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CEND-3107-261
Joint U.S. - Euratom
UC-25, Metals, Ceramics
and Materials
TID-4500, 47th Edition

PROGRESS REPORT
BURNABLE POISON ADDITIONS TO UO_2

For Period
October 1 to December 31, 1965

Prepared By:

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BURNABLE POISON ADDITIONS TO UO_2

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FOREWORD

The United States and the European Atomic Energy Community (EURATOM) on May 29 and June 18, 1958, signed an agreement which provides a basis for cooperation in programs for the advancement of the peaceful applications of atomic energy. This agreement, in part, provides for the establishment of a Joint U.S. -Euratom research and development program which is aimed at reactors to be constructed in Europe under the Joint Program.

The work described in this report represents the Joint U.S. -Euratom effort which is in keeping with the spirit of cooperation in contributing to the common good by the sharing of scientific and technical information and minimizing the duplication of effort by the limited pool of technical talent available in Western Europe and the United States.

BURNABLE POISON ADDITIONS TO UO_2

by

R. C. Brayer and S. S. Christopher

ABSTRACT

A 6% enriched UO_2 fuel element, E-1, containing 500 ppm B_4C was fabricated at Combustion Engineering and was irradiated in the Saxton Reactor to a peak burnup of ~ 1070 MWD/MTU at a peak specific power of 17.2 kw/ft. A second rod, E-4, will be reinserted in the Saxton Reactor during February and irradiated to a burnup of approximately 5000 MWD/MTU. Nondestructive evaluation of element E-1 has been completed and no defects or dimensional changes were found. The nondestructive evaluation included optical examination, dimensional analysis, gamma scanning and plenum radiography. Sample locations for the destructive evaluation have been established.

The limits of accuracy and precision of boron analysis have been established for both the 1-1' dianthrimide colorimetric and mannitol titration methods. The effects of such variables as preoxidation, sample size, furnace loading temperature, type of boron compound and effect of contaminants have been determined.

Experimental facilities are being prepared to perform a study on UO_2 - B_4C compatibility.

I. SUMMARY

Two 6% enriched UO_2 fuel elements containing 0.05 wt% B_4C were fabricated using the process described in CEND-3107-238*. The elements were irradiated in the Saxton Reactor between July 12 and August 16, 1965. The burnup indicated for the elements was 1070 MWD/MTU at a peak specific power of 17.2 kw/ft. One element, designated E-1, has been sent to the hot cells for post-irradiation examination; the second element, E-4, will be reinserted into the reactor during February and irradiated to a burnup near 5000 MWD/MTU.

During this report period a nondestructive evaluation which included visual examination, dimensional analysis, gamma scanning and plenum radiography, was conducted on Rod E-1. The visual examination confirmed earlier results indicating that the element surface was clean and free of defects. The dimensional measurements were in good agreement with pre-irradiation values. Plenum radiography to determine fuel movement by measuring expansion markers was unsuccessful due to the poor quality X-ray radiograms obtainable in the hot cell. Additional effort on this phase of the evaluation has been suspended on the basis that this information is not vital for the successful completion of the objectives. Single channel and multi-channel gamma scans were performed at specific locations along the length of the element; the gamma spectra obtained were typical of those anticipated for irradiated fuel and irradiated stainless steel. Destructive evaluation of element E-1 is planned for the next reporting period.

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* Brayer, R.C., and W.P. Chernock, "Burnable Poison Additions to UO_2 ", CEND-3107-238, May 1965.

