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**ZIRCALOY-CLAD UO_2 FUEL ROD
EVALUATION PROGRAM
QUARTERLY PROGRESS REPORT No. 2
FEBRUARY 1968 - APRIL 1968**

COMPILED BY
F.H. MEGERTH

U.S. ATOMIC ENERGY COMMISSION
CONTRACT AT(04-3)-189
PROJECT AGREEMENT 41

NUCLEAR FUELS DEPARTMENT
GENERAL ELECTRIC
SAN JOSE, CALIFORNIA

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ZIRCALOY-CLAD UO_2 FUEL ROD
EVALUATION PROGRAM

MASTER

Quarterly Progress Report No. 2
February 1968 - April 1968

Compiled by
F. H. Megerth

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SECTION I

INTRODUCTION

At the time of the final shutdown of the Vallecitos Boiling Water Reactor (VBWR) and the termination of the AEC Fuel Cycle Program [Contract AT(04-3)-189, Project Agreement 11] the leading Zircaloy-2-clad, UO_2 fuel rods of the program's Type J basic assemblies had attained rod-average exposures as high as 3×10^{20} fissions/cc (10,000 MWd/STU). To demonstrate at the earliest possible time the capability of Zircaloy-clad fuel to achieve average burn-ups of at least 6×10^{20} fissions/cc (20,000 MWd/STU) and to take advantage of the available lead time in achieving these burn-up targets, 98 of the higher exposure Fuel Cycle Program rods were incorporated in a special assembly, designated SA-1, and inserted in the Dresden reactor for additional irradiation. The assembly consists

of two 7-by-7 subassemblies stacked one on top of the other in a channel of standard dimensions for the Dresden reactor.

Assembly SA-1 has been operated continuously from June 1964 to the present time with the exception of reactor shutdowns for refueling and maintenance.

To investigate in detail the performance of the SA-1 fuel rods and to determine the isotopic composition of the highly exposed fuel, a two-phase program of destructive examination and analysis was drawn up. For the initial phase of the program, 10 rods were removed from the assembly in January 1967 and were shipped to the Vallecitos Nuclear Center. The examination of these 10 rods is now in progress.

SECTION II

SUMMARY

2.1 ASSEMBLY OPERATION

The operation of Assembly SA-1 during this period was limited to the first three days of February. On February 3 the Dresden reactor was shut down for refueling and general maintenance. This outage is scheduled to continue until about June 1, 1968.

Assembly SA-1 was leak tested by the sipping technique in March; no indication of fuel rod failure was detected. Hence, it is planned the assembly will be in operation during the next reactor cycle (Cycle 6).

The estimated average and peak exposure values for SA-1 remain at 7.5×10^{20} fissions/cc (27,200 MWd/STU) and 12.2×10^{20} fissions/cc (44,500 MWd/STU), respectively.

2.2 EXAMINATIONS AND ANALYSES
OF FUEL RODS

The following examinations and analyses were conducted on the 10 fuel rods which were permanently removed from Assembly SA-1 in January 1967:

Relative burnups and power on both a rod-to-rod basis and axial location on a given rod

basis were determined from gamma scans on all rods. Data obtained with a germanium (lithium) detector system indicated that the rod-average burnups of the highest exposure rods in the group (A13 and A41) were about 20 percent greater than those of the lowest exposure rods (A4 and B70). The axial gamma profiles of the 10 rods were of similar shape. The ratios of axial peak burnup to average burnup for the different rods ranged from 1.15 to 1.30.

Neutron radiographs taken of six rods revealed a few 1 to 2 mm (0.04 to 0.08 in.) gaps in the fuel columns at pellet interfaces. Fracturing or marked cracking was detected in about 5 percent of the pellets and occurred to varying degrees in all rods at random locations. The neutrographs did not reveal

any anomalies in either the end plugs or the heat transfer portion of the tubing; however, uninterpretable dark spots were noted within the plenums of three rods.

