1 of 1

DECLASSIFIED

DOCUMENT NO.

DUN-4847

Hanford Category C-44

SERIES AND COPY NO.

//

DATE

October 15, 1968

(CLASSIFICATION)

DOUGLAS UNITED NUCLEAR, INC.

RICHLAND, WASHINGTON

☒

RESTRICTED DATA

THIS DOCUMENT CONTAINS RESTRICTED DATA AS DEFINED BY THE ATOMIC ENERGY ACT OF 1954. ITS TRANSMITTAL OR THE DISCLOSURE OF ITS CONTENTS IN ANY MANNER TO AN UNAUTHORIZED PERSON IS PROHIBITED.

☐

OTHER OFFICIAL CLASSIFIED INFORMATION

THIS MATERIAL CONTAINS INFORMATION AFFECTING THE NATIONAL DEFENSE OF THE UNITED STATES WITHIN THE MEANING OF THE ESPIONAGE LAW, TITLE 18, U. S. C., SECS. 793 AND 794, THE TRANSMISSION OR REVELATION OF WHICH IN ANY MANNER TO AN UNAUTHORIZED PERSON IS PROHIBITED BY LAW.

TITLE

QUARTERLY REPORT

CONTAMINATION CONTROL - COLUMBIA RIVER

APRIL - JUNE 1968

AUTHOR

R. G. Geier

THIS DOCUMENT MUST NOT BE LEFT UNATTENDED WHERE AN UNAUTHORIZED PERSON MAY HAVE ACCESS TO IT. WHEN NOT IN USE, IT MUST BE STORED IN AN APPROVED LOCKED REPOSITORY WITHIN AN APPROVED GUARDED AREA. WHILE IT IS YOUR POSSESSION AND UNTIL YOU HAVE OBTAINED A SIGNED RECEIPT FROM CLASSIFIED FILES, IT IS YOUR RESPONSIBILITY TO KEEP IT AND ITS CONTENTS WITHIN THE LIMITS OF THIS PROJECT AND FROM ANY UNAUTHORIZED PERSON. ITS TRANSMITTAL TO, AND STORAGE AT YOUR PLACE OF RESIDENCE IS PROHIBITED. IT IS NOT TO BE DUPLICATED. IF ADDITIONAL COPIES ARE REQUIRED, OBTAIN THEM FROM THE RELATED ISSUING FILE. ALL PERSONS READING THIS DOCUMENT ARE REQUESTED TO SIGN IN THE SPACE PROVIDED BELOW.

ROUTE TO:

PAYROLL NO.

LOCATION

FILES ROUTE DATE

SIGNATURE AND DATE

BNW			OCT 21 1968	
ATTN: C. J. Touhill	30908	713	OCT 22 1968	
D. E. Olsen		2224		
D. E. Olsen	32887	2224	NOV 5 1968	
D. E. Olsen	32887	154	FEB 24 1975	
D. B. Carlock	30894			

MASTER

DECLASSIFIED

DECLASSIFIED

DUN-4847
Hanford Category C-44

RESTRICTED DATA

This document contains restricted data as defined in the Atomic Energy Act of 1954. Its transmittal or the disclosure of its contents in any manner to an unauthorized person is prohibited.

This document consists of
14 pages, No. 11 of 37
copies. Series 6.

QUARTERLY REPORT

CONTAMINATION CONTROL - COLUMBIA RIVER

APRIL - JUNE 1968

R. G. Geier
Program Manager
Process and Programs Section

Classification Cancelled And Changed To

DECLASSIFIED

October 15, 1968

By Authority of Mr S E Gydesen,
11-27-89 RLO-06-4
By J Stone 11-30-89
Verified By B Roberts
11/30/89

DOUGLAS UNITED NUCLEAR, INC.
RICHLAND, WASHINGTON

Classification Cancelled (Change to

~~CONFIDENTIAL~~

By Authority of Assistant

11-24-74
By L Pope 2/24/75

NOTICE

THIS REPORT WAS PREPARED FOR USE WITHIN DOUGLAS UNITED NUCLEAR, INC. IN THE COURSE OF WORK UNDER ATOMIC ENERGY COMMISSION CONTRACT AT(45-1) - 1857, AND ANY VIEWS OR OPINIONS EXPRESSED IN THE REPORT ARE THOSE OF THE AUTHOR ONLY. THIS REPORT IS SUBJECT TO REVISION UPON COLLECTION OF ADDITIONAL DATA.

