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POSTIRRADIATION ANALYSIS OF HIGH-TEMPERATURE, LOW-BURNUP TUNGSTEN - URANIUM DIOXIDE TEST PLATES

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16. Abstract

Problems in determining fuel burnup of high temperature tungsten - uranium dioxide fuel plates were resolved by using tungsten-184 as a thermal flux monitor. Methods for determining fission product releases from fueled test plates were evaluated. The use of fission product cesium-137 as a burnup monitor was also evaluated

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SUMMARY

The objective of these irradiations was to evaluate the uranium retention and iodine and xenon release qualities of test fuel plates. The plates contained uranium oxide dispersions which were fission-heated to anticipated rocket fuel element operating temperatures and neutron flux levels.

We found the following problems when we tried to use conventional methods for determining fission product releases and fuel burnup. Fission product recovery by leaching methods was unreliable. Burnup values obtained from the U^{236}/U^{235} uranium ratio change were in many cases so small that results by mass spectrometry analysis were inaccurate. The high temperature of the irradiations precluded the use of standard thermal flux dosimeters.

Problems in measuring the fuel burnup were resolved by using tungsten-184 in the cladding of the test plates as a thermal neutron dosimeter. This measure of burnup then provided a sound basis for judging the effectiveness of fission product recovery and for determining the fission product release.

Cesium-137 and iodine-131 were the fission products of interest. The fueled test plates were dissolved and capsule components leached for complete accountability of these fission products.

Based on the number of fissions calculated from the tungsten-185 activity and from the mass spectrometry analysis, only 70 percent of the cesium-137 produced was recovered by the leach methods. The analysis of the fuel plates for iodine-131 indicated that approximately 99 percent of the iodine-131 produced had left the plates, whereas the sodium hydroxide leach method indicated that only 20 percent of the iodine-131 produced had left the plates. The failure to recover all of the cesium-137 and iodine-131 can be based on two reasons, both unique to this type of irradiation:

(1) The leaching and dissolution methods employed were not efficient because of the types of iodine and cesium compounds formed at the high temperatures.

(2) The cesium-137 and iodine-131 as they deposited on the cooler interior portions of the capsule wall were covered by a layer of tungsten metal which had vaporized from the fuel test plates. Some test plates lost as much as 38 percent of their original weight including fuel.

INTRODUCTION

The irradiations discussed in this report were part of a program to demonstrate the feasibility of refractory-metal-clad fuel elements for nuclear rocket application. The report does not consider any aspects of the overall program or the programmatic results of the irradiations described here. Rather, we will talk about the methods we used and tried to use to measure fuel burnup and fission product release.

The total irradiation program consisted of approximately 40 capsule irradiations. All of these capsules were processed using the methods described in this report for determining fission product release and fuel burnup. The data on fission product release from these irradiations were quite scattered. To determine the cause of the data scatter we initiated the work described in this report.

In addition to the tests scheduled for the program, we irradiated four fueled plates to evaluate our methods for fission product recovery and for uranium burnup. This report treats only the work on these four irradiations. We will discuss

(1) The use of tungsten-184 as a thermal flux dosimeter in high-temperature irradiations to determine the total fissions which occurred in fueled test plates.

(2) The use of fission-product cesium-137 recovered by radiochemical methods as a measure of total fissions which had occurred.

(3) The efficiency of various leaching methods used remotely to recover fission product iodine, cesium, and fission gases.

The development work related to establishing tungsten-184 as a thermal neutron dosimeter is described in references 1 and 2.

EXPERIMENTAL DETAILS

The fueled test plates were 2.54 by 2.54 by 0.127 centimeter thick. The fuel was enriched uranium oxide in a matrix of metallic tungsten. The fuel content and homogenity of the test plates were checked and verified by chemical analyses. The experimenter supplied the fuel loading information. All test plates were encased in a picture frame of tungsten and clad with 0.013 centimeter of tungsten. Each fueled test plate was surrounded by a thermal radiation shield and suspended inside an evacuated stainless steel capsule (see fig. 1). Each of the four capsules was irradiated separately in the HT-1 test hole of the Plum Brook Reactor. The actual thermal neutron fluxes which the test plates saw varied from 10^{13} to 10^{14} neutrons per square centimeter per second.

