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BAW-3765-3
Joint U. S.-Euratom
R&D Program
August 1967

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H.C. \$ 3.00, MN 65

ING

OF ZIRCALOY

— Quarterly Technical Progress Report No. 3 —

April 1 to June 30, 1967

B. W. Whitaker
Project Manager

C. R. Johnson
Technical Manager

AEC Contract No. AT(30-1)-3765
B&W Contract No. 59-3101

Performed for
THE UNITED STATES ATOMIC ENERGY COMMISSION
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SUMMARY

The principal objective of this program is to provide a better definition of the operational limitations for Zircaloy fuel cladding. During service in water-cooled reactors, Zircaloy cladding will corrode and absorb some of the hydrogen released by the corrosion reaction. The corrosion rate of Zircaloy in high-purity water at the temperatures of pressurized and boiling water reactors is not excessive, and therefore is not considered a limiting criterion. However, hydrogen does adversely affect some of the mechanical properties of Zircaloy; consequently, excessive hydrogen absorption during service could degrade these mechanical properties to the extent that either the operational conditions or the life of the cladding would have to be curtailed.

The program is divided into two tasks: (1) corrosion and hydriding tests of Zircaloy under heat transfer conditions to determine the quantity of hydrogen absorbed during service, and (2) mechanical property tests of hydrided Zircaloy to determine the effect of hydrogen on cladding properties.

The loop was first operated during the second quarter of the program. The first specimen was removed from the loop May 3, 1967, after a 60-day exposure, and a second specimen was removed June 12, 1967, after slightly more than 32 days' exposure. At the end of this report period the loop contained four specimens that had been exposed for more than 100 days and one specimen that has been exposed for about 20 days. We expect to continue operation with pure water until early September. The second test series will begin operating with boric acid water late in September or early in October.

Gaseous hydriding and oxidation in air have been successful in preparing specimens to simulate fuel cladding after 1000 days service. The hydriding method, however, introduces about twice as much hydrogen in the open-ended tubular and flat strip specimens as in the sealed-

tube heat transfer specimens. The reason for this discrepancy in hydrogen absorption has not been established, but it could be due to a surface area relationship. The hydriding operation is being controlled to introduce the desired quantity of hydrogen in the heat transfer specimens.

The specimens removed after the 60-day exposure have been sectioned for hydrogen analyses. The analytical data indicate that the heat transfer and isothermal loop specimen absorbed considerably more hydrogen than did the autoclave specimens. Weight change data are not yet available, so that the ratio of the hydrogen absorbed by the Zircaloy to that released from the oxidation reaction has not been calculated. Some alteration in the order of specimen exposure was required because of delays in preparing specimens; however, the overall schedule remained firm throughout this report period.

All the mechanical property specimens were hydrided by the end of this report period. The tensile tests of cold-worked material have been completed, and all annealed specimens have been tested except those containing over 350 ppm hydrogen. The tensile data show that hydrogen, at concentrations up to 1000 ppm, has no significant effect on the tensile strength of cold-worked Zircaloy-4 tubing over a temperature range from 70 to 800 F. At temperatures below 250 F and hydrogen concentrations above 900 ppm, the tensile elongation of cold-worked tubing is significantly reduced. The elongation values were reduced from 16% for as-received material to less than 6% for material containing 1000 ppm hydrogen. At temperatures above 250 F, elongation is insensitive to hydrogen content, and below 250 F there is no appreciable effect for less than 300 ppm hydrogen on any of the mechanical properties measured.

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1. INTRODUCTION

The fuel cladding specified for current pressurized and subcritical-pressure boiling water reactors is usually one of the zirconium-tin alloys, Zircaloy-2 or Zircaloy-4. These alloys have the strength and corrosion resistance necessary to withstand the rigors of reactor service, and their nuclear properties are much more desirable than those of other available materials.