Analyses of the chemically removed crud solutions from the six rods indicated that nickel, iron, copper, and chromium were the principal metallic elements present. The average deposition of these four elements combined was approximately 275 mg/dm^2 of fuel rod surface. Nickel constituted approximately 65 to 75 percent of this combined weight.

Five of the fuel rods were punctured and the contained free gases were collected. The gas contents ranged from 4.7 to 9.5 cc (0.28 to 0.58 in.³). The internal void volumes of these rods ranged from 2.6 to 3.8 cc (0.16 to 0.23 in.³).

SECTION III

ASSEMBLY OPERATION

The operation of Assembly SA-1 during the report period (February 1, 1968 through April 30, 1968) was limited to the first three days of February. The Dresden reactor was shut down February 3 for refueling and general maintenance. This outage is scheduled to continue until about June 1, 1968.

Assembly SA-1 was leak tested by the sipping technique in March; no indication of fuel rod failure was detected. Hence, it is planned to operate the assembly in the next reactor cycle (Cycle 6).

The estimated exposure values for SA-1 at the end of Cycle 5 are presented in Table 1. These values include both the VBWR and Dresden operations.

TABLE 1
SA-1 EXPOSURES AT END OF
DRESDEN REACTOR CYCLE 5
FEBRUARY 3, 1968

SA-1 Exposures	Fissions/cc $\times 10^{-20}$ (MWd/STU)
Assembly Average	7.5 (27, 200)
Lead Rod Average	9.3 (34, 100)
Lead Rod Peak	12.2 (44, 500)

Reactor water chemistry data for December 1967 and January 1968 are given in Table 2. These data, extracted from monthly reports of the Commonwealth Edison Company on Dresden operation, were not available at the time of issue of the preceding progress report.

TABLE 2
DRESDEN REACTOR WATER CHEMISTRY

	December 1967			January 1968		
	Minimum	Average	Maximum	Minimum	Average	Maximum
pH	5.8	7.0	8.9	5.0	7.4	8.7
Conductivity (μ mhos)	0.32	0.46	0.68	0.32	0.45	0.64
Turbidity (APHA)	3.0	5.3	13.0	1.8	6.9	12.0
Chloride (ppm)	0.020	0.038	0.160	0.001	0.029	0.068

SECTION IV

EXAMINATIONS AND ANALYSES OF FUEL RODS

During this period the following work was carried out on the 10 fuel rods that were permanently removed from Assembly SA-1 in January 1967.

4.1 GAMMA SCANNING

4.1.1 Procedure

The 10 rods were gamma scanned to provide data on relative burnup and power both on a rod-to-rod basis and with respect to axial location on a given rod. Both germanium (lithium) and sodium iodide (thallium) detectors were employed.

The 4-cc Ge(Li) detector was located outside the hot cell, directly in front of a 0.051-cm (0.020-in.) slit in the 91-cm (36-in.) cell wall. Thus, as a fuel rod was drawn past the opposite end of the slit, the detector received a collimated beam of gamma rays. The pulses from the detector were passed through a solid state preamplifier and the pulses shaped and amplified further in a Tennelec amplifier (Figure 1). The amplifier output pulses were routed to a 256-channel pulse height analyzer and to two single-channel analyzers. One of the single-channel analyzers was set to accept pulses at

the energy corresponding to that of the cesium-137 line, and the other was set at slightly higher energies, i. e., approximately 710 keV, for a "background" signal. The output pulses from each of these analyzers were fed to a scaler and also to a linear count rate meter, from which the signals were fed to a chart recorder in such a way that the background signal was subtracted from the total count rate at the Cs-137 position, to yield a net Cs-137 count rate. The multi-channel analyzer had to be operated with the discriminator set at approximately 480 keV to minimize dead time losses. The system was balanced by adjusting the energy and window width of the single-channel analyzer so that the counts accumulated per unit time on the respective scaler agreed with those accumulated by the multi-channel analyzer (correcting for the 10⁶ percent deadtime).

