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TITLE

INTERIM REPORT I
PRODUCTION TEST IP-728
HALF-PLANT SODIUM SILICATE TEST

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INTERIM REPORT I
PRODUCTION TEST IP-728
HALF-PLANT SODIUM SILICATE TEST

By Authority of DS Lewis
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Research and Engineering Section
Irradiation Processing Department

September 16, 1965

HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

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INTERIM REPORT I
PRODUCTION TEST IP-728
HALF-PLANT SODIUM SILICATE TEST

INTRODUCTION

The radionuclides in reactor effluent are produced almost exclusively from the irradiation of parent elements which are adsorbed on the film which forms on the in-reactor surfaces. The nuclides are subsequently desorbed into the effluent water. One of the areas of activity in the research and development program directed toward reducing reactor effluent radioactivity has been to find some means of minimizing the adsorption of parent materials and/or desorption of radioisotopes. In the course of these investigations, it was shown in laboratory coupon tests that sodium silicate added to process water reduced the adsorption of arsenic by a factor of five.¹ A single-tube in-reactor test was run using a silicate addition of 20 ppm SiO_2 to give some indication of the effect of flux and other reactor variables on the behavior of the silicate. The results from the test showed that the concentration of P^{32} , As^{76} , and Np^{239} in the effluent were reduced by a factor of two to three. Lesser reductions were obtained for other isotopes, and the Na^{24} concentration increased by a factor of two.²

The cost of feeding sodium silicate at 20 ppm SiO_2 to the coolant for the five Irradiation Processing Department reactors is estimated to be \$2,000,000 per year (sodium silicate cost of \$0.01 per pound of 28 per cent SiO_2 material). Although such a cost is believed to be prohibitively high, it was considered necessary to confirm the reductions in effluent radioactivity on a larger scale as well as to investigate the effects of silicate on filming and corrosion. To accomplish the above, a half-plant test was initiated at D Reactor³.

This report summarizes the data obtained and observations made during the six months the test has been in operation.

SUMMARY

Effluent Activity

Results from both the routine effluent monitoring and from the analyses of samples supplied to Battelle-Northwest personnel indicate that reductions in effluent

1. R. W. Perkins and D. E. Robertson. The Effect of Chemical Additives and Coating Materials on the Adsorption of Radionuclide Parent Elements on Aluminum Surfaces, HW-75976. December 26, 1963.
2. D. E. Robertson and R. W. Perkins. Reduction of Effluent Water Radionuclides by the Addition of Sodium Silicate to Process Water, HW-75782. December 10, 1962.
3. R. G. Geier. Production Test IP-728, Half-Plant Sodium Silicate Test, RL-REA-50. October 26, 1964.

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radioactivity at least as high as predicted from the single-tube tests are being achieved, and for some isotopes the reductions are greater than predicted. In some instances, notably P^{32} , the improvement was slow in coming. Whether this is due to an equilibration period or changing river conditions cannot be stated at this time.

Filming

It was found that during the period February through April (the months of cold inlet water), film formation on fringe columns loaded with thoria target elements caused Panellit pressure increases averaging 0.5 psig per day. Although this is not sufficient to abrogate the early detection of a tube operating abnormally, it adds complexity to operation of the reactor and the interpretation of data.

The phenomenon was alleviated when the inlet water temperature increased to 10 C.

Rear Face Exposure

An unexpected occurrence has been the consistently higher reactor rear face radiation levels on the test side. The levels have been from two to threefold higher. This has resulted in increased exposure to personnel and is regarded as adding to the cost of the present test and a serious problem if additional silicate usage is attempted.

The high radiation levels appear to be due to La^{140} , and an investigation of the source is in progress.

Corrosion

Preliminary assessment of corrosion data from coupons and corrosion probes indicate no deleterious effects on the uniform corrosion of X8001 and 1100 aluminum and carbon steel. In-reactor tube wall thickness measurements have not indicated an effect of the silicate, but a difference would not be seen at this time unless it was one of manyfold. More data are required to define precisely the effect of silicate on corrosion.

