RADIOISOTOPE CONCENTRATIONS IN THE PRTR PRIMARY COOLANT
AND HELIUM SYSTEM DURING OPERATION WITH A FAILED FUEL ELEMENT(S)

By

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HW--70980

DE92 005513

Radiological Chemistry
Chemical Research
Hanford Laboratories Operation

September 7, 1961

HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

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INTRODUCTION

Fission product measurements in the PRTR primary D_2O and helium systems during the startup tests in May 1961, showed the presence of a defective fuel element(s).⁽¹⁾ Reactor operation was discontinued during the latter part of May and during June for reactor maintenance and fuel element inspection. On resumption of operation on July 1, a sampling and analytical program was initiated to study the radioisotope concentrations resulting from this defective element(s) which were present in the primary D_2O and helium systems during the various phases of reactor operation. The data accumulated during this study are to serve as background information in future fuel element rupture identification and location studies as well as serving as a guide in setting up the rupture monitor for the individual tubes of the PRTR reactor. A summary of the analytical results from this study and their theoretical interpretation is presented in this paper.

SUMMARY AND CONCLUSIONS

Measurements of the fission products in the PRTR primary D_2O and helium system during the July 1 startup and subsequently have reaffirmed the presence of a defective fuel element(s). Bursts of fission gases following reactor startup or increases in power level were observed and under the reactor's operating conditions were equivalent to that produced in 150 grams of uranium (or 870 mg Pu). These activity bursts frequently increased the specific activity of the helium system by two orders of magnitude (and an order of magnitude in the D_2O), and were clearly defined by the presently operating gross gamma monitors. The gross activity as recorded by these monitors has a very sharp climb for about 30 minutes and a peak duration of approximately 6 hours. Calculations based on the observed activity increase in the helium system during bursts indicate that during the first minute of the burst the gas activity from a tube containing a failed element would be about 2 orders of magnitude greater than normal.

The activity differential would drop to about a factor of 5 in 5 minutes and to about 1 percent in 30 minutes. It is therefore evident that a rupture monitor must be designed to provide almost instantaneously comparison of the reactor's 85 tubes if it is to be successful in pinpointing defective fuel element locations.

Theoretical and Discussion

The presence of a failed fuel element(s) in the PRTR reactor demonstrated the usefulness of the presently operating "gross-gamma" rupture monitors for the total helium and primary D_2O systems. It also permitted the measurement of the gross activity changes and of specific radioisotope concentration changes during the various phases of reactor operation. The normal observed radioisotope levels, and the expected time, magnitude, and duration of activity bursts determine the requirements for a successful process tube rupture monitoring system.

Previous observations were confirmed that essentially the only fission products in the helium system were the fission gases (and their daughters) and in the primary D_2O system were the fission gases and the iodine radioisotopes.⁽¹⁾

