reactivity associated with the irradiation of K pile splines will be measured in the PCTR.

Resonance activation traverses will be made in a large I&E fuel element (2.5" x 1.6") to obtain the relative surface absorption with variable water inside and outside the fuel element. Several foil traverses will be made to compare to thermalization and transport codes.

Measurements of the diffusion length will be made in a light water-graphite system, both parallel and perpendicular to the water channels. Such measurements will be helpful in determining the diffusion coefficient for a reflector and lattice.

Measurements of the differential scattering of slow neutrons from water will be continued. Preliminary emphasis will be placed on the study of large energy change scattering as a function of water temperature (20° to 100° C).

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Attempts will be made to provide a new beam hole facility to obtain differential scattering cross sections by rotating crystal time of flight techniques during FY-1963. This facility will extend the range of present measurements to lower neutron energies and larger momentum exchanges.

2. Old Reactors - FY-1964

The bucklings of the C-pile mockup with present fuel will be measured with 0 and 6 control rods for air cooling.

Analysis of the data taken in FY-1963 on loss of coolant from a K-pile lattice with zirconium tubes will be completed.

Neutron thermalization and spectra in a graphite-water absorbing rod system will be studied, supported in part by funds from other programs.

The possible methods of obtaining cross section information on U²³⁶ will be studied. If any methods are usable and foil material is available, this information will be obtained.

Diffusion length measurements in water-graphite systems will be continued during FY-1964 and will possibly include measurements with aluminum or iron rods inserted in a lattice array.

Measurements of neutron scattering from water at elevated temperatures will be continued.

3. N Reactor - FY-1963

Data from previous exponential pile runs will be re-analyzed with best analytical methods to obtain best values of the pile bucklings. The control rod data will be analyzed to obtain the extrapolation distances into the rod.

Data will be obtained so that the strengths of the control rods in the flooded exponential pile may be obtained. Flooding will be simulated by placing polyethylene around the process tubes and masonite in the steam vents.

A report on the first N Reactor measurements will be issued early in the year. A second report on the control rod strength and the buckling of the flooded pile will be issued late in the year.

4. N Reactor - FY-1964

Measurements of bucklings with Mark II fuel elements will be made with 0, 1, and 6 control rods in the wet flooded pile, with process tube flooding and with flooding also in the steam vents. The same measurements will be repeated for the wet pile with control rods, and with 1 and 6 columns of ceramic balls.

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B. Applied Physics

The following programs in Applied Physics are applicable to the old reactors and the N Reactor during FY-1963 - 1964.

1. Neutron Spectrum Studies

Theoretical investigations of the neutron spectrum in both homogeneous and heterogeneous systems will continue. Description of crystalline and molecular binding effects by use of the modified heavy gas scattering kernels will be further explored. Application of a workable generalized multi-group method of analyzing a heterogeneous cell containing multi-temperature regions will be demonstrated in FY-1963. This method will be applicable to calculations of temperature effects and thermalization properties of different regions. This work will be partially supported by O4 program funds.

2. Digital Computer Codes for Reactor Analysis

Efforts will continue toward providing, maintaining, and improving a comprehensive system of digital computer codes for physics analyses of reactors. Evaluation of these codes and of cross section sets for the codes will continue. Potential HAPU users will be provided with these evaluations and with instructions and assistance in operating the codes.

3. Computational Programming Services

Digital computer programming services in support of the efforts of other physics components, in particular the Nuclear Physics Research Operation of HLO and Operational Physics and Reactor Physics of IPD, engaged in O2 Program work will continue. These services include preparing codes for processing and correlating experimental data, modifying codes for special studies, and assisting personnel of other components in the use of these codes.

C. Reactor Physics

1. Old Reactors

Analytical studies will continue on improving the reactor kinetics models used in determining speed-of-control limits, nuclear instrumentation response times and trip settings, and safety rod inoperability limits. Hazards studies associated with reactor modifications such as overbore, retubing with Zircaloy-2, E-N loadings, etc., will be conducted as required. Basic analytical physics studies will continue to evaluate temperature coefficients, reactor control, and other physics aspects related to nuclear safety.

In-reactor tests will be conducted to evaluate reactor loadings to accommodate partial or total xenon override.

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Final analytical results will be obtained on the conversion ratio and product buildup from the E-N demonstration loading at H Reactor, and analytical studies will continue on the recycle program.

Analytical studies directed toward a consistent set of criteria for nuclear instrumentation will be continued. Analog computation services will be provided in support of physics studies relating to nuclear safety and process improvement.

Shielding studies will be conducted utilizing the DR Bulk Shielding Facility, to evaluate shielding types which may be of interest to Hanford. Experimental data will be correlated with theoretical considerations.

2. N Reactor

Analytical work required for N Reactor Process Standards, startup physics tests, kinetics studies, and safety aspects of improved Phase I fuel elements will be continued.

Experimental lattice physics measurements will be analyzed and results correlated with present theoretical models. Extrapolation to a Mark II fuel design based on these correlations will be made and checked by further experiments. In-pile conversion ratio measurements on heavier fuel elements will be made after N Reactor startup.

Simulator studies will be carried out on the N Reactor Simulator to study the performance of the complex N Reactor Systems. These engineering and physics studies will be designed to improve operating methods and to determine consequences of various operator actions and errors.

D. Heat Transfer and Hydraulics

1. Old Reactors

- a. Visual studies performed in the heat transfer laboratory during FY-62 revealed that the self-support devices used to center fuel elements within the process tubes can adversely affect heat transfer conditions from the fuel elements. For example, with a test section representing the case where a self-support was crushed against the fuel jacket, it was found that the boiling burnout heat flux was reduced significantly.

During FY-1963 these studies of the effects of self-support design on pressure drop, flow distribution, and boiling will be continued and expanded. Using electrically heated test sections in a glass process tube, high speed motion pictures will be taken of the heat transfer processes from the fuel elements with various types of self-supports. These will be compared with the case of fuel element support ribs and with the ideal case of fuel situated concentrically within the process tube without any supporting devices.

The results of this study will include a visual and physical description of the heat transfer effects from all the different methods of

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supporting fuel elements in a process tube. Flow conditions studies will be those for a typical central zone tube and for a fringe zone tube. Heat fluxes studies will be those of normal power levels, 1-1/2 times normal power level, and those resulting in boiling burnout. Test section orientation with respect to the process tube will be concentric and 70 per cent eccentric where possible.

- b. The present methods of establishing maximum outlet water temperatures for individual process channels will be verified for self-supported K fuel elements in Zircaloy process tubes. Laboratory experiments using full scale electrically heated test sections will be performed under both steady state and transient conditions of heat generation and coolant flow.

The steady state experiments will measure pressure drop and temperature while the flow is slowly decreased at constant power generation until film boiling conditions are reached. These experiments will be run at power levels of 700, 1000, 1500, 2000 and 2500 KW and the results will allow a description of steady state heat transfer conditions for nearly any possible operation condition.

The transient experiments will define the flow, pressure, and temperature conditions within the process tube and fuel element for conditions of partial flow blockage, for complete flow loss to a single tube, and for power surges. These experiments will aid in defining the conditions which can lead to excessive fuel temperatures and in determining the reactor control response necessary to detect such conditions.

- c. Current studies of the transient and steady state capacity of the reactor effluent piping systems will be continued. Further model tests will be conducted by Washington State University which will be supported by analytical studies by the General Engineering Laboratory. These results will be supplemented by on-reactor tests and will be applied to the three general types of piping arrangements (B, D, F; DR, H, C; and K).
- d. Of prime importance in the design and operation of any nuclear reactor is assurance that fuel temperatures always will be maintained low enough to prevent reactor damage. As a consequence, careful consideration is given to avoiding those process conditions which result in boiling burnout and the associated high fuel temperatures. For the Hanford production reactors, most flow decreases or power surges that could cause boiling burnout first result in a hydraulic instability condition. Therefore previous work has been concentrated on defining and detecting hydraulic stability conditions and only secondary considerations have been given to defining boiling burnout conditions. However, a more exact knowledge of boiling burnout conditions is necessary to assure that the reactors are always operating within safety specifications.

Using electrically heated test sections, experimental data will be obtained where necessary to define boiling burnout conditions for all possible cases of reactor operation under steady state conditions. The program will include an examination of the effects of such things as abnormal flow distribution, fuel positioning, flux peaking and fuel supports.

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2. N Reactor

- a. Boiling burnout data have been obtained in the laboratory with test sections representing each of the three flow channels in the NPR tube-in-tube fuel elements. These test sections include ones for the center hole, the middle annulus with two heated surfaces, and the outer annulus with one heated surface.

For all of these test sections, data exist for pressures of 1500 psia and 1000 psia and for wide ranges of flows, coolant enthalpies, and heat generation rates.

The application of these data to a full length charge of NPR fuel elements will be verified by use of a 17 foot long test section containing all three flow channels. This full size section represents the downstream half of a charge of fuel elements and has a cosine heat generating distribution. The test section will be operated during carefully chosen experiments at conditions approaching boiling burnout. The results will be compared with those determined using test sections of the individual channels.

From the results of these experiments the conditions leading to excessive temperatures as caused by boiling burnout will be defined for a wide variety of operating conditions.

- b. The hydraulic stability characteristics of single NPR coolant channels will be determined. Experience with reactor channels at low pressure has shown that when flow is decreased at constant tube power, the pressure drop increases due to boiling and the flow conditions become unstable. This situation is known to be less severe at higher pressures but the exact conditions must be determined experimentally.

Pressure drop-flow relations will be investigated first for the fuel elements at constant tube power using an electrically heated test section in the laboratory. Second, two phase pressure drop through NPR jumpers and hydraulic fittings will be determined installing these components in the heat transfer apparatus. Next, these data will be combined and treated using analytical techniques for application to all possible reactor conditions.

- c. The investigation of the effects on heat transfer conditions of the non-co-axial positioning of NPR tube-in-tube fuel element components will be continued. Electrically heated test sections will be used to determine the boiling burnout conditions when the supports have been collapsed or removed from either component of the fuel element. In addition, visual studies of the boiling conditions around the normal and collapsed supports will be made using an electrically heated test section in a glass tube.
- d. The effects on fuel element temperatures of gross pressure and flow changes will be investigated in a series of transient heat transfer experiments. An electrically heated test section has been built which

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is a full scale mockup of the downstream half of a full tube charge of NPR fuel elements. Using this test section in the laboratory heat transfer apparatus, various transient experiments will be performed including those simulating a sudden rupture of the reactor piping. In these experiments a reservoir of hot water at high pressure will provide coolant to the test section after the outlet piping from the test section has been opened to atmosphere. In addition, the conditions resulting from a power surge will be investigated briefly.