Since there is a continuing effort to increase the thermal performance of water reactors, the Zircaloy fuel cladding is required to withstand more severe operating conditions for longer periods of time. Intermittent relocation of fuel elements in the reactor core also adds to the severity of service conditions by introducing a potentially high-strain-fatigue factor over and above the cyclic thermal stresses of normal service. Although operating experience with Zircaloy has been excellent, some failures of experimental Zircaloy-clad fuel elements indicate that operational limitations should be defined more precisely before subjecting a Zircaloy-clad core to the demanding conditions of advanced reactor designs. The goal of this program is better definition of operational limitations for Zircaloy fuel cladding.

The program is divided into two experimental tasks:

1. Corrosion and hydriding tests of Zircaloy under heat transfer conditions.
2. Determination of the effects of hydrogen and hydride distribution on the mechanical properties of Zircaloy.

During the third quarter, the corrosion and hydriding loop continued operation. Two specimens were removed after exposures of about 30 and 60 days. At the end of the quarter all five test stations in the loop were occupied. The 60-day specimen was sectioned, and samples were analyzed for hydrogen content.

Specimens simulating fuel cladding after 1000 days of service are being prepared by a gas hydriding method followed by oxidation in air at 950 F. One such specimen was inserted in the loop on June 12, 1967, and it operated satisfactorily throughout the remainder of the report period.

The remaining tensile tests of cold-worked specimens were completed. Elevated temperature testing of annealed specimens will be completed during the fourth quarter.

The first test series in the loop should be completed during the fourth quarter, and the second series should be started early in the fifth quarter. All remaining tensile testing and most of the burst testing will be completed during the fourth quarter. Work on a topical report covering the mechanical properties portion of the overall corrosion and hydriding program will be initiated during the fourth quarter.

2. CORROSION AND HYDRIDING OF ZIRCALLOY UNDER HEAT TRANSFER CONDITIONS (D. W. Koch and M. M. Rubright)

2.1. Task Objective

This task will provide quantitative data on corrosion and hydriding rates for zirconium alloy fuel cladding under conditions simulating those expected in advanced water reactor service. Zircaloy-2 and Zircaloy-4 hydriding characteristics will be compared, and an abbreviated test in borated water will be performed to determine whether the chemical poison addition has any gross effects on corrosion and hydriding.

2.2. Task Description

Under heat transfer conditions prevalent in current and advanced water reactor designs, fuel cladding surface temperatures can exceed 650 F, and the temperature gradient through the cladding can be as high as 150 F; thus the inside surface temperature of the cladding can be in excess of 800 F. Corrosion of the cladding under a temperature gradient might proceed at a rate somewhat greater than that predicted by isothermal tests at the bulk water temperature. Similarly, absorption of some of the hydrogen released during the corrosion reaction should increase with the increased corrosion rate. However, hydrogen migrates down a temperature gradient in zirconium and concentrates in the lower-temperature region. In the case of fuel cladding operating under high heat flux conditions, the lower-temperature region is also the corroding surface where hydrogen is being introduced. This activity tends to promote hydrogen concentration at the outside surface of the cladding, and zirconium hydride will precipitate in this location when the solubility limit is reached. The high concentration of hydrogen at the surface of the cladding should inhibit further hydrogen absorption, and the precipitated zirconium hydride should have some effect on

the mechanical properties of the cladding. This program is designed to provide quantitative answers to these suppositions.

In a previous program at B&W, Zircaloy-4 specimens exposed in 630 F, 2200 psi water at a calculated surface heat flux of 450,000 Btu/hr-ft² (about 650 F surface temperature) corroded at a slightly higher rate than did the 630 F isothermal specimens exposed in the same corrosion loop.¹ Hydrogen absorption by the heat transfer specimens, however, was appreciably higher than that under isothermal conditions. This result led to speculation that (1) the hydrogen absorption rate of Zircaloy-4 under the local boiling test conditions is much greater than the rate under isothermal or non-boiling conditions, (2) the presence of a hydride layer or zone of high hydrogen concentration enhances rather than inhibits hydriding, or (3) there existed an extraneous source of hydrogen (such as a contaminant in the helium cover gas used inside the tubular specimens). The current program is expected to resolve the question.