An evaluation was performed on the pyrohydrolysis-1-1' dianthrimide and mannitol titration techniques for boron analysis. These techniques can be used to obtain boron analysis within $\pm 9\%$ and $\pm 11.2\%$ at a 95% confidence level for the dianthrimide and mannitol techniques. The effect of such variables as sample size, furnace loading temperature, preoxidation, type of boron compound and contamination on pyrohydrolysis separation and subsequent analysis has been performed. It has been found that preoxidation can be used and results in better pulverization of the sample. Due to vapor loss of H_3BO_3 , it was concluded that the furnace loading temperature should be held near $200^\circ C$. Sample size was not found to be a factor in analyzing the fuel mixtures for boron. Samples containing various boron compounds were analyzed and the techniques were satisfactory for all compounds except the $UO_2-UB_2-UB_4$ composition which gave low values of boron. However, this is not attributed to the analytical technique but to the heterogeneity* of the reference UB_2+UB_4 compound mixture used to prepare the standard samples. The effects of lithium on boron analysis was determined and found to have little influence on the accuracy of the determination. The analytical portion of this program is complete and no additional studies will be performed.

A UO_2+B_4C compatibility program has been defined and is based on past studies performed at CEND. This task will determine the reactions taking place between UO_2 and B_4C by performing tests in both open and closed systems and analyzing both solid and gaseous reaction products.

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* It was later determined by x-ray analysis of the UB_2+UB_4 reference compound that a wide variation of boron content and impurities was present and was dependent on the particle analyzed. Thus we feel that the variations obtained on the boron analysis of the standard sample can be attributed to the heterogeneity of the reference compound.

II. POST IRRADIATION EVALUATION

A. Introduction

Two rods, 41 in. long and 0.600 in. diameter, were irradiated in a 2 x 2 assembly in the core of the Saxton Reactor. Each rod contains 6% enriched UO_2 , vibratory compacted with 500 ppm B_4C to a density in excess of 86% of theoretical. The objective of the irradiation is to determine whether gross axial migration of boron occurs during irradiation in an operating fuel element. A secondary objective is to determine whether gross radial migration of boron occurs as a result of the steep radial thermal gradients encountered in an operating element. It was believed that radial migration, if it occurred, could be tolerated, and would have little influence on the future applications of this method of burnable poison addition. However, gross axial migration, if it occurred, could not be tolerated since hot spots would become hotter as a result of local boron depletion.

Non-destructive evaluation is being conducted (according to the flow chart shown in Figure 1) on fuel rod E-1 which was irradiated to a peak burnup of 1070 MWD/MTU at a peak specific power of 17.2 kw/ft. A record of the power history of the Saxton Reactor during the irradiation of this element is shown in Appendix I together with estimates of fuel centerline temperatures at each recorded power level.

The second rod, E-4, will be reinserted in the center of the Saxton Reactor in February and will be irradiated to a burnup of about 5000 MWD/MTU. Comparison of data from rods E-1 and E-4 should provide a reasonable insight into the influence of burnup and time on the potential for boron migration.

FIGURE 1 Post-Irradiation Non-Destructive Testing Scheme for Irradiated Rod E-1

B. Non-Destructive Evaluation

Stereoscopic Examination

Stereoscopic examination of element E-1 at 30X magnification confirmed the results of earlier examinations which revealed that the element surface was clean with no abnormalities. Figure 2 shows a representative surface area on the rod. A slight spotted appearance was noted on the rod in the region of the maximum heat rating; this appearance is typical of the beginning of crud deposition. Two small surface imperfections were found, Figure 3 and 4, at locations four and fifteen inches from the bottom of the element. The scratches and imperfections are probably associated with the spacers used to hold the elements in the 2 x 2 assembly.

The fuel element was cleaned using an alkaline permanganate ammonium citrate solution. Specific attention was paid to this operation since boron shims have been used in Saxton and it was believed necessary to insure that residual surface boron did not influence later results of boron migration analyses. No adherent scale remained on the surface of the element after cleaning. No undissolved scale was found in the resulting solution. The amount of dissolved crud in the cleaning solution was too small for further evaluation.