- e. The flow requirements to provide adequate cooling of the NPR fuel elements during reactor shutdown conditions will be determined in the laboratory. When cooling a long train of horizontal fuel elements at low flow rates there is concern that should boiling take place, the steam and water will stratify and leave the uppermost part of the fuel to be cooled with a steam atmosphere. In this case the top of the fuel element could run considerably hotter than the bottom. Using the full scale electrically heated mockup of the NPR fuel elements, heat transfer experiments will be run to define fuel temperatures at various heat generation rates for low flows.
- f. During the investigation of the hydraulic stability characteristics of NPR coolant channels, considerable data will be obtained concerning the pressure drop of steam-water mixtures through pipe fittings. Two-phase pressure drop is difficult to predict with accuracy from theoretical considerations when the validity of existing correlations for pipe fittings is not known. Therefore these data will be analyzed in detail to extend the present knowledge of the pressure drop of two-phase flow.
- g. Fundamental studies of the critical flow of steam-water mixtures from pipes and orifices will be continued. The high pressure water storage tank in the 189-D heat transfer laboratory will be used to supply hot water at pressures up to 2000 psi for measurement of flow rate and pressure drop through the test sections. The data will be compared with theoretical predictions and the results will be of considerable value in hazards calculations and design of pressurized water reactors.
- h. Experiments have been started to investigate heat transfer conditions beyond boiling burnout in the film boiling region. It is the practice today to assume that boiling burnout will always result in fuel melting and thus must be avoided at all costs. The results of this study will define the actual fuel temperatures expected after boiling burnout. There is a good possibility that reactor operation is feasible in the film boiling region under some conditions without fuel element damage and a definition of such conditions will be most valuable.

E. Graphite

1. Old Reactors

a. Contraction

The contraction of the graphite samples exposed in C and K Reactors will

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continue to be measured as a function of increasing exposure as part of the work toward evaluating and understanding the distortion of the reactor moderator stacks. NPR graphites will be included.

A series of irradiations will be performed to quantify the tentative conclusion that full cross section graphite bars contract at a higher rate under irradiation than do small samples. This work should provide a better understanding of the relation of data from small samples to moderator stack distortion.

Work to determine the effect of compressive loads in the range 50 to 350 psi on dimensional changes in graphite under irradiation at about 600 C has been underway for some time and initial data are being obtained. Results to date show an effect of loading on initial growth. The effect on long-term contraction is still to be determined.

b. Stored Energy

The moderator graphite in all reactors will be sampled as appropriate to permit a more exact analysis of the possible effects of stored energy release in the stacks. Renewed emphasis on this program is appropriate, since the accumulated fast-neutron exposure of the moderator graphite has doubled or trebled since the last samples were taken and analyzed. This program will include measurement of total stored energy content and stored energy release spectra on core samples cut from the reactor moderator. Measurements of the stored energy accumulation in graphite specimens irradiated under known conditions will also be made, with emphasis placed on samples exposed at high temperature. The total stored energy measurements will be made at the National Bureau of Standards using oxygen-bomb calorimetry. Measurement of the stored energy release spectra will be performed at Hanford.

c. Burnout

Monitoring the potential for graphite oxidation during operation will be continued at all reactors using small graphite test samples in bare process channels. In the past data from this program served as the basis for allowing increases of moderator temperature by showing that the burnout of the graphite did not exceed specified limits. Data from this program have demonstrated that in recent operation most of the burnout of graphite has been due to in-leakage of air into the reactor gas atmosphere rather than due to reaction of the graphite with carbon dioxide.

The location of the maximum burnout due to the carbon dioxide - graphite reaction is near the stack center line; in contrast, the oxygen - graphite maximum burnout rates are believed to occur approximately 80 to 100 in. from the front face of the graphite stack. Rates of burnout of the small monitors due to reaction with oxygen from air in-leakage has greatly exceeded two per cent per thousand operating days. At these high rates burnout does not occur uniformly throughout large graphite bars. Efforts will be made to establish the relation of burnout rates determined in monitoring with small graphite specimens to the rates and nature of oxidation of full size bars in the moderator stack.

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To provide an improved understanding of burnout of graphite for an improved statement of technical bases for reactor process standards on burnout control, information will be developed on the specific reaction rates of carbon dioxide, oxygen, and water vapor with graphite singly and in mixtures as a function of concentration and temperature for the range of conditions encountered in the Hanford reactors. Special attention will be given to the inhibiting effects of carbon monoxide and hydrogen on the reaction rate. The work will be at least primarily an examination of existing data on a theoretical basis rather than an extensive experimental program. It may be desirable, however, to conduct certain confirmatory physical experiments.

It is appropriate to phase out the work on plastics and elastomers since much of the needed information has been obtained. Unless other problems appear, such work will be discontinued during FY-1963. A limited number of qualified tests and quality control work may be continued on a work-order basis.

2. N Reactor

a. Contraction

Studies will continue to provide a basis for calculations of the maximum amount of graphite contraction that will occur during the operating lifetime of N Reactor. Graphite samples will be irradiated at an accelerated rate in the GETR to an exposure equivalent to the predicted lifetime exposure of N Reactor.

V. 11/1/64

It is of special interest to determine, 1) whether the rate of contraction with exposure decreases as exposure goes to relatively very high levels and 2) whether a (saturation) limit of irradiation-inducible contraction is reached at exposure levels less than that corresponding to maximum exposure of full power, intended-lifetime operation of the N Reactor. Secondary objectives of the program are measurements of physical properties such as modulus of elasticity, X-ray parameters, and helium density with increased measurements will provide additional information for use in evaluating effects of irradiation on the NPR graphite stack.

During FY-1963 one first-generation and three second-generation instrumented capsules will be removed from the GETR, disassembled and measurements of the samples will be made. Additionally, two second-generation and two third-generation capsules will be fabricated and installed in the GETR in the latter part of FY-1963. This test irradiation program is planned to continue through FY-1966.

F. Gas Atmosphere

1. Old Reactors

*RWB 11/1/64
To be done
by 7PD?*

Tests will be continued through FY-1964 to evaluate reactor operation and environmental conditions on graphite and process tubing materials with

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the use of reactor gas atmosphere consisting of nitrogen and helium mixtures. The requirements for additional gas atmosphere instrumentation and gas composition control will be determined by March, 1963.

The study to determine the amount of Cl^4 which is generated in the reactor atmosphere and released to exhaust gases and liquid effluent, as the result of using nitrogen as the gas atmosphere, will be brought to a conclusion by December, 1962. Identification of the chemical forms of Cl^4 from the effluents will be undertaken only if it is necessary to control the amount of Cl^4 released.

2. N Reactor

The conditions of reactor gas atmosphere composition and other factors in N Reactor for compatible operation with graphite and Zircaloy pressure tubes will be determined as a part of a continuing program.

While the major factors which determine whether or not zirconium will hydride under specified conditions ~~has~~ been established, two areas of some importance remain unstudied. The first of these is the degree to which reliance on the reverse water gas reaction to inhibit hydrogen pickup can be made:

Studies are required in which dried carbon monoxide and hydrogen are mixed in the presence of Zircaloy and graphite at representative NPR temperatures to determine rates of oxidation of the Zircaloy.

Under some conditions carbon monoxide and carbon dioxide are also capable of oxidizing zirconium. While they do not prove able to inhibit completely the hydrogen absorption by zirconium, they might contribute in some part to the inhibition process. Limited studies of zirconium oxidation rates in carbon monoxide and carbon dioxide, with and without the presence of water vapor, will be made.

The major compatibility problem arises from the destruction of the protective qualities of the zirconium oxide film by the neutral or reducing atmosphere that is most compatible with the graphite moderator. A pilot study has shown that water vapor may diffuse through hot graphite at a rate approximately commensurate with maintaining a continuous zirconium oxide film. It is urgently important along with other graphite burnout program objectives to study the effect of water vapor on NPR graphite at elevated temperatures. Studies include a measurement of the rate of reaction of small quantities of water vapor with large graphite bars and the effect of gas composition on diffusion kinetics.

In-reactor monitoring of the protective properties of the zirconium oxide film may be possible by measuring the changes in electrical conductivity of the oxide. Initial experiments have shown that before zirconium can absorb hydrogen from a gas atmosphere the oxide becomes highly conducting. Additional studies are required to establish reproducibility of measure-

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ments, methods of making electrical contact at the oxide-gas interface, alloy effects, temperature dependence of measurements, and the effect of irradiation on film conductance.

While the practical implications of such a development have been emphasized, it is obvious that these studies will be quite informative about zirconium oxidation and hydriding mechanisms.

G. Zirconium Alloys

The performance characteristics of N Reactor, Zircaloy pressure tubes will be determined through an extensive pre- and post-irradiation testing program. The task is complicated by the wide variety of metallurgical histories of the pressure tubes. As an aid in extrapolating relatively short-term tests on sections of pressure tubes, long-term creep tests (up to 30,000 hr) are being performed at Battelle Memorial Institute on tensile samples cut from rolled strip and from longitudinal sections of N Reactor pressure tubes. A comprehensive interpretive analysis of the data obtained to date will be published during FY-1963. Short-term burst tests and stress rupture tests will be performed on sections of N Reactor pressure tubes having representative fabrication histories. Initial elevated temperature burst tests will be performed on sections of irradiated KER pressure tubes. Examination and testing of at least one KER, Zircaloy-2 pressure tube is planned during FY-1963.

The characterization of the brittle crack arrest properties of Zircaloy-2 pressure tubes will be emphasized. The effect of crack length, wall thickness, stress level, degree of cold work and reactor environment on the crack arrest properties will be measured. The amount and distribution of hydrogen in Zircaloy-2 can drastically change its properties. Techniques will be developed to introduce known quantities of hydrogen into Zircaloy-2 and control its distribution throughout the cross section of the tube wall. Both burst, creep rupture and crack arrest tests will be performed to determine the effect of the hydride.

H. Chemistry, Corrosion and Decontamination

1. Old Reactors

To better predict and reduce corrosion of aluminum process tubes and fuel cladding, a more complete understanding of its mechanism is required. To this end tests will be made on the effect of reactor radiation on corrosion, the effect of heat flux without radiation in electrically heated samples, and corrosion in single-pass deionized water compared to process water. Cladding temperature will be measured with thermocouple slugs and related to corrosion.

Quachromglucosate will be evaluated in in-reactor tests as a replacement for dichromate as a corrosion inhibitor. This substitution has the potential for reducing the level of Cr-51 and total chromium in the reactor effluent. Other replacements for dichromate will be investigated. Fuel element corrosion measurements will be extended to higher heat fluxes and

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surface temperatures to predict hot-spot behavior as a function of pH and inhibitor levels. Changes in water quality proposed for reduction of effluent activity will be evaluated as to their effect on corrosion. The use of carbon dioxide to reduce pH will be studied.

New reagents for decontaminating both in-reactor and out-of-reactor primary piping systems will be investigated with regard to their efficiency of decontamination and corrosion attack on piping metals. Activity buildup throughout the reactor system as a result of crud deposition will be studied as a function of water treatment.