During the initial reactor startup for this period (July 1, 1961) the primary D_2O and helium samples were rushed to the laboratory to measure their first gamma-ray spectra 3-4 minutes after sample collection. The object of these rapid measurements was to determine if short-lived gamma-emitting fission products (half lives of minutes) were present in significant amounts in these systems. If they were, a monitor set to channel their energies might provide a very sensitive fuel element rupture detector. The gamma-ray spectra measured during this initial startup were not typical of those previously observed from the helium gas system. A considerable amount of air had been trapped in the helium system during the previous maintenance period and this resulted in the production of a comparatively large amount of A^{41} (from the A^{40} normally present in air) which essentially masks the gamma ray spectra (see Figure 1). The A^{41} concentration dropped off rapidly during succeeding reactor operation due to the continuous addition of helium to replace helium leakage and A^{41} was a very minor constituent after one week of operation. After the air had been "purged" from the helium system, further measurements on helium samples were performed. Measurements of the gamma ray spectra on 4 minute old samples and 30 minute old samples did not show a significant difference. This would indicate that the contribution from short-lived fission products (half lives of minutes) is very small. However, if a sample were obtained and analyzed immediately when a burst of activity was being released, its spectrum might appear significantly different. A gamma-ray spectrum of 30 minute old gas is presented in Figure 2. It is evident that the Xe^{133} (5.2d), Xe^{135} (9.2 hr) and Kr^{88} (2.8 hr) are major constituents in the helium system. This gamma ray spectrum of the fission gases is very similar to that obtained from the off gases of bare UO_2 (measured after a 30 minute decay period).⁽²⁾ The gases Xe^{133} , Xe^{135} and Kr^{88} were also the major constituents of the primary D_2O system,⁽¹⁾ however, the iodine radioisotopes were sometimes present in rather large amounts. As an index to fission product release, the radioisotopes Xe^{133} , Xe^{135} and Kr^{88} were measured routinely in the helium system while I^{131} , I^{133} and I^{135} were measured in the primary D_2O system. These data have been tabulated for the period July 1 through July 24 in Table I. In Figures 3 to 6 these data are plotted on graphs of reactor power level to permit a direct evaluation of the effect of reactor operation on radioisotope release. The radioisotope concentrations were measured only three times per day in most cases and the lines connecting these points are not intended to infer concentrations at the intermediate locations. However, it is evident that high concentrations (bursts) of radioisotopes generally do coincide with a reactor power level increase. With this infrequent sampling, it was not possible to obtain reasonable definition of the activity bursts. During the July 15 startup and operation, samples were collected hourly for analysis and they provided much better "peak" definition. Following this startup, a "burst" of activity was released which increased the Xe^{133} (5.2d) and Xe^{135} (9.2 hr) by factors of 30 and 450 respectively in a one hour period.

Gross gamma monitors on the total primary D_2O and helium effluents have been recording the relative gamma intensities (but with some recorder and other instrument difficulties) during reactor operations. The activity bursts recorded by these instruments confirmed those observed by spot sampling and gave a much better definition of the relative magnitude and the width of the bursts. Data obtained from the gross-gamma chart readings showed that the total gamma level of the helium system can be expected to "jump" by one to two or more orders of magnitude from two to five hours after startup to a high power level (15-20 MW), or after a substantial increase in power level. A similar but lower relative increase was observed in the primary D_2O system. The rapid increase in activity occurred during about a 30 minute period and the peak width was usually about 6 hours. The amounts of natural uranium or Pu^{239} necessary to produce the peak (burst) concentration of the fission gas Kr^{88} (see Table I)

under the reactor's operating conditions have been calculated and are tabulated in Table II. For these calculations, 6000 standard ft³ was used as the volume of the primary helium system⁽³⁾ and the reactor neutron flux was taken as 7×10^{11} neutron/cm²/sec/megawatt.⁽⁴⁾ To place these values in proper perspective, they are also tabulated in Table II as the fraction of the total Kr⁸⁸ present in a single uranium fuel rod (2500 grams⁽⁵⁾). From these data it is evident that some of the activity bursts release a rather large fraction of the fission gas (about 6 percent) assuming that it comes from a single faulty element.

The problem of locating a failed element in PRTR is complicated by the rather high recirculation rate of the primary coolant (3 - 4 times per minute). It can be readily calculated that if a radioisotope with a half life of one hundred minutes or more is released at a steady state during reactor operation that the activity in the effluent from its tube would not be significantly higher (less than 20 percent higher) than the activity in the total effluent. Under these conditions the probability of detecting a faulty fuel element, either from a radiochemical analysis of all the process tubes' gas or liquid effluents or from periodic measurements of the effluents' gamma-ray emission, would be very small. An approach which does appear promising, however, is to continuously compare the gamma-ray emission rate of the 85 process tubes. The occurrence of an activity burst could be very easily identified under these conditions. Assuming that the emission of an activity burst lasts 30 minutes (about 100 passes of the primary D₂O through the reactor) and the resulting specific activity of the primary helium system is increased by two orders of magnitude, then the effluent from the faulty tube would be higher than the average effluent by approximately the following amounts:

<u>Time after initiation of burst - minutes</u>	<u>Gas from failed element tube Gas from total reactor</u>
0	1
0.25	100
1	30
5	5
30	1.01

From the above data it is evident that a failed element could be very readily pinpointed if the rupture monitoring system were adjusted to provide instantaneous comparison of the gaseous effluents from the 85 reactor process tubes.