DISCUSSION

A. General

1. Water Treatment

The half-reactor sodium silicate test was initiated at D Reactor on January 15, 1965. The silicate concentration was increased gradually, and test conditions of 20 ppm SiO_2 were established on January 25. The sodium silicate was added as a dilute solution to the lines entering the far side 190 Tanks. Additional acid was added to the far side 190 Tanks to compensate for the basicity of the sodium silicate. During shutdowns silicate additions were continued but the concentrations were reduced to 5 to 10 ppm SiO_2 .

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The sodium silicate used was "N" grade and contained a SiO_2 to Na_2O weight ratio of 3.20. It was obtained from the Philadelphia Quartz Company.

Throughout the test period other water treatment parameters have remained constant and common to both sides of the reactor as follows:

Alum feed rate - 18 ppm
Sodium dichromate feed rate - 1.0 ppm
Coolant pH - 6.6

2. Reactor Loading

The reactor was charged with varying quantities of thorium target elements during the test period. Inasmuch as the filming, to be discussed later, appeared to occur particularly on the target columns, the reactor loadings are described below:

- a. Fringe loadings of thorium targets were in place in accordance with PITA-31⁴ prior to the start of the test and have remained throughout.
- b. The central zone was charged with thorium and supporting E-metal columns during the outage beginning February 18, in accordance with PITA-34⁵. The thorium columns were discharged and replaced with a second charge during the outage beginning April 1. The second thorium charge and the E-metal were discharged during the outage beginning May 18, and the central zone was returned to normal operation.

B. Effluent Radioactivity Results

Samples of reactor effluent are taken every weekday during reactor operation from each side of the reactor for routine monitoring purposes. Normally the samples are combined and one analysis made. During the test period, separate analyses were made for P^{32} and As^{76} . The results are shown below:

4. P. D. Gross and K. L. Hladek. PITA-31, Fringe-Blanket Irradiation of Thorium Oxide, HW-84021. September 11, 1964. (Secret)
5. P. D. Gross and K. L. Hladek. PITA-IP-34, Central Zone Irradiation of Thorium Oxide, Smaller Reactors, RL-REA-488. January 20, 1965. (Secret)

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	Phosphorus-32		Arsenic-76	
	pc/ml		pc/ml	
	<u>Test</u>	<u>Control</u>	<u>Test</u>	<u>Control</u>
August	1.2	2.7	20	80
July	2.0	4.6	23	91
June	5.9	7.7	28	74
May	7.9	13.4	56	165
April	8.6	11.6	23	86
March	10.1	10.7	30	56

It appears as though a lengthy equilibration period is required before improvement in the effluent P³² concentration begins. Erratic daily results were also obtained during the early months. However, during July and August, the silicate produced greater than a factor of two reduction. Over the period March through August, the P³² concentrations were an average factor of 1.7 lower on the test side. The As⁷⁶ concentration appeared to respond sooner and more consistently to the silicate. However, here again continual improvement is evident with the test side being a factor of four lower during July and August. The As⁷⁶ concentration averaged a factor of 3.2 lower from March through August. Insufficient data are available to say whether the early results were actually an equilibration period or the fact that silicate is more effective during certain seasonally dictated water conditions.

In addition to the analyses of the routine monitoring samples, analyses were also carried out on samples supplied to Battelle-Northwest personnel. A preliminary evaluation of the results showed that during July the test side concentrations of As⁷⁶ and Np²³⁹ were about a factor of five lower, Sb¹²⁴ were about a factor of four lower, and P³² were about a factor of three lower. No reductions were obtained for Zn⁶⁵, Mn⁵⁶, and Cr⁵¹. The anticipated increase in Na²⁴ concentration by a factor of two was found⁶. These results for P³² and As⁷⁶ are quite comparable to the July and August routine monitoring results.

An over-all assessment of the effluent results obtained to date indicates that the sodium silicate addition is reducing the test side effluent activity at least to the extent forecast by the single-tube tests, and in the case of some isotopes even greater reductions are being obtained.

