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RADIOCHEMISTRY FOR THE RUPTURE OF ZIRCALOY-2 CLAD HEAVY-WALLED TUBULAR FUEL ELEMENT IN KER LOOP-3.

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RADIOCHEMISTRY FOR THE RUPTURE OF ZIRCALOY-2 CLAD HEAVY-WALLED TUBULAR FUEL ELEMENT IN KER LOOP-3

INTRODUCTION

On the 0000 - 0800 shift, December 9, 1960, the delayed neutron monitor on KER Loop 3 gave a high coolant activity signal indicating a possible fuel element failure in this loop. KE Reactor was shut down immediately thereafter.

This report is being written to summarize the events pertinent to this KE Reactor scram and to discuss the results and significance of data from analyses on coolant and coupon samples taken from the KER Loop 3 system.

SUMMARY AND CONCLUSIONS

A high delayed neutron signal on the coolant in KER Loop 3 on December 9, 1960 indicated the possibility of a fuel element rupture. The KE Reactor was shut down. The fuel elements in this loop were discharged during the outage. A crack was found near the end cap weld on one fuel element in the 105 KE viewing pit. Further investigations in the 300 Area showed that several of the fuel elements had cracks at the base of the end caps.

Coolant samples were drawn from the KER Loop 3 system and subjected to radiochemical analyses. Higher than normal concentrations of fission products were discovered.

DISCUSSION

There were two test objectives in the irradiation of the Zircaloy-2 jacketed heavy-walled tubular fuel elements in KER Loop 3.[1,2] The first was to determine the behavior of these fuel elements during high-temperature operation. The second was to obtain irradiated, predefected, capped, heavy-walled Zircaloy-2 clad tubular fuel elements for in-reactors failure testing in this ETR after irradiation to a desired exposure in the KER Loops. The operating conditions in KER Loop 3 for this test were as follows:

1. Equilibrium effluent temperature - 270-280 C.
2. Temperature at time of rupture - 40 C.
3. Coolant pH - 9.9 maintained by flow through a lithium based ion exchange resin and LiOH additions.
4. Loop flow - 60 gpm.
5. Cleanup flow - 2-3 gpm.
6. System pressure - ~ 1600 psi.
7. Degasification rate - ~ 0.25 gpm.

KER Loop 3 is a stainless steel system with a Zircaloy-2 in-reactor tube. The operational chronology pertinent to this fuel failure is as follows:

1. July 3, 1960 - Charged Zircaloy-2 clad heavy walled tubular fuel elements in KER Loop 3.
2. October 27, 1960 - Charged 1 Zircaloy-2 clad predefected capped element in KER Loop-3.

3. ~0350 Dec. 9, 1960 - KER Loop 3 delayed neutron monitor began to rise.
4. ~0450 Dec. 9, 1960 - KER Loop 3 delayed neutron monitor went full scale. KE Reactor shut down. KER Loop 3 began depressurization cycle.
5. ~0457 Dec. 9, 1960 - KER Loop 3 began single pass operation.
6. 0000 - 0800 shift, Dec. 9, 1960 - discharged KER Loop 3 fuel elements.

No noticeably discolored liquid passed from the process tube at the time of the discharge. Visual inspection of the heavy-walled tubular fuel elements in the KE viewing facility revealed that all were covered with dark adherent, comparatively thin oxide coatings along all heat generating surfaces. A 1/16" crack was found during this inspection on Element Number 17, a heavy-walled fuel element (not a capped defect specimen).* The uranium core was not visible. Inspection in the Radiometallurgical Facility 300 Area showed that five elements from the charge experienced a "necking-down" effect of the Zircaloy-2 at the base of the end cap. Cracks were found on all of these five.** The uranium cores were however not visible. All this information indicates that only a small amount of uranium was corroded from the fuel element responsible for the high activity signal. Shut down, discharge, handling and transfer doubtlessly aggravated the already deteriorated condition of these fuel pieces at the base of the end cap. Likewise for all of the cracks and "necking-down", there was only one piece from the charge that caused the high delayed neutron indications that caused the shut down of the KE Reactor. For cracks to appear simultaneously on two different fuel elements does not seem highly likely.