LEGAL NOTICE

THIS REPORT WAS PREPARED AS AN ACCOUNT OF GOVERNMENT SPONSORED WORK. NEITHER THE UNITED STATES, NOR THE COMMISSION, NOR ANY PERSON ACTING ON BEHALF OF THE COMMISSION:

A. MAKES ANY WARRANTY OR REPRESENTATION, EXPRESSED OR IMPLIED, WITH RESPECT TO THE ACCURACY, COMPLETENESS, OR USEFULNESS OF THE INFORMATION CONTAINED IN THIS REPORT, OR THAT THE USE OF ANY INFORMATION, APPARATUS, METHOD, OR PROCESS DISCLOSED IN THIS REPORT MAY NOT INFRINGE PRIVATELY OWNED RIGHTS; OR

B. ASSUMES ANY LIABILITIES WITH RESPECT TO THE USE OF, OR FOR DAMAGES RESULTING FROM THE USE OF ANY INFORMATION, APPARATUS, METHOD, OR PROCESS DISCLOSED IN THIS REPORT.

AS USED IN THE ABOVE, "PERSON ACTING ON BEHALF OF THE COMMISSION" INCLUDES ANY EMPLOYEE OR CONTRACTOR OF THE COMMISSION, OR EMPLOYEE OF SUCH CONTRACTOR, TO THE EXTENT THAT SUCH EMPLOYEE OR CONTRACTOR OF THE COMMISSION, OR EMPLOYEE OF SUCH CONTRACTOR PREPARES, DISSEMINATES, OR PROVIDES ACCESS TO, ANY INFORMATION PURSUANT TO HIS EMPLOYMENT OR CONTRACT WITH THE COMMISSION, OR HIS EMPLOYMENT WITH SUCH CONTRACTOR.

DECLASSIFIED

DUN-4847

-2-

DISTRIBUTION

- 1-3. AEC-RL: Attn: D. G. Williams
4. AEC-RL: Attn: R. L. Plum
5. AEC-Wash: Attn: A. Perge
Division of Operational Safety
6. AEC-Wash: Attn: W. J. Lindsey
Division of Production
7. BNW: Attn: J. F. Honstead
8. BNW: Attn: J. P. Corley
9. BNW: Attn: R. W. Perkins
10. BNW: Attn: W. B. Silker
11. BNW: Attn: C. J. Touhill
12. T. W. Ambrose
13. R. S. Bell
14. C. D. Corbit/J. H. Soehnlein
15. R. B. Crum
16. E. L. Etheridge
17. J. W. Frymier
18. R. G. Geier
19. R. W. Hallet, Jr.
20. C. D. Harrington
21. H. F. Jensen
22. P. C. Jerman
23. R. T. Jessen
24. W. K. Kratzer
25. C. W. Kuhlman
26. R. J. Meyer
27. R. Nilson
28. D. W. Peacock
29. R. S. Peterson
30. R. W. Pitman
31. R. W. Reid
32. J. W. Riches
33. O. C. Schroeder
34. G. W. Wells
35. W. G. Westover
36. DUN File
37. DUN Record

DECLASSIFIED

DECLASSIFIED

DUN-4847

-3-

QUARTERLY REPORT
CONTAMINATION CONTROL - COLUMBIA RIVER
APRIL - JUNE 1968

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 weekday 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. C Reactor

The alum feed rates at C Reactor were 18.3, 17.5, and 18.7 ppm during April, May, and June, respectively. The sodium dichromate feed rate was held at 0.5 ppm during the period, and pH of the coolant was maintained near 6.6. 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>
June, 1968	14.9	105
May, 1968	23.7	95
April, 1968	20.8	195
April-June 1967*	19.1	115
April-June 1966*	5.9	100

*Three month average

DECLASSIFIED

DECLASSIFIED

DUN-4847
-4-

Both the P-32 and As-76 values in June are comparable to the 1967 period.