In the test program covered by this task, Zircaloy-2 and Zircaloy-4 tubing will be exposed to 640 F water flowing past the specimen at approximately 13 feet per second. The specimens will be internally heated over 6 inches of their length with special single-ended electrical heaters producing a calculated surface heat flux on the specimen of 600,000 Btu/hr-ft². An intentional hot zone will be installed at the end of the heated length to inhibit hydrogen migration to the cooler, unheated portion of the specimen. Precautions will be taken to inhibit introduction of hydrogen to the specimens by means other than corrosion during the test.

Five test stations are situated in parallel legs of the test loop so that a test specimen can be isolated and removed from test without interfering with the other four specimens. Metallographic examination will be used to determine oxide film thicknesses and hydride distribution, and the hydrogen concentration of the specimens will be determined by vacuum-extraction. Samples for hydrogen analysis will be taken from various axial positions to evaluate axial migration of hydrogen and selective hydriding of the specimens. Radial hydrogen distribution will be determined by metallographic methods and by analyzing samples of machine chips removed from the surface of the specimen during successive turnings. Weight change and hydrogen absorption values will be compared

with similar data from previous experiments and with data from isothermal control specimens exposed in the loop and in a connected autoclave. The autoclave will be operated at a temperature close to that of the surface of the heat transfer specimens.

Specimens will be removed and examined at intervals during the 180-day exposure to permit evaluation of both corrosion and hydrogen absorption as a function of exposure time. To evaluate the insulating effect of a substantial corrosion film on corrosion and hydriding rates, several heat transfer and control specimens will be pre-corroded to simulate the end-of-life condition of fuel cladding after service in a pressurized water reactor. The corrodent conditions during this initial 180-day test series will be similar to those of an unpoisoned pressurized water reactor.

The second series will be tested for 30 days. Specimens from the 180-day test will be continued in the 30-day test, and some virgin specimens will be added. The corrodent conditions for the second series will simulate those of a pressurized water reactor with borated water. Evaluation of the specimens will be similar to that of the 180-day test, with emphasis on determining any gross effects on corrosion and hydriding owing to the altered corrodent conditions.

2.3. Work Accomplished During Report Period

2.3.1. Summary

The corrosion and hydriding loop has operated with heat transfer specimens in place since March 9, 1967, except for a nine-day scheduled outage for loop maintenance. During the quarter, two specimens were removed after exposures of 30 and 60 days. The 60-day specimen was sectioned, and samples were analyzed for hydrogen content.

A gas-hydriding and air-oxidation method was developed to prepare specimens simulating the fuel cladding condition after 1000 days of service in a pressurized water reactor. Approximately 250 ppm hydrogen is introduced into the specimen, and an oxide coating of about 250 mg/dm² is added in the separate operations. Some modification in the order of exposing specimens in the loop was necessary because of

delays encountered in preparing the specimens. However, the overall program schedule remained firm throughout the report period.

2.3.2. Loop Operation

One specimen was removed from the loop on May 3, 1967, after being exposed for 1450 hours (≈ 60 days). At the same time, corresponding isothermal specimens were removed from the autoclave connected to the loop. Operation of the loop was then suspended until May 9, 1967, to permit necessary maintenance and repair operations. A new Zircaloy-4 specimen was inserted into the loop and operation was resumed. On June 12, 1967, a second specimen was removed after 785 hours of exposure (≈ 30 days); this removal was made without interrupting the main loop operation. A new Zircaloy-4 specimen was inserted on the same day and exposure begun. As of July 1, 1967, the specimens in the loop had accumulated exposures ranging from 464 to 2598 hours. The associated isothermal specimens in the connected autoclave have accumulated from 441 to 2499 hours of exposure time.

