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GENERAL ELECTRIC

HANFORD ATOMIC PRODUCTS OPERATION – RICHLAND, WASHINGTON

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TITLE
RADIOCHEMISTRY FOR THE RUPTURE OF A TUBE IN TUBE FUEL ELEMENT IN KER LOOP 3

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L. D. Perrigo

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November 5, 1959

RADIOCHEMISTRY FOR THE RUPTURE OF A TUBE IN TUBE FUEL ELEMENT IN KER LOOP 3

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INTRODUCTION

On the 0000 - 0800 shift, September 30, 1959, the delayed neutron monitor on KER Loop 3 gave a high coolant activity signal. Events occurred rapidly that tended to substantiate the possibility of a fuel element failure in this loop. KE reactor was shut down immediately thereafter.

This report is being written to summarize the sequence of events prior to and those following the KE reactor scram and to discuss the results and significance of data from analyses on coolant and coupon samples taken from KER Loop 3.

SUMMARY AND CONCLUSIONS

A high delayed neutron signal on the coolant in KER Loop 3 on September 30, 1959 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. An examination in the radiometallurgical facility showed that there was a rupture on the inner tube of a tube and tube fuel element.

A coolant sample was drawn from the KER Loop 3 system and subjected to a radiochemical analysis. Concentrations of fission products many fold higher than normal were discovered. Np^{239} and La^{140} were found also to be adsorbed on the surfaces of metal coupons removed from this loop. This information, from a radiochemical standpoint, indicates a rupture and it concurs with the indications seen on the delayed neutron monitor.

DISCUSSION

KER Loop 3 is a high-temperature, high-pressure recirculation system constructed of stainless steel with a zircaloy-2 process tube. This loop, since February 1959, has operated with its coolant pH maintained at 4.5 by the addition of phosphoric acid. This water quality was chosen since it appears to be the optimum for aluminum corrosion studies.

A. Operation Conditions

At equilibrium the following are the operating conditions on KER Loop 3:

1. Temperature - 245 - 285°C
2. Coolant pH - 4.5 with H_3PO_4 additions
3. Loop flow - 60 gpm
4. Cleanup flow - 2 - 3 gpm
5. System pressure - ~1625 psi
6. Degasification rate - ~.25 gpm

Figure 1 shows a plot of the process tube effluent temperature for KER Loop 3 for the period in which the tube in tube fuel elements were in this loop. There were twenty days of operation at temperature and pressure.

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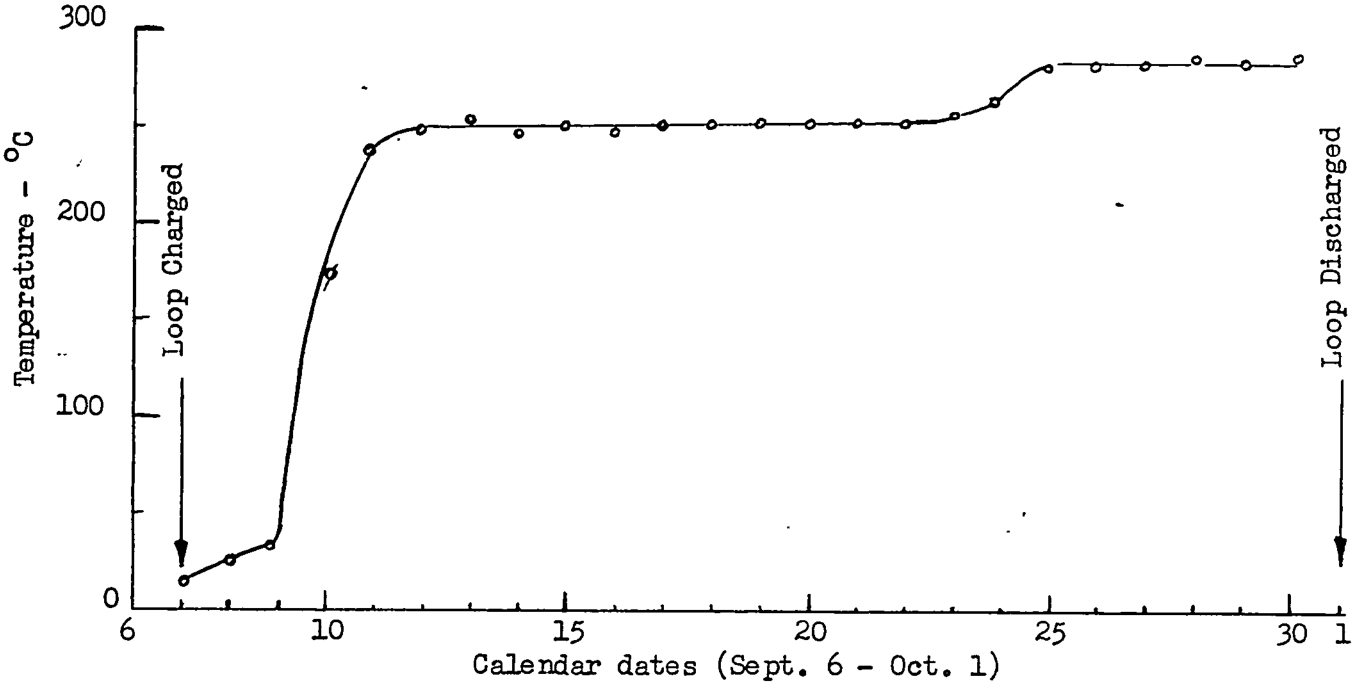

