OFFICIAL USE ONLY

PENDING PATENT CLEARANCE

This Report may not be released to General Distribution until such time as patent clearance has been obtained from EURATOM.

DISCLAIMER

CEND-3107-216
Joint U. S. -Euratom
UC-25, Metals, Ceramics
and Materials
TID-4500, 29th Edition

PROGRESS REPORT

BURNABLE POISON ADDITIONS TO UO_2

Period
April 1 to June 30, 1964

Prepared by:

R. C. Brayer
W. P. Chernock

Contract AT(30-1)-3107
U. S. Atomic Energy Commission

Issued July 1964

Unclassified
Classification

W. H. Simon 7/20/64
Authorized Classifier Date

NUCLEAR DIVISION
COMBUSTION ENGINEERING, INC.
WINDSOR, CONNECTICUT

CEND-3107-216
Joint U. S. -Euratom
UC-25, Metals, Ceramics
and Materials
TID-4500, 29th Edition

EXTERNAL DISTRIBUTION:

No. of Copies

Mr. M. Plisner, Contract Administrator — 22
U. S. Atomic Energy Commission
New York Operations Office
376 Hudson Street
New York, New York 10014

Including copies for:

U. S. -Euratom Joint Research and Development Board — 20
51 Rue Belliard
Brussels, Belgium

Mr. J. M. Simmons, Chief — 2
Fuels and Materials Development Branch
U. S. Atomic Energy Commission
Division of Reactor Development
Washington, D. C. 20545

Mr. W. R. Voigt — 3
Water Reactors Branch
U. S. Atomic Energy Commission
Washington, D. C. 20545

Dr. R. C. Dalzell, Assistant Director for Foreign Activities — 2
U. S. Atomic Energy Commission
Division of Reactor Development
Washington, D. C. 20545

U. S. Atomic Energy Commission — 3
Division of Technical Information Extension
Post Office Box 62
Oak Ridge, Tennessee 37831

Mr. Harmon S. Potter, Chief — 1
New York Patent Group
Brookhaven National Laboratory
Upton, L. I., New York 11973

EXTERNAL DISTRIBUTION (Continued):

No. of Copies

Dr. J. Frye
Metallurgy Division
Oak Ridge National Laboratory
Post Office Box X
Oak Ridge, Tennessee — 1

Dr. Stephen Lawroski
Argonne National Laboratory
9700 South Cass Avenue
Argonne, Illinois — 4

Mr. T. J. Pashos
General Electric Company
Atomic Power Equipment Department
Post Office Box 1131
San Jose, California — 1

Dr. R. Allio
Westinghouse Electric Corporation
Atomic Power Department
Post Office Box 1075
Pittsburgh, Pennsylvania — 1

Dr. Ersel A. Evans
General Electric Company
Hanford Atomic Products Operation
Richland, Washington — 1

CEND-3107-216
Joint U.S.-Euratom
UC-25, Metals, Ceramics
and Materials
TID-4500, 29th Edition

BURNABLE POISON ADDITIONS TO UO_2

TABLE OF CONTENTS

BURNABLE POISON ADDITIONS TO UO_2

LIST OF FIGURES

CEND-3107-216
Joint U.S.-Euratom
UC-25, Metals, Ceramics
and Materials
TID-4500, 29th Edition

BURNABLE POISON ADDITIONS TO UO_2

LIST OF TABLES

INTRODUCTION

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.

ACKNOWLEDGMENT

This program, sponsored by the Joint U.S.-Euratom Research and Development Board is being conducted by Combustion Engineering, Inc. Significant contributions have been made by the following personnel:

Project Engineer	R. C. Brayer
Chemical Analysis	W. C. Taylor
Thermal Testing	C. E. Burdg
Sample Preparation	R. C. Brayer
Irradiation Program	W. P. Chernock

CEND-3107-216
Joint U. S. -Euratom
UC-25, Metals, Ceramics
and Materials
TID-4500, 29th Edition

BURNABLE POISON ADDITIONS TO UO_2

By

R. C. Brayer and W. P. Chernock

ABSTRACT

Severe boron segregation has been observed in vibratory compacted fuel rods containing larger (20/25 mesh), self-shielded B_4C particles. The higher boron concentrations found in the upper portion of the rods are the result of B_4C migration through the vibrating bed. These results are in direct contrast to those obtained with smaller (minus 325 mesh), B_4C particles. Incremental compaction is being pursued as a possible solution to this problem.