During the period it was possible to increase the filter effluent turbidity on one-half the plant in accord with PTA-128* for about a month. There was no difference in effluent activity between the two sides of the reactor.

b. K Reactors

The alum feed rates were 17.2, 19.1, and 15.9 ppm during April, May, and June, respectively, at KE Reactor and 16.9, 16.5, and 14.5 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 0.5 ppm. The P-32 and As-76 concentrations in the K Plant effluents during recent months were:

	<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>
June, 1968	4.1	14.6	65	80
May, 1968	7.5	21.7	55	65
April, 1968	18.2	17.5	120	115
April-June 1967*	12.5	6.1	90	115
April-June 1966*	5.3	7.1	60	95

*Three month average

The P-32 concentrations in recent months have been quite erratic. The As-76 values are in the same range as in prior years.

c. 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 45, 39, and 15 curies per day during April, May, and June, respectively. The 12-month average release rate at the end of June was 27 curies per day. This can be compared to a 12-month average of 28 curies per day as of the end of December 1967.

*DUN-3973, "Production Test Authorization-128, Half Plant Test of Process Water Turbidity," G. W. Wells and R. G. Geier, March 18, 1968 (Unclassified)

DECLASSIFIED

DECLASSIFIED

DUN-4847
-5-

2) Arsenic-76

The release rates of As-76 averaged 320, 160, and 140 curies per day during April, May, and June, respectively. The 12-month average release rate at the end of June was 250 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 June was 270 curies per day as compared to 280 curies per day at the end of December 1967.

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 June 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. No changes were made during the period.

III. RESEARCH AND DEVELOPMENT

A. Water Treatment Variables

"Cat Floc," a positively charged polyelectrolyte, has been used as the coagulant in the Water Treatment Pilot Plant for one full fuel cycle. The substitution of this material for aluminum did not result in any change in the effluent concentrations of Sc^{46} , Cr^{51} , Co^{60} , Zn^{65} , La^{140} and Np^{239} from either the aluminum or zirconium tubes. The

DECLASSIFIED

DECLASSIFIED

DUN-4847

-6-

effluent concentrations of P^{32} and As^{76} were increased 3.5 and 4.1 times, respectively.

This loss of effectiveness is likely the result of removal of less of the parent material from the raw water. The two parents of interest are anionic and form insoluble compounds with aluminum and/or are removed by association with hydrous aluminum oxide in the standard treatment process. The corresponding association with "Cat Flocc" probably does not form such an insoluble compound and, therefore, arsenate and phosphate retention may be reduced by mass action effects of other more abundant anions such as sulfate. Conversely, if a strong bond is formed, removal is governed by the performance of the filters.

In an attempt to improve the filtered water turbidities, 5 ppm of bentonite was added to the raw water to provide an artificial turbidity and promote floc growth to facilitate removal by filtration. This was successful in providing turbidities of 0.04-0.07 Jackson unit, which were maintained throughout the filter run. This operation did cause in-reactor filming and required a purge periodically to reduce the pressure drop across the process tubes.

The filming problem continued throughout the period of its use. In addition, filter runs were shortened by rapid breakthrough to the extent that the water supply had to be supplemented by addition of process water to the clearwell.

Effluent activity was quite low during the initial portion of the run, but increased dramatically when the filming started. The effluent concentration of the As^{76} from the aluminum tube was double that observed when the cooling water was treated with Cat Flocc alone and an order of magnitude higher than found when using process water as the coolant. The same increase was seen in the effluent from the zirconium tube, but the initial activity levels were lower than would have been present with process water and thus the subsequent higher concentrations resulted in nearly equivalent values. These values were also a factor of three lower than obtained with water treated with Cat Flocc alone.

The treatment process was recently modified to include five ppm of alum in addition to Cat Flocc and bentonite in an attempt to provide more efficient removal of inert arsenic. No effluent data have yet been obtained; however, operational characteristics such as filter run and filter effluent turbidity are satisfactory.

B. Deionized Water Studies

Operation of two single pass tubes, charged with zirconium-clad fuel instead of aluminum and cooled with deionized water, resulted in reduced concentrations of effluent radioisotopes. These reductions ranged from 60 percent to 90 percent of the concentrations obtained

DECLASSIFIED

DUN-4847

-7-

during operation with aluminum clad fuel and verified the previous results obtained during similar operational sequences.

These results are significant because the additional zirconium surface did not provide similar reductions when process water was used to cool the tubes. This re-emphasizes the importance of the impurities introduced with process water in providing both the primary source of the parent isotope and the material from which the in-reactor film is derived.

The deionized water studies have been discontinued temporarily due to failure of the exchange beds. The situation will be rectified shortly, and work will be started on the reconstitution of process water experiment.

