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QUARTERLY REPORT
CONTAMINATION CONTROL - COLUMBIA RIVER
JANUARY - MARCH 1968

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July 22, 1968

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DUN-4477

-2-

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-3-

QUARTERLY REPORT
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I. INTRODUCTION

It is the purpose of this report to present data obtained from the routine reactor effluent radioactivity monitoring program as well as the results obtained from the research and development program directed toward the reduction of effluent radioactivity.

II. PERTINENT ASPECTS OF REACTOR OPERATION

A. General

Samples of effluent are obtained each week day during operation and analyzed for P-32, As-76, Np-239, and Cr-51. The Zn-65 concentration in the effluent from each reactor is obtained from a weekly composite sample. The above nuclides are monitored because:

1. Phosphorus-32 and Zn-65 are the nuclides of Hanford origin which are of significance in fish, shellfish, and foodstuffs.
2. Arsenic-76 and Np-239 are responsible for most of the dose received by drinking water derived from the Columbia River.
3. Chromium-51 is the nuclide released in the greatest quantity downstream of the Hanford plant, but its contribution to dose is relatively small.

B. Phosphorus-32 and Arsenic-76

1. Concentration in Reactor Effluent

a. B Reactor

The alum feed rate at B Reactor was 11.4 ppm during January. The coolant pH was maintained at 6.6. The sodium dichromate feed rate to the near side of the reactor was 0.5 ppm, and the far side feed rate was 1.0 ppm. The average P-32 and As-76 concentrations in the effluent are shown in the following table.

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	<u>Phosphorus-32</u> <u>pCi/ml</u>	<u>Arsenic-76</u> <u>pCi/ml</u>
January, 1968	7.6	115
January, 1967	6.5	95
January, 1966	14.8	95

Both the P-32 and As-76 results are in line with those of the previous year.

b. C Reactor

The alum feed rates at C Reactor were 12.7, 11.3, and 15.3 ppm during January, February, and March, respectively. The sodium dichromate feed rate was held at 0.5 ppm during the period. The pH of the coolant was 6.2 on the far side and 6.6 on the near side until January 22, 1968. Following that date the pH was 6.6 on both sides. The P-32 and As-76 concentrations in the reactor effluent during recent months were:

	<u>Phosphorus-32</u> <u>pCi/ml</u>	<u>Arsenic-76</u> <u>pCi/ml</u>
March, 1968	19.1	210
February, 1968	15.6	165
January, 1968	18.3	255
January-March 1967*	11.6	115
January-March 1966*	10.2	100

*Three month average

The values in recent months are higher than for the corresponding period in previous years.

c. K Reactors

The alum feed rates were 12.8, 12.3, and 14.4 ppm during January, February, and March, respectively, at KE Reactor and 11.9, 11.8, and 14.7 ppm at KW Reactor. The coolant pH was maintained at 6.6, and the sodium dichromate concentration remained at 1 ppm throughout the period at KW Reactor. At KE Reactor the sodium dichromate concentration was reduced to 0.5 ppm on March 21, 1968. The P-32 and As-76 concentrations in the K Plant effluents during recent months were:

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DUN-4477
-5-

	<u>Phosphorus-32</u>		<u>Arsenic-76</u>	
	<u>pCi/ml</u>		<u>pCi/ml</u>	
	<u>KE</u>	<u>KW</u>	<u>KE</u>	<u>KW</u>
March, 1968	16.5	11.2	135	130
February, 1968	14.4	10.0	105	80
January, 1968	8.9	5.3	125	105
January-March 1967*	6.0	6.0	65	70
January-March 1966*	7.1	8.6	50	130

*Three month average

Both the P-32 and As-75 concentrations found in recent months are somewhat higher than during the corresponding period of 1967.

During the period a number of efforts were made on one or two KW Plant filters to use activated silica as a filter conditioner. However, it was not possible to obtain reproducible results and the test was abandoned.

d. Long-Term Trends

The long-term trends in P-32 and As-76 concentrations are shown in Figures 1 and 2.

1. Phosphorus-32

The release rates of P-32 averaged 21, 20, and 36 curies per day during January, February, and March, respectively. The 12-month average release rate at the end of March was 29 curies per day. This can be compared to a 12-month average of 28 curies per day as of the end of December 1967.

2. Arsenic-76

The release rates of As-76 averaged 320, 230, and 360 curies per day during January, February, and March, respectively. The 12-month average release rate at the end of March was 290 curies per day. This compares to a 12-month average of 270 curies per day at the end of December 1967.