Capsules A and B were longer irradiations (>12 hr) so that fuel burnup could be determined from the change in the U^{236}/U^{235} ratio. The temperatures for capsules A and B ranged from 2600 to 3000 K. From this the thermal flux could be determined and

compared with thermal flux values calculated from tungsten-185 activity. Capsules C and D were irradiated for 4 hours at 3300 K.

Capsules A and B, irradiated for burnup analysis, were stored for about 2 months to allow short-lived fission products to decay. Capsules C and D were processed in the hot cell within 24 hours after irradiation using the puncture rig described in reference 3. Gas samples were collected and analyzed for xenon-135 to determine the iodine-135 release. Both capsules were leached with 0.1 normal sodium hydroxide (NaOH) solution to obtain the release of iodine-135 from the fuel.

After a decay period of approximately 2 months, we cut open all four capsules remotely in the hot cell and removed each component. We leached or dissolved the inside wall of each capsule with aqua regia in a glove box located in the cell. The exhaust from the glove box was passed through liquid-nitrogen-cooled carbon traps and then into a radioactive gas collection system to collect iodine-131.

We partially dissolved the molybdenum-tungsten thermal radiation shields from each capsule with a mixture of hydrofluoric acid and nitric acid. This operation was also performed in the glove box connected to the radiation gas collection system.

Each leach solution was analyzed separately for cesium-137, iodine-131, and uranium. The carbon traps and radioactive gas collection tanks were sampled for iodine-131.

We weighed each fuel specimen after irradiation. Then we removed the corners of the picture frame cladding for tungsten-185 analysis.

We dissolved the tungsten corners in a mixture of hydrofluoric acid and nitric acid $(HF)(HNO_3)$, and then determined the tungsten-185 activity for each fuel plate.

Each specimen was then dissolved entirely in a mixture consisting of two parts phosphoric acid (H_3PO_4) and one part perchloric acid ($HClO_4$). The dissolution was performed in a reflux condenser connected to a sodium hydroxide scrubber trap and a carbon trap to retain any iodine. The resulting solutions were analyzed by radiochemical methods for cesium-137 and iodine-131. Total uranium was determined colorimetrically.

Mass spectrometric analyses for uranium burnup were performed on fuel specimens A, B, and C using the change in the U^{236}/U^{235} ratio.

DISCUSSION OF RESULTS

Burnup Analysis

Cesium recovery. - One object of this study was to recover all of the cesium-137 produced during irradiation. If complete recovery is possible, then the number of fissions and hence, the burnup, can be calculated. The total cesium-137 found and the percentage of recovery for each irradiation are listed in table I. These recoveries are

TABLE I. - CESIUM-137 FOUND FROM LEACHING OF CAPSULES

	Capsule A	Capsule B	Capsule C	Capsule D
Cs^{137} total found, disintegrations/min	--------	8.9×10^9	4.5×10^9	4.3×10^9
Cs^{137} produced (calculated from W-185 activity), disintegrations/min	10.0×10^9	12.2×10^9	6.6×10^9	5.7×10^9
Cs^{137}, percent found of total produced	(a)	73	68	76
Cs^{137}, percent in fuel specimen of total produced		4	6	7
Cs^{137}, percent in baffle leach of total produced		39	15	19
Cs^{137}, percent in capsule acid leach of total produced		30	11	15
Cs^{137}, percent in 0.1N NaOH leach of total produced	Not leached	Not leached	36	35

[a]Solution lost.

based on fissions calculated from the tungsten-185 activities. None of the recoveries for cesium-137 exceeded 76 percent.

Most of the cesium-137 was found in the 0.1 normal sodium hydroxide solutions of the two capsules (C and D) which were caustic leached. In the other two capsules most of the cesium-137 was found in the leaches of the thermal radiation shields. As can be seen from table I, the fuel specimens contained less cesium-137 than the leaches. This means that during irradiation most of the cesium-137 escaped from the fuel.