C. River Water Trace Element Composition

During the last several years the three new Columbia River Dams may have produced a significant effect on the trace element behavior of the Columbia River by providing both large areas of river bottom for leaching and for sediment deposition. Also, the increased use of pesticides and fertilizers may have contributed additional quantities of trace materials.

Increased concentrations of several radioisotopes in reactor effluent water prompted an investigation to determine any changes that might have occurred in the past several years in the trace element concentrations in Columbia River water. Preliminary results indicate 13 ppb of Mn and 120 ppb of Zn, both of which are about three times higher in concentration than existed in 1963. An arsenic concentration of 2.9 ppb was comparable to previous values. The antimony concentration was about 3.7 ppb; however, no historical data are available for this element.

In general, the observed concentrations of Cu, Mn, As, U, Zn, Co, Sc, Sb, Br, and Cs were comparable to those measured in 1963. Exceptions were Zn, Co, and Sc which were higher by respective factors of 4, 2, and 10 than the previous values.

D. Film Studies

Concurrently with the increased effluent concentrations, the dose rates on the downstream face have increased. To gain information on downstream deposition of radioisotopes, film was removed from an unidentified downstream spacer from one of the K Reactors and analyzed. Concentrations of iron, cobalt, and zinc appeared to be comparable to those measured previously on D Reactor elements. Zr^{95} and Hf^{181} were two orders of magnitude higher, and there seemed to be a significant increase in surface deposition of the rare-earth nuclides.

DECLASSIFIED

DUN-4847
-8-

Two additional downstream spacers from a KW zirconium process tube were received shortly after a reactor shutdown. The film was removed, acid-washed, and filtered. Analysis of the two fractions showed that the activity per unit surface area was lower by one to three orders of magnitude than the previous analysis of dummy film had shown. It is postulated that this was due in part to the fact that the current samples were taken from a zirconium tube, while the initial sample had been exposed in an aluminum tube. The radioisotopes Nb⁹⁵, Sb¹²⁴, and Pa²³³ were associated with the insoluble fraction, while the remainder of the isotopes found were soluble in hot concentrated nitric acid.

E. Water Filtration Studies

1. Rapid Mix Studies

Studies were conducted to evaluate the effect of rapid mix intensity on system performance of the 200 W Area water treatment pilot plant. Tests were run at mixer propeller speeds from 170 to 500 rpm. Initial results obtained are presented in the following table.

<u>Mixer rpm</u>	<u>Filtration Time, (min)</u>	<u>Effluent Turbidity, (JTU)</u>	<u>Total Headloss, ft - H₂O</u>
170	300	5.5	3.5
340	300	5.0	3.7
500	270	2.8	7.1

2. Plant Filter Media Tests

A vacuum system was assembled and tested successfully for in-place sampling of the reactor water treatment plant filter beds. Initial test samples were taken from the west half of filters No. 5 and No. 10 at K West. Following are the data from these samples:

<u>Mesh Size (mm)</u>	<u>Weight Percent Less Than Mesh Size</u>			
	<u>Filter #5</u>		<u>Filter #10</u>	
	<u>Anthracite</u>	<u>Sand</u>	<u>Anthracite</u>	<u>Sand</u>
0.35	--	1.5	--	0.26
0.50	0.02	17.1	--	12.0
0.60	0.09	45.2	0.08	22.3
0.71	1.3	94.9	0.7	68.0
0.84	3.1	100.0	1.9	91.5
1.00	15.1		8.8	100.0
1.41	51.7		45.2	
1.68	67.2		65.3	
2.00	81.3		86.5	
2.38	88.3		96.2	
3.36	100.0		100.0	

DECLASSIFIED

DUN-4847

-9-

<u>Filter</u>	<u>Effective Size</u>		<u>Uniformity Coefficient</u>	
	<u>Sand</u>	<u>Anthracite</u>	<u>Sand</u>	<u>Anthracite</u>
5	0.44	0.98	1.50	1.54
10	0.49	1.02	1.39	1.54

The filter material from filter #10 was placed in a glass column with 24 inches anthracite and 6 inches sand to study the effect of temperature on bed expansion during backwashing. Following are the results from these experiments.