C. Filming

As indicated above, both the reactor core and fringe were being used for thorium irradiations during the initial portion of the test. Throughout February and

6. Private communication, D. E. Robertson, BNW, August 6, 1965.

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March the Panellit pressures on the control side tubes were normal. The following observations were noted on the test side during the same period:⁷

1. Film buildup on the fringe thoria columns (operating with a ΔT of 5 C) was sufficient to cause an average 0.5 psig per day increase in the Panellit pressure for each column.
2. Film buildup on the central zone thoria columns (operating at a ΔT of 10 C) was sufficient to cause an average 0.2 psig per day increase in Panellit pressure for each column.
3. Negligible film buildup was observed on the tubes charged with uranium.
4. The film buildup on fringe tubes 0173 and 4673 (not included in the average given above) was sufficient to cause an increase in Panellit pressure 1.2 psig per day per tube. The columns were discharged on March 7, after total Panellit pressure increase of 51 psig had been reached. The discharged target elements were shiny and gave no visual indication of being heavily filmed. The Panellit pressure increase following recharging averaged 2.3 psig per day, and the tubes were discharged again on March 25.
5. Purging was not always an effective method of removing the film.

The filming continued in a manner similar to that described through April. In May the rate of Panellit pressure increases had dropped substantially and by mid-June slight decreases in Panellit pressures were being observed. The decreases, however, have not nearly compensated for the pressure increase which had already been accumulated.⁸

A possible explanation for the filming phenomenon is the temperature of the inlet water. The filming took place during the cold water period and appeared to alleviate as soon as the inlet temperature warmed up. Such an explanation would indicate a change in solution viscosity, possibly the precipitation of colloidal SiO_2 polymer. It is apparent, however, that whatever change took place occurred in a very narrow temperature range.

Panellit pressures are a fundamental part of the reactor safety system as well as providing indications of ruptures on fuel and target element

7. Letter, D. L. Hirschel to J. T. Baker, "Filming Observations - D Reactor," April 1, 1965.

8. Private communication, D. L. Hirschel.

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swelling. The rates of film formation observed in the early portion of the test were not great enough to abrogate the early detection of an abnormal tube. However, complications such as those introduced by the film formation trend make accurate interpretations of operational data more difficult.

D. Rear Face Radiation Levels

It has been apparent from the start of the test that the radiation levels on the test side rear face have been higher than on the control side rear face. This unexpected result has increased personnel exposure. The difference in radiation levels can be seen clearly from Figures 1 and 2, which show the rear face survey maps for the outages of March 25 and April 1. Both maps show that the radiation level is between a factor of two and three higher on the test side of the reactor. This is true at about eight hours after shutdown as well as at about twenty hours after shutdown. Thus, although nearly a factor of two decay occurs in twelve to fifteen hours, both sides appear to be decaying at about the same rate.

Gamma scans on rear nozzle Van Stone flange inserts indicate about the same quantities of most of the usual effluent isotopes, e.g., Na^{24} , Cr^{51} , Mn^{56} , etc., on both sides of the reactor. A slightly higher amount of Np^{239} and a significantly higher amount of La^{140} are present on the test side⁹. It would appear that the La^{140} is the source of the increased radiation level. Investigations into the source of the isotope and remedial measures are underway.

It has been estimated that 50 per cent or more of the exposure received by personnel in the Irradiation Processing Department is the result of rear face work. Thus, an increase in rear face dose rates is regarded as a very real problem because it is translatable into increased manpower with the attendant additional costs.

E. Corrosion

In order to monitor the effects of the silicate on the corrosion of reactor components' coupons, corrosometer probes, weighed fuel, and in-reactor tube wall thickness measurements have been used. The status of the corrosion monitoring is as follows:

1. Coupons

Coupon holders containing coupons of 1100 and X8001 aluminum as well as carbon steel were exposed in the downstream end of process tubes on both

9. Private communication, D. E. Robertson, BNW, August 10, 1965.

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the test and control side of the reactor. Weight loss measurements indicate about the same weight loss for the X8001 and carbon steel on both sides of the reactor, although the carbon steel on the test side showed evidences of pitting attack. The weight loss of the 1100 aluminum was less in the silicate treated water.

2. Corrosometer Probes

Carbon steel corrosion probes installed in the inlet water to D Reactor indicated no difference in uniform corrosion rate between the test and the control side. Recent measurements indicate that pitting is occurring on the test side.

Corrosion probes of both X8001 aluminum and 1020 carbon steel were exposed in hot reactor effluent. Neither the aluminum nor the carbon steel showed a difference in uniform corrosion between the two sides of the reactor. However, it appears that pitting is occurring on the carbon steel in the silicate treated water¹⁰.