Dimensional Evaluation

A dimensional evaluation conducted on element E-1 indicated that swelling had not occurred. The length of the element was determined to within 0.001 in. and the diameter measurements in the hot cell were made at both 0° and 90° at six inch intervals along the length of the rod using a Bausch and Lomb Model 25 optical gauge. The results

FIGURE 2
Typical Surface Condition of Fuel Element
E-1

3.3X

FIGURE 3 3.3X
Surface Marking on Fuel Element E-1
Located 15 in. from the bottom of the element

FIGURE 4 3.3X
Surface Marking on Fuel Element E-1
Located 4 in. from the bottom of the element

of the dimensional measurements are listed in Table I and are compared with the pre-irradiated measurements that were performed at CEND. The data are in good agreement and no anomalies were noted.

Gamma Scanning

Element E-1 was gamma scanned using a single channel analyzer for gamma radiation of energies greater than 0.5 Mev. The gamma scan curve for the element is shown in Figure 5. The gamma scan curves indicate a shift of about three inches upward in the position of maximum burnup from that anticipated in Saxton. The gamma scan curve will be used to locate sample positions and evaluate temperature distributions in the fuel. Also shown in the figure is a plot of the ion chamber current recorded on a continuous basis during the rod scanning.

In addition to the single channel gamma scan, multi-channel gamma scans were performed at specific locations along the length of the fuel element; these are noted by the arrows on the gamma scan curve of Figure 5. The gamma spectra in Figure 6 are typical of irradiated fuel with fission product activity while those in Figure 7 are typical of the activity from irradiated stainless steel.

Plenum Radiography

Expansion markers* were located in the plenum of the element prior to irradiation. These markers were arranged so that maximum fuel column expansion and contraction could be measured. It was believed that

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* Brayer, R.C. and S.S. Christopher, "Burnable Poison Additions to UO_2 ", CEND-3107-249, November 1965.

FIGURE: 5 Gamma Scan and Sampling Position of Enriched Fuel Rod E-1.

FIGURE: 6 Typical Fission Spectrum (17 inches from bottom of rod) - 20 Kev/
Channel - 0.1 minute count.

FIGURE:7 Typical Activation Product Spectrum (35 1/4 inches from bottom of element) - 20 Kev/ Channel - 2 minute count.

TABLE I
DIMENSIONS OF FUEL ELEMENT E-1

A. Fuel element length (in.)

Pre-Irradiation

40.956

Post Irradiation

40.960

B. Fuel element diameter (in.)

Axial Position (Inches from bottom of rod)	Pre-Irradiation			Post Irradiation		
	0°	90°	Avg.	0°	90°	Avg.
Weld	0.6008	0.6023				
2				0.5993	0.5998	0.5996
3	0.5990	0.5990	0.5990			
8				0.5994	0.5991	0.5993
9	0.5993	0.5990	0.5992			
14				0.5992	0.5995	0.5994
15	0.5993	0.5992	0.5993			
20				0.5991	0.5993	0.5992
21	0.5992	0.5990	0.5991			
26				0.5993	0.5997	0.5995
27	0.5993	0.5990	0.5992			
32				0.5995	0.5990	0.5993
33	0.5994	0.5991	0.5993			
38				0.5998	0.5992	0.5995
Weld	0.6020	0.6018	0.6019	0.6022	0.6018	0.6020
40				0.5990	0.5994	0.5992

this information would add to the available technology on the performance of vibratory compacted fuel elements. Although these data are not vital to this program, it was initially believed that such data could be obtained with little additional effort. Initial attempts, described below, to radiograph the marker locations were only partially successful. On the basis that the costs for this evaluation were becoming disproportionally high and the marginal utility of such data in this program, work in this area has been discontinued.

Radiographic techniques were employed to determine the location of the expansion markers. Initial results using a Co^{60} source were unsuccessful and an attempt to use a conventional portable x-ray machine in the hot cell was undertaken. However, the quality of the X-ray radiograms, although improved, was marginal and further development of the technique was required. On the basis of the discussion in the previous paragraph further effort was discontinued. Those indications which are readable on the radiograph obtained show that no appreciable fuel movement had occurred during irradiation.