FIGURE 1. KER Loop 3 Process Tube Effluent Temperatures

B. Operation Chronology

The operational chronology pertinent to this fuel failure is as follows:

1. September 7, 1959 - charged three zircaloy-2 clad tube in tube fuel elements into KER Loop 3.
2. September 27 - 30, 1959 - pressure drop across process tube increased 4 psi.
3. ~0600. September 30, 1959 - delayed neutron monitor jumped to 30% of full scale and held momentarily. Then it increased to 60%. There were no other monitor indications from the other KER loops.
4. ~0602, September 30, 1959 - Started mounting portable gamma counters on KER Loop 3.
5. ~0605, September 30, 1959 - KER Loop 3 delayed neutron monitor went 100% scale. Scale factor was increased by 500 but the indicator remained at the full scale position. Sympathetic responses were noted on delayed neutron monitors on loops 1, 2, and 4. KE reactor shut down. KER Loop 3 began depressurization cycle.
6. ~0608, September 30, 1959 - KER Loop 3 began single pass operation.
7. ~0600, October 1, 1959 - discharged tube in tube fuel elements from KER Loop 3 process tube.

The discharge of the fuel elements was accompanied by the passing of a considerable quantity of darkly colored liquid. This liquid was described as "looking like mud".[1] The liquid in the pickup tray was so murky that one could not see the fuel elements until the loop coolant was displaced by fresh water.

Visual inspection of the tube in tube fuel elements in the KE viewing facility revealed that all had random deposits of crud of appreciable thickness. The failure in the zircaloy-2 cladding could not be positively identified at KE Reactor. Their geometries are such that inner surfaces are not accessible for inspection in most viewing facilities. The crud on the heat transfer surfaces varied from black to light brown in color. However, inspection in the Radiometallurgical Facility, 300 Area, has shown that there was a defect on the inner surface of the inner rod approximately at the mid point in length.[2]

C. Radiochemical Analyses

A water sample was drawn from the KER Loop 3 emergency storage tank approximately three hours after the loop depressurization operation was completed. This sample was subjected to certain radiochemical procedures to determine fission product activity loadings. These loadings are, however, probably lower by a factor of greater than 2.5 than those that actually were in the loop. This is the result of loop depressurization that occurs when the reactor scrams with an indication of a fuel failure in the loop's process tube. The depressurization cycle results in the coolant being dumped into the emergency storage tank with the simultaneous forcing of an equal volume of demineralized water from this storage tank into the recirculation system. Since the emergency storage tank volume is five hundred gallons and the KER Loop 3 volume is approximately two hundred gallons there was a dilution of approximately 2.5 before the water sample was drawn for analysis. Some settling of particulate matter probably, also, occurred in the tank prior to sampling. This would also influence the data and the results from this radiochemical analysis shown in TABLE I should, therefore, be lower than the undiluted unsettled coolant.

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(1) W. K. Kratzer - Personal Communication

(2) HW-62428, Examination of Ruptured KER Tube in Tube Fuel Element from IP-250-A, Supplement B, G. A. Last and G. T. Geering.

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TABLE I

Radioanalysis of KER Loop 3 Emergency Storage Tank Water

Analysis	$\mu\mu$ Curies/ml
Total Beta	460,000
I_{total}	120,000
I^{131}	23,000
Ru^{106}	2,300
Zr^{95}-Nb^{95}	34,000
Np^{239}	50,000
Cs^{137}	310
Cu^{64}	10,200
Ba^{140}	~ 5,000
Sr^{89}	~ 2,000

Exposure time- 511 equivalent full power hours.

All activities calculated to four hours after beginning of KE outage on 9-30-59.

