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WANL-TME-1878
November 1968

Westinghouse Astronuclear Laboratory

WESTINGHOUSE ASTRONUCLEAR LABORATORY
CONTRIBUTION TO THE
TWENTY-SEVENTH HIGH TEMPERATURE FUELS
COMMITTEE MEETING
10,11,12 DECEMBER 1968
(TITLE UNCLASSIFIED)

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WANL-TMF 1878
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SECTION 1

(CRD) RESULTS OF THE NUCLEAR ROCKET REACTOR TEST NRX-A6 (U)

1.1 (CRD) REACTOR OPERATING CONDITIONS (U)

(CRD) The reactor was brought quickly to full power from a brief hold for a systems checkout at an exit gas temperature of about 2000°R (850°C). The power ramp was controlled by feedback from the thermocouple measured gas temperature in the nozzle chamber. Other instrumentation indicated that the controlling temperature measurement was erroneous and the power was trimmed accordingly. However, a short duration power "spike" occurred, taking the reactor to a higher than planned power level. The power level was then held at 1150 to 1160 megawatts for the duration of the 60-minute test.

(CRD) Figure 1 shows the reactor power as a function of time. Also plotted is the control drum position, indicative of reactivity change. The change in reactivity arose from four principal sources: loss of graphite moderator by corrosion; loss of fissile material; fission product xenon buildup; and loss of molybdenum protective coating. The first three decreased the reactivity, the Mo loss increased it giving a net decrease. The Mo coating was deposited over the NbC primary protective coating. Some of the Mo penetrated the cracks in the NbC filling in between the NbC and the fuel substrate and filling the cracks. An appreciable fraction of the Mo deposited over the surface of the NbC. This distribution is illustrated by the fuel section in Figure 2 in the post test condition. From the Mo coating remaining on the surface it can be seen that Mo loss was gradual, occurring throughout the course of the test.

(U) The temperatures in the reactor were determined in several ways. Thermocouples measured the temperatures of instrumented but non-fueled fuel element shapes. The data from these were used to calculate the temperatures of adjacent fuel elements. This type of information is used in Figures 4 to 9; the data points indicate measured temperatures, the lower curves are the calculated fuel temperatures based on the measurements. Additional thermocouples measured the exit gas temperature and the temperature of the mixed gas (cooler gas came from structural component cooling streams) in the nozzle chamber.

(CRD) Additional checks on temperature limits were made by studies of fuel particle migration, thermal capsule melting*, and fission product distribution. Fuel particle migration, as shown in Figure 3, indicated a maximum fuel temperature of about 4800°R (2390°C). This agrees well with the data taken during the power spike. Figure 4 gives the axial temperature distribution for an element near the center of the core at the time of the spike. Figure 5 is a similar temperature distribution for a peripheral element at the same time.

(U) After the power was leveled, the maximum fuel temperatures and exit gas temperatures were lower than those during the spike as shown by Figures 6, 7, and 8 for elements in the central, mid-radial, and peripheral regions, respectively. These conditions were held for the remainder of the test except for an increase in peripheral temperatures as shown in Figure 9 representing conditions just before shutdown. The overall thermal history is presented in Figure 10 in terms of nozzle chamber temperature and flow rate. These data can be converted to fuel temperatures at various radial and axial positions by reference to the previous plots.

1.2 (CRD) TEST RESULTS (U)

1.2.1 (CRD) Summary (U)

(CRD) The NRX-A6 test was extremely successful and exceeded its primary objective which was to operate at rated conditions for 60 minutes or to a predetermined loss of reactivity. The total run time of 62 minutes above a nozzle chamber temperature of 4100°R more than doubled the full power and temperature endurance of previous reactors. The fuel element time rate of corrosion (gross weight loss) was 20 to 25 percent of that observed in the NRX/EST and NRX-A5 reactors.