2. N Reactor

Corrosion products will tend to deposit on the in-reactor surfaces of NPR, resulting in possible flow restriction, resistance to heat transfer from the fuel elements, and activation of radioisotopes and their subsequent release causing high activities in ex-reactor portions of the primary system. Studies will be continued on the deposition of iron oxides and other corrosion products on fuel and piping surfaces, the chemical and physical nature of this deposit, and the adsorption and activation of isotopes on it. The effect of deposition on heat transfer and fuel element cladding temperature will be studied with thermocouple slugs in normal operation, following decontamination, and at below normal pH. Possible flow restriction due to film deposition in the graphite cooling system will be evaluated. The deposition of radioactivity on various components of the external loop will be mapped in tests in KER. Crud and radioactivity will be studied as a function of water quality, and the results of poor control of water quality evaluated. Fouling of heat exchanger surfaces under NPR conditions will also be studied.

New decontamination processes which have shown promise of significant advantages over presently recommended processes will be tested more thoroughly with respect to both effectiveness in decontamination and corrosion. Oxalate-peroxide solutions will be tested as more effective alternates to bicarbonate-peroxide to dissolve uranium oxide and fission products. The present decontamination for once-through cleaning of the piping to and from the reactor is not fully satisfactory in that it is corrosive to couples of stainless-to-carbon steel and contains phosphorus which presents a serious disposal problem. Inhibited ammonium citrate and other solutions will be tested as replacements. The corrosion of valve facing materials by alkaline permanganate, which has been found to be a serious problem, will be studied further. Decontamination assistance will be given the NPR rupture loop in PRTR.

Dynamic corrosion studies of zirconium alloys will be extended to higher temperatures (320 C to 600 C) in the new TF-8 and TF-15 loops, to study breakaway corrosion at hot-spot conditions on fuel elements or process tubes. The behavior of Zircaloy-2 during and after breakaway (accelerated) corrosion will aid in predicting the corrosion of N Reactor process tubes. Newer alloys will be tested as well as Zircaloy-2. Fretting of process tubes due to chattering of fuel elements and spacers will be evaluated. The effect of off-standard water quality will be investigated.

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Long-term evaluation of carbon steel corrosion will be continued, and corrosion in off-standard water studies. Problems in non-uniform corrosion will be evaluated, particularly caustic and chloride stress corrosion of stainless steel. Ammonium-hydroxide will be evaluated as an alternate for lithium hydroxide for pH control.

Tests will be conducted in the new TF-23 loop, which mock-up the N Reactor secondary system, to identify corrosion and water quality problems that may be encountered during operation of N Reactor. The use of hydrazine and ammonium hydroxide for control of oxygen and pH will be evaluated, and alternate methods of control will be studied. Corrosion of copper alloys in the system by ammonia, and crevice corrosion of the stainless-to-carbon steel junction in the steam generator will be studied.

N Reactor components will be exposed to the atmosphere for extended periods of time before and during construction and before start-up. The corrosion during such periods will be studied, and protective coatings will be evaluated.

Automatic instrumentation of gas and chemical analysis of the coolant streams will be evaluated. The proposed use of hydrogen and hydrazine to remove oxygen in the primary and graphite cooling systems will be evaluated. The problems in deoxygenation of back-up primary coolant will be studied. The problems encountered in the control of water quality will be studied.

I. Radioisotope Reduction - Reactor Effluent

The concentrations of several radioisotopes discharged to the Columbia River in the reactor effluent water have decreased by a factor of two as a result of the adoption of the high alum feed process for treating reactor coolant. However, inasmuch as future production planning anticipates no effluent discharge capable of producing physiological effects on downstream water users, further reductions are desirable. This program has been undertaken to develop processes capable of achieving a substantial reduction in the biologically significant radioisotope content of reactor effluent. A ten-fold reduction appears to be a reasonable goal.

The research and development program encompasses the following areas: 1) development of methods for removal of parent materials responsible for radioisotope formation from the process water, 2) fundamental studies to define methods for eliminating or reducing the time of adherence of radioisotope parent materials on the films by changes in the chemical constitution of water, chemical form of parent materials or changes in the adhering film itself, 3) studies to provide radioisotope monitoring, and 4) studies to provide geologic and hydrologic data as may be necessary for radioisotope removal. Specific areas of activity are listed below.

The water treatment pilot plant at 1706-KE will be ready to supply specially prepared water to two single pass reactor tubes in September 1962. The first study will be directed toward optimizing the aluminum sulfate flocculation process insofar as radioisotope parent removal is concerned. The second

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study will determine the effects of aluminum nitrate as the flocculation agent. Following these, studies will be made: 1) using alternate flocculating agents such as ferrofloc and iron-potassium permanganate, 2) other additives such as sodium silicate or polyelectrolytes, 3) changing the addition points of the chemicals, 4) changing holdup time, and 5) changing filter media or flow rates.

Laboratory and in-reactor tests will be continued to determine the effect of sodium silicate as a water treatment additive for radioisotope reduction. Other chemicals having the potential to inhibit film formation or radioisotope parent adsorption will be investigated.

Studies with deionized water which will provide fundamental information on the adsorption kinetics and the influence of radioisotope parent concentrations are underway. These are expected to continue with a variety of chemical additives at different concentrations to determine their effect on the effluent activity.

Both laboratory and plant tests will be undertaken to define the applicability of zeta potential measurements for water plant control. Additional efforts will be given to other means for controlling water treatment plants to minimize effluent activity.

The above activities will be directed to define the most promising approach to reducing the radioisotope concentrations in reactor effluent by December, 1962 and to demonstrate a process for obtaining a factor of ten reduction by July, 1963.

The details remaining in connection with the disposal of NPR decontamination waste will be completed. Studies will continue as necessary to determine the impact of over-all NPR wastes on the river in order to provide technical bases for process standards assuring compliance with NPR disposal criteria.

The prototype of the As⁷⁶ monitor will be calibrated, the effects of background determined, and the unit put into service. The feasibility of a P³² monitor will be established.

Geology and hydrology studies will be conducted to permit: 1) the development of a crib site criteria, 2) effects of unusual discharges in present facilities, and 3) to clarify ground water movement and temperature patterns.

The effluent activity reduction research and development program will be accompanied by engineering studies embracing economic analyses, full reactor testing and process equipment scoping and development as required. In addition to laying the ground work for process adoption, such engineering studies will also serve as a guide to economically feasible research and development approaches.

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J. Instrumentation

1. Old Reactors

a. Automatic Control Techniques

Analytical and experimental work will continue to develop automatic control techniques which may have practical applications. Planned experiments at 105-D and 105-KW will provide data for determining dynamic characteristics of the reactors. Additional equipment will be needed for these experiments for rod oscillation tests. The accompanying analytical work is aimed at derivation of appropriate reactor transfer functions and an equivalent model for control systems studies. The continuing long range objectives are to determine the feasibility of automatically controlling the reactor and the characteristics of an optimal control system.

b. In-Core Flux Monitors

Development work will continue on in-core flux monitors and other equipment required for period monitors, power-rate-of-rise indicators, and intermediate level monitoring devices. In-core chamber development will continue through FY-1963 and FY-1964 to obtain an in-core sensor which is sufficiently reliable for use in the Hanford reactors.

c. Instrumentation for Effluent Control Studies

Instrumentation as described in topic I, Radioisotope Reduction, will require development efforts during FY-1963. The prototype of the As⁷⁶ monitor will be calibrated, the effects of background determined, and the unit put into service. The feasibility of a P³² monitor will be established.

d. Radiation Monitoring Instrumentation

Development work will be continued on radiation monitoring instrumentation, and emphasis will be placed on the development of instrumentation which will result in reduced exposure to monitors, operators, and maintenance personnel. Development of alarming dosimeters which alert workers when a predetermined dose or dose rate is reached will continue. The refinement of dose rate telemetering will be carried out. A practical system for integrating dose rate to dose will be advanced, together with compatible television and communications systems for use in remote monitoring and supervising techniques.

Instruments will be developed which will be capable of precisely determining the emission rate of radiation through the primary and secondary reactor shields.

e. Reactor Monitoring Instrumentation

Instrumentation will be developed in support of the Pile Life Studies

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for the old reactors. This will include development of instruments for measuring the displacement and the internal dimensions of process tubes, for measuring the depth of surface scratches inside process tubes, and for analysis and control of the reactor atmosphere.

f. Others

Special instruments will continue to be developed to support the nuclear physics cross-section program.

2. N Reactor

a. N Reactor Analog Simulator

Activities in support of the comprehensive N Reactor simulator will be accelerated during FY-1963 and will reach the full level of effort by mid-FY-1964. Specific design work and development of detailed simulation models and circuits will be carried out. Initial problem applications for the simulator will be planned in detail, and operating and maintenance personnel will be trained so as to provide beneficial use of the simulator as rapidly as possible.

b. Defect Test Loop Instrumentation

Development, design, and installation of special instrumentation for fuel failure detection studies at the PRTR Defect Test Loop will be completed during FY-1963. Experiments will be planned and conducted to determine optimum methods for operating the NPR Fuel Failure Monitor, and to obtain basic data relevant to fuel failure detection and indicative of failure mechanisms.

c. Others

Instrumentation, design, development, and testing activities in support of the N Reactor project will be continued as required. Consultations and technical advice will be provided on a variety of instruments and instrument system problems.

Special instrumentation will be designed and provided as required for N Reactor startup.

K. Design and Mechanical Development

1. Old Reactors

Development efforts will be required in support of establishing technical requirements for improvements, modifications, and maintenance to the reactor mechanical systems. This work will be accomplished from both R&D and Process Technology funds. Following is a summary of the tasks to be undertaken during FY-1963 - 1964.

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a. Reactor Overbore Studies

Evaluation and refinement of current overbore process tube assembly designs will continue in anticipation of FY-1964 project authorization. The adequacy of the reactor piping system and components at 105-C and 105-H will be reviewed for operation at projected overbore process conditions. Criteria for overbore fuel handling systems will be prepared and the development of supplementary control systems to operate in smooth-bore tubes will be undertaken. Studies of graphite channel distortion effects on process tube stresses and fuel handling will be performed to determine graphite channel configuration. Prototype systems will be fabricated and tested both off-pile and on the C Reactor overbore test facility. Among the equipment and components to be evaluated are reactor gas and water seals, alternate nozzle configurations, donut shielding train material effectiveness, rolled tube joints and collars, biological shield loadings, thermal shield and Ball 3X entry, and reactor process instrumentation.

Design of the water plants at the C and H Reactors will be reviewed and the requirements established for supporting an overbored reactor. The design criteria and technical requirements for replacement process pumps at 190-H will be developed and procurement of a prototype pump initiated by mid-FY-1963. Testing of the prototype pump will be performed at the 100-F Area pump test stand in FY-1964.