Even with these diluting factors there were much greater concentrations of Np^{239}, I^{131}, I_{total}, Sr^{89} and Ba^{140} than are encountered during normal operation of a low pH system (TABLE II). This trace of a fission product spectrum found in the KER Loop coolants during normal operation is generally believed to be the result of the fissioning and resulting recoil of the products from the uranium impurity found in the zircaloy-2 process tubes. Probably there is also some contribution from diffusion from the fuel elements to the loop coolants.

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TABLE II

Fission Product Spectrum in Low pH System During Normal Operation

Sample	Exposure eq full power hrs.	Total Beta	I^{131}	I_{total}	Cu^{64}	Np^{239}	$Ba^{140}-La^{140}$	$Sr^{89}-Sr^{90}$
		(all activities in $\mu\mu$ curies/ml)						
1	81	108,00	.24	17	42,000	-	12	3.2
2	106	60,600	.78	7.5	8,200	-	7.0	5.6
3	36	18,200	.30	6.1	370	770	4.6	.75
4	205	521,000	-	-	4,200	620	13	.26
5	301	1,089,000	-	-	186,000	< 610	16	5.7
6	79	394,000	-	-	2,400	< 18	14	3.9
7	267	570,000	-	-	1,700	< 5.6	36	-
8	489	745,000	-	-	800	< 5	44	34
9	225	945,000	-	-	330	< 83	150	73
10	340	570,000	-	-	280	< 3.9	120	32
11	125	420,000	-	-	2500	< 2.8	20	26

All activities calculated to four hours after sampling time.

The data shown in TABLE II were accumulated on a low pH system operating with no cleanup. This would tend to maximize the activities found in a loop during normal operation (no defected fuel element). At the time of the rupture in KER Loop 3 a phosphate regenerated mixed bed ion exchange resin was being operated continuously in parallel with the loop for cleanup purposes. This constitutes another sink in the coolant system in addition to the adsorption on pipe walls and leakage. For comparable activity sources, a loop with a cleanup system in operation should have lower coolant activity loadings.

The presence of copper in the KER Loops has been attributed to the dissolution of copper compounds used in sealing threaded pipe joints. The coolant loading of this short half life isotope would, then, be a function of the frequency of this type of maintenance operation. However, there is a strong possibility that some of the Cu^{64} activity found in the water sample drawn from the emergency storage tank on September 30, 1959 came from the copper lubricant used during the coextrusion of the zircaloy-2 clad tube in tube fuel elements.

[redacted]

D. Coupon Analyses

Several inconel-X, monel, and stainless steel washer type coupons were in the KER Loop 3 mockup tube at the time of the KE outage. These were removed and subjected to a gamma scan analysis. Significant quantities of Np^{239} La^{140} were found to be adsorbed on the coupon surfaces. Quantitative data are not available at this time.

After consideration of this analytical information from two sources, the liquid and metal samples, and other associated factors already mentioned, there is no doubt from a radiochemical standpoint that there was a rupture in KER Loop 3 on September 30, 1959.

E. Delayed Neutron Monitor Response

The first indication of a fuel failure in KER Loop 3 was given by the delayed neutron monitor jumping to 30% of full scale. This indication and the following fluctuations are shown in FIGURE 2. Only one delayed neutron emitter daughter, Cs^{137}, was sought in the radiochemical analysis on the liquid sample. No direct correlation between the height of the delayed neutron signal and specie concentration has been, therefore, attempted. The presence of Cs^{137}, however, shows that there were appreciable quantities on hand to cause a delayed neutron monitor response.

The accompanying effluent loop temperature variations are shown in FIGURE 3.