(CRD) Total reactor reactivity loss due to carbon, uranium, and molybdenum was 0.70\$ which was significantly lower than previous reactors, i. e., 3.2\$ for NRX/EST and 2.1\$ for NRX-A5. Change in control drum cold critical angle was from 97.0 degrees at

*(U) Thermal capsules are refractory metal alloys encapsulated in graphite. The metal-metal-carbon eutectic temperature for each composition used has been determined. Thus, the melting or lack of melting of specific capsule compositions establishes both upper and lower limits to the maximum temperature to which they were exposed.

initial criticality to 100.5 degrees following the power run as compared with 99.1 degrees to 147.6 degrees for NRX/EST and 99.3 degrees to 137.3 degrees for NRX-A5. A comparison of the variations in reactivity in these three reactors is given in Figure 11.

(U) The results of the disassembly and post-operational examinations confirmed that the NRX-A6 reactor can operate at or above rated conditions for 60 minutes without deleterious effects on reactor system components. The performance of the core was the most optimistic result obtained from the NRX-A6 test. The interior fuel element and the central elements survived the full hour run with no structural damage and a smaller than expected corrosion weight loss.

(CRD) Performance of the NRX-A6 fuel elements typically was characterized by inter-element bonding, mild surface corrosion, low pinhole densities, low mid-band weight losses, relatively higher hot end weight losses and excellent corrosion resistance and integrity of the aft unfueled tip. The inter-element bonding was caused by pyrolytic graphite deposition at the core mid-plane. Except for creating disassembly problems, this effect is not harmful.

(CRD) Mid-band weight losses of typical NRX-A6 fuel elements appeared to be approximately one-tenth that of the NRX-A5 fuel elements. The hot end weight losses for both the NRX-A6 and NRX-A5 fuel elements were about the same. NRX-A6 demonstrated excellent corrosion resistance and integrity in the aft non-fuel tip region. Only 30 fuel elements exhibited corrosion in the region of the tip joints. Some variation in corrosion performance was noted between the various matrix-coating batch types and with respect to core radius. These variations were not nearly as pronounced, however, as had been observed in previous NRX reactor tests.

(CRD) Notch-type corrosion patterns similar to those observed on peripheral NRX-A5 elements were again observed on the peripheral faces of some NRX-A6 peripheral elements, although the fraction of peripheral NRX-A6 elements affected was lower. The post test analysis on the peripheral fuel elements show that this notching was due to thermal stress at the peripheral corners of the fuel elements. This high thermal stress occurred during startup and was due to the combined effects of the heat flow from the core to the hot buffer filler strips

and the cooling effect of a high leakage flow between the filler strips at locations where shifting of the element filler strips created large flow gaps. It is this leakage flow that caused the element notching to occur only at certain points in this case. More recent NERVA designs eliminate both these stress causes. The heat flow to the filler strips is eliminated by moving the filler strips outside the thermal barrier and the large leakage flow path is eliminated by placing the wrapper directly against the fuel.

1.2.2 (CRD) Corrosion Behavior (U)

(CRD) The corrosion weight loss in NRX-A6 fuel elements averaged 11.2 grams of which 10.5 grams were estimated to have been due to corrosion of the coolant channel bores. The radial distribution of bore weight loss for two axial fuel element regions is shown in Figure 12. These data indicate a low, fairly uniform loss from the cold end of the elements (0 to 30 inches), and a hot end (30 to 52 inches) loss which is a factor of 7 to 10 higher and varies with core radius.

(CRD) In previous NRX reactor tests, 70 to 90 percent of the bore corrosion weight loss occurred in the 15 to 25 inch region. The low level of loss in this region of the NRX-A6 fuel constitutes a major improvement which can be largely attributed to changes in coating technique. The typical post-test condition of the NRX-A6 coolant channels in this region is depicted in Figure 13. The fuel slice illustrated is inch 20 through inch 25. Portions of the NbC coating were flaked away to show molybdenum depositon patterns corresponding to cracks in the NbC.