The specimen removed on May 3, 1967, was photographed (Figure 2-1) and then sectioned for chemical and metallographic analysis. Table 2-1 lists the preliminary hydrogen values for the various segments of the specimens. These values, obtained with a Leco Analyzer, are averages for duplicate runs. Additional analytical determinations scheduled for this specimen involve the measurement of the hydrogen gradient across the tube wall and a separate check on the bulk hydrogen content utilizing a vacuum fusion approach. Such analyses have not yet been performed. No analytical work on the specimen removed on June 12, 1967, has been performed.

2.3.3. End-of-Life Specimen Preparation

The specimen inserted into the loop on June 12, 1967, had been treated so that the surface was covered with an oxide coating simulating the surface condition expected on Zircaloy-4 cladding after 1000 days of reactor operation. This type of surface, referred to as an end-of-life condition, incorporates a hydrogen content of about 250 ppm and an oxide thickness equivalent to about 250 mg-dm². Preliminary tests have shown that this surface is best formed by a two-step operation involving gaseous hydriding of the surface followed by oxidation.

No unusual problems have been encountered with this specimen, the first of its kind to be exposed in the loop. Table 2-2 lists the hydrogen content of the various components of the specimen.

2.3.4. Operating Schedule

As a result of operating considerations, certain changes in specimen numbers and the sequence of specimen exposures have been made to the schedule listed in the previous quarterly report. The amended schedule is shown in Figure 2-2. The specimen identification now used and the current status of specimens are listed in Table 2-3. There is no change in the overall exposure plan or in the operating schedule.

2.4. Planned Work for Next Report Period

Operation of the loop will continue. Specimen 4-4, together with its control specimens from the loop and autoclave, will be removed on July 12, 1967, after an exposure of 120 days. Specimen 4-16, the first end-of-life specimen, will then be removed after a 45-day exposure.

Flow in the loop is not expected to be curtailed during these specimen changes, and no loop outages are planned during the remainder of Test Phase I.

Table 2-1. Hydrogen Content of Zircaloy-4 Specimen
Zr-4-13, Prefilmed, 60 Day Exposure

Sample type	Original H ₂ content, ppm	Final H ₂ content, ppm
Heat transfer	16	55; 66
Unheated bulk water	16	33
Isothermal (flat)	19	41
Isothermal (flat)	19	41
Isothermal (tube)	19	42

Table 2-2. Hydrogen Content of End-of-Life Zircaloy Specimens Before Exposure
(Oxide Coating Thickness = 270 mg-dm²)

Specimen	Zircaloy type	Corresponding specimen type	Hydrogen content, ppm
C-16	Zircaloy-4	Heat transfer, sealed tube	254
H-16	Zircaloy-4	Isothermal, open tube	649
CC-8	Zircaloy-2	Heat transfer, sealed tube	254
HH-8	Zircaloy-2	Isothermal, open tube	683

Table 2-3. Specimens for Corrosion and Hydriding Tests

Specimen	Pretest condition	Exposure, days	Current status
Zircaloy-4 Series			
Zr-4-1	750 F prefilm	180 - test 1	Undergoing exposure
Zr-4-2	750 F prefilm	180 - test 1 30 - test 2	Undergoing exposure
Zr-4-4	750 F prefilm	120 - test 1	Exposure completed 7-12-67
Zr-4-13	750 F prefilm	60 - test 1	Exposure completed 5-3-67
Zr-4-5	750 F prefilm	30 - test 1	Exposure completed 6-12-67
Zr-4-16 ^(a)	End-of-life	45 - test 1	Undergoing exposure
Zr-4-17 ^(a)	End-of-life	30 - test 1	Not yet exposed
Zr-4-18 ^(a)	End-of-life	15 - test 1	Not yet exposed
Zr-4-19 ^(a)	End-of-life	30 - test 2	Not yet exposed
Zr-4-20 ^(a)	End-of-life	30 - test 2	Not yet exposed
Zircaloy-2 Series			
Zr-2-1	750 F prefilm	180 - test 1 30 - test 2	Undergoing exposure
Zr-2-8 ^(a)	End-of-life	45 - test 1	Not yet exposed
Zr-2-9 ^(a)	End-of-life	15 - test 1	Not yet exposed
Zr-2-10 ^(a)	End-of-life	30 - test 2	Not yet exposed

^(a) New specimen number designation.