Studies of work sequencing and methods and their relationship to component design in support of the reactor overboring process will also be undertaken. Work will also be undertaken in FY-1963 - 1964 to provide improvements to the equipment and procedures utilized in the overboring operations including removal and replacement of tubes and gunbarrels.

b. Mechanical Problems Associated With Graphite Distortion

Graphite distortion data collection in the form of borescope and television examination of vertical and horizontal rod channels will continue to be supplemented by displacement measurements in these channels. Better criteria definition for vertical channel sleeves will be established by installation of test specimens at both C and K Reactors which will enable estimates of forces applied to an installed sleeve.

Off-reactor mockup tests of alternate sleeving materials for vertical rod channel liner application will continue, and in-reactor evaluation of the best candidate materials will be initiated on a two channel basis at C Reactor.

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Off-reactor tests of flexible vertical rods will include speed of insertion measurements in sleeved channels as well as verification that the proposed assembly is compatible with the existing Ball 3X system.

Development of equipment to remove saddle blocks from horizontal rod channels will be completed and will be available for application at the K Reactors. A prototypic flexible horizontal control rod will be fabricated and tested in both an off-reactor mockup and a one channel in-reactor installation.

Continued support will be given to reactor operation in development and evaluation of equipment necessary for interim vertical rod channel corrective measures. Tools for controlled graphite removal will be required in conjunction with interim vertical rod replacement. Development of equipment to enlarge vertical rod channels prior to sleeve installation will continue on a prototype basis.

Efforts will also be continued to establish improved methods of process channel rehabilitation. This will include improvements to the graphite drilling methods, improvements to expanding cutters for fringe graphite channel relief, and improvement of methods and equipment to permit efficient preparation of process channels for zirconium tube installation at the K Reactors.

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VI. METALLURGY PROGRAMS

The "Metallurgy Research and Development Program"* has been developed by the Fuels Preparation Department and presents a description of development efforts to be performed and sponsored by FPD. This program has been reviewed by Irradiation Processing Department personnel and there is general agreement on program content. Since it is nearly impossible to separate reactor and fuel performance, it is necessary that the reactor and fuels programs be carefully integrated, and it is expected that such practices will necessarily continue. The Metallurgy program has been issued separately; however, it is included in this document along with the Reactor program to present a complete picture of the reactor-fuel research and development programs and how they are integrated.

A. Present Reactors

The program efforts on present reactor fuels will include (1) development of alternate cladding processes designed to produce fuel superior in quality to that produced by the AlSi process, (2) basic surface chemistry studies in support of alternate process development, (3) a fuel quality evaluation program, plus (4) supplementary work including efforts to develop and improve projection fuel types for use in either ribbed or smooth-bore process tubes.

1. Alternate Cladding Process Development

As the severity of operating conditions in the present Hanford reactors increases, improved fuel element canning processes must be developed or the present AlSi process must be improved in order to prevent the fuel quality from limiting the power and operating efficiency of the reactors. Extrapolation of the performance of present fuel elements to projected future operating conditions indicate that AlSi bonds may become embrittled to the point that failure rates will become excessive. For example, examination of irradiated elements in the Radiometallurgy Laboratory frequently uncover considerable bond cracking. Further, pre-irradiation total external bond count has been shown to be strongly related to side failure rate. Therefore, it is a reasonable conclusion, qualitatively, to expect deteriorating performance with AlSi fuels as operating severity is increased. On the other hand, direct experimental verification of such a conclusion is required before any major change in canning process can be firmed. Thus, the first thing that is dictated by these considerations is a test of AlSi-processed elements under the projected severe conditions. The overbore test in C Reactor should provide valuable data since the operating conditions of the higher power elements in this test are in the range of interest.

Concurrently, analytical and preliminary development work has been initiated to develop and evaluate nickel-plated, diffusion-bonded fuel elements made by various processes.

Four processes were selected for evaluation at HAPO.

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*HW-74134, "Fuels Preparation Department Sponsored 02 Metallurgy Research and Development Program - FY-1963", L. M. Loeb, August 29, 1962 (CONFIDENTIAL)

a) Sylcor Hot Press

In this process the electro-nickel plated uranium fuel core is placed in an aluminum jacket. The assembly is then bonded by applying heat and mechanical pressure to the assembly in a containment die.

Test elements have been fabricated by Sylcor by the process which they use for the elements for SRP reactors. About 1000 normal uranium elements were received, ex-reactor evaluated, and 192 elements charged to the reactors under production test in FY 1962. The post-irradiation evaluation of these elements will be completed in FY 1963. Plans are to procure, ex-reactor test, in-reactor test, and evaluate about 1000 enriched CIVE type elements in FY 1963.

b) HAPO Hot Press

In this process the electro-nickel plated uranium fuel core is placed in an aluminum jacket. The assembly is then bonded by applying heat and mechanical pressure to the assembly in a containment die. The process in many respects is similar to the Sylcor hot press process but has some unique differences such as a worked closure.

The HAPO hot pressing facility was renovated and the bonding conditions optimized in FY 1962. In FY-62, 192 elements were charged to the reactor. FY 1963 plans for HAPO hot press elements involve only the post-irradiation examination of these elements.

c) Gas Pressure Bonding

In the gas pressure bonding process the electro-nickel plated uranium core is placed in the aluminum jacket and closed by vacuum welding. The fuel assembly is then bonded in a heated autoclave using a gas (e.g., helium) to pressure bond the assembly.

During FY 1962 tests were run by SRL to bond HAPO type elements by gas pressure bonding. The bonding conditions in the SRL autoclave facility were not adequate to produce satisfactory bonding. Preliminary pressure bonding tests at BMI have been promising. More extensive testing is planned in FY 1963. If these tests are successful, plans are to produce 500 fuel elements later in the year and reactor test them if the element quality warrants.

d) Hot Die Sizing

This is a new bonding technique initially developed by the Savannah River Laboratory (SRL). The electro-nickel plated uranium core is placed in an aluminum jacket and preheated in excess of 1000°F. The assembly is then forced through cold sizing dies and a bonded fuel element is obtained.

During FY 1962 hot die-sizing equipment was installed and tested at HAPO. During FY 1963 plans are to optimize the process and produce reactor quality elements for irradiation testing. During the second quarter of FY-63, a two to five tube feasibility test will be reactor

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charged following out-of-pile evaluation. It is expected that shortly after the first of the calendar year a 10-20 tube statistical test will be reactor charged.

An independent development and irradiation program at HAPO on diffusion-bonding processes is necessary for several reasons:

1. The differences in fuel operating conditions between SRP and HAPO are too large to permit direct extrapolation of fuel performance at the two sites.
2. In a comparative evaluation such as we are pursuing, especially in the fuels business, it is highly desirable, if not mandatory, that as many extraneous (i.e., non-technical) variables as possible be eliminated from the program.

Basic development effort is required to establish a process which will give consistently high bond strengths and which will provide a cap-can interface suitable for subsequent closure welding. A major problem in the latter area may be lubricant contamination of the closure area, while the program on uranium surface chemistry discussed below will be critical to meeting the former goal.

Since the integrity of any fuel element can be assumed to be no greater than the proven and properly calibrated testing techniques indicate it to be, a concurrent nondestructive testing development, described later, is underway on diffusion bonded elements.

2. Uranium Surface Chemistry

As development programs in nickel-plating of internally and externally cooled (I&E) cores, optimization of AlSi canning cycles for dingot metal, cathodic etching and copper plating of uranium cores, etc. have progressed, it has become increasingly evident that uranium surface chemistry plays a major role in determining final product quality. Therefore, we have initiated a program in this area. Initially, the work will be aimed at developing a better basic understanding of the important process parameters in the plating of nickel on uranium. The results of these basic studies will then be used as bases for improving the current plating process.

A variety of electrochemical measurement techniques will be applied to analysis of phenomena occurring in the surface preparation and nickel plating steps. It is hoped that these studies may allow correct inferences to be drawn regarding the critical parameters in these steps and that these inferences can be tested under process conditions.

Electrochemical techniques such as potential polarographic, chrono-potentiometric, and electrode impedance measurements will be applied to characterization of the uranium surfaces produced in systematically varied surface treatments. Results of these studies will be correlated with measurable parameters in the subsequent nickel electrodeposition step and in turn with the quality of nickel diffusion bonds formed with the electrodeposited nickel film.

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Then the more promising measurement techniques for practical application will be selected and evaluated under a range of conditions of practical importance for a production application. The object in these studies will be to provide quantitative control techniques useful in obtaining and maintaining desirable operating conditions in a production process.

Studies will also be made to establish nondestructive methods for measuring the plate thickness and the quality of the uranium to nickel bond. The plate thickness will be measured by eddy current techniques and, assuming technical success, pilot plant equipment will be in use early in the second quarter of FY-63. The nickel to uranium bond evaluation will be much more difficult to effect and this development effort could run into FY-64.

3. Fuel Quality Evaluation Program

Several years ago, a program of routine and continuous monitoring of the present reactor fuel elements was begun, under the title of the "Fuel Quality Certification Program". At present, sample fuel elements from each lot are withdrawn from the production stream, subjected to careful measurement of bond count, warp, inside and outside diameter along their length and are specially charged to the reactor. Upon discharge, the similar dimensional measurements are made. Remote methods of measuring post-irradiation bond count are being developed as outlined in Section III, A, 6. In addition, the elements are examined for hot spots and other surface anomalies. Thus, we have a continuous record of quality trends as measured by warp, tube-filling capacity (an artificial volume statistic which helps characterize over-all dimensional change during irradiation), O.D. changes, per cent hot spots and bond count.

From these data, we have derived a number of valuable conclusions, among which are:

- a. There is a definite relationship between side hot-spot failure rate, total pre-irradiation external bond count and tube-filling capacity. Statistical analysis of the performance of I&E fuel elements canned between August, 1959 and April 1961, yielded the following significant relationship:

$$q_s = 1.07 - 1.40(\text{LBC}) - 0.39(\text{TFC}) + 0.51(\text{LBC})(\text{TFC})$$

where: q_s = metal performance level parameter for side failures

LBC = logarithm of the sample average (external bond count +1)

TFC = average tube filling capacity ($\sqrt{\text{mils}}$)

Practically speaking, in the ranges of interest, the relation indicates that the side failure rate approximately triples as external bond count or tube filling capacity doubles.

- b. Detailed analysis of data taken at NLO in late 1960 indicates that:

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- i. grain size of fuel cores varies with position along ingot length, with larger grains coming from either end of the ingot than from the middle.
- ii. carbon, oxygen and nitrogen segregate extensively along ingot length, while iron and silicon are uniformly distributed.
- iii. since grain size is changing rapidly near the ingot top, it can be hypothesized that cores from this position might be of less uniform grain size than cores from the center portion of the ingot. This non-homogeneity could be a driving force for irradiation-induced warp.

Practically speaking, these data do not bear on present performance in that changes have been made in many steps of the core fabrication process. However, from a scientific viewpoint, these conclusions are quite interesting, in that the i and ii conflict somewhat with present beliefs on the minimal role of oxygen and nitrogen in controlling grain size, while iii suggests a testing requirement.