The fact that cesium-137 escapes from the fuel tends to rule out its use as a method for determining the total fissions occurring in a specimen irradiation at high temperature. The partial dissolution and acid leaching of the components was quite complicated with conditions difficult to control. Total dissolution of the capsule or thermal radiation shields was out of the question because of the large amount of metal ions in solution. The metal ions interfere with the cesium recovery.

There are two plausible explanations for incomplete recovery of both the cesium-137 activity and the iodine-131 activity (discussed in a later section). Both reasons are unique to these irradiations. Our leaching methods may have been ineffective due to the types of cesium and iodine compounds formed at the high temperatures of the irradiations. Or at these high temperatures, the cesium and iodine left the fuel and deposited on the cooler interior surfaces of the capsule. These fission products were then covered by a layer of tungsten metal which vaporized from the fueled test plate and deposited on the same cooler surfaces. We found evidence to support this latter mechanism. Some of the test plates lost as much as 38 percent of their original weight, including fuel. Analyses of the irradiated fuel plates showed fuel loss from the test plates was 33 percent for capsule A,

61 percent for capsule B, 73 percent for capsule C, and 83 percent for capsule D. At the high irradiation temperatures, it is reasonable to assume the capsule contained a gaseous mixture composed of volatile fission products, including cesium and iodine, and vapors of tungsten and uranium dioxide. Hence either explanation is reasonable.

Flux determined by tungsten-185 activity. - We dissolved the tungsten corners of the specimens and determined the tungsten-185 activities. Using an effective cross-section of 1.94 barns (verified by cobalt-60 comparator in the Plum Brook Reactor and the Mock-Up Reactor) we calculated the integrated flux received by the tungsten metal. By using these data and the weight of uranium originally loaded, we calculated the total fissions for each fuel specimen. Table II lists the integrated flux from the tungsten-185, total

TABLE II. - COMPARATIVE TOTAL FISSIONS

	Capsule A	Capsule B	Capsule C	Capsule D
Neutron levels calculated from tungsten-185 activity	4.5×10^{18}	5.3×10^{18}	3.0×10^{18}	2.9×10^{18}
Fissions calculated[a] using tungsten-185 flux	3.5×10^{18}	4.2×10^{18}	2.3×10^{18}	2.0×10^{18}
Fissions using cesium-137 found (line 1, table I)	(b)	3.1×10^{18}	1.7×10^{18}	1.5×10^{18}
Fissions determined[a] by mass spectrometry (burnup analysis)	2.8×10^{18}	4.1×10^{18}	2.0×10^{18}	(c)

[a]Based on fuel loading.
[b]Solution lost.
[c]Burnup not determined.

fissions calculated, the fissions calculated from the cesium-137, and the fissions found by mass spectrometry.

The uranium in fuel specimen D did not experience sufficient burnup to allow a calculation based on the changes in uranium-236 content. The uranium-236 content of this specimen was high and could not be used for reliable burnup analysis.

Data from the analysis of tungsten-185 activity in the cladding was used for all calculations of fissions in this study. These calculated fissions were confirmed by the burnup analysis by mass spectrometry.

TABLE III. - IODINE-131 FOUND FROM LEACHING OF CAPSULES

	Capsule A	Capsule B	Capsule C	Capsule D
I^{131} total found, disintegrations/min	(a)	1.8×10^{12}	1.3×10^{12}	1.5×10^{12}
I^{131} produced (calculated using W-185 flux), disintegrations/min	6.0×10^{12}	7.3×10^{12}	4.0×10^{12}	3.4×10^{12}
I^{131}, percent found of total produced	(a)	24	34	44
I^{131}, percent in fuel specimen of total produced		1	1	1
I^{131}, percent in baffle leach of total produced		1	4	3
I^{131}, percent in capsule leach of total produced		1	1	2
I^{131}, percent in 0.1N NaOH leach of total produced	Not leached	Not leached	20	34
I^{131}, percent in gas collection tanks of total produced	34	22	8	5

[a]Solution lost.