3. Fuel

Twenty columns of weighed fuel, ten from each side of the reactor, were irradiated for a metal cycle, discharged, and sent to the metal examination facility for visual examination and weighing. The results of these tests are not available at this time.

4. In-Reactor Tube Wall Thickness Measurements

In-reactor tube wall thickness measurements have been made periodically during the course of the test. To date, no differences have been observed between the two sides of the reactor¹¹. However, the normal uniform corrosion rate of tubes in pH 6.6 coolant is low and considering measurement accuracy, differences would not be apparent in six months unless they were differences of manyfold.

While it is still too soon to state precisely the effect of the silicate on the corrosion of reactor components, the successful operation for six months and data obtained to date do not indicate problems of a catastrophic nature.

Specialist, Effluent Studies
Process and Reactor Development

RG Geier:nmd

10. Letter, "Corrosion of Carbon Steel Probes in Silicate," R. B. Richman to R. G. Geier, dated August 6, 1964.
11. Private communication, D. K. George.

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FIGURE 1RADIATION SURVEY OF REAR REACTOR FACE

All readings taken from the rear railing with CP window closed.
Reading mR per hour.

Reactor Down 3/25/65 0030 Hr

Second Survey* 3/25/65 0030 Hr

First Survey 3/25/65 0915 Hr

	<u>Test</u>			<u>Control</u>		
	<u>4286</u>	<u>4279</u>	<u>4273</u>	<u>4267</u>	<u>4260</u>	
		180 (100)		90 (60)		
<u>3692</u>	<u>3686</u>	<u>3679</u>	<u>3673</u>	<u>3667</u>	<u>3660</u>	<u>3654</u>
200 (100)		380 (200)	250 (150)	150 (100)		50 (50)
<u>2992</u>	<u>2986</u>	<u>2979</u>	<u>2973</u>	<u>2967</u>	<u>2960</u>	<u>2954</u>
<u>2392</u>	<u>2386</u>	<u>2379</u>	<u>2373</u>	<u>2367</u>	<u>2360</u>	<u>2354</u>
270 (150)		450 (250)	300 (200)	200 (150)		70 (50)
<u>1792</u>	<u>1786</u>	<u>1779</u>	<u>1773</u>	<u>1767</u>	<u>1760</u>	<u>1754</u>
			300 (200)			
<u>1092</u>	<u>1086</u>	<u>1079</u>	<u>1073</u>	<u>1067</u>	<u>1060</u>	<u>1054</u>
200 (130)		370 (230)		180 (130)		70 (50)
	<u>0486</u>	<u>0479</u>	<u>0473</u>	<u>0467</u>	<u>0460</u>	
	160 (100)		150 (100)		80 (60)	

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FIGURE 2
RADIATION SURVEY OF REAR REACTOR FACE

All readings taken from the rear railing with CP window closed.
Readings in mR per hour.

Reactor Down 4/1/65 1459 Hr

Second Survey* 4/2/65 0900 Hr

First Survey 4/1/65 2130 Hr

← Test			✱	Control →		
	<u>4286</u>	<u>4279</u>	<u>4273</u>	<u>4267</u>	<u>4260</u>	
		200 (100)		100 (50)		
<u>3692</u>	<u>3686</u>	<u>3679</u>	<u>3673</u>	<u>3667</u>	<u>3660</u>	<u>3654</u>
200 (130)		400 (230)	280 (180)	150 (120)		60 (35)
<u>2992</u>	<u>2986</u>	<u>2979</u>	<u>2973</u>	<u>2967</u>	<u>2960</u>	<u>2954</u>
<u>2392</u>	<u>2386</u>	<u>2379</u>	<u>2373</u>	<u>2367</u>	<u>2360</u>	<u>2354</u>
300 (170)		600 (280)	380 (210)	220 (130)		80 (40)
<u>1792</u>	<u>1786</u>	<u>1779</u>	<u>1773</u>	<u>1767</u>	<u>1760</u>	<u>1754</u>
<u>1092</u>	<u>1086</u>	<u>1079</u>	<u>1073</u>	<u>1067</u>	<u>1060</u>	<u>1054</u>
220 (120)		400 (230)		200 (120)		80 (50)
	<u>0486</u>	<u>0479</u>	<u>0473</u>	<u>0467</u>	<u>0460</u>	
	160 (90)		150 (90)		80 (45)	

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