The 4.5-cm $\times$ 5.1-cm (1-3/4-in. $\times$ 2 in.) NaI(Tl) crystal was located directly behind the Ge(Li) detector and was connected through conventional electronics to a linear count rate meter and chart recorder (Figure 1). This system was operated in the normal way, i. e., only those gamma rays with energies in excess of 0.60 MeV were accepted by a single-channel analyzer operated in the

FIGURE 1. SCHEMATIC DRAWING OF Ge(Li) AND NaI(Tl) GAMMA SCANNING SYSTEMS

integral mode, the output of which was fed to the count rate meter. Thus, the scan data could be accumulated simultaneously by both systems.

Each rod was scanned by drawing it vertically past the slit at a rate of 2.5 cm (1 in.) per minute. The axial count rate profile was thus recorded by each system on their respective chart recorders. In addition, for the Ge(Li) detector, the spectrum was accumulated and the counts stored by each of the scalers and by the multi-channel analyzer for the complete traverse of the fuel rod to provide integral data.

All rods were scanned in duplicate. It was found after initially scanning some of the fuel that appreciable day-to-day variations occurred. Thus, one rod, B76, was selected as a standard which was scanned at the start and end of each daily scanning period, and was used to adjust the data for any variation during the 5-day scanning period.

4.1.2 Results

A typical gamma ray spectrum obtained on the multi-channel analyzer is shown in Figure 2. The regions selected for the single-channel analyzers are depicted; the channel width was about 15 keV. In practice the background region was actually between the Pr-144 and Zr-95 lines. A profile scan of rod A13 obtained with the Ge(Li) detector system is shown in Figure 3.

The integral data on the relative burnups of the different fuel rods obtained by the use of the Ge(Li) detector system is shown in Table 3. These values imply that the range of burnups among the 10 rods is smaller than the 5.8×10^{20} f/cc (21,000 MWd/STU) to 8.5×10^{20} f/cc (31,000 MWd/STU) estimated by purely computational methods.⁽¹⁾ Based on previous experience, the values obtained with the Ge(Li) system are estimated to be

correct within ± 3 percent (1 σ). Specimens sectioned from four of the rods will be analyzed for burnup by the neodymium mass spectrometric method⁽²⁾ and by the normal radiochemical determination of Cs-137. These analyses will provide a means of verifying the nondestructively determined relative burnup values and will furnish absolute values of exposure.

TABLE 3
RELATIVE BURNUP DATA BASED ON
THE Ge(Li) DETECTOR SYSTEM

Rod	Rod - Average Burnup (Rod A4 = 1.00)
A4	1.00
A13	1.22
A28	1.04
A41	1.20
A45	1.07
A46	1.10
B70	1.00
B76	1.10
B88	1.10
B97	1.09

Figures 4 and 5 show the axial gamma profiles of rods A13 and B97 obtained with the NaI(Tl) detector system. Gross gamma scans of the 10 rods are given in Appendix A. All scans were of a generally similar shape and typically showed two peaks of approximately the same relative height. One peak was adjacent to the short end plug of each rod, which, in turn, was located at the middle of the full fuel assembly during operation; the second peak was located about one-quarter rod length away from the first.

The ratios of peak burnup to average burnup along the lengths of the rods were determined from the above scans and are listed in Table 4.

FIGURE 2. GAMMA RAY SPECTRUM OF ROD A13 TAKEN WITH 4cc Ge (Li) DETECTOR. ACCUMULATED FOR THE COMPLETE FUEL ROD TRAVERSE

FIGURE 3. AXIAL VARIATION OF THE COUNT RATE OF THE Cs-137 LINE TAKEN WITH THE Ge (Li) DETECTOR SYSTEM

FIGURE 4. AXIAL GAMMA PROFILE OF ROD A13

FIGURE 5. AXIAL GAMMA PROFILE OF ROD B97

TABLE 4
AXIAL PEAK-TO-AVERAGE BURNUP
RATIOS FOR SA-1 FUEL RODS

Rod	<u>Peak Burnup</u> Average Burnup
A4	1.18
A13	1.16
A28	1.13
A41	1.19
A45	1.16
A46	1.15
B70	1.25
B76	1.30
B88	1.25
B97	1.20

4.2 NEUTRON RADIOGRAPHY

4.2.1 Procedure

Fuel rods A13, A28, A41, B70, B76, and B97 were mounted vertically in an aluminum holder and transferred by cask from the hot cells to the neutron radiographic facility at the Nuclear Test Reactor (NTR).