An optimum energy discrimination for the rupture monitor would depend on previous reactor operation and maintenance history. If the primary helium system has been open to the air, large amounts of A⁴¹ (1.3 Mev gamma) will be produced (from the A⁴⁰ in the air) which will lower the sensitivity of any detection system based on energies below 1.3 Mev. If the rupture monitor is to be operated in the presence of A⁴¹ its discriminators should be set at 2 Mev to eliminate interference from this radioisotope. However, if the infrequent presence of large amounts of A⁴¹ and its attendant effect of lowering the rupture monitors' sensitivity can be tolerated, a generally higher sensitivity for the system can be obtained by using a low discriminator setting (just safely above normal instrument noise). If it

should be necessary to locate a faulty fuel element prior to the satisfactory operation of the rupture monitoring equipment, it is recommended that the method of coolant water stagnation and individual tube sampling⁽⁶⁾, which was developed at the Vallecitos Atomic Laboratory, be used.

ACKNOWLEDGEMENT

The authors wish to acknowledge the assistance of L. S. Kellogg of the Analytical Laboratories Operation who performed part of the analysis included within this report.

9-7-61

RW Perkins, CW Thomas:pct

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TABLE I

RADIOISOTOPE CONCENTRATIONS IN THE PRIMARY D₂O AND HELIUM SYSTEMS
(d/m/ml x 10⁻⁴)

<u>Date</u>	<u>Time Sampled</u>	<u>Helium System</u>			<u>D₂O System</u>		
		<u>¹³³Xe</u>	<u>¹³⁵Xe</u>	<u>⁸⁸Kr</u>	<u>¹³¹I</u>	<u>¹³³I</u>	<u>¹³⁵I</u>
5/17/61	1130	0.78	7.0	3.6	0.028	0.04	
7/1/61	2317	0.016	0.0064	0.0058	0.0008	0.0008	0.0028
7/3/61	2205	0.0028	0.0095	0.025	0.0026	0.0134	0.015
7/4/61	0215	0.0030	0.01	0.019	0.0005	0.004	
7/4/61	1730	0.0290	0.13		0.0005	0.017	
7/5/61	1015	0.0300	0.27	0.075	0.0017	0.15	0.12
7/5/61	1510	0.035	0.98		0.074	1.7	1.7
7/6/61	0940	0.050	0.52	0.02	0.019	0.3	0.15
7/6/61	2130	0.050	0.56	3.1	0.16	0.96	0.72
7/7/61	0245	0.11	1.3	0.37	0.42	5.0	5.1
7/7/61	0600	0.062	1.3		0.18	1.7	1.3
7/7/61	1822	0.029			0.02	0.45	
7/8/61	0345	0.095	2.1		0.6	6.4	4.3
7/8/61	1545	1.4	9.8		0.1	1.4	4.3
7/9/61	0230	0.72	2.1		< 0.01	< 0.1	< 0.1
7/9/61	1215	1.4	1.8		< 0.01	< 0.1	< 0.1
7/9/61	1700	2.6	3.9		< 0.01	< 0.1	< 0.1
7/10/61	0200	1.5	3.0	3.5	< 0.01	< 0.1	< 0.1
7/10/61	0945	1.6	3.5	8.5	< 0.01	< 0.1	< 0.1
7/10/61	1645	1.2	3.4		< 0.01	< 0.1	< 0.1
7/11/61	0100	2.0	6.8	14.8	< 0.01	< 0.1	< 0.1
7/11/61	0900	2.0	6.0	9.6	< 0.01	< 0.1	< 0.1
7/11/61	0950	1.7	4.2	6.1	< 0.01	< 0.1	< 0.1
7/11/61	1720	1.9	2.4	3.3	< 0.01	< 0.1	< 0.1
7/12/61	0100	2.1	2.5	3.5	< 0.01	< 0.1	< 0.1
7/12/61	0900	1.4	1.4	1.1	< 0.01	< 0.1	< 0.1
7/15/61	0136	0.08	0.01	0.35	0.0026		
7/15/61	0236	2.4	4.5	15	0.024		
7/15/61	0336	4.0	5.2	29	0.023		
7/15/61	0436	3.3	3.3	24	0.020		
7/15/61	0536	5.8	5.4	25	0.17		
7/15/61	0636	5.3	5.1	24	0.013		
7/15/61	1317	2.6	2.0	2.9	0.003		
7/15/61	1800	1.2	1.1	2.7	0.001		
7/15/61	1900	1.5	1.4	2.4	.0019		
7/15/61	2000	1.8	1.4	2.9	.0019		
7/15/61	2100	1.9	1.7	3.0			
7/15/61	2200	1.4	1.1	2.8	.0021		
7/16/61	0100	1.7	1.3	0.8	.0013		
7/16/61	0900	1.7	1.6	1.8	.0019		
7/16/61	1736	1.6	1.3		.0017		
7/17/61	0150	1.2	1.3	2.8	.0017		
7/17/61	0250	1.4	2.0	3.7	.0017		

