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STATUS REPORT: PRODUCTION REACTOR PROCESS TUB FILM
COMPOSITION AND RADIONUCLIDE INVENTORY STUDIES

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STATUS REPORT: PRODUCTION REACTOR PROCESS TUBE FILM
COMPOSITION AND RADIONUCLIDE INVENTORY STUDIES

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HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

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STATUS REPORT: PRODUCTION REACTOR PROCESS TUBE FILM:
COMPOSITION AND RADIONUCLIDE INVENTORY STUDIES

INTRODUCTION

The discharge of radioactivity from Hanford production reactors to the Columbia River has been studied for a number of years. The primary purposes of these studies have been to learn of the factors influencing production reactor effluent radioactivity concentrations and to develop methods for lowering the discharge concentrations of these radionuclides. In 1962 the Coolant Systems Development Operation (CSDO) began work on this program to determine the in-reactor film compositions and the inventory of radionuclides held in these films with the long-term objective of learning more about the complex in-reactor retention reactions that influence radionuclide effluent concentrations.

This report gives the present status of the work that CSDO is doing on present reactor process tubes.

SUMMARY

Aluminum process tubes have been removed from the KE and C-Reactors for film and radioactivity studies. Deposits and film from these tubes have been analyzed and pseudo boehmite, $\text{Al}_2\text{O}_3 \cdot 2-3 \text{H}_2\text{O}$, was found to be the major aluminum corrosion product constituent in the downstream section of these tubes. The corrosion product films from the upstream and central zone regions have not been fully identified. Iron in the films was found to increase by a factor of 4-5 from the inlet to the outlet of the C Reactor process tubes. The identity of the reddish-brown deposits on the surface of the aluminum oxides has not yet been determined. The major long-life radionuclides in the films were P^{32} , Sc^{46} , Cr^{51} , Co^{60} , Fe^{59} , and Zn^{65} .

Further study is needed to better define the composition and the radioactivity in process tube films.

DISCUSSION

Since March of 1962, three production reactor process tubes, one from the KE Reactor and two from the C Reactor, have been removed and used for film composition and radionuclide inventory studies. The first, the tube from the KE Reactor, was used primarily to develop suitable sampling techniques and scope the magnitude of the irradiated sampling problem.

In May 1962 two tubes were removed from C Reactor. Specimens were removed as 1" x 1" arc pieces from the tube segments with an underwater hand shearing device. Some difficulty was experienced with film spalling during this operation, especially on the downstream pieces. The specimens were processed in either an iodine-methanol apparatus to obtain the film or in a 0.9 M H_2SO_4 -0.3 M oxalic acid mixture to obtain dissolved film solutions. Samples of both types were used for composition and inventory studies.

1. Film Composition

Two different analytical techniques were employed in determining the composition of the deposits and the film on aluminum process tubes: infrared transmission spectra and standard wet chemical methods. Infrared analyses were made on the solid material remaining after treatment with a solution of iodine in methanol. The infrared data from the C Reactor specimens given in Table I show that the process tube film composition is different in the influent region from that at the effluent end of the tube.

TABLE I

Compounds in C Reactor Process Tube Films

<u>Tube 1283</u>	<u>(Rear Face)</u>	<u>Tube 2667</u>
Pseudo Boehmite	1'	Pseudo Boehmite
Pseudo Boehmite	3.1'	Pseudo Boehmite + unknown
Pseudo Boehmite + Boehmite	5.5'	Boehmite
Pseudo Boehmite + Boehmite	8.1'	Pseudo Boehmite
Pseudo Boehmite	11.0'	Unknown
Pseudo Boehmite + unknown	14.1'	Unknown
Pseudo Boehmite + Bayerite	17.5'	Pseudo Boehmite + unknown
Unknown	21.1'	Unknown
Pseudo Boehmite	25.0'	Pseudo Boehmite + unknown
Unknown	29.1'	Pseudo Boehmite + unknown
Unknown	34.0'	-
Boehmite + unknown	38.8'	-
	(Front Face)	

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The major constituent in the aluminum oxide near the reactor rear face is pseudo boehmite.* Unknown compounds tend to be predominate near the influent end. Some of the results designated as unknown showed some Bayerite** like infrared spectra but they were not well enough defined to definitely label them as such. Occasionally boehmite*** appears to be an important constituent of these aluminum oxide films. The reddish brown surface deposits on the aluminum oxides have not been characterized by infrared techniques.

Film dissolved in the 0.9 M H_2SO_4 -0.3 M oxalic acid mixture was analyzed by standard wet chemical techniques. Plots of the amount of iron and aluminum per square inch as a function of tube length are given in Figure 1. These data are rough and approximate only. On several occasions during sampling appreciable amounts of the reddish brown surface deposit and some base oxide were lost in the shearing operation. In addition, it was difficult to cut close to 1" x 1" arcs. Figure 1 is useful, however, in showing what appears to be a trend for more iron to be deposited in the downstream portion of the process tubes than is found upstream. The higher effluent than inlet temperatures are expected to cause more aluminum corrosion and consequently thicker oxide films downstream.

2. Radionuclide Inventory

Complete radionuclide loadings per sample are not now available and will probably not be ready for evaluation for two or three months. In general however the following appear to be the major long-lived radionuclide constituents in the process tube films:

P^{32} , Sc^{46} , Cr^{51} , Co^{60} , Fe^{59} and Zn^{65} .

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* Pseudo boehmite is a hydrous aluminum oxide with 2 to 3 waters of hydration, $Al_2O_3 \cdot 2-3 H_2O$.

** Bayerite is an aluminum oxide with 3 waters of hydration. It has a monoclinic crystal structure with two molecules per unit cell and in the American system of nomenclature is $\beta Al_2O_3 \cdot 3H_2O$.

*** Boehmite is an aluminum oxide with 1 water of hydration. It has an orthorhombic crystal structure with two molecules per unit cell. In the American system, it is $\alpha Al_2O_3 \cdot H_2O$.

a. Iron in Reactor Tube Film

b. Aluminum in Reactor Tube Film

FIGURE 1. Iron and Aluminum Content of Reactor Tube Films

3. General Comments

The data given in this report were acquired during the initial phases of the study on process tube film composition and radionuclide inventories. Further work is required to determine the composition of the corrosion product and deposited films. Refinements in sampling and handling techniques will be required before more reliable data can be obtained.

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