C. Neptunium-239

1. Concentration in Reactor Effluent

The monthly average concentrations of Np-239 in the small and K Reactor effluent are shown in Figure 3.

2. Reactor Output

The 12-month average release rate of Np-239 at the end of March was 290 curies per day as compared to 280 curies per day at the end of December 1967.

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DUN-4477

-6-

D. Chromium-51

1. Concentration in Reactor Effluent

The monthly average concentrations of Cr-51 in the small and K Reactor effluent are shown in Figure 4.

2. Reactor Output

The 12-month average release rate of Cr-51 at the end of March was 750 curies per day. This can be compared to a 12-month average release rate of 820 curies per day at the end of December 1967.

E. Water Plant Operating Conditions

The changes made in coolant treatment since mid-1961 are shown in Table 1.

III. RESEARCH AND DEVELOPMENT

A. Water Analysis

The analytical data obtained in January and February for the phosphate, iron, silica, and sulfate are shown in Table II. In addition, some zinc analyses are included. The analytical program was terminated with the shutdown of B Reactor.

B. B Area Reactor Effluent Infiltration Test

The infiltration test in the B Area trench located east of the 107 C retention basins continued. The trench is 500 ft. long, 40 ft. wide at the bottom, and 200 ft. wide at the top. This will result in a wetted surface of 52000 sq. ft. when the water level is 10 ft. above the bottom. The trench is 850 ft. from the river at the closest point. Effluent is introduced to the trench through two 42 in. diameter pipes which are tied to the 66 in. diameter line which runs from the west 107 C retention basin to the river.

B Reactor effluent was started into the trench on October 30, 1967 at a rate of 5000 gpm. During the first week of operation the rate was increased to 25,000 gpm. Succeeding flow increases brought the rate to 50,000 gpm on November 15, 1967. The rate has been maintained since. Again an infiltration rate of about 1500 gals/sq ft-day is indicated.

The height of the water table in the vicinity of the trench increased by as much as 15 feet and has apparently stabilized. The water level in the peripheral wells is continuing to increase. Wells as far as two to three miles from the trench have shown a water level increase of from three to five feet. The ground water under B Area had been hot (50 to 70 C) from effluent basin and effluent line leakage. These

temperatures have not changed a great deal. Ground water temperatures in the surrounding area are increasing slowly. While extensive new seepage areas formed along the river bank early in the test, they also appear to have stabilized. Radiochemical analyses of the river bank springs show I-131 and Cr-51 concentrations to have been reduced by factors of two to three. The Zn-65 and P-32 concentrations were reduced by factors in excess of 100.

C. Water Filtration Studies

Approximately 15 gallons of Columbia River water were passed through a continuous high-speed (25,000 rpm) centrifuge to remove particles down to 0.1 μ which produce turbidity. This material was compared by X-ray diffraction with sediment samples collected from the river bank at Richland. The analysis showed the materials to be similar so that the river bank sediment can be used to produce turbid water for the pilot plant experiments.

Using the water filtration pilot plant, experiments were conducted to determine alum requirements at various turbidity levels and the associated zeta potential. At least squares fit of the data yielded the following equation:

$$y = 10.22 + .752X$$

x = turbidity, JTU

y = Alum Conc., mg/l for 0 zeta potential.

This data is preliminary and will require additional information before it can be fully accepted because of the errors involved in the zeta potential determination. Data from continued laboratory work plus river samples and plant operating data will be added to check the above equation.

D. Streaming Potential

The streaming potential and associated currents are of interest to those concerned with in-reactor corrosion since they can be anticipated in reactor coolant channels. This current, if substantial, could cause significant corrosion. To determine whether such currents could be obtained in practice, a low temperature test was run in an out-of-reactor loop using a metal flow channel. The data obtained indicated that the potential obtained reached a limiting value with increasing flow. Since this behavior could be either harmful, if due to the shorting of the current through the walls of the channel, or relatively harmless, if due to the characteristics of the flow, further experiments were considered necessary to distinguish between the two cases. Preliminary tests using a plastic tube as the flow channel substantiate the latter conclusion. Because the data are not suitably reproducible, the work will be continued. There are also questions of the effects of temperature and coolant chemistry to be considered.