The examples noted are representative of a number of such items which led us to conclude that much would be gained through a greatly expanded analytical effort in fuel quality evaluation. For the first time, we are able to sift through past performance in a systematic manner and provide sound, significant direction to our uranium core development work. Indeed, we already have formulated potentially-significant tests in the areas of heat treatment, chemistry and fabrication techniques as a start.

Recent work at HAPO and SRP has indicated that "between ingot" differences may be responsible for a major portion of the observed variation in fuel irradiation performance. If "between ingot" variation is responsible for a portion of the remaining variation, there is a distinct possibility it can be controlled and the attending problems eliminated. The present efforts of the Quality Certification Program are devoted toward a better understanding of Fuel Effects. Specifically, the following programs should be completed during FY 1963.

1. Completion of Off-Site Uranium Data Processing System for both I&E and NPR fuels.
2. Irradiation of selected chemistry samples taken from "normal I&E production".
3. Development of models describing uranium distortion, considering the corrosion effects of can wall on I&E's.
4. A consideration of all factors, i.e. growth and dimensional instability toward the development of a fuel element to achieve the proper dimensions at the time of most severe service.
5. A complete characterization of as-produced fuel dimensions will be completed for all products.

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4. Fuel Support Development

Collapsed or broken projection rails have caused two fuel cladding ruptures to date in approximately 14,500 self-supported fuel elements irradiated. This experience has confirmed that the initial X-8001 aluminum alloy, collapsible-bridge rail design may be inadequate strength-wise to withstand mechanical charging and guarantee fuel self-centering in ribless process tubes. Strength of the original rail design has been improved on an interim basis by increasing the rail thickness. In order to insure proper support strength, collapsible features, and low resistance to coolant flow, a program has been initiated to develop and evaluate improved projection rail designs. Rails fabricated from materials of higher strength will also be investigated.

Priority is being given to optimize rail designs for fuels to be used in the K-Reactors after retubing these reactors with ribless Zircaloy tubes. Some work will also be done to evaluate rail designs utilizing nickel-plated supports and aluminum alloys other than X-8001. Evaluation of potentially improved rails for the "K" fuel models is scheduled for completion during 1963. Projection rail design optimization for overbore fuel elements and other fuel models as required will follow. Subsequent development effort is expected to consist of large scale irradiation testing and minor rail design modifications as may be warranted to further improve rail integrity.

5. Clad Corrosion Studies

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There will be a continuing effort in FY 1963 to study the corrosion of aluminum cladding in an effort to reduce the corrosion rate by developing new alloys or developing more corrosion resistant oxide films with revised autoclaving techniques.

Although there has been a reduction in clad associated, normal uranium fuel element failures, there was not a comparable reduction for enriched fuel. Presumably, higher heat fluxes make enriched elements liable to hot spot failures under conditions that do not affect normal fuel. Thus, special coating techniques to protect enriched fuel have a considerable economic incentive though only a portion of a reactor loading is enriched. While corrosion of normal fuel does not now limit reactor operation, a considerable corrosion allowance for the cladding of normal fuel is still required. Development of a corrosion resistant coating permitting a reduction in the clad thickness offers the benefit of more uranium per slug and/or more efficient slug cooling from an enlarged coolant annulus. The excellent corrosion record of current fuel is related to low pH water condition currently maintained in the reactors. Current data show that P^{32} activity released to the river is primarily associated with sulfuric acid pH control and alum water treatment. If activity levels of P^{32} released to the river come to be considered excessive, it might be necessary to reduce acidification of the coolant. Corrosion rates may be expected to increase, placing a greater premium on protective coatings even for the normal fuel.

Specific studies will include the use of higher temperature and/or dual

cycle autoclave cycles to produce more corrosion resistant oxide films or to provide a more stringent proof test of the integrity of the end closures. Other studies will include evaluations of the effect of cladding chemistry, cladding metallurgy, and water environment (i.e. temperature, hydrogen ion concentration, inhibitor concentration, velocity, and pressure) on the corrosion rate and corrosion mechanisms.

6. Nondestructive Testing Development

The bulk of the research and development work on nondestructive testing of present reactor fuels, exclusive of the substantial effort on process assistance, involves developing fuel testing methods in support of the alternate process development. Also work is underway in support of the Fuel Quality Evaluation Program to develop remote techniques for examining bond integrity, cladding thickness, core defects, and closure integrity on irradiated fuel elements.

B. New Production Reactor

The research and development program on fuel for the NPR will emphasize evaluating the characteristics of the fuel now under manufacture and continually improving the fuel components and fabrication steps to assure adequate fuel performance in support of the projected production goals for the reactor. Major emphasis in the program will lie in the areas of improved fabricating techniques and fuel component quality, performance testing, fuel element failure behavior, and basic metallurgy. Fabrication development and irradiation data supporting advanced fuel concepts will receive increased emphasis during the year. The research and development effort will be supplemented by a substantial process assistance program in FY 1963. There will be a continuing effort to reduce the cost of fabricating the present element and to develop a less expensive design.

1. Fuel Evaluation

Because of the shortage of irradiation experience on NPR fuel under prototypic conditions and because of the inherent variation in product properties from any newly-developed process, it is imperative that the product be characterized as precisely as possible. Therefore, there will be a quality analysis program to determine the pertinent metallurgical properties of the fuel product. This information will then be employed to modify raw material or process variables in order to improve the fuel quality as measured by its performance in-reactor.

This evaluation activity will range across the entire scope of the process. The newly established N-fuel quality data system will be used to inter-relate feed material data, coextrusion process data, and KER post-irradiation results. A strong effort will continue in the development and application of nondestructive testing techniques to obtain a more precise characterization of the product. Also the diffusion layers in the bond interfaces will be carefully studied with the use of an electron microprobe.

a. Quality Analysis Program

The N-fuel quality data system has been patterned after the "Fuel Quality Evaluation Program" described for present reactor fuels in

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item III.A.3 above. Since this data analysis program has been formulated concurrently with the firming up of both the specifications and the process itself, the results of the program will be forthcoming earlier than if the program were established after the process is in full production. It is expected that the data handling and analysis system will be in place and results forthcoming by the end of the second quarter of FY 1963.

b. Nondestructive Testing

Nondestructive testing research and development efforts during previous years have resulted in the development and application of test equipment which determines the quality of N-fuels in the following areas: (1) elements -- clad-core bonding, clad thickness and integrity, end closure integrity, uranium core integrity -- and (2) billets and zirconium -- the integrity and grain structure of billets and zirconium components prior to extrusion. FY 1962 efforts were mainly directed toward obtaining maximum utilization of this equipment through refinement of the tests and the procedures by which they are applied. Although much has been accomplished in improving the accuracy and precision of the data supplied by these tests, more work has to be done. Therefore, a large process assistance program on this area will continue through FY 1963.

The major research and development effort in FY 1963 will be devoted to the development of additional test methods that are needed to more completely measure N-fuel quality. A practical nondestructive test method of insuring adequacy of the heat treating process, for example, does not now exist. Exploratory investigations have revealed that the velocity of ultrasonic propagation is about three per cent higher in as-extruded fuel elements than it is in beta heat treated uranium. Effort will be made to develop a method of exploiting this fact to insure that each fuel element has been adequately beta heat treated.

The fuel supports are an important factor in obtaining satisfactory fuel performance. Tests are needed to insure that the supports are crack free. Also, the spot welds with which these supports are attached to the fuel tubes must be closely controlled in order to insure adequate weld strength, with no uranium penetration. Attempts will be made to develop electronic techniques of obtaining this control.

Investigations will also be made to determine the effect of resonant frequencies of the assembled fuel tubes on the fatigue life of the supports. As the elements have an exceptionally high mechanical Q (a term defining the ability of a material to absorb energy), physical displacements associated with resonant vibrations may be large enough to fatigue the clips in a relatively short period of time. Thus, attempts will be made to accurately determine fuel element natural resonant frequencies, and the effect of these on the fatigue life of the supports.

In assembly of inner and outer fuel tubes, the inner is locked into position by a special tool which crimps together clips in the inner-outer tube annulus. The rigidity of attachment is dependent upon

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tolerances of the mating assembly, and the positiveness with which the locking clips are crimped. Thus, a nondestructive test is needed to insure the tubes are fastened together with strength adequate to withstand reactor operation. Experiments reveal that this factor may influence to a measurable degree the resonant frequency of the inner fuel tube. This effect will be investigated further.

Uranium surface contamination on the fuel must be kept within limits in order that the effectiveness of the reactor rupture detection system will not be compromised. Successful alpha counting methods of measuring uranium contamination on the inner fuel tubes has been demonstrated on prototype equipment. Additional work is required to extend these measurements to outer fuel tubes, and to develop equipment more amenable to production line testing. In addition, other nuclear instrumentation is needed to determine the enrichment level of enriched uranium billets for nuclear safety reasons. The possibility of performing these measurements by gamma counting techniques will be investigated.

Considerable effort will be directed toward better instrumentation to control the brazing operation on the end closures. The brazing temperature must be controlled between close limits in order to insure adequate braze wetting without excessive melting of the uranium and subsequent contamination of the end closure. Adequacy of the present optical pyrometer temperature measuring system for this purpose is uncertain. More precise methods of measuring and controlling braze temperature will be investigated.

Preliminary investigations suggest that the flow and wetting action of the molten end closure braze be materially enhanced through the use of sonic vibration of the element during the brazing operation. To employ this technique as a regular step in the brazing process, much effort is yet required to develop practical methods of applying it as a production process.

c. Interface Diffusion Studies

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The successful fabrication of NPR fuel elements depends basically upon the controlled and reproducible interdiffusion of mating component materials in the various operations, i.e. in the coextrusion cladding operation and in the end closure operation.

To develop a good understanding of these processes and improve the quality and uniformity of the fuel product, a clear characterization of the diffusion zones in the interface areas must be developed. Because of the small cross-sectional dimension of these diffusion zones (generally one micron or less), they cannot be practically studied by normal metallographic and chemical techniques. Therefore, a program has been initiated to use an electron microprobe analyzer, having a 0.5-micron-wide, focused electron beam to study these diffusion zones.

The electron microprobe will therefore be used to study the diffusion zones (1) between the core and the cladding in as-extruded and beta-treated fuel, (2) between the braze and uranium or the braze and

Zircaloy in the brazed end closures, (3) between the components used in the various alternate end closures under development, and (4) between the copper can and the Zircaloy cladding in the coextrusion billets. An attempt will also be made to attempt to detect compositional changes in areas of the Zircaloy cladding which display anomalous thickening or thinning in the cladding operation.

