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PROGRAM FOR CRUD AND CORROSION STUDIES
FOR PROPOSED HANFORD REACTOR CONDITIONS

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PROGRAM FOR CRUD AND CORROSION STUDIES
FOR PROPOSED HANFORD REACTOR CONDITIONS

INTRODUCTION

This report presents a proposed test program to study the corrosion of aluminum-clad fuel elements under conditions expected at the more severe conditions which will be encountered in operating the present Hanford reactors at higher power levels.

SUMMARY

Fuel element corrosion will be measured at outlet temperatures up to 110 C and surface temperatures up to 150 C. Heat fluxes will range up to 340 watts/cm², over twice as much as for existing corrosion data. Self-supported solid elements will be used having cores of U²³⁵ - Al alloy clad in X-8001 aluminum. Thermo-couples will be included in some elements to measure jacket temperatures. Testing will include a comparison of pH 6.5, 7.0, and 7.5, and a study of reduced dichromate and of substitutes for dichromate. Nickel-plated elements will be included. This testing will use four of the 1706-KE single-pass tubes through 1962. Replacement of the present aluminum tubes with Zircaloy tubes is required.

DISCUSSION

Purpose

The proposed increases in the power levels of the existing Hanford reactors during the next few years, will necessitate a careful evaluation of the problems of fuel element corrosion which may be encountered at the more severe operating conditions expected.

It has been forecast⁽¹⁾ that by 1965 the reactors will be operating at a maximum specific power of 109 KW/foot, as compared to about 75 at present. Surface temperatures and flow rates are also expected to increase significantly. If, as expected, hot spots are eliminated or greatly reduced by the use of self-supported elements, then uniform corrosion will become a more important factor in limiting fuel element power. Past experience has demonstrated^(3,4) that such corrosion increases greatly with the heat flux through the corroding surface. In I and E elements, such as will be used in future loadings, very high heat fluxes are produced in the hole. Thus at 109 KW/ft, the heat-flux in the hole of an element of 0.410" ID and 31.1% heat split will be 340 watts/cm². Present usable data on fuel element corrosion, which were obtained with solid slugs, cover a range only up to about 160 watts/cm². Prediction of corrosion rates at the expected higher heat fluxes by extrapolation of data obtained over a considerably lower range of conditions is uncertain; widely different rates may be predicted depending on which of several corrosion correlations one chooses to use as the basis of extrapolation. A few recent tests (13) of enriched I and E elements have been of limited value in shedding light on this problem. It is therefore evident that corrosion measurements at higher

heat fluxes are required in order to predict the corrosion rates which will be encountered in anticipated reactor operation.

One important reason for the increase in corrosion rate with heat flux is believed to be the temperature drop across an insulating layer of oxide and externally deposited crud. The thickness of this layer is believed to depend on water quality, temperature, power level, and perhaps other variables. It is possible to measure this temperature drop and to study its dependence on the above variables by inserting thermocouples in the aluminum cladding.⁽⁵⁾ However, such measurements have not been made for active slugs in Hanford process water.

Non-uniform attack, such as groove pitting and erosion-corrosion, may also become more serious than under present conditions. It is therefore necessary to determine the conditions at which such attack will be serious and how it may be avoided.

When going to significantly more corrosive conditions, it is also necessary to re-evaluate the optimum water quality. The presence of more severe conditions will provide an added incentive to lower pH to reduce corrosion of the fuel elements, whereas the replacement of aluminum tubes with Zircaloy will eliminate one incentive for the lower pH. The reduction of aluminum corrosion by a given reduction in pH may well be different for corrosion under the more severe conditions. The effectiveness of corrosion inhibitors may also be somewhat different. There is also a need to find an inhibitor to replace all or part of the dichromate now being used, in order to reduce both cost and effluent activity. Therefore, corrosion data at the expected future reactor operating conditions is required over a range of pH and for different inhibitors and concentrations thereof.

Test Facilities

1. Single-Pass Mockup Facilities. The single-pass mockup facilities are being re-designed to provide more versatile use and more economical operation. The present system of 6 complete full-scale reactor flow systems⁽¹⁵⁾ will be changed to a system with 2 full-scale flow systems and 4 smaller facilities. The smaller facilities will use small samples and will be suitable for more accurately defining small differences in corrosion rates and water quality characteristics. The remaining mockup tubes will be used to confirm the small-scale results in the full-scale geometry prior to actual reactor testing.
2. Single-Pass In-Reactor Facilities. This program will use four of the present eight tubes which receive water from 1706 KE on a once-through basis.^(12,15) Water treatment for each pair of these tubes can be independently varied.