The NTR facility utilizes horizontal neutron beams obtained from the physical center of the NTR's annular core. One beam is allowed to leave the reactor building via a shuttered, conical hole in the 1-1/2-meter (5-foot) thick north wall. On the outside of this wall a large stationary concrete receiving cask has been constructed. A hole in one side is in alignment with the beam hole in the reactor wall and allows the neutron beam to enter a central cavity in the cask. The central cavity is open at the top of the cask where a lead and steel transfer cask can be positioned to discharge radioactive objects down into the neutron beam. A stepped plug, mounted on wheels, penetrates the receiving cask on the opposite side of the cask from the beam entrance. Neutron radiographic transfer plates up to 13 x 38 cm (5 x 15 in.)

are positioned on the inside face of the plug and are changed only when the beam shutter is closed and the radioactive object is withdrawn into the transfer cask.⁽³⁾ Dysprosium transfer plates were used to produce neutrographs of the above six rods. The neutron exposure time was 60 minutes. Three separate exposures were required to cover the full length of the rods. After irradiation the transfer plates were placed in contact with x-ray film for approximately 16 hours. Positive prints were then made from the exposed film. A variation in contrast was obtained by using two different developing times in the printing process.

4.2.2 Results

Portions of the positive prints from the neutrographs are shown in Figures 6 and 7. Gaps in the fuel columns at pellet interfaces as large as 1 to 2 mm (0.04 to 0.08 in.) were noted in approximately five instances (Figures 6B and 7C). Fracturing or marked cracking of pellets, as shown in Figures 6A and 6B, was detected in about 5 percent of the pellets and occurred to varying degrees in all rods in random locations.

One of the principal values of neutron radiography is the ability to detect regions of high hydride concentration in the cladding, which may be indicative of a defective fuel rod. These regions show up on the positive prints as dark areas. In the case of the SA-1 rods, no such areas were observed in the end plugs or heat transfer portion of the tubing. However, the following phenomena were noted:

- Rod A41 - a dark spot about 1 mm (0.04 in.) in diameter in the plenum next to the end plug (Figure 7B).
- Rod B70 - a dark spot about 0.5 mm (0.02 in.) in diameter in the plenum next to the wafer (Figure 6C).

A) RODS B97, B76, AND B70 - PLENUM ENDS. } PELLET CRACKING AND INTERFACIAL GAPS
 B) RODS B97, B76, AND B70 - MID-LENGTHS. } ARE INDICATED. (SHORT DEVELOPMENT TIME)
 C) RODS B97, B76, AND B70 - PLENUM ENDS. DARK AREAS AND LIGHT LINE ARE INDICATED. (LONG DEVELOPMENT TIME)

**FIGURE 6. NEUTRON RADIOGRAPHS OF
SA-1 FUEL RODS**

B97

B76 (A)

B70

A41

A28 (B)

A13

A41

A28 (C)

A13

- A) RODS B97, B76, AND B70 - SHORT END PLUG ENDS.
- B) RODS A41, A28, AND A13 - PLENUM ENDS. DARK AREA IS INDICATED.
- C) RODS A41, A28, AND A13 - SHORT END PLUG ENDS. INTERFACIAL GAP IS INDICATED.
(LONG DEVELOPMENT TIME, ALL CASES)

FIGURE 7. NEUTRON RADIOGRAPHS OF SA-1 FUEL RODS

- c. Rod B97 - a dark line about 3 mm (0.12 in.) long and parallel to the rod axis and a light line about 4 mm (0.16 in.) approximately perpendicular to the first (Figure 6C). The light line may indicate the presence of a crack.