2. Component Development

Although the NPR fuel fabrication process has been established and is in production, substantial effort is needed to study and improve the fabrication characteristics and the reactor performance of the major components, i.e. the uranium core, the Zircaloy cladding, and the supports. The end closures are discussed separately in Section III.B.3.

a. Uranium Core

There are three areas of concern in the uranium development program: fabricability, dimensional stability under irradiation (swelling, growth, bumping, and warp), and thermal cracking. Unfortunately, these problems are interrelated. Thus, the development program must take into account all factors which lead to long, expensive research and development efforts.

1) Fabricability

Acceptable coextrusions can now be made with uranium billets produced by casting 11-inch diameter ingots, triple-beta heat treating, alpha extruding to billet blank size, single-beta heat treating and machining to final size. The uranium contains about 150 ppm iron and 100 ppm silicon as additives for grain refinement. Substantial improvement is needed in the control of the metallurgical structure and the control of the dimensions, especially in the thickness of the inner cladding of the outer tube and the outer cladding of the inner tube. Work will continue through FY 1963 and into FY 1964 to continuously improve in these areas. As improvement in uniformity of cladding can be achieved, the cladding can be made thinner, initially, and substantial money saved in initial Zircaloy component costs. An attempt to optimize and/or justify the level of iron and silicon additives will be involved in the program for the coming year.

A study of possible alternate additives is needed to determine the optimum effects upon grain size, structure, surface chemistry and irradiation stability. In particular, the fabrication of uranium containing approximately 400 ppm iron and approximately 800 ppm aluminum will be studied and the comparative effects of Fe-Si and Fe-Al additives upon the fabrication of test fuel pieces will be determined. A charge of test elements for the KER loops will be made available by the 4th quarter FY-63. The long range goal of the uranium additive study is to develop enough basic metallurgy and practical technology data to permit us to custom design uranium alloy composition(s) and heat treatment(s) for fabrication and use in NPR.

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The U-2 w/o Zr alloy will also be evaluated for fabricability, irradiation performance, and defect performance. A charge of test elements for the KER loops will be made available by the 4th quarter FY-63. Previous tests have indicated that U-2 w/o Zr alloy has a slower oxidation rate than regular uranium and consequently the U-2 w/o Zr elements have a slower rupture rate. Further tests will be run to confirm these data and also to evaluate coextrusion performance and swelling performance of this alloy.

Other efforts will include evaluation of (1) the fabricability of normal uranium and recycle enriched uranium as compared to regular enriched material and (2) the effect of increased reduction ratios in the primary extrusion operation on the coextrusion behavior of the billets.

2) Fuel Behavior During Irradiation

The problem is to learn how the NPR fuel element will behave under all environmental conditions that it will be subjected to in the reactor. This information is needed for several reasons: (a) the dimensional changes in the fuel element during irradiation should be known so that the initial dimensions can be selected to allow for the changes, (b) at some (unknown) value of tensile strain in the outer cladding, a failure problem should be encountered, and (c) at some (unknown) value of compressive strain in the inner cladding, buckling will begin. Ideally, an expression is needed for the amount of swelling to be expected as a function of exposure, fuel temperature, coolant pressure and cladding thickness. This would take care of (a) above. With some knowledge of the cladding strain limits, failure criteria could be developed for the fuel to satisfy the other two needs.

There is not now in existence sufficient direct experimental data to permit prediction of the swelling rate of NPR fuel elements under the range of conditions that will exist in the N Reactor, nor has the theory been developed sufficiently to allow this prediction to be made. For use in the NPR fuel design studies, a limiting law was developed several years ago which was believed would predict an upper limit to the amount of swelling that will occur as a function of coolant pressure, exposure and fuel temperature:

$$\frac{\Delta V}{V} = 4.0 \times 10^{-6} E \text{ (solid fission products)} + 4.189 N r^3 \text{ (fission gas)}$$

$$N = 6.717 \times 10^{14} e^{-0.01167^\circ T}$$

$$E = MWD/T$$

$$^\circ T = \text{Temperature in Degrees Kelvin}$$

$$P = \text{Psi}$$

$$r^3 + \frac{0.02901}{P} r^2 - \frac{8.032 \times 10^{-6} E^\circ T}{P} = 0$$

The largest amount of swelling predicted for an NPR fuel element

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is 2.7 per cent for the outer tube and 2.0 per cent for the inner tube (assuming a coolant pressure of 1650 psi, peak goal exposures of 2300 and 1700 MWD/T and peak average core temperatures of 385°C and 405°C for the outer and inner tubes, respectively). The maximum uniform cladding strain corresponding to these swelling rates is estimated to be 0.5 per cent and 0.8 per cent for the outer and inner tubes, respectively, which is less than the expected "failure" limit of one to two per cent estimated from off-site data.

The completed prototype tests have provided some data; more data will be provided by planned tests. Data obtained to date are summarized below:

No. of Elements	Element Type	Exposure (MWD/T)	Avg. Fuel Temp. (°C)	Avg. Volume Increase, %	
				Measured	Predicted
4	KSE3 ⁽¹⁾	1200	460	1.3	1.6
4	"	2000	460	3.0	2.8
4	"	3600	410	5.0	4.6
5	NIE1 ⁽²⁾	500	240	0.2	0.3
4	NIN1 ⁽³⁾	1000	380	0.4	1.0

Peak NPR Conditions

Outer Tube	2300	385	-	2.7
Inner Tube	1700	405	-	2.0

- (1) A coextruded Zr-2 clad, single-tube element designed to approximate an N-fuel outer tube.
- (2) Enriched N-fuel inner tube.
- (3) Natural N-fuel inner tube.

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Much more data is needed to define the problem and to develop failure criteria. However, it is currently believed that, if we can avoid crud deposition, the NPR fuel element will operate below the critical cladding strain limit at the presently anticipated maximum operating conditions.

FY 1963 will provide the first opportunity to evaluate the performance of a complete N-fuel assembly. At the present time, two test charges of N Reactor fuel elements are being irradiated in the KER loops, and a single prototype element is completing its irradiation in the ETR. An accelerated testing program will be conducted in which pre and post-irradiation measurements and observations will be utilized to establish the general adequacy of N fuel performance, and to determine density and dimensional changes resulting from irradiation. Post-irradiation measuring equipment will be installed early in FY 1963.

Pre and post-irradiation dimensional measurements will be programmed into a computer and analyzed in terms of warp, wall thickness, O.D., I.D., and ovality. The changes in dimensions resulting from irradiation will be evaluated in terms of pre-irradiation dimensions, fabrication history and irradiation parameters. The results of this

evaluation should establish the effects of geometry variations and fabrication history on fuel performance.

Visual and radiometallurgical examinations of irradiated elements will be conducted to establish clad, fuel, closure, bond and support performances.

Density changes will be correlated with irradiation temperature and exposure to establish a swelling law in terms of irradiation parameters.

3) Thermal Stress Cracking

In reactor service, uranium behaves in a very peculiar manner, resembling "silly putty" in its mechanical behavior. During irradiation, under slow loading conditions, its behavior is essentially that of a viscous fluid, in that it will deform plastically under light loads and apparently will not support stress elastically. This behavior is seen when the fuel temperatures are comparable to or higher than those expected in the NPR. Irradiated uranium is brittle, however, in the absence of a neutron flux or under conditions of rapid loading. The presence of the fission products as unwanted alloying elements and the existence of internal cracks apparently are the cause of the loss of ductility. Internal cracks form as a result of the anisotropic thermal expansion characteristics of uranium. Microstrain in and between the grains occurs whenever the temperature of the metal is changed. As the fission product concentration increases, the metal's ability to flow plastically declines and microcracks begin to appear. There is reason to believe that the cracking occurs primarily during reactor shutdown when the embrittled fuel is quenched, with a tendency for the healing of the cracks during the subsequent operating period.

In the prototypic fuel tests performed to date we have not seen a fuel failure that resulted from a stress failure of the fuel core, but all uranium irradiated beyond about 1500 MWD/T has been found to have microcracks and, frequently, macrocracks. When the cracks extend to the cladding, there has been no sign that cladding failure was imminent or even likely. However, such failures have been observed in atypical tests and our data is so limited that one cannot now support the conclusion that thermal stress failure will not occur in NPR under unusual conditions of operation. Indeed, thermal stress failure constitutes a real technical limit to the operation of uranium metal fuel elements. A definition of the limit would depend on the geometry, specific power, cladding strength and the uranium quality and will be made by observing the cracking in the elements irradiated in KER loops by ultrasonic testers and some Radiomet examinations.

b. Zircaloy Cladding

Research and development on fuel cladding is centered around Zircaloy-2. Exploratory studies of other special zirconium alloys also are being pursued. The principal present-day problems involved in the use of

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Zircaloy-2 as a fuel cladding for metallic uranium are two in number: corrosion and hydriding, and clad stress failure.

1) Corrosion and Hydriding

Zirconium is a very reactive metal. The oxide coating formed on exposure to water or oxygen is protective under the expected clad exposure conditions in N Reactor operation (high purity water, adjusted to pH-10 with LiOH, at 275 to 300°C). After 72 hours in an autoclave at 400°C (anticipated nondestructive testing conditions) and 4 months at 300°C (approximate NPR service conditions), the anticipated corrosion penetration of good quality Zircaloy-2 is less than 0.1 mil. However, under abnormal conditions, as in the KER Loop fuel failure in low pH water in September 1959, crud deposits can lead to high temperatures and excessive corrosion. In the referenced case, 20 mils of cladding were corroded through in 20 days, a corrosion rate typical of temperatures near 500°C. A curve of corrosion rate versus temperature is shown in Figure 1. Note the rapid increase in corrosion rate in the temperature ranges of interest.

Hydrogen pickup and subsequent effects of cladding properties also is of concern. Of the hydrogen produced in the corrosion reaction, approximately 25 per cent finds its way into the cladding. Present indications are that 25-mil cladding will pick up about 35 ppm of hydrogen during the service life noted above. This amount, added to the 50 ppm allowed in the Zircaloy-2 billets, should not be enough to affect tensile properties under slow loading conditions. However, applicable data are scarce.

Thus, with the foregoing facts and tentative conclusions in hand, we are carrying out a program of study and development with the basic purpose of characterizing the behavior of Zr-2 in and around the environments expected during NPR operation. There is a growing realization that significant improvements over Zircaloy-2 are most likely to result from the development of a basic understanding of the corrosion and hydriding mechanisms of zirconium and its alloys. It is in this latter direction that we are aiming our work. Included in these studies are the development of data on the multiple rate transition phenomenon in zirconium, the effect of rate transition on hydrogen pickup, irradiation field effects on these rates and the influences of environment. Concurrently, studies of the electrical resistance of zirconium corrosion product as a measure of its lattice unsaturation and the ability of ions to diffuse through it and of the effect of stress pattern on distribution of hydride platelets within a corroding specimen are underway. The first of these latter studies has yielded information which strongly suggests that the ability of hydrogen to diffuse through the protective oxide coating is strongly dependent on the presence of lattice vacancies in the oxide. If this conclusion is correct, irradiation may serve simply as a means of introducing such vacancies, which would mean that the cause of the presently high (though not now dangerous) and unexplained hydrogen pickup rate in fuel jackets can be pinpointed. The stress pattern study was prompted by studies at

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Savannah River Laboratory which are interpreted by them to indicate that radially-oriented arrays of hydride platelets are formed as a result of tensile stresses in the fuel cladding and brittle fracture of thermally-cycled fuel elements follows. Formerly, it was believed that these hydride patterns resulted from metal texture.