TABLE I (Continued)

<u>Date</u>	<u>Time Sampled</u>	<u>Helium System</u>			<u>D₂O System</u>		
		<u>Xe</u> ¹³³	<u>Xe</u> ¹³⁵	<u>Kr</u> ⁸⁸	<u>I</u> ¹³¹	<u>I</u> ¹³³	<u>I</u> ¹³⁵
7/17/61	0350	1.3	2.3	4.2	.0013		
7/17/61	0450	1.3	2.6	3.7	.0014		
7/17/61	0550	1.4	2.6	4.2	.0015		
7/17/61	1120	0.46	1.2		.001		
7/18/61	2216	1.9	6.5		.0055		
7/19/61	0119	1.2	2.2	5.1	.003	.019	
7/19/61	1358	0.34	0.3		.0009	.019	
7/19/61	2130	0.75	0.4		.0009	.017	
7/20/61	0020	0.67	0.3		.0012	.017	
7/20/61	0920	0.48	0.3	0.38	.0009	.014	
7/20/61	1300	2.7	8.4	16	.009	.07	
7/20/61	1955	6.9	28	75	.01	.084	
7/21/61	0050	3	10	12	.05	.31	
7/21/61	0700	6.4	42	46	.044	.29	
7/21/61	1621	8.7	26	169	.05	.3	
7/21/61	1823	6.9	30	34	.045	.26	
7/22/61	0202				.036	.21	
7/22/61	0950			16	.017	.106	
7/22/61	1841				.0092	.058	
7/23/61	0430				.011	.083	
7/23/61	0920				.013	.1	
7/23/61	1700				.04	.08	
7/24/61	0120				.011	.088	
7/24/61	1244			.05	.0028	.014	

TABLE II

EQUIVALENT AMOUNTS OF NATURAL URANIUM AT HIGH Kr⁸⁸ CONCENTRATIONSIN PRIMARY HELIUM SYSTEM

<u>Date Measured</u>	<u>Natural U Equivalent (grams)</u>	<u>Kr⁸⁸ in Helium/Total Kr⁸⁸ in Fuel Rod (percent)</u>
7/7	18	0.73
7/8	> 31*	> 1.2*
7/11	22	0.9
7/15	48	1.9
7/16	4.8	0.19
7/17	6.4	0.25
7/18	32	1.3
7/20	108	4.3
7/21	155	6.2

* Estimate based on Xe¹³⁵ concentration

FIGURE 1

Gamma Ray Spectrum of a Gas Sample from the Primary Helium System Collected 3 Hours After the Initial July 1 Startup and Counted 5 Minutes Later

FIGURE 2

Gamma Ray Spectrum of a Gas Sample from the Primary Helium System Measured 30 Minutes After Collection

FIGURE 3

Radio-Iodine Concentration in the Primary D₂O System Projected on Reactor Power Level Graph

FIGURE 4

Fission Gas Concentration in the Primary Helium Gas System Projected on Reactor Power Level Graph

FIGURE 5
Fission Gas Concentration in the Primary Helium Gas System Protected on Reactor Power Level Graph

FIGURE 6

Radio-Iodine Concentration in the Primary D₂O System Projected on Reactor Power Level Graph

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