The present ribbed aluminum process tubes will be replaced by smooth-bore tubes of Zircaloy-2. The proposed use in these tests of solid elements operating at high heat flux requires the use of self-supported elements in smooth-bore tubes in order to reduce the danger of hot spots from poor alignment or flow channelling and in order to provide uniform and known corrosion conditions around the circumference. Since present plans

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call for the retubing the Hanford reactors with Zircaloy, it is preferable to replace the present ribbed tubes with Zircaloy rather than ribless aluminum. This will permit more realistic piloting of expected reactor conditions in future tests in the single-pass tubes.

Zircaloy tubes of C-Reactor diameter (1.681" I.D.) will be used. Six such tubes of K-Reactor length are now available, whereas K-diameter tubes would not be available before October. The C size has the additional advantages of (1) giving higher linear velocities and (2) having nearly the same ID as K-size aluminum tubes, so the same fuel elements could be used. Orifices will be installed in the outlet fittings to raise the outlet pressure and prevent surface boiling at high heat flux. A device to retain these orifices has been designed and approved for use on these tubes.

The replacement of four tubes with Zircaloy for this corrosion test program is part of a program for retubing all eight of the single-pass tubes supplied by 1706 KE. Of the other four tubes, two are to be replaced by C-diameter ribless Zircaloy tubes and two with K-diameter aluminum tubes. In connection with this retubing the two tubes now in the fringe zone will be moved to central locations. These other four tubes will be used principally for the study of effluent activity.

Fuel Elements

The measurement of corrosion rates at high heat fluxes imposes limits on the type of fuel elements which can be used. The use of solid elements with enriched metallic uranium cores of convenient diameter will produce excessive core temperatures. The use of I and E elements for corrosion measurement has been generally unsuccessful. The hole and annulus run at different conditions and therefore have different corrosion rates, neither of which can be measured independently from the weight loss of the element. The high heat flux of interest is obtained in the hole which is only a small fraction of the corroding surface. Direct determination of corrosion in the hole by sectioning and measuring remaining clad thickness is unreliable because of dimensional changes during irradiation, and uncertainty of the initial clad thickness. Measurement of clad thickness by an eddy current device appears to be impractical. For these reasons solid cylindrical elements with cores of Al-U²³⁵ alloy have been selected for corrosion measurements. Such cores have the additional advantage of greatly reducing the possibility of damage to the tube in the event of a rupture.

Except for the first loading, the fuel elements will be made of "J" cores (7.5% U²³⁵ in Al), 8.29" long, clad in regular X-8001 aluminum jackets of 1.44" O.D. These elements should provide the desired maximum heat flux (340 watts/cm²). Since the "J" cores must be obtained from off-site and will not be available in time, the first loading will use similar elements made from "C" cores (4.5% U²³⁵ in Al, 6.0" long).⁽¹⁴⁾ These cores are now available at Hanford, and are being made into fuel elements.

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The elements will have special supports, which will be welded to the ends of the elements and not contact the lateral surface. This will reduce flow irregularities on the heat-transfer surface, and prevent cladding hot spots due to formation of insulating oxide under the supports.

Cores of Pu-Al could be used as an alternate to U²³⁵-Al. Such elements could be fabricated at Hanford.

The thermocouple elements and the method of leading the thermocouple wires out the rear nozzle will be the same as have been used successfully in several tests in the KER loops. (5,6,8,9,10) Cores of U²³⁵-Al will be canned in X-8001 Al tubulars of 1.44" OD with 1/8" walls. Three holes for 1/16" diameter thermocouples will be drilled into the thick jacket from one end to a depth of 1 3/4".

Regular natural I and E elements will be used at the upstream end of the charges as heaters, since water temperatures are too low in the upstream half of the charge to measure significant corrosion. The C-IV-N elements of appropriate dimensions, designed for the initial loadings of the C-reactor pilot retubing program, are available. (2)

Loading Patterns

In general, two loading patterns will be used, with the thermocouple slugs operating at different power levels. Both loadings will have 19 C-IV-N elements at the upstream end, the nineteenth being a water-mix element. These will be followed by a 4" solid dummy. One loading will then have 9 "J" elements (overall length 9.3") or 12 "C" elements (overall length 7.0"), the last one being a thermocouple slug. The other will have 15 "J" or 20 "C" elements, the last one again being a thermocouple slug. These two loading patterns will place the thermocouple slugs in the No. 10 and No. 4 positions respectively (in reference to standard adjacent tube loadings), or at 76% and 39% of the peak flux in the tube, assuming a cosine distribution.