<u>Temperature,</u> <u>°C</u>	<u>Backwash Rate,</u> <u>gpm/ft²</u>	<u>Bed Expansion,</u> <u>%</u>
6	12	8.7
	14	12.3
	16	16.0
	18	19.7
	20	23.3
10	12	7.0
	14	10.7
	16	14.3
	18	18.0
	20	21.7
15	12	5.0
	14	8.7
	16	12.3
	18	15.7
	20	19.3
20	12	3.3
	14	6.7
	16	10.3
	18	13.7
	20	17.3

Samples were recently taken from all of the KW filter beds and size separation work was started.

F. 100-B Area Trench Disposal Test

The B Area trench was supplied from the 107-C West retention basin. Late in April the effluent was diverted to the 107-C East basin, and the test was shut down. It remained down for the remainder of the report period.

DECLASSIFIED

DECLASSIFIED

DUN-4847

-10-

G. Water Analyses

Following the shutdown of B Reactor, the analysis of Columbia River water was terminated for about a month. Sampling was then initiated at C Reactor on a less frequent basis. The analyses for phosphate, iron, silica, sulfate, and zinc are shown in Table II.

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

RGG:rn

DECLASSIFIED

TABLE I

WATER PLANT OPERATING CONDITIONS

Alum Feed Rate Changes, ppm

Initially all reactors variable from 3 to 14.

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

pH Changes

Initially all reactors 7.0

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

Dichromate Changes, ppm

Initially all reactors 1.8

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

DECLASSIFIED

DUN-4847

-12-

TABLE II

WATER ANALYSIS

183 C RAW WATER

	<u>Fe</u> <u>ppb</u>	<u>PO₄</u> <u>ppb</u>	<u>SO₄</u> <u>ppm</u>	<u>SiO₂</u> <u>ppm</u>	<u>Zn</u> <u>ppb</u>
2-29-68	196	154	19.2	7.3	-
3-11	144	147	27.8	4.5	210
12	182	180	19.8	6.8	190
13	205	418	20.8	6.7	190
14	165	220	19.6	6.8	140
20	125	127	22.0	8.3	56
28	268	190	21.0	9.9	140
4- 6	173	177	23.0	8.4	140
7	178	159	21.2	11.0	130
8	194	190	20.3	7.8	170
11	140	164	25.0	9.0	140
13	82	146	76.5	7.6	75
15	114	170	22.2	7.0	67
18	190	< 10	64.0	2.5	70
19	184	110	31.0	8.2	140
22	195	90	22.8	7.7	130
23	174	85	22.8	8.7	124
5- 3	103	91	22.0	6.1	85
4	70	72	23.7	5.4	76
5	78	143	23.7	5.4	84
6	100	104	34.7	4.9	95
8	170	56.2	22.7	5.1	96
10	124	< 10	38.5	7.4	3
11	170	30	22.2	9.5	15
12	116	27.5	22.2	10.4	4
14	36	< 10	26.8	10.1	5
16	50	< 10	19.0	5.8	5
17	45	33	18.8	5.0	45
20	36	25	19.6	4.8	47
21	196	83	17.6	4.8	92
23	92	< 10	19.7	4.8	91
31	164	14	18.8	4.0	92
6- 1	84	25	17.5	3.7	119
2	79	127	23.8	4.4	111
4	67	17	17.2	3.7	95
5	164	62	19.8	6.0	120
6	200	21	16.8	6.0	80
9	164	20	18.8	5.8	34
10	344	23	18.8	5.8	27
13	140	33	17.9	5.5	
14	186	25	15.0	5.2	
15	172	20	14.8	4.8	
28	171	99	16.0	3.7	
7- 1	204	102	23.6	4.3	
8	66	243	15.2	5.2	28

DECLASSIFIED

UNCLASSIFIED

DUN-4847

-13-

FIGURE I

RADIOACTIVITY IN REACTOR EFFLUENT

[REDACTED]

DECLASSIFIED

DUN-4847
-14-

FIGURE II

RADIOACTIVITY IN REACTOR EFFLUENT

NEPTUNIUM-239

CHROMIUM-51

DECLASSIFIED

[REDACTED]

AIM

Association for Information and Image Management

1100 Wayne Avenue, Suite 1100
Silver Spring, Maryland 20910
301/587-8202

Centimeter

Inches

MANUFACTURED TO AIM STANDARDS
BY APPLIED IMAGE, INC.

DATE

FILMED

12/22/94

END