Whether the above spots were caused by anomalies in the Zircaloy tubing or in the stainless steel plenum support tubes cannot be resolved at this time. Visual examination of the external cladding surfaces in these areas did not reveal any abnormalities.

4.3 CRUD ANALYSIS

4.3.1 Procedure

The general method employed to remove the crud from the fuel rods by dissolving it with hydrochloric acid was described in the preceding quarterly progress report.⁽¹⁾ The crud was removed from each of the rods in two steps, i.e., from approximately one-half of the length each time. Fresh acid was used for each of the steps. Bottom subassembly rods A4 and A13, also A28 and A41, were combined into pairs for crud removal. Top subassembly rods B70 and B76 were de-crudded separately. The resulting eight solutions were analyzed for metallic elements by wet chemical techniques. Portions of two of these solutions were evaporated to solid residues which were then analyzed by x-ray fluorescence.

4.3.2 Results

Analyses of the solutions, given in Tables 5 and 6, indicated that nickel, iron, copper, and chromium were the principal metallic elements present in the crud. Nickel constituted approximately 65 to 75 percent of the combined weight of these four elements, which is expressed in terms of milligrams deposited per square decimeter of fuel rod surface. The data in Table 5 are presented in the form of a top-to-bottom full-assembly

profile. These profiles indicate that in the upper three-fourths of the assembly the deposition of Ni, Fe, Cu, and Cr was, on the average, somewhat greater than 300 mg/dm², while in the lower one-fourth the deposition of these elements was approximately 100 mg/dm². The relative concentrations of the elements did not vary widely with elevation, although it appears that the relative concentration of copper increased somewhat at mid-assembly, while the relative concentration of iron increased slightly at the top and bottom of the assembly.

The wet chemical and x-ray fluorescent analyses on the two solutions from the A4 + A13 rod pair are not in complete agreement, particularly with respect to the relative amounts of iron and copper. The reason for this is unknown.

4.4 GAS COLLECTION AND VOID VOLUME DETERMINATION

4.4.1 Procedure

The gas sampling system consists of an in-cell puncture mechanism connected to an out-of-cell vacuum collection system. The top end of the rod was sealed in a set of aluminum puncture blocks lined with neoprene gaskets. The system was then evacuated to a maximum leak rate of 1 μ /min. The fuel rod was mechanically punctured and the released gas was allowed to diffuse into the evacuated system which had a known volume. When the system achieved equilibrium, the gas pressure was measured and recorded. The gas was then expanded into a pressure bomb for sampling.

After collecting the gas sample, the internal void volume was measured. The punctured fuel rod was back-purged with argon to a positive pressure in the rod. The rod was then removed from the puncture block and allowed to achieve equilibrium at

TABLE 5

CHEMICAL ANALYSIS OF CRUD SOLUTIONS

Rod	Portion of Assembly Length Measured from Top [cm (in.)]	Fe		Cu		Ni		Cr		Fe, Cu, Ni, Cr Total Deposition (mg/dm ²)
		Deposition (mg/dm ²)	Relative Concentration (%)	Deposition (mg/dm ²)	Relative Concentration (%)	Deposition (mg/dm ²)	Relative Concentration (%)	Deposition (mg/dm ²)	Relative Concentration (%)	
B70	0 to 46	69	18.8	18	4.9	274	74.7	6	1.6	367
B76	(0 to 18)	72	23.1	14	4.5	226	72.4	<1	-	312
B70	46 to 104	34	13.9	14	5.7	187	76.3	10	4.1	245
B76	(18 to 41)	61	14.8	50	12.2	295	71.8	5	1.2	411
A4 + A13	107 to 152	44	17.1	34	13.2	176	68.5	3	1.2	257
A28 + A41	(42 to 60)	71	17.3	69	17.2	255	63.8	5	1.2	400
A4 + A13	152 to 211	24	18.9	6	4.7	94	74.0	3	2.4	127
A28 + A41	(60 to 83)	22	22.0	8	8.0	69	69.0	1	1.0	100