E. Deionized Water Studies

Following the experiment in which unexpectedly high effluent activity was obtained when zirconium-clad fuel charges were exposed to process water, it was decided to see if the results from a previous experiment in which low effluent activities which resulted from the use of Zr-clad fuel and deionized water could be duplicated. The data from this experiment are incomplete; however, effluent activities are decreasing. The effluent from the zirconium tube contains more activity than that from the aluminum tube, which is contrary to the previous results. This is attributed to the fact that a new aluminum tube was put in service and has not developed a complete surface-deposited impurity layer. The difference in effluent activity is thus attributed to differences in the activation products formed in the available deposits.

F. Water Treatment Variables

"Cat Floc," a positively charged polyelectrolyte, is now being used as the coagulant in place of alum in the Water Treatment Pilot Plant. The main objective in this test is to eliminate the aluminum that passes through the filter. It has been theorized that the residual aluminum may furnish the bulk of the material that forms the film on in-reactor surfaces, and as such, furnished impurities either through codeposition or exchange. The initial experiment involved "Cat Floc" addition at a rate (ea 0.5 ppm) sufficient to maintain neutral surface potential. At first, Separan was added as a filter aid, but high filter effluent turbidities prompted elimination of this additive. Even so, the filter effluent turbidities remained higher than the normal 0.05 Jackson unit (J.U.). It increased after backwash to about 0.4 J.U. and decreased through the run to 0.1 J.U., probably due to the formation of a "schmutzdecke."

G. Computer Study

In previous reports we have discussed the use of the analog and digital computer in developing our model of the kinetics of activity buildup on reactor process tubes, fuel elements and in the coolant. In the initial work with the analog computer, we found that the coefficients relating to the deposition and release of material from the surfaces were interrelated.

The relationship obtained is

$$B = aA + b$$

where A and B are the release and deposition coefficients and a and b are assumed to be constant. Furthermore,

$$a = \frac{m}{ke}$$

and

$$b = \frac{m}{k_e} [k_e f k_i (1 - V) - \lambda_0]$$

where m is a geometry factor for the system, k_e is the equilibrium constant which relates the concentration of the material on the film to an equilibrium liquid concentration, $f k_i$ is the rate at which particles deposit on the surface by methods other than pure diffusion, $1 - V$ is the fraction of the material in particulate form and λ_0 is a rate coefficient for material leaving surface in particulate form.

In this particular study we have examined the activation of arsenic, present in concentrations of less than one part per million. The test facilities consisted of two tubes in KE Reactor charged with aluminum clad fuel. One tube was a zirconium alloy and the other aluminum.

We have found that good fit between experiment (which contained large unexplained variations in the data) and the theory, using the digital computer, was obtained when it was assumed that the arsenic could form two chemical species. The parameters calculated, based on this assumption, are:

For the aluminum tube:	<u>a</u>	<u>b</u>
(1)	40	85
(2)	4.3×10^{-10}	43

The data scatter was such that a , for species (2), ranged between 4.3×10^{-10} and 20.

For the zircaloy tube:	(1)	(2)
	1100	17
	38	40

The calculations also indicated that species (1) in each tube was the same. A similar conclusion was drawn for species (2).

Using the above data, while realizing that some of the numbers are of more qualitative than quantitative significance, the values for k_e , $f k_i$, and λ_0 were calculated. Additional assumptions made were that $f k_i$ and λ_0 are the same for both tubes for a given species and that $1 - V = 0.02$. The values calculated are:

Term	Aluminum Tube Species		Zircaloy Tube Species	
	(1)	(2)	(1)	(2)
k_e (cm ⁻¹)	0.2	1.9×10^{10} to 0.4	0.006	0.18
$f k_i$ (cm/sec)	0.15	0.08	0.15	0.08
λ_0 (hr ⁻¹)	0.05	0	0.05	0

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DUN-4477

-10-

All of the values calculated for k_e are much higher than the value of 0.0003 cm^{-1} obtained from an independent experiment for aluminum. Also, the k_e data for the Zircaloy tube are smaller than those for the aluminum tube which is contrary to expectation.

The kinetic deposition factors, fk_i , are of the order of calculated values. The calculated factors were obtained using the assumption that the particles were of the order of 0.2μ in diameter.

No evaluation of the λ_0 values will be made at this time owing to insufficient data.

R. G. Geier, Program Manager
Process and Programs Section

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TABLE I
WATER PLANT OPERATING CONDITIONS

Alum Feed Rate Changes, ppm

Initially all reactors variable from 3 to 14.