Fission Gas Analyses

The plenum of the element, E-1, was punctured and the released fission gases were collected. A mass spectrographic analysis of the gases is in progress.

C. Destructive Evaluation

Based on the gamma scan curves on element E-1, the sampling locations have been fixed to conform with the burnup distribution of the rod. The sampling scheme is shown in Figure 5; the destructive evaluation of the element will be in accordance with the flow chart presented in Figure 8.

FIGURE 8 Post-Irradiation Evaluation Process Flow Chart (Destructive Evaluation)

III. ANALYTICAL PROGRAM

A. Introduction

The optimization of the analytical methods for boron determination initiated during the previous period was continued during this quarter. Emphasis is being placed on an evaluation of pyrohydrolytic separation of boron followed by 1-1'-dianthrimide colorimetric and mannitol titration for analysis of the boron content. Synthetic samples containing a variety of mixtures of uranium oxide and boron compounds such as B_4C , H_3BO_3 , UB_2 and UB_4 were pyrohydrolyzed and analyzed for boron. Additional samples were also prepared which contained 2 to 3 mg lithium carbonate and up to 2 grams 304 stainless steel in order to mock up some of the compositions of the irradiated samples.

B. Discussion and Experimental Evaluation of Boron

Pyrohydrolytic Separation

Pyrohydrolysis has been used successfully with boron in materials such as Zircaloy, stainless steel, uranium, boron carbide and UO_2 . The technique is relatively simple and accurate and is readily applicable to remote operation. Pyrohydrolysis has initially been selected for both the irradiated and dummy fuel element analysis.

The evaluation of pyrohydrolysis as the reference separation method is currently in progress. The variables that are being investigated are:

- 1) Preoxidation (500° in air)
- 2) Initial temperature of furnace during loading
(200°C vs. 750°C)
- 3) Effect of sample size (1, 7, and 16 grams)
- 4) Type of boron compounds (B_4C , UB_2 , UB_4 , H_3BO_3)
- 5) Influence of contaminants (Li-304 stainless steel)

1) Pre-Oxidation

Air preoxidation at 500°C for one hour is normally used as a means of obtaining homogeneous samples for pyrohydrolysis and analyses. The effect of preoxidation on boron recovery was evaluated by exposing standard samples containing 390 ppm boron as B_4C to the preoxidation treatment. A portion of the same standard sample was pyrohydrolyzed and analyzed for boron without the preoxidation step. The data obtained for the two samples are reported in Table II.

The data show that preoxidation does not result in boron loss due to the thermal treatment when the boron is in the form of B_4C . Additional data concerning the effect of preoxidation on samples containing H_3BO_3 are being evaluated.

TABLE II
EFFECT OF PREOXIDATION
(Pyrohydrolysis-Mannitol Technique)

		% Recovery per lg Aliquot
Sample A	No preoxidation	76.7, 93.8, 92.6, 91.0 91.0 Average: 89%
Sample B	Preoxidation 1 Hour 500°C in air	100.8, 83.8, 85.4, 86.2, 100.8, 85.4, 105.6. Average: 92.6%

2) Effect of Furnace Temperature During Loading

The effect of furnace loading temperature on boron recovery was evaluated as a means of reducing the amount of time required for pyrohydrolysis. (A 750°C furnace loading temperature compared to 200°C would reduce the separation time by a factor of two.)

The furnace temperature at loading was evaluated at two temperatures, below 200°C and 750°C. Samples containing boron as B_4C and H_3BO_3 were pyrohydrolyzed and analyzed to determine the effect of temperature on boron recovery. No loss in boron content was observed when boron was loaded in the form of B_4C . However, the samples containing boron in the form of H_3BO_3 showed a loss of boron when loaded at 750°C. These results are shown in Table III; the H_3BO_3 is probably lost by vapor phase transport. Based on these results and the possibility of having B_2O_3 in the irradiated fuel elements, the 750°C furnace loading temperature step will not be used for analyzing either the depleted or irradiated elements.