Fission Product Iodine Release

Iodine recovery. - Iodine-131 concentrations and the percentages in each of the leach solutions are shown in table III. Also listed are the percent recoveries for iodine-131 based on the iodine-131 produced. No recovery exceeded 44 percent. Most of the iodine-131 was probably lost during the leaching operations. The loss was due to deposition on the metal surfaces of the pipes leading from the glove box to the radioactive gas collection system, and on the inside surface of the gas tanks.

During the leaching of the capsules and baffles, strong oxidizing acids had to be used to attack the metal to ensure complete release of fission products. In this environment, iodine will become elemental and volatilize. This was evident when capsules and baffles were acid leached on capsules A and B (table III). Most of the iodine-131 found (90 percent) was found in the radioactive gas collection tanks. We found that acid leaching (with aqua regia) liberates large amounts of iodine.

Table III also shows that only a small percentage of iodine produced remained in the test specimens at these high temperatures.

Evaluation of 0.1 normal sodium hydroxide leaching and gas evolution methods. - We were interested in evaluating our two methods for determining the iodine-135 release from fuel specimens during irradiation. Of primary interest, the caustic leach method gave a 15 percent release for iodine-135 and a 20 percent release for iodine-131 on capsule C (table IV). The gas evolution method to determine iodine-135 gave a release of 7 percent for capsule C. The results for capsule D were similar to those for capsule C.

TABLE IV. - IODINE-131 AND IODINE-135 RELEASES BASED ON FISSIONS

CALCULATED FROM TUNGSTEN-185 FLUX VALUES

	Capsule A	Capsule B	Capsule C	Capsule D
I^{131}, percent released with 0.1N NaOH leach	Not leached	Not leached	20	34
I^{131}, percent released considering I^{131} remaining in fuel specimen	98	99	99	100
I^{135}, percent released with 0.1N NaOH leach	--	--	15	28
I^{135}, percent released with gas evolution method	--	--	7	5

By considering the ratio of iodine-131 remaining in the fuel specimens to the iodine-131 produced using the fissions calculated from the tungsten-185 activities, the releases are of the order of 99 percent for all specimens. There was approximately 1 percent iodine retention in the specimen.

CONCLUSIONS

We were able to account for approximately 70 percent of the cesium-137 produced based on the fissions calculated from the tungsten-185 and mass spectrometer burnup analysis. We could not recover all the cesium produced because the leaching methods we employed were not efficient in freeing the deposited fission product cesium-137.

Considering the cesium recovery and the problems in leaching, handling, and analysis of the leach solutions, it is our opinion that the method of determining the number of fissions from cesium-137 produced would be unreliable.

We found that the 0.1 normal sodium hydroxide leach used for iodine recovery in this particular application did not remove all of the iodine isotopes from the capsule interior as we had assumed. After analyzing the fuel specimens for iodine-131 remaining, we found that approximately 99 percent of the iodine-131 had left the fuel specimens. Our 0.1 normal sodium hydroxide leach method indicated that only 20 percent of the iodine-131 produced had left the specimen. In future work to obtain more realistic iodine release data, the fuel specimen should be removed from the capsule and analyzed directly for iodine isotopes.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, September 22, 1969,
120-27.

NASA-Langley, 1969 —— 22 E-5229

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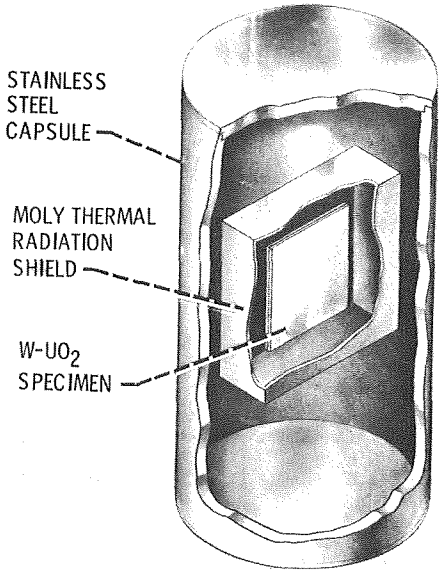

Figure 1. - In-pile capsule.

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