A. Coolant Radiochemical Analyses

A water sample was drawn from K-3 during the loop depressurization and dump cycle. Another was later drawn from the K-3 emergency storage tank. These samples were subjected to certain radiochemical procedures to determine fission product loadings in the water.

The data in Table I should be the best radiochemical data to date from a KER Loop rupture. Even though the loop was in its depressurization cycle there should not be very many diluting factors to modify the loadings that are actually present from this rupture.

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* W. K. Kratzer - Personal Communication.
** G. T. Geering - Personal Communication.

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TABLE I

Radioanalysis of KER Loop-3 Coolant During The Rupturing Of A 16" Heavy-Walled Tubular Fuel Element

Isotope	Activity (in $\mu\mu$c/ml)	Isotope	Activity (in $\mu\mu$c/ml)
I^{131}	5.7	Sc^{46}	8
La^{140}	310	Co^{60}	12
Ce^{143}	410	P^{32}	150
Np^{239}	.2	Cu^{64}	140
Sm^{153}	130	Na^{24}	ND*
Ce^{144}	970	Mn^{56}	ND
Cs^{137}	310	As^{76}	ND
Zn^{65}	27	Sr^{89}	80
Zr^{95}-Nb^{95}	270	Sr^{90}	26

* ND means nondetectable.

Following other ruptures, a sample was taken from the emergency storage for a radiochemical analysis.(3,4,5,6) Certain modifying factors always influenced this data, however. The loadings in the emergency storage tank are lower by a factor of $\sim$2.5 than those in the loop because the depressurization and dumping operation dilutes the coolant. Some settling of relatively large particulate matter probably, also, occurs in the tank before sampling. The thermal shock of the loop system seems to have caused some activity fluctuations such as the large increase of Cu^{64} activity over normal conditions. These data are presented in Table II.

[redacted]

TABLE II

Radioanalysis Results of K-3 Emergency Storage Tank Water

Isotope	Activity (in μμc/ml)	Isotope	Activity (in μμc/ml)
As^{76}	200	P^{32}	16
Cr^{51}	57	Zn^{65}	3.7
Np^{239}	.2	Sc	.9
Sm^{153}	210	Co^{60}	3.2
Eu^{152}	35	Zr^{95}-Nb^{95}	56.0
La^{140}	41	Sr^{89}	73
Ce^{143}	50	Sr^{90}	1.8
I^{131}	8.3		
Cu^{64}	2100		
Ce^{144}	220		
Cs^{137}	19		

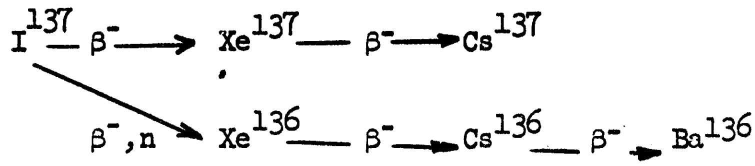
A comparison of the data in Tables I and II with data from the coolant under normal conditions shows that most of the fission product activities were higher than normal. A few such as I^{131} were of comparable levels. From a radiochemical standpoint there is, however, little doubt that there was a rupture in KER Loop 3. The difference in the fission product spectrum for the rupture in this case can be attributed to the type of fuel element failure. Examination of the fuel pieces revealed that there was little corrosion of the uranium core. Most of the species that were in the coolant were the daughters of gaseous or very soluble species and could therefore be easily transferred to the coolant with only small portions of the uranium seeing the coolant.

A small amount of fission products is observed in the coolant during normal operation.[3,4,5,6] This is presumed to be primarily due to fissioning of the uranium impurity in the Zircaloy-2 process tube and the resulting recoil of the products into the coolant stream. Some diffusion from the fuel elements probably also occurs.