TABLE III

EFFECT OF FURNACE LOADING TEMPERATURE ON THE BORON RECOVERY OF SAMPLES
CONTAINING H_3BO_3

(Pyrohydrolysis - Dianthrimide Technique)	
	% Boron Recovery
Loading at 750°C	71.7 - 72.0 - 73.1 Average: 72.3%
Loading below 200°C	85.0 - 88.7 Average: 86.9%

3) Effect of Sample Size

The influence of sample size on boron recovery was evaluated in several steps to insure that the separation and analyses techniques being used at CEND and Westinghouse give accurate and consistent results.

The sample size for the irradiated rods can vary from 1 to 7 grams and it was necessary to determine the effects, if any, of sample size on boron recovery and analysis. Since the pyrohydrolysis - mannitol technique is being used for analysis of the irradiated rod, this technique was used to evaluate the samples. Prior to the separation step, all samples were subjected to a pre-oxidation treatment of one hour at 500°C followed by blending to insure good homogeneity. The results of the investigation are shown in Table IVA and indicate excellent agreement between the one gram and seven gram samples. Based on these results the range between one and seven grams can be used for boron analysis.

Since there was a question of the effect of boron recovery as a function of a larger sample size, (the larger sample size would be used for out of pile analyses) one and sixteen gram samples were prepared for pyrohydrolysis; both samples contained boron in the form of B_4C . The data,

presented in Table IV B indicate no effect of sample size on recovery. The difference in per cent boron recovery, namely 89 vs. 88.1%, are within the limits of precision* for this operation.

TABLE IV
EFFECT OF LOADING SIZE

A. Comparison of 1g and 7g loading (B_4C Compound)

Type of Analysis	Boron Content of Sample (ppm)	
	Sample 1	Sample 2
1g loading (Pyrohydrolysis-Mannitol Technique)	209) 231) Average 229 246)	353) 361) Average 352 341)
7g loading (pyrohydrolysis-Mannitol Technique)	233) Average 225 216)	350) Average 346 342)

B. Comparison of 1g and 16g loading (B_4C Compound)

Type of Analysis	% Boron Recovered
1g loading (pyrohydrolysis-Mannitol technique)	76.7, 93.8, 92.6, 91, 91 Average: 89%
16g loading (pyrohydrolysis-1-1' dianthrimide technique)	93.3, 84.0, 88.4, 93.1, 81.5 Average: 88.1%

TABLE V
EFFECT OF TYPE OF BORON COMPOUND USED
(Pyrohydrolysis-1-1' Dianthrimide Technique)

	Loading Temperature	% Boron Recovered
B_4C	200°C	87.8
$B_4C + H_3BO_3$	200°C	86.9
H_3BO_3	200°C	85.0 88.7
$UB_2 + UB_4$	200°C	70.7

* Mannitol: $\pm 11.2\%$ @ 95% confidence level
1-1' Dianthrimide: $\pm 9.0\%$ @ 95% confidence level

4) Effect of Type of Boron Compound

During irradiation, reactions may occur between UO_2 and B_4C resulting in the formation of new boron compounds such as B_2O_3 , UB_2 and UB_4 . The analytical techniques developed for this program must be able to accurately reflect the boron composition independent of the boron compound present. A series of samples containing various boron compounds was evaluated by the pyrohydrolysis-1-1' dianthrimide technique. The sample compositions prepared and evaluated were $\text{UO}_2\text{-B}_4\text{C}$, $\text{UO}_2\text{-H}_3\text{BO}_3\text{-B}_4\text{C}$, $\text{UO}_2\text{-H}_3\text{BO}_3$ and $\text{UO}_2\text{-UB}_2\text{-UB}_4$. (H_3BO_3 was substituted for B_2O_3 since this material has a tendency to pick up moisture during handling).