(CRD) The hot end (30 to 52 inches) weight losses in the NRX-A6 core were about the same as those in NRX-A5. Thus, the time rate of corrosion in this region for NRX-A6 elements was about 50 percent of the corresponding rate in NRX-A5 elements. The principal mechanism for hot end weight loss was limited by carbon diffusion through the NbC coating. However, other types of corrosion were noted in some NRX-A6 fuel elements which have been correlated with differences in NbC coating parameters. In the following discussion, fuel elements characterized by diffusion controlled corrosion will be designated as having "Type W" coatings; coatings giving rise to corrosion pockets and pinholes will be designated "Type Y".

TABLE 1
(CRD) COMPARISON OF NbC DEPOSITION PARAMETERS* (U)

	Type Y	Type W
NbC Deposition Temperature, °C	1750 ± 25	1700 ± 20
Coating Time, hrs	17	14
NbC Thickness, mils	2.0 - 2.1	1.9 - 2.0
Crack Density, per inch	41 - 58	60 - 90
Mo Coating, Mg/in	10 - 30	30 - 50

* Values given correspond to an axial position of 45 inches, the position of maximum frequency of pinhole occurrence

The post-corrosion appearances of the two types of element are contrasted in Figure 14. Table 1 gives the nominal coating parameters for the two types. Fuel elements with Type Y coatings were found to have a higher hot end weight loss than those with Type W coatings (Rf. Table 2 in the next section). Further discussion of the hot end corrosion performance of Type Y coatings will be given in the next section.

(CRD) The axial variation in post-reactor flexure strength, averaged for the 79 elements tested, is plotted in Figure 15. The average and range of flexure strength for Quality Control testing of NRX-A6 production elements, representing all fuel loadings and various manufacturing histories, has been superimposed on this figure. Also included in Figure 15 are the core average, axial variations in flexure strength obtained from the post-operational testing of 56 fuel elements from NRX-A5.**

(CRD) Flexure strength changes from the QC average in NRX-A6 were characterized by an increase in the cold end region, practically no change in the midband region and a decrease in the hot end region. With the exception of the cold end increase, these flexure strengths correspond to the axial variations in fuel element incremental weight loss. There

** (CRD) The Quality Control average flexure strengths for NRX-A5 production elements was nominally about 13 percent lower than that of NRX-A6 elements; 280 pounds for NRX-A6 production elements and 244 pounds for NRX-A5 production elements.

is a dramatic difference between the NRX-A6 and NRX-A5 flexure data in the 20 to 25 inch axial region where the higher corrosion damage and correspondingly greater weakening of the NRX-A5 elements occurred.

1.2.3 (CRD) Pinholes (U)

(CRD) The occurrence of pinholes in the surface of NRX fuel elements has been of major concern with respect to both bore corrosion and surface corrosion performance. The formation of a pinhole implies a localized region of accelerated bore corrosion (a "corrosion pocket") which when intersecting the surface of the fuel element can produce corrosion of the adjacent fuel element surface. The pinhole frequency (pinholes/element) in NRX-A6 fuel elements was approximately 50 percent lower than that observed in NRX-A5 as shown in Figure 16. Pinholes in the A6 fuel elements typically occurred in the hot end axial region (40 to 50 inches) while those in NRX-A5 were found predominantly in the 22 to 25 inch and 38 to 41 inch regions. The typical appearance of pinholes in NRX-A6 elements is shown in Figure 17.