Such loadings are expected to give the following conditions for the corrosion test pieces (using "J" elements):

Water temperature at first test element	50-60 C
Water temperature at tube outlet	90-110 C
Surface temperature	130-150 C
Heat flux	140-340 Watts/cm ²
Water velocity	33-40 feet/sec

The U²³⁵-Al (or Pu-Al) cores which will be used in the downstream loadings have a high reactivity compared to normal reactor loadings. It will therefore be necessary to charge some Mint or depleted uranium elements in adjacent tubes to prevent an excessive local power level.

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Testing Programs

This testing is divided into three scheduled phases, with additional supplemental phase welded as Phase 4. The first step in the program is to determine the corrosion and film deposition characteristics of the present water quality, and determine how these vary with fuel element heat flux and surface temperature. After present water quality effects have been measured, second phase tests will define the effects of inhibitor types and concentrations. Third phase of this program will be to prove the optimum process water pH values at the proposed conditions of temperature, power level, and flow velocity. Supplemental testing may be required to complete the program. These tests may be needed to study other variables or conditions which may be indicated from results of the scheduled tests, or may be needed to retest certain conditions.

Operating conditions for each phase of this program are shown in Table I. Each part of the testing program is described in detail below.

1. Initial Testing-Phase I. The first phase will require one satisfactory exposure period. This testing will begin as soon as the Zircaloy tubes are installed, and the fuel elements ready. This will probably be October, 1960. Approximately three months operation at equilibrium reactor level will be required to provide good corrosion measurements. This would bring the first test to completion in January, 1961.

The water used during this initial testing will be the present process water at pH 7.0, and this same water with pH reduced to 6.5. All water will be supplied by the 1706 KE facility.

The number, and type, of test elements are listed in Table I for Phase I, and are described above under Fuel Elements and Loading Patterns. The corrosion test pieces and thermocouple slug will be made of "C" alloy (4.5 w/o U²³⁵ in Al), and therefore operate at lower heat fluxes than those used in Phases II and III.

2. Phase II Testing. The second in-reactor testing phase will include three exposure periods, with each exposure period to cover three months of equilibrium reactor operation. This second phase will start in-reactor immediately after completion of the first phase testing; that is, January, 1961. The second phase testing will continue through December, 1961.

This phase of the test program will compare different concentrations of corrosion inhibitor, and different inhibitor chemicals. Inhibitor testing will be underway in the single-pass mockup facilities prior to and during the initial in-reactor testing of this second phase of the program. The purpose of this testing will be to determine a combination of inhibitor chemicals which will permit reduction of dichromate concentration without significant cost increase.

Hopefully, a combination of inhibitors may be found which can replace dichromate altogether. Inhibitor combinations which show some promise include:

- (1) Nitrate with dichromate
- (2) Nitrite with dichromate
- (3) Silicate with dichromate
- (4) Quachrom glucosate
- (5) Sodium chrome glucosate

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The one, or two, most promising combinations will be tested in-reactor.

During all three exposure periods in this second phase, all tubes will be cooled with water at pH 7.0. Two tubes will be cooled with regular process water with 2 ppm of dichromate. Two tubes will be cooled with water containing only 1 ppm of dichromate during the first and second exposure periods, and then cooled with water containing the recommended concentrations of inhibitor combinations as determined from out-of-reactor testing. This phase of the testing will also compare the performance of nickel plated elements with bare aluminum-jacketed pieces.

3. Phase III Testing. The third-phase testing in this program will define optimum process water pH for cooling the reactors at the proposed operating conditions. Two exposure periods, of about three months each, will complete this phase of the program. This final phase will start about December, 1961 and will be complete by August, 1962.

For this testing, two tubes will continue to operate as controls, with pH 7.0 process water, while two tubes will operate at different pH values. However, the corrosion inhibitor for this phase of the testing will be that combination and concentration of inhibitors which were determined by the second phase testing to be most effective. This inhibitor combination will be used in all four tubes during this third phase testing. The two tubes on test conditions will be cooled by pH 6.5 water during the first exposure period of this phase of the program, and will be cooled by pH 7.5 water during the second exposure period.