The results of these analyses are shown in Table IV. The samples were loaded into a furnace operating at a temperature below 200°C . This temperature was considered optimum from past studies. The values obtained ranged from 85 to 88.7% boron recovery for all the samples except the $\text{UO}_2\text{-UB}_2\text{-UB}_4$, which was only 70.7%. The low value is attributed to non-homogeneity of the $\text{UB}_2\text{-UB}_4$ reference compound.*

5) Influence of Contaminants

Since lithium will be present in the irradiated fuel as a result of boron transmutation by an $n\text{-}\alpha$ reaction; its influence on the analytical determination of boron was evaluated. A fuel burnup of 5000 MWD/MTU would result in a lithium concentration in the fuel of about 20 ppm. Samples containing 20 and 30 ppm lithium were prepared and evaluated. A U_3O_8 standard sample containing 140 μg of lithium carbonate (30 ppm lithium) was pyrohydrolyzed; it was found that less than one per cent of the lithium was carried over with the boron vapors, the rest remained in the U_3O_8 residue. Additional samples were then prepared of $\text{UO}_2\text{-B}_4\text{C}$, $\text{UO}_2\text{-B}_4\text{C-H}_3\text{BO}_3$, $\text{UO}_2\text{-UB}_2\text{-UB}_4$

* The standards for UB_2 and UB_4 were later analyzed and found to contain large quantities of impurities distributed in a heterogeneous manner.

containing 20 ppm lithium as lithium carbonate to determine the effect of lithium on boron analysis. These samples were analyzed for boron by both the mannitol titration and 1-1' dianthrimide techniques. Both techniques gave satisfactory results for boron and were within the precision limits established for the type of boron compound present in the sample.

C. Selection of Method of Boron Analysis and Statistical Study

The pyrohydrolysis-1-1' dianthrimide method of analysis has been selected to analyze the depleted rods at CE. This analysis will be conducted under the following conditions:

- 1) No preoxidation step will be used.
- 2) The initial temperature of the furnace at loading will be $\leq 200^{\circ}\text{C}$ and pyrohydrolysis will be carried out for $1\frac{1}{2}$ hour at 1350°C .
- 3) The sample size will be 16 grams of fuel maximum.

An evaluation was conducted of all previously analyzed samples which contained B_4C and H_3BO_3 and meeting the above conditions. The results are shown in Table VIA. These indicate that if a bias of $\frac{1 \times 100}{87.6} = 1.14$ is applied to the analytical results, the boron content of the samples will be known within $2 \times (+4.5\%) = 9\%$ at a 95% confidence level. Therefore, a sample containing 390 ppm boron (500 ppm B_4C) will then be reported in a range from 355 to 425 ppm $\pm 9\%$ at a 95% confidence level.

The pyrohydrolysis-mannitol method of analysis has been selected to analyze the enriched rods at Waltz Mill. This analysis will be conducted under the following conditions:

- 1) The initial temperature of the furnace at loading will be $< 200^{\circ}\text{C}$ and pyrohydrolysis will be carried out for one hour at 1350°C .
- 2) The sample size will be 7 grams of fuel, maximum.
- 3) If the results of the preoxidation study on $\text{UO}_2\text{-H}_3\text{BO}_3$ mixtures indicate that boron loss does not exceed 10% of the total boron in the sample, a one hour preoxidation step will be included in the analysis as a means of fuel pulverization. Since the amount of boron present in the fuel in the form of oxide is anticipated to be much less than 30% of the total boron present the 10% loss for the boron present in this form will then introduce an error of less than 3%.

The results of an evaluation of test samples containing B_4C analyzed under the conditions listed above including the pre-oxidation step is shown on Table VIB.

These indicate if a bias of $\frac{1 \times 100}{91} = 1.10$ is applied to the analytical results, the boron content of the samples will be known within $2 \times (+5.6) = 11.2\%$ at the 95% confidence level. A sample containing 390 ppm boron ($500 \text{ B}_4\text{C}$) will then be reported in a range from 346 to 434 ppm $\pm 11.2\%$ at 95% confidence level.