(CRD) As introduced in the previous section, there was a difference in the hot end bore corrosion patterns of the Type Y and Type W bore coated fuel elements in NRX-A6. This difference is reflected in the axial bore weight loss distribution and pinhole frequency data for the two types given in Table 2. The pinhole frequency in the Type Y elements is

TABLE 2

(CRD) BORE WEIGHT LOSS AND PINHOLE FREQUENCY IN NRX-A6 FUEL (U)

	Average Bore Weight Loss Grams/Element		Pinhole Frequency Pinholes/Element
	<u>0 to 30 inches</u>	<u>30 to 52 inches</u>	
Type W	0.9	8.3	1.6
Type Y	1.3	9.3	7.7
Type W - NbC Additive	1.8	11.3	2.0
Type W - Nb Resinate	1.1	10.4	1.0
All Core	1.2	9.0	4.9

about five times that of the Type W elements. However, the magnitude of this difference is not reflected in the differences observed in hot end weight losses. That is, the pinholes observed cannot account for the entire difference in hot end weight loss between the two types. Therefore, the existence of sub-surface corrosion pockets is implied. This is in accord with the observed existence of corrosion pockets in the Type Y elements as shown in Figure 14 and previously described.

(CRD) It appears that the most significant difference between the two types of coatings is in their crack densities (Cf. Table 1). Lower crack densities require larger crack widths and imply poorer coating adherence. This condition existed in the downstream end of the Type Y elements and is believed to have allowed the hydrolysis of fuel particles at the coating-fuel interface before the reactor test. The swelling of hydrolyzed fuel particles is known to cause coating defects as shown in Figure 18. Coating defects of this type would be expected to result in corrosion pockets like those observed. In support of this hypothesis, a strong correlation existed between pinhole frequency and NbC crack density in NRX-A6 fuel elements as shown in Figure 19.

1.3 (CRD) CONCLUSIONS (U)

(U) The operation of the NRX-A6 reactor was highly successful. By comparison with previous NRX tests, NRX-A6 displayed high performance for a longer period of time and under more stringent conditions. The operating data and results for the NRX reactor series are summarized in Table 3.

(CRD) The corrosion performance of the NRX-A6 fuel elements showed that a major improvement has been realized in the elimination of severe corrosion in the 0 to 30 inch region. The hot end performance indicated that extended fuel element life in high temperature operation and multiple restart capability will require further improvement.

TABLE 3

(CRD) NRX-A2 THROUGH NRX-A6 MAJOR REACTOR PERFORMANCE SUMMARY (U)

<u>Reactor</u>	<u>NRX-A2</u>	<u>NRX-A3</u>	<u>NRX/EST</u>	<u>NRX-A5</u>	<u>NRX-A6</u>
Chamber Temperature (°R)	Time within chamber temperature interval (minutes)				
3700-3800	0.8	0.2	3.6	0.7	0.1
3800-3900	---	13.4	6.1	3.0	0.1
3900-4000	---	3.0	5.3	11.3	0.1
4000-4100	---	---	7.3	12.8	0.6
4100-4200	---	---	7.8	2.3	54.6
4200-4300	---	---	---	---	6.8
4300-4400	---	---	---	---	0.6
Equivalent Full Power Radiation (Minutes)					
a. Entire Test Series	5.7	26.8	54.4	32.2	66.9
b. Single Run	5.1	14.3	14.5	16.4	66.6
Maximum Flow Rate (Lb/Sec)	75.6	73.5	87.9	71.9	72.0
Maximum Power (MW)	1096	1100	1155	1120	1200
Number of Startup and Shutdown Cycles Above 2500°R	1	3	6	2	1
Core Average Corrosion Weight Loss, Grams Per Element	1.0	10.6	32.4	27.0	11.2
Observed Total Reactivity Loss, (\$)	0.1	0.9	3.2	2.1	0.7

SECTION 2

(CRD) DEVELOPMENT STUDIES (U)

2.1 (CRD) ADVANCED COATED PARTICLE EVALUATION AT WANL (U)

(CRD) Data presented by R. Bard, LASL, during the Thirteenth Coated Particle Conference in December 1967, demonstrated the feasibility of producing pyro-carbon coated uranium dicarbide fuel beads resistant to thermal migration in the 2600°C to 2800°C range when fabricated in a specified manner and given a preliminary thermal re-organization cycle of 2300°C for four hours.