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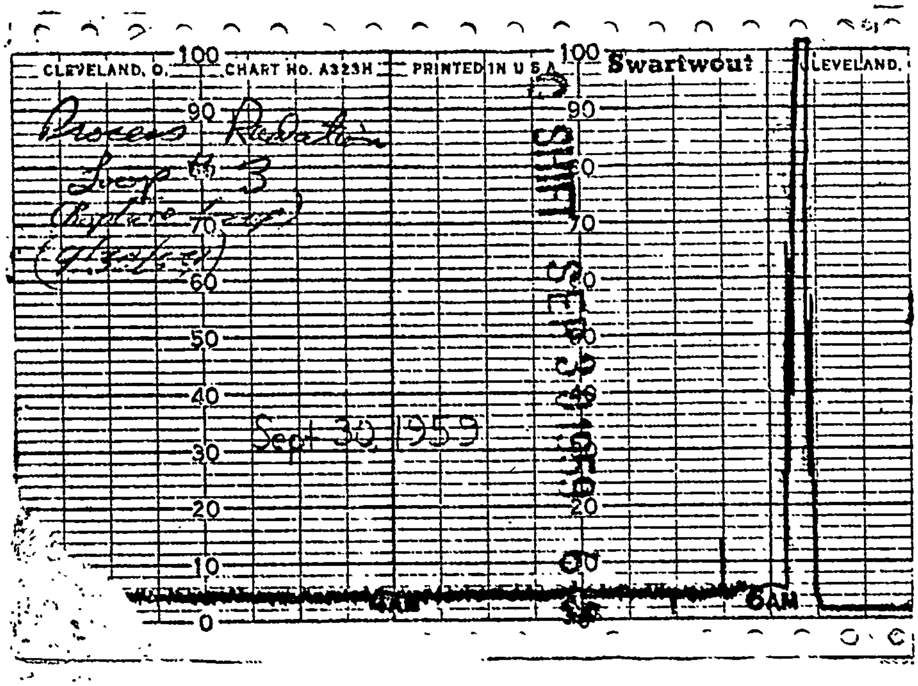

FIGURE 2. KER Loop 3 Delayed Neutron Monitor Indications

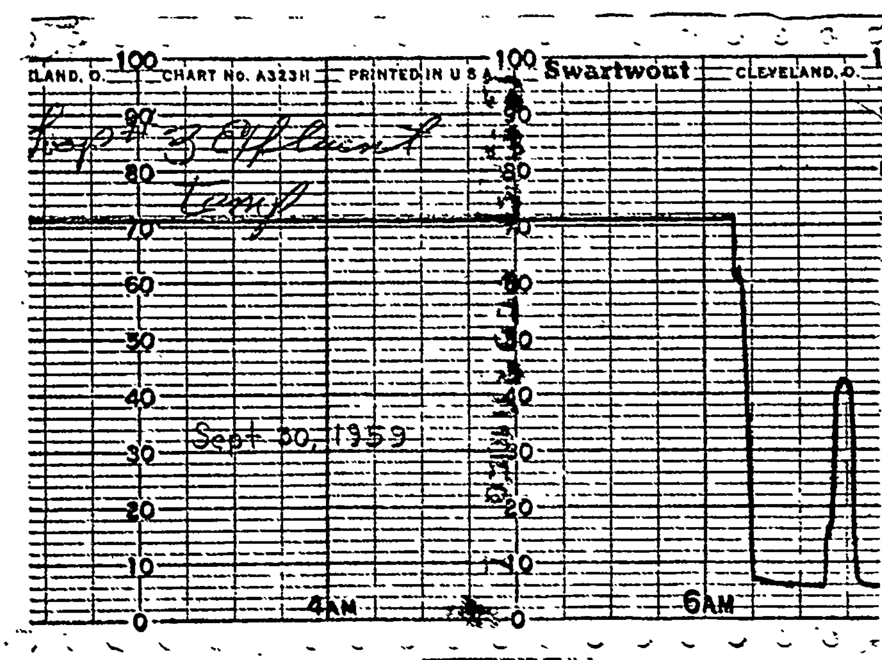

FIGURE 3. KER Loop 3 Process Tube Effluent Temperature

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F. Loop Radiation Levels

High activity levels were noted in the north corridor of the 1706 KE facility shortly after the first delayed neutron signal. Flushing after shutdown reduced gamma activity levels to the point that there was little appreciable increase in cell or piping activity over the normal when operation was resumed with a dummy charge.

Several days later a gamma heating test was conducted on this loop. Coolant flow was reduced to 15-20 gallons per minute and all cooling water was valved out of the heat exchanger. Over a period of time the system temperature climbed to a limit of 197°C. After reaching this temperature flow was increased in incriments. Accompanying these flow fluctuations was an increase in the delayed neutron activity over the normal background indications. Loop gamma activity began to climb also. There are two apparent possibilities: (1) A deposit of uranium dioxide in the loop was disloged so that was available for irradiation; (2) Some uranium dioxide that had been adsorbed in the flux zone was dislodged by velocity fluctuations.

Coolant samples were taken for radio analysis. A fuller explanation should be available when data from these samples are available.

ACKNOWLEDGEMENT

The efforts of R. W. James, W. H. Zimmer and their coworkers in the Purex and Redox Analytical Operations are gratefully acknowledged. The helpful suggestions and comments of F. E. Sharp and R. V. Dulin, IPD, on the sections covering KER Loop 3 operation are appreciated. KER Loop 3 is operated by the Coolant Testing Operation, IPD.

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HANFORD LABORATORIES OPERATION

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