A tungsten clad UO_2-B_4C element has been fabricated and will be used to verify the stability of UO_2-B_4C mixtures in a closed system at temperatures of 2100 to 2200°C. The relative potential for axial and radial boron migration is being investigated by inducing UO_2-B_4C reactions in an open system containing a test element operated at a fuel centerline temperature of about 2500°C with a radial/axial thermal gradient ratio of about 250. Boron analyses are in progress.

Defect clad samples have been fabricated. Testing has been deferred until the fabrication of a loop, as part of another Atomic Energy Commission program, has been completed.

Negotiations for irradiation testing of two full length fuel rods in Saxton have been initiated.

I. SUMMARY

A program was initiated to determine whether vibratory compaction methods, developed earlier in this program could be applied to UO_2-B_4C mixtures in which the particle size of the B_4C was increased from minus 325 mesh to the range of 20 to 25 mesh (0.071 to 0.084 cm). The larger sized B_4C particles are desirable for some water reactor concepts on the basis of their better self-shielding characteristics. Results of boron analyses for these rods indicated that the local boron concentrations were well outside the limits of $\pm$20% of the nominal loading, established as a target for this program. In all of the rods, there was a concentration of the B_4C in the upper half of the fuel column, indicating particle migration through the vibrating bed. These results for rods containing the larger, self-shielded B_4C particles were in direct opposition to those attained earlier in the program for rods containing minus 325 mesh, non-self-shielded B_4C. Attempts will be made to solve the problem of particle migration in the vibrating bed by utilizing incremental compaction.

A combination of the evaluations conducted to date on the potential for the formation of volatile boron species has indicated that less than 10^{-5} and 10^{-2} of the B_4C present will react or decompose in a closed system at 2000 and 2500°K. An attempt to verify this conclusion has been initiated in which a sealed tungsten clad test sample, containing a mixture of UO_2-0.05 w/o B_4C will be heated for 100 hours at 2100 to 2200°C with an axial gradient of 10 to 20°C/cm and with no imposed radial gradient.

A number of unsuccessful attempts to induce a radial/axial thermal gradient of about 250 at a fuel centerline temperature greater than 2000°C were aborted. A new test sample design has been evolved and an element has been operated with the desired radial/axial thermal gradient and at the required fuel centerline temperature of about 2500°C. Boron analyses are now in progress. In this test, an attempt was made to induce the formation of large quantities of volatile boron species by providing an open system. In this manner, the relative tendency for axial versus radial boron migration can be assessed under a radial/axial thermal gradient typical of that encountered in water reactor fuel elements.

Six rods, three containing UO_2 and three with UO_2-0.05 w/o B_4C, have been vibratory compacted to 83 to 86% of theoretical density and both pinhole and slit defects have been introduced in the cladding. The defected rods will be tested in a pressurized water loop to determine the extent, if any, of preferential leaching of the boron. Testing has been deferred until the construction of a loop, for another Atomic Energy Commission program, has been completed.

Negotiations have been initiated for the irradiation of two full size elements in Saxton. These oxide elements will contain 0.05 w/o of minus 325 mesh, non-self-shielded B_4C. One element will be irradiated to the range of 900 to 1800 MWD/MTU whereas the second element will be irradiated to 5000 MWD/MTU. Emphasis on post-irradiation examination will be concentrated upon axial boron distributions.

II. INTRODUCTION

This program is aimed at developing methods for fabricating fuel elements containing burnable poisons homogeneously distributed in UO_2.

The principal objectives of the program are:

A. To develop vibratory powder compaction methods for producing a uniform distribution of a burnable poison in high density UO_2 fuel elements.

B. To investigate the potential for redistribution of the burnable poison within the fuel element by means of out-of-pile thermal gradient tests.

C. To investigate the potential leaching of the burnable poison under defected clad conditions by means of loop testing of defected rods.

D. To irradiate test rods containing burnable poison to determine the potential in-pile boron redistribution.

E. To develop fabrication procedures for fuel elements containing large self-shielded poison particles..

During the first year's effort, electromechanical vibratory compaction methods were utilized to produce UO_2 fuel rods containing 0.05 w/o B_4C additions to densities of 88 to 89% of theoretical. The local boron contents were maintained within $\pm$20% of the nominal loading.

