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IODINE AND TELLURIUM RELEASE FROM UC GRAPHITE FUEL

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1.0 INTRODUCTION

An estimation of the quantity of radioiodine released from NERVA fuel, under various conditions, is required because of its potential biological hazard to both on site and off site occupants during normal reactor operations and also in the event of a reactor excursion. The amount of tellurium released is also important because it is the immediate precursor to iodine and much of the iodine activity to be found in test effluents will be the result of tellurium diffusion.

The present method employed to perform radioiodine analysis of this type involves dissolving the fuel specimen in a flask with an attached reflux condenser using fuming nitric and perchloric acids. The flask is heated until condensation forms on the inside wall of the condenser. The reaction is then interrupted and the wall dried by passing air through the system. This process is repeated until the sample is dissolved. Although this method is satisfactory, it becomes prohibitive when processing large numbers of samples since the dissolving time is approximately 48 hours per sample. An alternate method, which involves burning the fuel with pure oxygen, was developed in this laboratory to dissolve the sample in much less time and still retain the iodine.

The results of this experiment show that iodine is released at a faster rate than tellurium at the higher temperatures studied. There was no release observed for either element, however, at 2000°C in vacuum for 20 minutes. It was also found that release for both elements was retarded by increasing pressure.

2.0 EXPERIMENTAL

2.1 MATERIAL

The material used in this experiment was samples of NERVA fuel which is a homogeneous dispersion of pyrocarbon coated UC_2 beads within a graphite matrix. The beads are nominally 100 microns in diameter with a 25 micron coating. The sample loading was 0.4 g/cm^3 of uranium, 93% enrichment as U^{235} . The samples were 1/4 inch cylinders and 1/4 inch in length with a 0.095 inch coolant channel in the center. Each pellet weighed about 0.37 grams. These samples were machined from actual NERVA fuel elements which did not have the NbC lining.

2.2 IRRADIATION

The samples were irradiated at the Westinghouse Reactor Evaluation Center in a 100 watt Reactor for one hour. The irradiation capsule consisted of three graphite holders, 1/2 inch thick and about 3/4 inch diameter, stacked in an aluminum cylinder with a screw cap. The three holders each had three wells, equally spaced and large enough to contain the fuel specimens. Nine samples were irradiated in a flux to produce approximately 10^{10} fissions per pellet.

2.3 POST IRRADIATION TREATMENT

The experiment was designed to observe the release of iodine and tellurium from fuel specimens under different environmental conditions, keeping a constant heating time of 20 minutes. Samples 1, 2 and 3 were to be heated in a vacuum (about 4×10^{-4} mm Hg) at 2000°C, 2200°C, and 2400°C respectively. Samples 4, 5 and 6 were to be heated in helium (about 1

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FIGURE 1

CONTROLLED TEMPERATURE DIFFUSION APPARATUS

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atmosphere pressure) at 2400°C, 2600°C and 2800°C respectively. Samples 7 and 8 were used as control samples and number 9 was retained as a spare.

2.4 HEATING APPARATUS

The post-irradiation heating apparatus is shown in Figure 1. The chamber is a bell jar 18 inches in diameter and 24 inches high. A co-axial column enters the chamber through the base plate and supports the heating coil. The coil is one inch inside diameter and is constructed from 0.3125 inch copper tubing. The samples are supported on a graphite pedestal machined in such a manner that there is a minimum of contact area with the sample surface. The samples are heated in either a helium atmosphere or in a vacuum. A two inch vacuum system including a cold trap and oil diffusion pump serves to evacuate the bell jar system. A 15 KW radio frequency generator supplies power to the heating coil. The generator, heating coil, and transformers are cooled by a water recirculation and heat exchanger unit. Temperature measurements were made with a double wave length Milletron optical pyrometer having a range of 1500 to 3500°C. The sensing head was mounted in a position directly over the bell jar which permitted unobstructed sighting of the sample. A Veeco ionization guage and a thermocouple guage were used to monitor the pressure within the bell jar.

2.5 EXPERIMENTAL PROCEDURE

Each sample was gross gamma counted, at constant geometry, to determine relative count rates. This was done three days after irradiation to permit the very short lived fission product activity to decay out. Since the gross gamma activity of the samples at that time was only about a

Figure 2
 Fuel Combustion Apparatus

factor of five above the initial activity caused by the uranium, a correction for background had to be made. These samples were then heated as shown in Table 1. Gross gamma measurements were made before and after heating each sample to determine the fractional release of total activity. After the heat treatment, the samples were analyzed radiochemically for iodine and tellurium.