(U) In March 1968, WANL entered a purchase order with Gulf-General Atomic to establish the feasibility of volume production with commercial equipment by producing an iteration of the LASL Laboratory scale results. It was desired to obtain significant quantities for evaluation in NERVA fuel elements within the design guidelines existent for R-1 and subsequent series of test reactors.

(CRD) The R-1 guidelines were defined:

- 1) Nominal fuel element matrix temperature, 2300°C.
- 2) Maximum fuel element matrix temperature, 2500°C.
- 3) Maximum as-coated particle size not to exceed that of fuel beads previously applied to the NRX reactor series (250 microns maximum, "as-coated").
 - a) UC_2 core size before coating 105 to 149 microns.
 - b) "As-coated" particle size range 177 to 250 microns.
 - c) The pyrocarbon coating thickness to be 40 microns rather than the previously used 25 microns. (The total spread of "as-coated" particle distributions has a smaller range and the average size a larger nominal value. This results in a lowered uranium density and requires a larger number of particles per unit volume of fuel element matrix for any given uranium loading).

- 4) Minimum internal void following the 2300°C, four-hour thermal re-organization cycle to be larger than the relative size change between the high expansion core and low expansion pyro-carbon coating at reactor operating temperature.
 - a) The internal void is to provide relief from internal tensile stresses in the pyro-carbon coating arising from mechanical loading by the more highly expanded UC₂ core as the bead is heated.

(CRD) Fuel beads were delivered in both the "as-coated" and thermally reorganized conditions by General Atomic in June 1968. The migration resistance was rated by the supplier as less than 3 percent totally penetrated following a thermal re-organization cycle of 2300°C for four hours and a thermal migration cycle of 2600°C for 15 minutes.

(CRD) Both the "as-coated" and thermally re-organized materials have been extruded into NERVA fuel elements with a nominal loading of 400 mg/cc. All fuel elements have been manufactured and all scheduled manufacturing operations are presently completed. Samples have been taken and distributed to the cognizant testing groups.

(CRD) Evaluation of graphite matrix-fuel bead interactions is underway and will consist of:

- 1) Effect of graphitization temperature over the temperature range of 2300°C to 2500°C.
 - a) Evaluate bead damage by Xenon release following in-pile activation.
 - b) Metallographic examination of fuel element cross sections.
 - c) Radiography of fuel element cross sections cut as thin wafers.
 - d) Mechanical and physical properties, including creep studies.
- 2) Direct comparison of both re-organized and "as-coated" advanced beads to fuel beads previously applied to the NERVA program.

- a) Fuel elements are graphitized at 2300°C for four hours.

The thermally re-organized beads are dimensionally stable during graphitization.

The "as-coated" fuel bead coatings undergo thermal re-organization concurrently with the fuel element binder. Both the binder and the fuel bead coating assume their final dimensions and densities concurrently leading to the potential of developing the maximum residual stress situation throughout the fuel element matrix.

- b) An evaluation is made, following machining bores and external surfaces and hot gas leaching (gaseous HCl), of:

Physical and thermal properties.

Interactions between gaseous HCl used to leach fuel elements and molybdenum brazed joinings to non-fueled sections.

Mechanical properties, including creep studies.

(U) Following evaluation of this first phase, a second phase iteration will be conducted leading to final bore coated elements evaluated with the final hydrogen corrosion tests performance specifications applied to NERVA fuel elements. Second phase elements may be candidates for inclusion in the Pewee 2 core test.

(U) Completion of the first and second phase evaluations of advanced performance fuel beads in fuel elements and results from the loose bead studies being conducted by R. Bard at LASL will provide sufficient data to write purchase contracts for subsequent iterations of commercial production of further advancements in pyro-carbon coated UC_2 fuel beads.