2). Cladding Stress Failures

The cladding must be able to strain uniformly without fracture or necking in order to accommodate fuel swelling. Work by SRD and KAPL has indicated that Zircaloy-2 should undergo a uniform strain of one to two per cent before failure when used as a bonded cladding for metallic uranium fuel elements. The maximum anticipated strain in NPR fuel is less than one per cent, so no problem was anticipated. However, three elements, two rods and one tube, have failed in KER Loop tests by a mechanism in which localized deformation (necking) of the cladding occurred, longitudinally, to an extent of several inches over the surface of the cladding. Striations other than the one in which the failure occurred also were seen in each case. The uniform strain in the failures was less than one per cent in the two rod elements and about 2.5 per cent in the tubular element.

While the amount of uniform strain which a metal will withstand under creep loading generally is less than that seen under loadings typical of a tensile test, a limit of less than one per cent in Zircaloy-2 was quite unexpected. Therefore, it was postulated that other factors might be contributing: (a) nonuniform cladding thickness, (b) fast neutron radiation damage (KAPL tensile tests of sheet Zircaloy-2 have shown that a substantial loss occurs in the ability of sheet Zircaloy-2 to strain uniformly after irradiations equivalent to 1000 to 2000 MWD/T), (c) strain cycling (the NPR will subject the Zircaloy-2 cladding to strain cycling to a small degree).

As an immediate remedy, the clad thickness in the N fuel design was increased from 20 to 40 mils on the outer surface of the inner tube and to 25 mils on all other surfaces (the inner tube is felt to be more vulnerable than the outer tube because of the geometry, an equivalent amount of swelling will lead to greater cladding strain in a smaller tube). Since clad nonuniformity to some degree appears to be inherent in the coextrusion process, thickening the cladding reduces the relative variation in clad thickness, as well as strengthening the cladding.

Subsequently, a series of Zircaloy-2 clad rod specimens which had been irradiated in NaK capsule tests in the MTR to obtain swelling data were examined. The sixteen fuel rods in these tests yielded the following results:

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<u>Clad Thickness (mils)</u>	<u>No. of Elements</u>	<u>No. of Failures</u>	<u>No. with Striations (Including Failures)</u>
30	7	1*	0
20	9	6**	9

*brittle failure

**ductile failures

The rods were made with unalloyed uranium; a comparison of their operating conditions with those of the three element failures is shown in Table I.

TABLE I

<u>Variable</u>	<u>Rod Elements</u>	<u>Tubular Elements</u>	<u>Capsule Specimens</u>
Maximum Central Metal Temperature, °C	500	430	485-640
Clad Surface Temp., °C	260	287	195-365
Exposure, MWD/T	2500	3200	1035-2081
Clad Thickness, Mils	20	20	20-30

The following observations resulted from detailed examinations of the various pieces:

1. There is no direct evidence that there is a cladding temperature effect on the susceptibility to thinning or fracture in the cladding temperature range of 195 to 365°C.
2. Both cladding thickness and thickness variability are factors which influence the susceptibility of Zircaloy-2 cladding to localized instability.
3. The total cladding strain on bonded fuel elements apparently is unrelated to the fracture mechanism. The strain measurements and observations most needed are those immediately adjacent to the fractures and thinning.
4. If irradiation damage has any effect in the exposure range of interest, the threshold exposure for this effect is less than 1100 MWD/T (approximately 10^{20} nvt).

Another program to explore the clad thinning phenomenon is underway. Further capsule irradiation tests will be charged during the second quarter FY-63. Laboratory studies of the notch-sensitivity of Zircaloy-2, both before and after irradiation, are being pursued and analytical studies of Zircaloy-2 deformation as a result of swelling are in progress. However, considerable data must be obtained to support the above-noted observations and to lay the foundation for the establishment of design and material limits. Our goal is to improve our understanding and our fabrication techniques to the point that substantial reductions in Zircaloy-2 cladding thickness, with their attendant reductions in fuel costs, can be realized without a deterioration in fuel performance.

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c. Supports

The fabrication, precision sizing, attachment, and proof testing experience on fuel element supports and locking clips, as well as irradiation test experience, have outlined some serious problem areas. Some research and development effort, primarily in the area of Zircaloy physical properties and fabricability, will be carried out in conjunction with a strong process assistance effort.

Studies will be continued to establish adequate specifications for vendor supplied support hardware and to establish the adequacy of current supports to withstand both charge-discharge and irradiation requirements. New supports and locking clips will be designed and subjected to laboratory evaluation in order that backup support hardware will be available for possible use in the production of N fuel. Both strength and fatigue properties of present supports and new supports will be established and evaluated in terms of fabrication, handling, and service requirements.

3. End Closure Development

a. Present Closure

The present closure design requires chemical milling of uranium to provide an annular recess in the ends of the fuel tubes, brazing a Zircaloy ring in place with a 5 Be:95 Zircaloy-2 braze, and TIG welding the proposed braze line. Closure fabricated by this process have performed satisfactorily in irradiation tests to date. However, this particular closure process and closure product have several unique shortcomings. These are:

- 1) The brazing operation, at about 1050 C, raises the uranium into its gamma phase and the Zircaloy into its beta phase and thus makes the metallurgical structure of the core, clad, and bond, at the ends of the element different from the center of the element. These structural differences probably lower the performance potential of the elements.
- 2) The alloy braze ring is extremely expensive.
- 3) The temperature control is very critical -- low temperatures cause poor braze bonds; high temperatures cause melting of the uranium -- yet the accuracy of the controlling optical pyrometer has not been well determined.
- 4) The welded closure is not of the same chemical composition as the remainder of the fuel element -- it contains varying concentrations of beryllium and uranium contamination. This composition anomaly causes two problems: (1) potential fission product contamination of the primary coolant of the reactor which would reduce the effectiveness of the rupture detection instrumentation and (2) variation in corrosion resistance of the weld bead which causes white oxide formation during processing and potential high corrosion rates in-reactor.

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- 5) The beryllium is toxic and therefore close process control is required in the fabrication operations.

In addition to the above unique problems, there also exists the problem which must somehow be reckoned within any closure design on zirconium-clad uranium metal fuel, i.e. the large thermal incompatibility between zirconium and uranium. The thermal expansion coefficients of uranium and zirconium from room temperature to 400 C are approximately 18 and 6 microinches per inch-°C, respectively. During operation, the uranium is hotter than the zirconium which further increase the degree of incompatibility. One effect of this situation is the step in the cladding that exists at the core-end cap interface after brazing. On heating, the uranium expands more than the end cap, plastically deforming the cladding. The braze melts, fills the void space and freezes while the outer gap between the end cap and the cladding is expanded. On cooling, the end cap and braze contract less than the uranium and a step forms. At room temperature, the metals are deformed plastically to accommodate the relative dimensional changes and are loaded elastically. During operation strain cycling occurs. In the worst case, shear failure of the cladding at the cap-core junction would result.

Both research and development and process assistance efforts will be made during FY-1963 to improve the quality of the closure and reduce its cost. Primary efforts will be to improve the temperature control of the operation to reduce the price of the braze rings and to improve the braze joint design.

b. Alternate Closures

To date, we have not encountered shear failures in prototypic N fuel elements. However, irradiation experience is quite limited. We have had failures in unbonded pieces and in elements brazed with a Zr:12 w/o Fe:5 w/o Be alloy, at conditions quite unlike NPR operating conditions. This latter material gives more brittle braze layers than does the Zr:5 w/o Be alloy. However, the pertinent conclusion is that such failures of brazed closures are ever-possible, and, further an element with a cracked bond or no bonds at the cap-core interface may be more prone to shear failure than an equivalent element of unbonded design.

Strain cycling of uranium elements is not new. However, the experience with zirconium cladding has been mainly with ceramic fuels where the degree of strain cycling is far less. Most metal fuel performance data have involved the flat plate geometry. Thus, reasonable extrapolation of the experience of others to NPR fuel is not possible. Further, the first test of the braze closure which will produce results with significant statistical validity will be the first load operation of N Reactor.

Because of the potential shortcomings of the present closure design and the lack of experience with it, alternate closure studies are necessary. At present, a diffusion-bonded closure, using either gas pressure or

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mechanical bonding and a hot-headed and projection-welded closure are under development. The former involves cathodic etching of the uranium surface, deposition of a thin copper plate on the uranium and inner cladding surfaces and pressure bonding. The latter uses a projection-welded cap with a braze filler. In the long range, an integral closure, formed during the coextrusion, would be desirable if it would prove technically and economically feasible. Samples of one or more alternate closures should be under irradiation by mid-FY-1963.

Work will be carried out on a gas-pressure bonding technique which it is believed will produce an excellent closure. The pressure bonding of Zircaloy to uranium has been demonstrated in the uranium alpha phase, and the bonding of Zircaloy to Zircaloy at uranium alpha phase temperatures has been shown to be feasible utilizing a thin layer of another interface material. Continued activity will be concentrated now at utilizing these developments to produce a satisfactory end closure.

Another alternate closure is termed "hot-headed and projection welded". In previous work such a closure was developed. It required that the fuel tube be cut to length, the cut ends of the fuel simultaneously back extruded in order to cause the bonded cladding to flow back over the major portion of the fuel ends, and the fuel ends subsequently machined flat. A Zircaloy ring was projection welded to the fuel tube to complete the closure. This closure design is intrinsically strong; produces no metallurgical damage to the fuel clad, or bond; and performed well in small scale irradiation tests. The fabrication technique eliminates most of the tolerance problems which arise in solid-state bonding a Zircaloy ring into a recessed fuel tube. However, the closure in its original form was not bonded across the full end face of the fuel. Work is in progress to modify the projection welding step, either by the use of braze material between the cap projections or by modification of the projection design in such a way as to make possible a continuous bond across the face of the welded ring. When this objective is met, the closure produced will be thoroughly evaluated by ex-reactor non-destructive and destructive examination, test elements for irradiation will be fabricated, and an irradiation test charged as soon as test space is available.

Another alternate is the copper brazed and welded concept. A closure is under development in which the uranium at the ends of the cut fuel tube is recessed, a Zircaloy ring is electron beam welded in place and the bonding of the ring completed by application of heat and mechanical pressure. Bonding of the ring, particularly to the Zircaloy clad, is promoted by the use of copper as a bonding agent. Some additional work on the dies for the application of mechanical pressure is required. Closures fabricated by this process will be nondestructively and destructively evaluated, and if the process achieves quality and uniformity of product, test elements for irradiation will be evaluated.