7/1/61	B and C to 18	10/30/63	1/2 B to 11
7/6/61	KE and KW to 18	1/23/64	1/2 B to 7
9/6/61	KW to 8	5/7/64	1/2 B to 18
10/6/61	KW to 12	2/1/65	KW to 8
12/6/61	KW to 18	8/20/65	KW to 3
3/9/62	KE and KW to 10	10/15/65	KW to 6
4/12/62	KE to 13	1/12/66	KW to 9
4/30/62	KW to 18	3/27/66	KW to 12
5/21/62	KW to 20, KE to 18	9/15/66	KW to 15
9/1/62	KW to 18	9/1/67	B and C to zeta potential control
2/15/63	KE, KW Variable 6 to 18		
4/1/63	KE, KW to 12	11/8/67	KE and KW zeta potential control
5/1/63	KE, KW variable 12 to 18		

pH Changes

Initially all reactors 7.0

6/23/61	KW to 6.6	12/5/64	KW to 6.6
9/26/61	KE to 6.6	12/6/64	KE to 6.6
3/1/63	C to 6.6	11/21/66	1/2 KW & KE to 6.8
6/20/63	B to 6.6	1/5/67	1/2 KW & KE to 6.6
9/25/64	KE, KW to 7.0	9/19/67	1/2 C to 6.2

Dichromate Changes, ppm

Initially all reactors 1.8

9/29/60	1/2 C to 1.0	4/14/64	B, 1/2 C, KE, KW to 1.0
7/1/61	B, KE to 1.0	3/24/67	1/2 B to 0.5
9/26/61	KE to 1.8	9/19/67	C to 0.5
9/5/63	B, KE, KW to 1.4	3/21/68	KE to 0.5

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DUN-4477

-12-

TABLE II
WATER ANALYSIS
183 B RAW WATER

	<u>SO₄</u> <u>ppm</u>	<u>SiO₂</u> <u>ppm</u>	<u>PO₄</u> <u>ppb</u>	<u>Fe</u> <u>ppb</u>	<u>Zn</u> <u>ppb</u>
11-15-67	*	*	*	*	19
16	*	*	*	*	16
17	*	*	*	*	16
23	*	*	*	*	48
24	*	*	*	*	32
25	*	*	*	*	29
26	*	*	*	*	27
27	*	*	*	*	27
28	*	*	*	*	29
29	*	*	*	*	27
30	*	*	*	*	33
12- 1-67	*	*	*	*	31
3	*	*	*	*	28
4	*	*	*	*	23
12	*	*	*	*	86
13	*	*	*	*	26
14	*	*	*	*	21
15	*	*	*	*	25
18	*	*	*	*	25
19	*	*	*	*	24
20	*	*	*	*	24
21	*	*	*	*	26
22	*	*	*	*	23
23	*	*	*	*	6
24	*	*	*	*	24
25	*	*	*	*	16
26	*	*	*	*	24
27	*	*	*	*	21
28	*	*	*	*	16
29	*	*	*	*	100
30	*	*	*	*	24
31	*	*	*	*	17
1- 1-68	11.5	6.8	87	113	27
4	18.5	5.7	98	130	29
8	15.2	4.6	125	26	110
9	15.8	4.5	87	27	43
10	16.0	4.7	87	13	45
12	16.0	4.4	95	22	44

*Data reported previously in DUN-3935.

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DUN-4477

-13-

Table II (continued)

	<u>SO₄</u> <u>ppm</u>	<u>SiO₂</u> <u>ppm</u>	<u>PO₄</u> <u>ppb</u>	<u>Fe</u> <u>ppb</u>	<u>Zn</u> <u>ppb</u>
1-15-68	17.8	5.5	114	29	44
16	15.8	5.0	106	25	40
17	16.7	4.2	106	23	30
19	13.0	5.4	95	25	29
20	15.7	6.2	95	40	30
21	16.5	6.7	73	65	25
23	15.2	3.9	158	53	40
24	18.0	7.2	117	43	28
25	17.7	6.2	112	41	19
26	13.8	6.4	128	90	35
27	17.3	7.8	131	225	23
28	21.5	6.7	131	137	16
29	16.7	5.0	120	90	26
30	14.8	6.2	103	88	44
2- 1-68	16.5	6.2	117	61	37
2	16.3	7.3	112	65	44
6	21.2	6.9	120	78	39
7	19.2	8.0	144	47	35
8	19.1	6.6	128	45	36
9	20.3	6.4	142	98	39

RADIOACTIVITY IN REACTOR EFFLUENT

NEPTUNIUM-239

CHROMIUM -51

FIGURE 1

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RADIOACTIVITY IN REACTOR EFFLUENT

PHOSPHORUS-32

ARSENIC-76

FIGURE II