Figure 2-1. Zircaloy-4 Specimen, Group Zr-4-13, After
60-Day Exposure

Figure 2-2. Heat Transfer Corrosion Test Schedule

3. EFFECT OF HYDROGEN AND HYDRIDE
DISTRIBUTION ON THE MECHANICAL
PROPERTIES OF ZIRCALOY
(A. L. Lowe, Jr.)

3.1. Task Objective

This task will determine the effect of hydrogen, in the concentration range from 150 to 1000 parts per million, on the tensile and burst properties of Zircaloy tubing. Tensile tests will be performed at temperatures ranging from room temperature to 800 F (427 C) on hydrided specimens of cold-worked and annealed Zircaloy-4. A similar series of specimens will undergo burst testing at room temperature.

3.2. Task Description

The Zircaloy tubing specified for fuel cladding service in pressurized and boiling water reactors is expected to exhibit and retain tensile properties above those of annealed material. During operation, however, the fuel cladding is subjected to the hardening effects of radiation, the annealing effects of prolonged exposure at elevated temperature, and the ill-defined effects of increased hydrogen content caused by absorption of part of the corrosion-released hydrogen. Operational and lifetime limitations have been placed on Zircaloy-clad elements because of the possible effects of hydrogen on the mechanical properties necessary to sustain cladding integrity. The limitations are artificial in the sense that they did not result from failure experience or from systematic experimental determinations of the effects of hydrogen on these properties.

During a previous experimental program at B&W it was shown that hydrogen, in the form of a hydride case up to about 0.001 inch thick or uniformly distributed at a concentration up to about 350 ppm, has no significant effect on the tensile or burst strength of 0.420-inch-diameter, 0.025-inch-wall, cold-worked Zircaloy-4 tubing.² Tensile

elongation values for the hydrided specimens were not significantly lower than the elongation values for unhydrided specimens. The innocuous effect of hydrogen on the strength and longitudinal ductility indicates that limitations on allowable hydrogen concentrations have little experimental or practical basis. However, during burst testing, the ductility was appreciably less for the uniformly hydrided specimens than for the unhydrided specimens. The appearance of the fractures in the burst specimens clearly showed the embrittling effect of precipitated hydride in Zircaloy. In the cases of the unhydrided specimens and the electrolytically hydrided specimens, where the hydrogen is concentrated in a layer or case at the outside surface, considerable deformation occurred before fracture. Very little deformation was evident in the specimens with uniformly distributed hydride, and the fracture propagated nearly the full length of the specimen. However, it is impractical to base operational limitations on the fact that transverse ductility at room temperature is appreciably decreased by precipitated zirconium hydride, because of the considerable successful operating experience with the cladding material which contains more than 100 ppm hydrogen.³

The goal of this series of tests is to provide more precise data on the effects of hydrogen on the tensile and burst properties of currently available Zircaloy-4 fuel cladding tubing. Specimens containing up to 1000 ppm hydrogen will be tested, and the resultant data will be evaluated with respect to hydride morphology and the original condition of the specimen. These tests will provide data for evaluating strength and ductility as functions of hydrogen content and temperature.

3.3. Work Accomplished During Report Period

3.3.1. Summary

All the specimens have been hydrided, and tensile testing is almost complete. All the cold-worked tensile specimens have been tested, and all the annealed specimens have been tested except the specimens containing more than 350 ppm hydrogen (to be tested at elevated temperature). At concentrations up to 1000 ppm, hydrogen does not significantly affect the tensile strength of cold-worked Zircaloy-4 over the temperature range from 70 to 800 F. At test temperatures above 250 F and at concentrations up to 1000 ppm, hydrogen has

little effect on tensile elongation. However, at temperatures below 250 F and at hydrogen concentrations above about 300 ppm, there is a significant loss of tensile elongation in cold-worked Zircaloy-4 tubing.