The method for boron analysis was developed and was based on fusion of UO_2-B_4C mixtures in sodium carbonate at 1000°C in order to transform the contained boron to a water-soluble salt which was then used to produce boron quinalizarin complex solutions. These solutions were subsequently analyzed colorimetrically for boron.

The potential for axial migration of boron under exaggerated axial thermal gradient conditions and decreased radial thermal gradient conditions at centerline temperatures approaching the melting point of the fuel was investigated. Fuel rods, containing an axially positioned tungsten heater and compacted to 85 - 86.5% of theoretical density were tested in a helium and argon atmosphere and were subjected for two hours to an axial* thermal gradient of 40 to 60°C/cm. The radial thermal gradients were minimized by maintaining a clad temperature of 800 to 1150°C. A slight trend toward lower local boron concentration in higher temperature regions of the fuel was observed, although the local concentration of ±20% of the average boron value for the rod was maintained.

During the early part of this contract year, the potential for axial and radial boron redistribution was evaluated under an axial thermal gradient** from 8 to 12°C/cm at a centerline temperature up to 2000°C for approximately two hours. The geometry and method of preparation of these test rods were similar to those used in the first series of tests described previously.[1][2]

The results of this later series of tests indicated that:

A. Significant axial boron migration had not occurred.

B. Significant radial boron migration had not occurred.

C. A higher boron concentration was present in the outer 20 v/o of the fuel in both the as-compacted and thermally tested conditions. There was no significant increase in the content in this "skin" layer as a result of testing under thermal gradient conditions. Furthermore, there was no significant

---

* Emphasis was placed on studies of axial migration early in the program on the basis that preferential loss of boron from regions of the fuel in power peaks would increase the peaks and lead to potentially unsatisfactory conditions.

** Typical of that encountered in power reactors.

decrease in boron content in the other 80 v/o of the fuel as a result of thermal gradient testing. The higher boron content at the "skin" of the fuel was attributed to the increase in fines in this region resulting from the decreased average interstice size found when large particles and physical boundaries (cladding) intereact.

In order to explain the slight trend toward lower local boron concentration observed in the higher temperature region of the fuel under the 40 to 60°C/cm axial thermal gradient, during the first test series, and the absence of radial boron movement under a 3000 to 4000°C/cm radial thermal gradient, during the second test series, a B_4C-UO_2 stability study was initiated. Two series of experiments were conducted under isothermal conditions in flowing argon. Samples containing 3 w/o B_4C-97 w/o UO_2 mixtures were held for 2 hours at temperatures ranging from 1600 to 2200°C. UB_4 was formed at temperatures as low as 1600°C. UB_2 was identified at higher temperature and appeared to be an intermediate stage in the dissociation of UB_4. Samples containing 0.2 w/o B_4C-UO_2 mixtures were heated for up to 2 hours in flowing argon to temperatures in the range of 1600 to 2130°C and showed a loss of 50 w/o of the boron present at 1600°C and a retention of less than 6 w/o of the boron at temperatures in excess of 1800°C. A comparison of observed weight losses with those predicted by various possible reactions indicated that the reaction of UO_2 plus B_4C to form CO_2 (or CO plus 1/2 O_2) plus UB_4, followed by dissociation of UB_4, provided good correlations. Thus, in a flowing argon system, where gaseous reaction products can be continually removed, a mechanism for boron volatilization and subsequent migration appeared to have

been established. An evaluation of the free energy of the UO_2-B_4C reaction between 2000 and 2500°K indicated that the equilibrium pressure of CO_2 varied from about 7×10^{-5} to 4×10^{-2} atmospheres between 2000 and 2500°K, respectively. The amount of B_4C which will be reacted in an operating element to produce this pressure was found to be negligible. The formation of a volatile boron species was dependent upon whether an "open" or "closed" system was used for testing. Since fuel elements are closed systems, it was concluded that boron volatility should not be a problem.

At this point the information obtained in this program required reorientation of the future experimental effort. The conclusions regarding the stability of the UO_2-B_4C system in a closed system needed verification. In addition, in the unlikely event that volatile boron species could be produced, it was required to establish, on a firm basis, that the boron would not move axially under the axial and radial thermal gradients found in an operating fuel element. The potential for axial redistribution must be negated if this method for burnable poison addition is to be adopted. Major emphasis has been placed in this area. In addition, work on self-shielded burnable poison additions has been initiated.