2.6 SAMPLE DISSOLUTION

The samples were dissolved in a dry combustion apparatus, as shown in Figure 2. The sample was placed in the ignition chamber and heated externally with a Fisher burner, while a flow of pure oxygen was passing through the system. When the burning became self-sustaining, the Fisher burner was removed and the reaction continued until the sample burned itself out. This took about five minutes. The oxygen flow was stopped and the ignition chamber cooled. The resulting uranium oxide was dissolved in the ignition chamber with 10ml 6N HNO_3 containing 3 drops of 30% H_2O_2 . Gentle heating was applied during the dissolving, while oxygen was again flowing through the system. Ice water was used as a coolant in the cold trap. The liquid trap was composed of 20 ml 3M NaOH and 1 ml 1M NaHSO_3 . When the reaction was finished the solution was cooled and drained from the ignition chamber. The entire system was then washed successively with 10 ml 1N HNO_3 , 10 ml H_2O , the basic solution from the liquid trap, and 10 ml H_2O . The rinsing was done very carefully to insure solution contact with the entire inside surface of the apparatus. The fractions were all combined and diluted to a known volume for radiochemical analysis.

2.7 RADIOCHEMICAL ANALYSIS

The heated samples were analyzed, along with the control samples, for iodine and tellurium using standard radiochemical purification procedures. Since the fresh fission product solutions contained several isotopes of each of the elements of interest, comparative count rates had to be used to determine fractional release instead of absolute disintegration rates. The counting of the samples was delayed 24 hours after separation. This permitted the major portion of the 2.3 hour I-132 to decay out of the iodine samples and grow into the tellurium samples. Counts were taken on later dates to make sure all of the samples, including the controls, were decaying at the same rate. Satisfactory results were obtained.

2.8 CALCULATION OF FRACTIONAL RELEASE

The fractional release was determined by comparing the treated samples with the controls and relating the radiochemically pure samples with the gross gamma activity. The purified samples were all counted in a standard geometry, as were the untreated pellets. This was done within a short period of time so that a decay correction between counts was unnecessary. A ratio of purified activity to gross gamma activity was determined for the control samples. An average value, of this ratio, was then used to establish the initial activity (A_0) in the treated samples. The following form was used.

$$R_I = \frac{I7/\text{gross } 7 + I8/\text{gross } 8}{2}$$

Therefore $IX = (R_I) (\text{Gross } X)$

where: $I7$ = iodine activity in control 7

$I8$ = iodine activity in control 8

Gross 7 - gross gamma activity in control 7

Gross 8 = gross gamma activity in control 8

R_I = average ratio for iodine

Gross X = gross gamma activity in sample X

I_X = initial iodine activity in sample X

The final activity (A_F) was obtained by counting the purified samples, correcting for yield and aliquot. The fraction retained, therefore, is A_F/A_O and the fraction released is $1 - A_F/A_O$. The gross release was determined simply by gamma counting the sample before and after heating, using constant geometry.

3.0 RESULTS

The results are summarized in Table 1 and shown graphically in Figure 3. The graph does not include the data from sample number 6 because of a system malfunction during that run. Air got into the bell jar and caused about 25% weight loss through oxidation. Another attempt was made at 2800°C in He using sample number 9. However, the generator failed after 12 minutes. The data obtained from the 12 minute run is included in Table 1 and Figure 3 along with an estimation, by extrapolation, of fractional release for a 20 minute run at 2800°C in He.

4.0 DISCUSSION

It appeared that quantitative retention of iodine, in the dissolving apparatus, was achieved because of satisfactory agreement between control samples. Also, some preliminary experiments, with the apparatus, showed that a negligible amount of iodine got beyond the cold trap.

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The results of this experiment show that pressure had a definite effect on the release of the elements studied. At 2400°C for 20 minutes, there was 35.8% iodine release in vacuum and only 8.4% in He. The tellurium followed the same pattern, under the same conditions, showing no release in He and 9.1% release in vacuum. The gross release, under the same conditions showed 13.4% in vacuum and 1.5% in He. This will change, however, depending on the age of the sample since the composition changes with time.

The experiment provided only preliminary release data for iodine and tellurium. Future experiments will include the effects of heating as well as sample size, geometry and concentration.

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TABLE 1
 FRACTION OF ACTIVITY RELEASED DURING
 20 MINUTE HEATING PERIODS AT VARIOUS TEMPERATURES

Sample Number	Temp. °C	Time Min.	Environ- ment	Gross Retained	Gross Released	Iodine Retained	Iodine Released	Tellurium Retained	Tellurium Released
1	2000	20	VAC.	.991	.009	1.00	0	1.00	0
2	2200	20	VAC.	.929	.071	.968	.032	1.00	0
3	2400	20	VAC.	.866	.134	.642	.358	.909	.091
4	2400	20	He	.985	.015	.916	.084	1.00	0
5	2600	20	He	.928	.072	.806	.194	.901	.099
6	2800	16	He (air)	.244	.756	.306	.694	.383	.617
9	2800	12	He	.781	.219	.454	.546	.713	.287
9*	2800	20	He	.635	.365	.090	.910	.522	.478

9* an estimation by extrapolating 12 minute data to 20 minutes

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