The burst-test apparatus was assembled during this quarter. Testing will begin next quarter and should be completed during the fifth quarter. The overall schedule for this task remains firm. A topical report covering the mechanical-property testing will be prepared after the burst testing is completed.

3.3.2. Tensile Test Results

Cold-worked specimens in the as-received condition and containing up to 1000 ppm hydrogen were tested at temperatures up to 800 F. The strength data from these tests are plotted in Figures 3-1 to 3-6. Data at test temperatures below 650 F and at hydrogen concentrations of 350 ppm and less were reported in the previous quarterly report. The extreme values for all the hydrogen concentrations are plotted as a function of test temperature in Figure 3-7. The extreme variations in the data as compared to the mean value for as-received cold-worked specimens are within approximately 10% at any test temperature. For all practical purposes, hydrogen contents up to 1000 ppm have no effect on the strength properties of cold-worked Zircaloy-4.

Ductility data, as measured by the percentage of elongation in a prescribed gage length, are plotted in Figures 3-8 to 3-13. These data, representing an expansion of the data reported in the last quarterly report, include data obtained from tests carried out during this quarter.

The maximum variations in the total elongation data as measured over a 2-inch gage length are plotted in Figure 3-14. The curves for the mean values of the as-received material and the material containing 1000 ppm hydrogen are shown for reference. At temperatures above about 300 F the increase in hydrogen content causes a reduction in the total elongation to approximately 60% of the value for as-received material. Below 300 F the elongation decreases with decreasing temperature, and at room temperature the elongation of the hydrided material with 1000 ppm hydrogen is approximately one-

third of the value for as-received material. The observed effect of the hydrogen content on the room-temperature ductility is similar to that reported by other investigators.^{4,5}

The interrelationship of the effect of the hydrogen content and the temperature on the ductility of cold-worked Zircaloy-4 is plotted in Figure 3-15. Temperature appears to compensate for the effect of increased hydrogen content in the temperature range above 200 F. To maintain the material in a condition in which the ductility is always greater than 10%, the temperature must be kept above 250 F, or the hydrogen concentration must be less than 350 ppm.

3.3.3. Planned Work for Next Report Period

The experimental work should be completed during the next quarter with the possible exception of some burst tests. Metallographic evaluations of the hydrogen distribution will be initiated, and a topical report draft will be prepared.

Figure 3-1. Tensile Strength of Cold-Worked Zircaloy-4 as a Function of Temperature (20 ppm Hydrogen — As-Received)

Figure 3-2. Tensile Strength of Cold-Worked Zircaloy-4 as a Function of Temperature (150 ppm Hydrogen)

Figure 3-3. Tensile Strength of Cold-Worked Zircaloy-4 as a Function of Temperature (350 ppm Hydrogen)

Figure 3-4. Tensile Strength of Cold-Worked Zircaloy-4 as a Function of Temperature (500 ppm Hydrogen)

Figure 3-5. Tensile Strength of Cold-Worked Zircaloy-4 as a Function of Temperature (750 ppm Hydrogen)

Figure 3-6. Tensile Strength of Cold-Worked Zircaloy-4 as a Function of Temperature (1000 ppm Hydrogen)

Figure 3-7. Total Variation in Ultimate and Yield Strengths of Cold-Worked Zircaloy-4 as a Function of Temperature for all Hydrogen Contents Tested

Figure 3-8. Tensile Elongation of Cold-Worked Zircaloy-4 as a Function of Temperature (20 ppm Hydrogen — As-Received)

Figure 3-9. Tensile Elongation of Cold-Worked Zircaloy-4 as a Function of Temperature (150 ppm Hydrogen)

Figure 3-10. Tensile Elongation of Cold-Worked Zircaloy-4 as a Function of Temperature (350 ppm Hydrogen)