TABLE 6
X-RAY FLUORESCENT ANALYSIS OF
CRUD SOLUTION RESIDUES

Rod Pair	Portion of Assembly Length, Measured from Top [cm(inches)]	Element Detected	Relative Amount
A4 + A13	107 to 152 (42 to 60)	Ni Fe Cu Cr	Strong Major Minor Weak Minor Weak Trace
A4 + A13	152 to 211 (60 to 83)	Ni Cu Fe	Major Minor Weak Minor

atmospheric pressure. The rod was rotated 180 degrees from the original position and the hole was sealed with the neoprene on the back-up plate in the puncture block. The system was evacuated and the rod was repunctured. The system pressure was measured and recorded after it had reached equilibrium. From the pressure in the rod and pressure and volume of the evacuation system, the void volume of the rod was then calculated.

Five fuel rods were selected for these operations. The rods differed from each other with respect to total burnup, heat flux, as-fabricated pellet density, and UO_2 powder lot. The gas samples collected will be analyzed by gamma counting, chromatography, and mass spectroscopy.

4.4.2 Results

The total free gas contents and internal void volumes of the rods are given in Table 7.

The gas contents ranged from 4.7 to 9.5 cc (0.28 to 0.58 in.³). The relatively greater amounts of gas came from rods with relatively higher burnups and heat fluxes as well as medium to low pellet densities. The pellets in these rods were fabricated from either UO_2 powder lot JO1DD alone or a combination of lots JO1DD and JO2DD. At this time, there is no obvious reason for the comparatively low void volume measurement on rod B76; however, additional measurements are being taken.

4.5 FUEL ROD SECTIONING

Specimens for isotopic analysis, cladding hydrogen analysis, and metallographic examination were sectioned from the rods. Preparation of mechanical properties test specimens was started.

TABLE 7
GAS COLLECTION DATA

Rod	Relative Average Burnup*	Estimated Peak Heat Flux [watts/cm ² (Btu/h-ft ²)]	As-Fabricated Pellet Density (Average Percent of Theoretical)	UO ₂ Powder Lot	Volume of Gas Collected (STP) [cc (in. ³)]	Rod Void Volume [cc(in. ³)]
A13	1.22	132(418,000)	94.4	JO1DD/ JO2DD	7.68(0.469)	3.79(0.231)
A41	1.20	132(418,000)	94.8	JO1DD	9.48(0.578)	3.69(0.225)
A46	1.10	105(333,000)	96.0	JO5HB	4.67(0.285)	3.71(0.226)
B70	1.00	111(351,000)	95.1	JO3HB	4.66(0.284)	3.68(0.225)
B76	1.10	137(435,000)	94.4	JO1DD/ JO2DD	7.09(0.433)	2.57(0.157)

*From Table 3

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APPENDIX A

GAMMA SCAN TRACES
NaI(Tl) DETECTOR SYSTEM

FIGURE A-1. GAMMA SCAN OF SA-1 ROD A4

FIGURE A-2. GAMMA SCAN OF SA-1 ROD A13

FIGURE A-3. GAMMA SCAN OF SA-1 ROD A28

FIGURE A-4. GAMMA SCAN OF SA-1 ROD A41

FIGURE A-5. GAMMA SCAN OF SA-1 ROD A45

FIGURE A-6. GAMMA SCAN OF SA-1 ROD A46

FIGURE A-7. GAMMA SCAN OF SA-1 ROD B70

FIGURE A-8. GAMMA SCAN OF SA-1 ROD B76

FIGURE A-9. GAMMA SCAN OF SA-1 ROD B88

FIGURE A-10. GAMMA SCAN OF SA-1 ROD B97

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The application of the Ge(Li) detector to nondestructively determine relative fuel rod burnup, as described in Section 4.1, is a technique developed under a G. E. sponsored program by C. P. Ruiz.

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