D. Lithium Analysis

Due to the transmutation of boron to lithium it will be necessary to determine the amount of lithium present in order to obtain a boron balance. It is proposed to analyze three axial and four radial samples from element E-1 for lithium to obtain a boron balance.

Previous investigations on this program have shown that during pyrohydrolysis practically all the lithium remains in the U_3O_8 residue. The analytical technique used to determine the amount of lithium in the residue consists of dissolving the residue in nitric acid and passing the solution through a Dowex 50 resin to extract the lithium. The lithium is then analyzed by flame photometry. Data indicate that 98% of the lithium present is recovered by this means. Final evaluations are being performed on unknown qualification samples.

TABLE VI
STATISTICAL EVALUATION OF BORON ANALYSIS

Type of Analysis	Number of Evaluations	Average Boron Recovery(%)	Coefficient of Variation (%)
A. Pyrohydrolysis + 1-1' Dianthrimide Analysis (No preoxidation)			
1) Boric acid control (No pyrohydrolysis)	15	---	± 2.01
2) UO_2-B_4C Mixture	6	88.	± 5.3
3) $UO_2-B_4C-H_3BO_3$ mixture	9	87.6	± 4.5
4) $UO_2-UB_2-UB_4$	5	77.*	$\pm 14.8^*$
B. Pyrohydrolysis + Mannitol Titration (with preoxidation)			
1) $HKC_8 H_4O_4$ control (no pyrohydrolysis)	6	---	$\pm .22$
2) UO_2-B_4C	6	91	± 5.6

* The results include in addition to the analytical precision, the heterogeneity factor for the UB_2-UB_4 samples.

IV. UO₂-B₄C COMPATIBILITY

A. Introduction

Previous investigations under this program indicated that volatile boron species form as a result of the UO₂-B₄C reaction at temperatures as low as 1600°C in a flowing argon system (open system). UB₄ is also formed at the lower temperatures and UB₂ plus UB₄ at higher temperatures.

In a closed system it was shown by calculation that the postulated UO₂+B₄C reaction, which forms UB₄ and CO₂ as the reaction products, ceased when the pressure of CO₂ exceeded 6.7×10^{-5} and 4.0×10^{-2} atmospheres at 2000 K and 2500 K. Further, it was calculated that decomposition of B₄C in a closed system would cease when 0.002% and 0.1% of the original B₄C content had decomposed at 2000 K and 2500 K.

In an attempt to verify the above conclusions, a series of out-of-pile closed system experiments were designed where UO₂-B₄C mixtures were enclosed in tungsten tubes and heated in vacuum to 2200°C and 1800°C for times in excess of 100 hours. The capsules operated with no induced radial thermal gradient and with an axial thermal gradient of 16°C/cm and 21°C/cm for the lower and higher temperature capsules. When the furnace was opened to remove the specimens, it was found that both capsules had failed, probably due to the poor thermal shock resistance of recrystallized tungsten. Thus the planned collection and identification of gaseous reaction products was not possible. The loss of cladding integrity converted the planned test of a closed system into one typical of an open system. An x-ray diffraction analysis was run on samples found deposited at the top of the plenum in the

coldest portion of one of the capsules. This analysis revealed two lines of B_2O_3 as one of the reaction products. Since B_2O_3 was not found in previous evaluations, it is the purpose of this task to determine the reactions taking place in the $UO_2 + B_4C$ system by performing tests in open and closed systems and analyzing the reaction products.

B. Experimental Program

The experimental program is aimed at obtaining information on the reactions taking place between $UO_2 + B_4C$ in open and closed systems. The results of these out-of-pile studies will be correlated with the analyses of the in-pile elements. The in-pile results will be based on fuel sample analysis and fission gas analyses on elements being irradiated in Saxton. As an example, the presence of CO_2 in the fission gas would lead us to believe that the reaction is behaving in accordance with past postulated mechanisms.