III. FABRICATION OF FUEL ELEMENTS CONTAINING SELF-SHIELDED B_4C

Reactor designs, based upon utilization of burnable poison control throughout core lifetime, require the use of larger boron-containing particles than those used to date on this program. The minus 325 mesh particles, used as a basis for this program, will provide effective control for those designs based upon boron depletion well before the end of reactivity lifetime. A program was initiated, during this reporting period, to develop a fabrication process for incorporating larger, self-shielded, B_4C particles into UO_2 fuel elements utilizing vibratory compaction methods previously developed. Particle diameters, consistent with a self-shielding factor of 0.5 and a loading of 0.06 w/o B_4C, were selected as the basis for this portion of the program. An approximate relationship between B_4C particle diameter and self-shielding factors for a typical water reactor design is shown in Table I. Selection of a self-shielding factor of 0.5, required the use of particles with diameters near 0.076 cm. These particles were obtained by utilizing the minus 20 (0.084 cm), plus 25 (0.071 cm) mesh fraction screened from Grade 14F "Norbide," which is a high boron, fused B_4C manufactured by the Norton Company.

As in the past, the B_4C particles were mixed with the UO_2 particles of one particle size fraction prior to final blending before compaction. In this case, the minus 30, plus 100 fraction of the particle size distribution, shown in Table II, contained all of the B_4C.

TABLE I

Self-Shielding Factors for B_4C Spheres

Average Particle Diameter (cm)	Self-Shielding Factor
0.003	0.98
0.013	0.90
0.025	0.79
0.051	0.63
0.076	0.50

TABLE II

Particle Size Distribution

Material	Particle Size (U.S. Standard Sieve Size)	Weight Percent
UO_2	-6, +16	60.0
B_4C	-20, +25	0.06
UO_2	-30, +100	15.0
UO_2	-200	25.0

A. Fabrication

An outline of the procedures used for vibratory compaction is presented below.

1. The minus 200 mesh UO_2 powder was sieved to separate the 200/325 and minus 325 mesh fraction. These particle sizes were combined in the ratio of 85 to 15, on a weight percent basis, by dry blending to provide the minus 200 mesh UO_2 used for these studies.

2. The boron homogeneity was optimized during loading by utilizing a series of twenty-six, 30-gram charges, of UO_2-0.06 w/o B_4C powder mixture. Each charge was prepared by first tumbling a mixture of the 20/25 mesh size B_4C particles and the 30/100 mesh size UO_2 particles. The preblended mixture of the medium size particles (20/100 mesh size) was blended with the coarse (6/16) and fine particles (minus 200).

The process used for loading the tube was identical to that used previously on the program. Each 30-gram charge was spatula-loaded into a stainless steel tube to minimize segregation of the blended powder. The tube was loaded in the horizontal position, placed upright to insure uniform layering of the charge, and the process was repeated.

3. A steel ram was inserted into the tube in direct contact with the top of the fuel column to minimize powder segregation during handling. The tube was compacted on a Model C-10 Vibration Exciter, driven by a Model T-35 Power Amplifier, manufactured by the MB Corporation. During compaction, the fuel column was axially restrained by a 7 kg (15 pounds) spring loaded ram. An acceleration force about 45 G, with a frequency continuously

oscillating between 200 and 500 cycles per second, was applied for a period of approximately fifteen minutes.