B. Instrumentation Response

The first indication of a fuel element failure in KER Loop 3 was given by a rise in the activity indicated on the delayed neutron monitor at approximately 0350 December 9, 1960. By changing the scale from 0 - 250 neutrons/sec to 0 - 500 neutrons/sec the increase in instrument readings was only momentarily checked. At 0450 the readings were such that the KE Reactor shut down. These fluctuations are seen in Figure 1.

The three precusors known with certainty that give delayed neutrons are Br^{87}, t 1/2, 54 sec; Br^{89}, t 1/2, 5.8 sec; and I^{137}, t 1/2, 22 sec. The I^{137} isotope decays according to the following scheme:



The Cs^{137} isotope, a I^{137} daughter, was determined in the two samples by radio-chemical analyses. These concentrations were higher than normal. No attempt was made to correlate delayed neutron monitor signal height with concentration and fuel element irradiation: but the presence of Cs^{137} at these higher concentrations suggests that there were significant quantities of delayed neutron emitters in the coolant to cause a higher than normal instrument response.

Only very weak sympathetic delayed neutron responses were noted on KER Loops 2 and 4.

Another interesting factor can be noted by comparing the delayed neutron response on KER Loop 3 with a rupture (Figure 1) and the response on KER Loop 1 with a rupture (Figure 2). Figure 1 shows a rise in activity occurring over a period of nearly an hour while Figure 2 shows an abrupt rise that could not be checked even by an instrument scale change.(4) This behavior sheds light on the difference in rupture mechanisms. The response in KER Loop 3 resulted from the exposure of a small amount of the uranium core to the coolant at low temperatures while the KER Loop 1 failure resulted from the rupture of a blister that discharged at high temperature, relatively large quantities of rupture products very rapidly into the coolant.

ACKNOWLEDGEMENT

The efforts of W. C. Johnson and his coworkers in the Purex Analytical Operation are gratefully acknowledged. C. L. Buckner, IPD, assisted in obtaining KER charts and rupture data. KER Loop 3 is operated by the Coolant Testing Unit, IPD.

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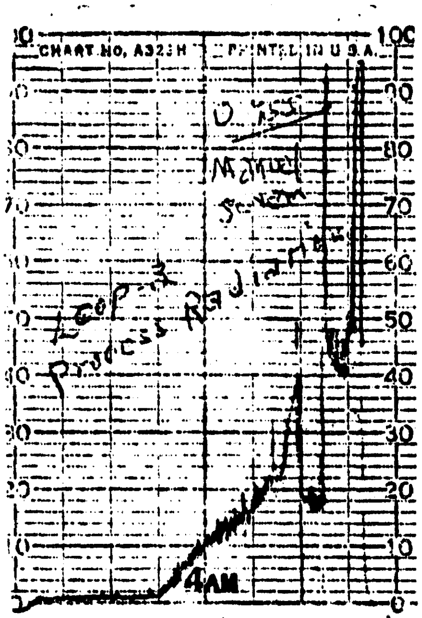

FIGURE 1. KER Loop 3 Delayed Neutron Indications.

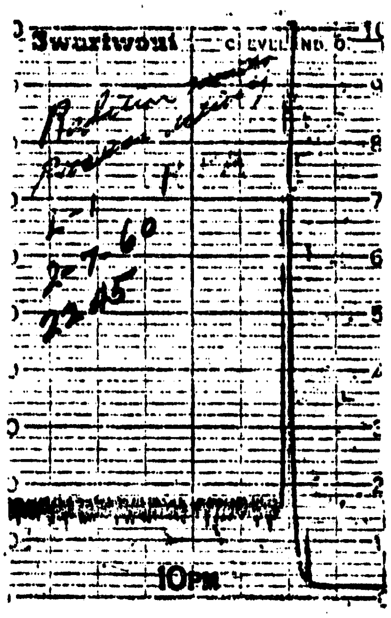

FIGURE 2. KER Loop 1 Delayed Neutron Indications

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