The testing program is presented in Table VII. Temperatures selected included the lowest temperature at which a reaction could start and a temperature typical of that encountered in the rods being irradiated in Saxton. In all cases, gas analyses will be performed and the fuel mixture will be evaluated for weight gain or loss, x-ray analysis, metallography and chemistry.

Reaction products will be evaluated to establish the most realistic reaction equations. The result will then be correlated with the data obtained on the irradiated fuel elements.

Currently, equipment is being fabricated and set up to undertake this study. The specimens for the open case will be subjected to a flowing atmosphere of helium. The gases of interest, CO and CO₂, will be collected by traps located in the system. The samples for the closed case will be run in evacuated sealed capsules which will be subsequently punctured for gas analyses.

TABLE VII

TEST SCHEDULE FOR COMPATIBILITY TESTS OF UO₂-B₄C

System	Time (Hrs.)	Temperature (C)
Open	25	800- 1300
Open	250	800- 1300
Open	25	1800- 2200
Open	250	1800- 2200
Closed	25	800- 1300
Closed	250	800- 1300
Closed	25	1800- 2200
Closed	250	1800- 2200

V. FUTURE WORK

The second element, E-4, will be inserted into the Saxton Reactor during February. Element E-1 will be destructively evaluated according to the flow chart presented in Figure 8. Four dummy elements will be destructively evaluated and samples taken for axial and radial boron analyses. These results will be compared with the data obtained on element E-1.

The program on $\text{UO}_2 + \text{B}_4\text{C}$ compatibility will be initiated.

APPENDIX I

SAXTON REACTOR POWER HISTORY

A. POWER HISTORY OF THE SAXTON REACTOR DURING IRRADIATION OF ROD E-1

<u>Date</u>	<u>Hours</u>	<u>Power Level</u>		<u>Cumulative</u>
		<u>(MWT)</u>	<u>MW Hrs</u>	<u>MW Hrs</u>
July 12	3	20	60	60
13	24	20	480	540
14	15	20	300	840
14	9	23.5	211.5	1051.5
15	24	23.5	564	1615.5
16	24	23.5	564	2179.5
17	8	23.5	188	2367.5
20	11.5	20	230	2597.5
21	15	20	300	2897.5
21	9	23.5	211.5	3109
22	24	23.5	564	3673
23	24	23.5	564	4237
24	24	23.5	564	4801
25	24	23.5	564	5365
26	24	23.5	564	5929
27	24	23.5	564	6493
28	24	23.5	564	7057
29	24	23.5	564	7621
30	24	23.5	564	8185
31	24	23.5	564	8749
Aug. 1	24	23.5	564	9313
2	24	23.5	564	9313
3	16.25	23.5	382	10259

<u>Date</u>	<u>Hours</u>	<u>Power Level (MWT)</u>	<u>MW Hrs</u>	<u>Cumulative MW Hrs</u>
Aug. 4	24	23.5	564	10823
5	24	23.5	564	11387
6	24	23.5	564	11951
7	7	23.5	164.5	12115
7	17	10	170	12285
8	24	10	240	12525
9	24	10	240	12765
10	24	10	240	13005
11	24	10	240	13245
12	24	10	240	13485
13	24	10	240	13725

B. Appropriate Fuel Peak Centerline Temperature:

<u>Power Level (MWT)</u>	<u>Peak Centerline Temperature (°C)</u>
23.5	2200
20	2030
10	1410

C. Summary

68.5 Hrs @ 20	MWT = 1370	MW HRS = 57.1	MWD
457.25 Hrs @ 23.5	MWT = 10745	MW HRS = 447.1	MWD
<u>161 Hrs @ 10</u>	<u>MWT = 1610</u>	<u>MW HRS = 67.1</u>	<u>MWD</u>
686.75	13725	571.9	