2-2 (CRD) OTHER DEVELOPMENT AREAS (U)

2-2.1 (CRD) Fuel Matrix Studies (U)

(CRD) Several experimental fuel elements in NRX-A6 were made with resin or carbide impregnated fuel matrices. The performance of these experimental elements showed no significant improvement over production NRX-A6 elements so this approach will be down-graded.

(CRD) The most promising area of matrix improvement experimentation is the use of high expansion graphites. By this means, it is hoped that the thermal expansion mismatch between the fuel substrate and the protective carbide coatings can be minimized. Thus, crack size and crack density in the coatings would be reduced.

2-2.1 (CRD) Coating Deposition Studies (U)

(CRD) The principal area of coating deposition development involves the addition of methane to the coating vapor. By this means carbon to form the carbide is provided from the vapor, not taken from the substrate. The advantages of this process are more adherent coatings -- (the substrate surface needn't recede by donating carbon to the coating)--, and higher deposition rates giving shorter production times -- (carbide formation rate is controlled by the vapor composition, not by carbon diffusion through the carbide).

(CRD) In addition, the deposition of ZrC over NbC in the hot end of the fuel element bores is being studied. The ZrC layer is intended as a barrier to diffusion controlled corrosion since the diffusivity of carbon in ZrC is lower than that in NbC.

2-2.3 (CRD) Advanced Fuel Studies (U)

(CRD) The concept of a fuel element composed of a fuel dispersion in graphite with a homogeneous region at the hot end is being explored. The homogeneous fuel material under consideration is a solid solution carbide, (U, Zr) C, with Zr as the major metal component. The advantages of this concept are: decreased hot end corrosion and the ability to operate the hot end of the reactor at higher temperatures. These effects would produce a rocket with a higher specific impulse.

Figure 1. (CRD) NRX-A6 -- Thermal Power and Control Drum Position (U)

Figure 2. (CRD) Molybdenum Coating Over NbC Primary Coating After Test (250X) (CRD)

Figure 3. (CRD) UC_2 Migration in Coated Fuel Particle (250X) (U)

Figure 4. (U) Central Region, Axial Temperature Distributions During Power Spike

Figure 5. (U) Peripheral Region, Axial Temperature Distributions During Power Spike

Figure 6. (U) Core Central Region, Axial Temperature Distributions During Power Hold

Figure 7. (U) Mid-Radial Region, Axial Temperature Distributions During Power Hold

Figure 8. (U) Peripheral Region, Axial Temperature Distributions During Power Hold

Figure 9. (U) Peripheral Region, Axial Temperature Distributions at End of Power Hold

Figure 10. (CRD) NRX-A6 Chamber Temperature and Flow Rate (U)

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Figure 11. (CRD) Variation of Cumulative Reactivity Loss with Time for the NRX-A4, NRX-A5, and NRX-A6 Reactors (U)

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Figure 12. (CRD) Radial Distribution of Coolant Channel Weight Loss (U)

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Figure 13. (CRD) Typical Cold End Fuel Section -- NRX-A6 (U)

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(a) Element with Type W Coating

(b) Element with Type Y Coating

Figure 14. (CRD) Comparison of Hot End Corrosion Behavior in NRX-A6 (U)

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Figure 15. (CRD) Core Average Axial Variation in Flexure Strength for 79 NRX-A6 and 63 NRX-A5 Fuel Elements (U)

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Rad. Inc.
 *NRX-A5
 *NRX-A6

*Percent Element Examined Which Pinholed

612540-5

Figure 16. (CRD) Radial Distribution of Pinhole Frequency and Density in NRX-A5 and NRX-A6 (U)

Figure 17. (CRD) Typical Appearance of Pinholes in NRX-A6 Fuel Elements (U)

Figure 18. (CRD) Illustration of the Formation of a NbC Coating Flake by UC_2 Hydrolysis (CRD)

Figure 19. (CRD) Correlation of Pinhole Frequency with NbC Crack Densities in NRX-A6 Fuel Elements in Region of Highest Pinhole Density (U)

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