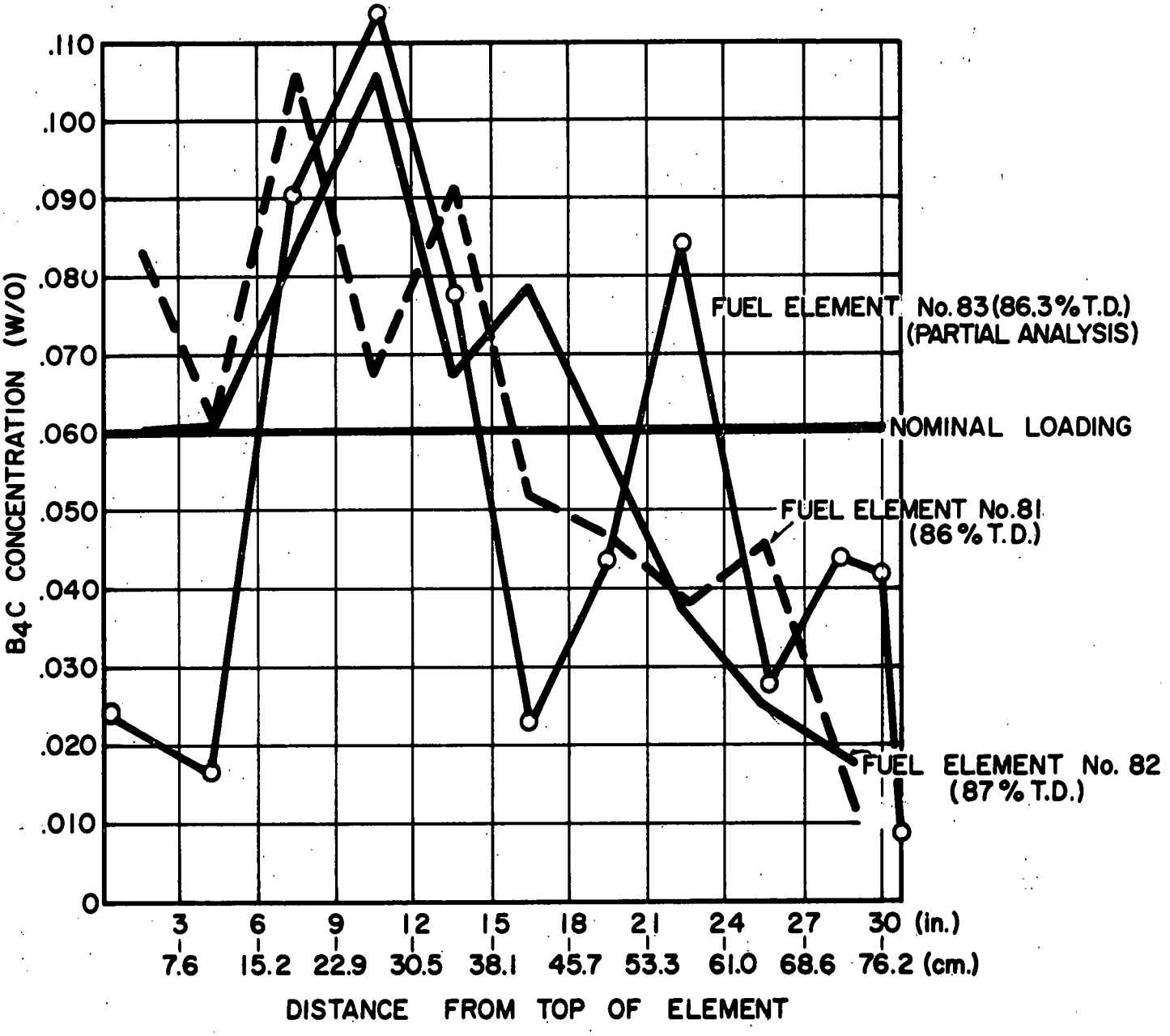
B. Evaluation

The compacted fuel density was determined from the weight of fuel, the fuel compaction height in the element, and the internal volume of the cladding. Densities up to 87% of theoretical were attained in the 76 cm long by 1.2 cm diameter fuel rods.

The fuel column was cut along its axis into one-inch long transverse sections for boron analyses. Two elements were analyzed by the HNO_3 solution method, previously reported.[1] The carbonate-fusion-quinalizarin technique[1] was used for boron analysis of a third element. Results of boron analyses were normalized to an average of 0.06 w/o B_4C for each rod, based on controlled weighing operations during loading. Consistent with previous results[1] of boron analyses for the non-self-shielded rods, all results were reported as an average for each three-inch increment.

Results of boron analyses for these rods are shown in Figure 1. In all rods the local boron concentration is well outside the limit set for the program of $\pm$20% of the nominal loading. These results are in direct contrast with those found[1] earlier in the program for similar rods containing non-self-shielded B_4C particles of minus 325 mesh size. Rods compacted with these smaller B_4C particles exhibited an axial boron variation well within the desired $\pm$20% of the indicated value.