Zircaloy jacketed fuel elements will also be tested during this third phase of the program. The corrosion and film formation rates will be measured for the Zircaloy jackets at the three different pH values, and the surface temperatures will also be measured for the Zircaloy jacketed elements. Fuel elements autoclaved in chromic acid are also candidates for testing in this program. (7,11) Chromic acid autoclaving significantly reduces uniform corrosion rates for aluminum. A severe disadvantage to this process has been localized (pitting) attack at points where the autoclave coating has been scratched or dented. With self-supports attached to the elements, the element surfaces will no longer be susceptible to scratching from normal handling and charging. Localized corrosion of chromic acid autoclaved elements should then be negligible on self-supported elements. Further tests may be required to finish this program.

Complete determination of inhibitor combinations and/or concentrations may require several additional exposure periods in-reactor, even though much

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preliminary development will be done in mock-up systems out-of-reactor. And, results of the scheduled tests may show different directions which the tests should go: such as tests of different aluminum alloys, tests of stressed samples, etc. Also, operational problems may spoil a test, so that retesting is needed. Fuel element ruptures, unusual reactor operation (such as heat cycles), or ambiguous test results may require a re-run of one of the scheduled tests.

R B Richman / DRD

R. B. Richman
Coolant Systems Development Operation
HANFORD LABORATORIES OPERATION

D R Dickinson

D. R. Dickinson
Coolant Systems Development Operation
HANFORD LABORATORIES OPERATION

RER: DRD: mf

TABLE I

	Charge Date	Discharge Date	pH	Inhibitor	Inhib. Dosage ppm	Thermo-Couple Element Position	Number of Test Elements	Jacket Material	Core Material
PHASE I									
Tube 1	October 1960	January 1961	7.0	Dichromate	2	#10	12	X-8001 Aluminum	4.5% U ²³⁵
" 2	"	"	7.0	"	2	4	20	" "	"
" 3	"	"	6.5	"	2	10	12	" "	"
" 4	"	"	6.5	"	2	4	20	" "	"
PHASE II									
First Loading									
Tube 1	January 1961	May 1961	7.0	Dichromate	2	#10	9	X-8001 Aluminum	7.5% U ²³⁵
" 2	"	"	7.0	"	2	4	15	" "	"
" 3	"	"	7.0	"	1	10	9	" "	"
" 4	"	"	7.0	"	1	4	15	" "	"
Second Loading									
Tube 1	May 1961	September 1961	7.0	Dichromate	2	10	9	X-8001 Al & Ni	"
" 2	"	"	7.0	"	2	4	15	" " Plate	"
" 3	"	"	7.0	"	1	10	9	" "	"
" 4	"	"	7.0	"	1	4	15	" "	"
Third Loading									
Tube 1	September 1961	December 1961	7.0	"	2	10	9	" "	"
" 2	"	"	7.0	"	2	4	15	" "	"
" 3	"	"	7.0	Combination	?	10	9	" "	"
" 4	"	"	7.0	"	?	4	15	" "	"
PHASE III									
First Loading									
Tube 1	December 1961	April 1962	7.0	Combination	?	10	9	X-8001 & Ni-Plate	7.5% U ²³⁵
" 2	"	"	7.0	"	?	4	15	X-8001 & CrO ₃ auto.	"
" 3	"	"	6.5	"	?	10	9	X-8001 & Ni-Plate	"
" 4	"	"	6.5	"	?	4	15	X-8001 & CrO ₃ autoclave	"
Second Loading									
Tube 1	April 1962	August 1962	7.0	Combination	?	10	9	X-8001 & Ni-Plate	"
" 2	"	"	7.0	"	?	4	15	X-8001 & Zr	"
" 3	"	"	7.5	"	?	10	9	X-8001 & Ni-Plate	"
" 4	"	"	7.5	"	?	4	15	X-8001 & Zr	"
SUPPLEMENTAL									
First Loading									
Tube 1	August 1962	December 1962	?	?	?	?	?	?	?
" 2	"	"	?	?	?	?	?	?	?
" 3	"	"	?	?	?	?	?	?	?
" 4	"	"	?	?	?	?	?	?	?

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