4. Warp and Straightening

Warp of the as-fabricated fuel tubes and of the fuel components during processing will continue to be characterized. This information may be

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essential to characterization of irradiated fuel specimens. A hot roll straightening process has been developed which permits recovery of warped pieces while simultaneously promoting stress relaxation. The use of this process may produce improved irradiation stability and permit recovery of that portion of the output which currently is rejected for warp beyond recovery limits. Warped fuel components will be selected for recovery by this process, straightened, and completed for irradiation testing. A comparative test of elements which remained within warp limits during all fabrication steps and of elements straightened by the hot roll process may be in order. Such a test may help to define the warp problem in general as well as determine the practicability of charging straightened elements.

5. Heat Treatment

At present, the salt bath process of heat treating appears to be satisfactory, but poses substantial operational problems related to safety, and requires subsequent vapor blasting operation which costs money and removes a small layer of Zircaloy. Some work will be performed to determine the desirability of developing alternate processes. Among these alternates is induction heat treating which has the advantage of cleanliness and fluid bed processes which have the attributes of bed temperature uniformity with high rates of heat transfer.

6. Testing of Ruptured Fuel

An extensive ex-reactor defect testing program has led to a fairly good characterization of the failure process when irradiation effects are missing. It was realized early that irradiation effects, including internal heat generation and radiation damage, would be important. Hence, the ETR rupture testing program was started to scout problems and the PRTR rupture loop has been planned so that NPR fuel failure characteristics and the rupture monitor response can be studied thoroughly.

Fuel element failures are a source of concern because of the possibilities that (a) failures will occur in such a way as to i) cause tube failure or ii) require frequent tube replacement, either because of hydriding or mechanical damage, and (b) failures will cause a reduction in coolant flow and precipitate other failures before the reactor can be shut down. Thorough study of the failure behavior problem requires careful integration of the fuel and the monitor system aspects of the problem in tests carried out under prototypic conditions. Repetitive testing is required as is an imaginative approach to simulation of failures. The PRTR Rupture Loop Program should have all these elements.

a. ETR Tests

In the ETR, a series of KSN1 elements at 0, 300 and 1700 MWD/T, have recently been failed. This loop cannot be cooled slowly, in simulation of NPR shutdown conditions, so tests have been run at full power until the coolant activity limit is reached. This produces a failure which is worse than would be expected in the NPR but is regarded as the most productive testing method for this loop. No further fuel rupture tests

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per se are planned for the ETR as the PRTR rupture loop will be in operation during FY-1963. A test or tests of in-reactor rupturing mechanisms will be carried out in the ETR during FY-1963.

b. IRP Tests

The IRP loop is used for subjecting defected, irradiated elements to different time-temperature programs. The irradiated element is defected by drilling a hole in the cladding in the Radiometallurgy Laboratory. Thus all irradiated elements are potentially available for testing in this loop. The test element is exposed to 300 C water until a radiation detector shows that fission products are being released to the coolant. Then the loop water may either be maintained at 300 C or cooled down at different rates, simulating various NPR cool-down options. In several respects, this is a better test than we can do in the ETR 3x3 loop. In two recent tests, an unalloyed uranium tubular element and a uranium-2 per cent zirconium element, both irradiated to 3200 MWD/T in KER-2, were tested under identical conditions. The gross corrosion rate of the unalloyed element was about 20 times that of the U:2 w/o Zr element. In corrosion tests of unirradiated fuel, the unalloyed uranium corrodes three to five times faster than U:2 w/o Zr.

The principal results of the failure behavior studies to date, in addition to providing a broad background of experience and data on corrosion rates and the data on unalloyed uranium versus U:2 w/o Zr given above, have been: (1) demonstration that radiation damage in uranium, after about 1000 MWD/T, leads to cracking or a disposition to cracking which results in a significantly higher gross corrosion rate, (2) demonstration, in a limited number of tests, that when an annular space of 100 mils or greater is maintained, the failing fuel element does not cause hydriding of nor damage to nearby zirconium components, and (3) demonstration, in IRP loop tests, that with cool-down rates typical of an NPR scram, very little corrosion of uranium occurs compared to what is seen after a test with the normal shutdown" rate of cooling.

Although extrapolation of IRP loop test results to N conditions is not unambiguous, the low cost and flexibility make further testing in this facility attractive. Twelve tests are planned for FY-1963.

c. PRTR Tests

Although considerable in-reactor and ex-reactor testing has been completed, additional in-reactor study using prototypic NPR fuel is needed to determine the effects of exposure, cooling rates and defect location on the post-failure corrosion damage incurred by an operating fuel element. These data will be used to assess the potential hazards which may be associated with a fuel element failure.

Development and in-reactor testing of an improved defecting device, capable of defecting any surface of an N fuel element, will be completed prior to the startup of the PRTR rupture loop in the second quarter of FY-1963. A test program will be initiated in the PRTR immediately following

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loop startup. This program, using fuel pre-irradiated to exposures up to 2000 MWD/T, will extend into FY-1964 and will provide data on both N Reactor rupture instrumentation performance and fuel element rupture characteristics. An unirradiated element will be failure tested late in the second quarter to test the equipment and shakedown the loop. An N outer tube, irradiated to 2000 MWD/T will be failure tested during the third quarter of FY-1963.

7. Advanced Development

In addition to the development efforts in FY-1963 to improve the quality of the present tube-in-tube NPR element and the effort to more clearly characterize the quality of the present design, a substantial effort will be devoted to (1) developing elements of more advanced design and (2) developing advanced coextrusion technology.

a. Single-Tube Element

The fabrication cost of NPR fuels can be markedly reduced by substituting a single-tube geometry for the present tube-in-tube design. Calculations indicate that a single-tube element will operate in NPR with acceptable heat fluxes but with moderately higher fuel temperatures. Data are needed on the performance of uranium fuel elements irradiated at high alpha-phase temperatures.

A dual-enrichment, single-tube fuel element of NPR geometry is currently being irradiated in the ETR under NPR conditions. This irradiation will be completed and the irradiation performance evaluated in FY-1963. Uniformly enriched, single-tube elements may be fabricated for irradiation in KER loops. These elements will be designed to operate at fuel temperatures and heat fluxes which will simulate those anticipated for a single-tube NPR fuel element. Post-irradiation evaluation of these test components will provide data on swelling and high temperature fuel performance which will be utilized in the design and testing of prototype single-tube NPR fuel elements.

b. Fluted-Tube Element

Plastic strain of NPR fuel cladding will occur as a direct result of fuel swelling, with the total observed strain increasing with increased exposure and fuel operating temperatures. As a consequence, the ultimate life of a tubular element may be limited by clad performance, e.g., either inner bore buckling or localized plastic deformation and failure of the outer clad. A fluted outer clad potentially eliminates clad performance as a limiting factor by allowing swelling of the fuel to occur without creating high tensile stress in the outer clad or high compressive stress on the inner clad. Fluted tubular elements of approximately NPR-inner geometry have been fabricated and will be irradiated and examined during FY-1963.

An NPR size fluted single-tube element has also been designed, and fabrication and irradiation of this element will be completed during

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FY-1963. Irradiations will be conducted on an interim measurement basis. Test results and post-irradiation examination results will be used to evaluate fuel performance and as a basis for design modifications and larger scale testing.

c. Coextrusion Technology

During FY-1963 extrusion development work will be directed toward:

- 1) Developing materials and techniques for fabricating extremely thin (.005 to .015 inch) clad by coextrusion,
- 2) developing improved extrusion lubricants, e.g., glass,
- 3) evaluating the use of steel cans in place of the present copper cans in order to improve clad uniformity and/or reduce costs,
- 4) determining the cause and nature of "end effects" in the end billets in primary extrusions.
- 5) developing methods of fabricating integral end closures on NPR fuel elements, and
- 6) evaluating the effects of basic uranium and Zircaloy-2 metallurgical properties on the quality of the coextruded products.

8. Irradiation Effects in Zircaloy-2

NPR process tube life and fuel element cladding behavior is dependent upon the combined effects of neutron irradiation and a hot water environment on the mechanical properties of Zircaloy-2. To be assured of safe operation of the NPR, surveillance of changes in the pertinent properties is being maintained. In order that integrated exposures may be obtained in advance of their occurrence in the NPR, irradiation of test coupons is being performed in the core of the ETR. In this high flux reactor, the entire operating life exposure of Zircaloy-2 process tubes in NPR can be simulated in approximately five years. Environmental conditions are quite similar to those expected in the NPR since the irradiations are being performed in a water-cooled high temperature loop.

Tensile properties as a function of orientation are being measured before and after irradiation on coupon specimens with cold work levels characteristic of NPR process tubing. Post-irradiation ductility and strength will be the major criteria for measuring deleterious effects in this alloy. In addition, notched tensile specimens are being irradiated and tested to measure the tendency of Zircaloy-2 to fail in a brittle catastrophic manner after irradiation.

9. Thermocouple Crud Probe

The possibility exists that crud deposition may occur on operating NPR fuel elements. It is necessary, both for experimental loop decontamination

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studies and for N Reactor use, that reliable thermocoupled crud probes be developed. Hopefully the present probe geometry is adequate, however, additional effort will be required to develop reliable techniques for inserting a thermocouple into the probe in such a manner that integrity of the assembly is not degraded. The use of Zircaloy-clad thermocouples should alleviate the basic problems which have been encountered in the use of stainless steel thermocouples in the past. During FY-1963, probes with Zircaloy-clad thermocouples will be evaluated. Test probes will be fabricated for use in loop decontamination studies and for irradiation evaluation once performance has been demonstrated. The first probe with Zircaloy-couples will be ready for reactor charging during the second quarter of FY-1963. Volume testing of crud probes of this design (with stubbed-off thermocouples) will be carried out during the third quarter of FY-1963. A fabrication and inspection process will be developed by the fourth quarter of FY-1963. Ten crud probes will be fabricated, inspected and ready for reactor use during the first quarter of FY-1964.

Backup designs will be developed concurrently to the extent that our experience indicates is prudent and to the extent that our testing facilities will permit.

10. Friction and Galling Tests

Friction and galling charging tests carried out using self-supported fuel elements in aluminum and Zircaloy process tubes during FY-1962 have shown that changes in the process tube or fuel support materials require re-evaluation of the charging forces to be anticipated, and of the incentives for the use of charging oil. Preliminary results from the studies are informative, however, additional work is needed in at least three specific areas:

- 1) Further definition of the relationship of charging oil to the sliding friction between similar and dissimilar materials. This study will bear directly on charging practices both in the older reactors and in N Reactor.
- 2) Evaluation of the coefficient of friction associated with specific aluminum self-support designs.
- 3) Discharge forces associated with purposely defected self-supported fuel elements.

With the exception of determining the coefficient of friction associated with specific self-support designs, this program should be complete by the end of FY-1963.

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