Figure 3-11. Tensile Elongation of Cold-Worked Zircaloy-4 as a Function of Temperature (500 ppm Hydrogen)

Figure 3-12. Tensile Elongation of Cold-Worked Zircaloy-4 as a Function of Temperature (750 ppm Hydrogen)

Figure 3-13. Tensile Elongation of Cold-Worked Zircaloy-4 as a Function of Temperature (1000 ppm Hydrogen)

Figure 3-14. Total Variation in Total Elongation in 2-Inch Gage Length of Cold-Worked Zircaloy-4 as a Function of Temperature for all Hydrogen Contents Listed

Figure 3-15. Iso-Total Elongation Curves for Cold-Worked Zircaloy-4 as a Function of Temperature and Hydrogen Concentration

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9. U. S. Atomic Energy Commission
Oak Ridge Operations Office
P. O. Box E
Oak Ridge, Tennessee 37830
Attn: C. Matthews
10. U. S. Atomic Energy Commission (3)
Savannah River Laboratory
P. O. Box A
Aiken, South Carolina 29802
Attn: R. P. Marshall (2)
S. Rideout
11. U. S. Atomic Energy Commission
c/o Atomic Energy of Canada, Limited
Chalk River, Ontario, Canada
Attn: G. W. Perry
12. U. S. Atomic Energy Commission
c/o Whiteshell Nuclear Research
Establishment AECL
Pinawa, Manitoba, Canada
Attn: A. Sawatzky
13. Oak Ridge National Laboratory (4)
Union Carbide Corporation
AEC Operations
P. O. Box X
Oak Ridge, Tennessee 37830
Attn: P. Patriarca
M. L. Picklesimer
J. E. Cunningham
P. L. Rittenhouse
14. Battelle Memorial Institute (5)
Pacific Northwest Laboratory
P. O. Box 999
Richland, Washington 99352
Attn: J. J. Cadwell
R. Dillion
A. Bement
R. G. Wheeler
D. de Halas

15. Argonne National Laboratory (2)
9700 South Cass Avenue
Argonne, Illinois 60439
Attn: S. Greenberg
J. E. Draley
16. Atomics International
P.O. Box 309
Canoga Park, California 91304
Attn: C. E. Weber
17. Battelle Memorial Institute (2)
Columbus Laboratories
505 King Avenue
Columbus, Ohio 43201
Attn: S. Paprocki
W. Berry
18. Westinghouse Electric Corporation (4)
Bettis Atomic Power Laboratory
P.O. Box 79
West Mifflin, Pennsylvania 15122
Attn: B. Lustman (2)
W. J. Babyak (2)
19. Combustion Engineering, Inc. (3)
Nuclear Division
Prospect Hill Road
Windsor, Connecticut 06095
Attn: S. S. Christopher
W. Chernock
J. Tobin
20. General Electric Company (4)
Vallecitos Atomic Laboratory
P.O. Box 846
Pleasanton, California 94566
Attn: H. Klepfer (2)
K. Cohen (2)
21. Knolls Atomic Power Laboratory
P.O. Box 1072
Schenectady, New York 12301
Attn: A. E. Bibb
22. General Electric Company (2)
Nuclear Materials & Propulsion Operation
P.O. Box 15132
Cincinnati, Ohio 45215
Attn: F. White
S. Naymark

23. General Electric Company
Atomic Power Equipment Department
P.O. Box 254
San Jose, California 95125

Attn: D. H. Imhoff

24. Superior Tube Company
Morristown, Pennsylvania

Attn: C. Matthews

25. United Nuclear Corporation
5 New Street
White Plains, New York 10601

Attn: W. S. Mott

26. Westinghouse Electric Corporation (3)
Atomic Power Division
P.O. Box 355
Pittsburgh, Pennsylvania 15230

Attn: R. Allio (2)
P. G. DeHuff

27. Wah Chang Corporation
P.O. Box 460
Albany, Oregon 97321

Attn: S. Worchester

28. The Babcock & Wilcox Company (36)
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