As noted in Figure 1, there is a concentration of these larger boron particles at the upper end of the rod. This trend was observed on all three rods



B_4C AXIAL DISTRIBUTION AFTER COMPACTION OF SELF SHIELDED PARTICLES – UO_2 MIXTURE

Figure 1

and could be related to the large difference in density between UO_2 and B_4C particles. The difference in results obtained as a function of the B_4C particle size employed is probably associated with the ability of minus 325 mesh B_4C and UO_2 particles to agglomerate, whereas the larger 20/25 mesh B_4C particles are free to move through the vibrating bed. The problem of boron migration during compaction may be resolved through the use of incremental compaction. Rods will be fabricated by loading and compacting five 16 cm long increments. Post-compaction boron analyses will indicate whether this approach has been successful in minimizing the B_4C migration problem, observed for large B_4C particles, during compaction. The basis for the above approach is found in earlier work[1] on this program which indicated gross local axial density variations at low average densities and fairly uniform local axial densities at high average densities. Thus, the tendency for particle migration in the bed to occur decreases as the average rod density increases. On this basis, incremental densification appears to be a feasible solution to the boron distribution problem.

Results of recent B_4C irradiations[3] have indicated that large boron carbide particles, with diameters greater than 0.05 cm (self-shielding factor of 0.6), break down into particles which have average diameters near 0.025 cm and 0.011 cm, corresponding to self-shielding factors of 0.8 and 0.9. On this basis, there is some question concerning the ability to maintain a truly self-shielded particle diameter during irradiation. Although it is improbable that cracking of the larger isolated B_4C particles will adversely influence self-shielding, on the basis that the individual sections of the particles probably remain in registry, such a phenomenon will have to be demonstrated via in-pile testing if the concept of self-shielded boron is to be further pursued.

IV. B_4C-UO_2 STABILITY

Previous work on this program has demonstrated that volatile boron species form as a result of the UO_2-B_4C reaction at temperatures as low as 1600°C in a flowing argon system (open thermodynamic system). UB_4 was formed at the lower temperatures and decomposed to UB_2 and boron at higher temperatures.

In a closed system, it was shown (by calculations) that the UO_2-B_4C reaction ceased when the pressure of one of the reactants, CO_2, exceeded 6.7×10^{-5} and 4.0×10^{-2} atmospheres at 2000°K and 2500°K. It was further shown that decomposition of B_4C in a closed system would cease when 0.0002% and 0.1% of the original B_4C content had decomposed at 2000°K and 2500°K. However, dissolution of oxygen (produced by CO_2 decomposition) in the UO_2 lattice could alter these equilibrium gas pressures.

A previous thermodynamic assessment had indicated that the potential for B_4C dissociation into monoatomic boron and graphite should be inhibited at 2373°K when a monoatomic boron vapor pressure of 0.5×10^{-6} atmosphere is attained after reaction of less than 10^{-8} of the amount of B_4C present in a fuel element. More complete data concerning the dissociation of B_4C according to the formula $0.25\ B_4C_{(s)} \rightarrow 0.25\ C_{(graphite)} + B_{(g)}$ have been recently published.[4][5] These data show that the B_4C dissociation pressure at 2000 and 2500°K are 4.9×10^{-8} and 4.4×10^{-5}. Then the amount of B_4C decomposed before the dissociation process is halted is $\sim 10^{-6}$ and $\sim 10^{-3}$ of the amount of B_4C present at 2000 and 2500°K. Therefore, the potential for forming volatile species from this source is remote.

In an attempt to verify the above conclusions by means of out-of-pile tests, a series of closed system experiments have been designed where UO_2-B_4C mixtures are enclosed in tungsten tubes and heated to 2200°C for times in excess of 100 hours.

Tungsten was selected as a container material on the basis of its high temperature properties, low vapor pressure and good compatibility with UO_2. The original concept of lining the tungsten tube with a UO_2 sleeve was abandoned on the basis that it offered little advantage.

The first test sample, shown in Figure 2, has been conservatively designed to provide for a large gas plenum to provide adequate space (without producing an internal pressure which would exceed the 100 hour stress rupture life of tungsten at 2200°C) in the unlikely event that all of the B_4C will react and form gaseous products.

One test sample has been prepared and testing, at 2200°C with an axial thermal gradient of 10 to 20°C/cm with no imposed radial thermal gradient, will be completed during the next reporting period.

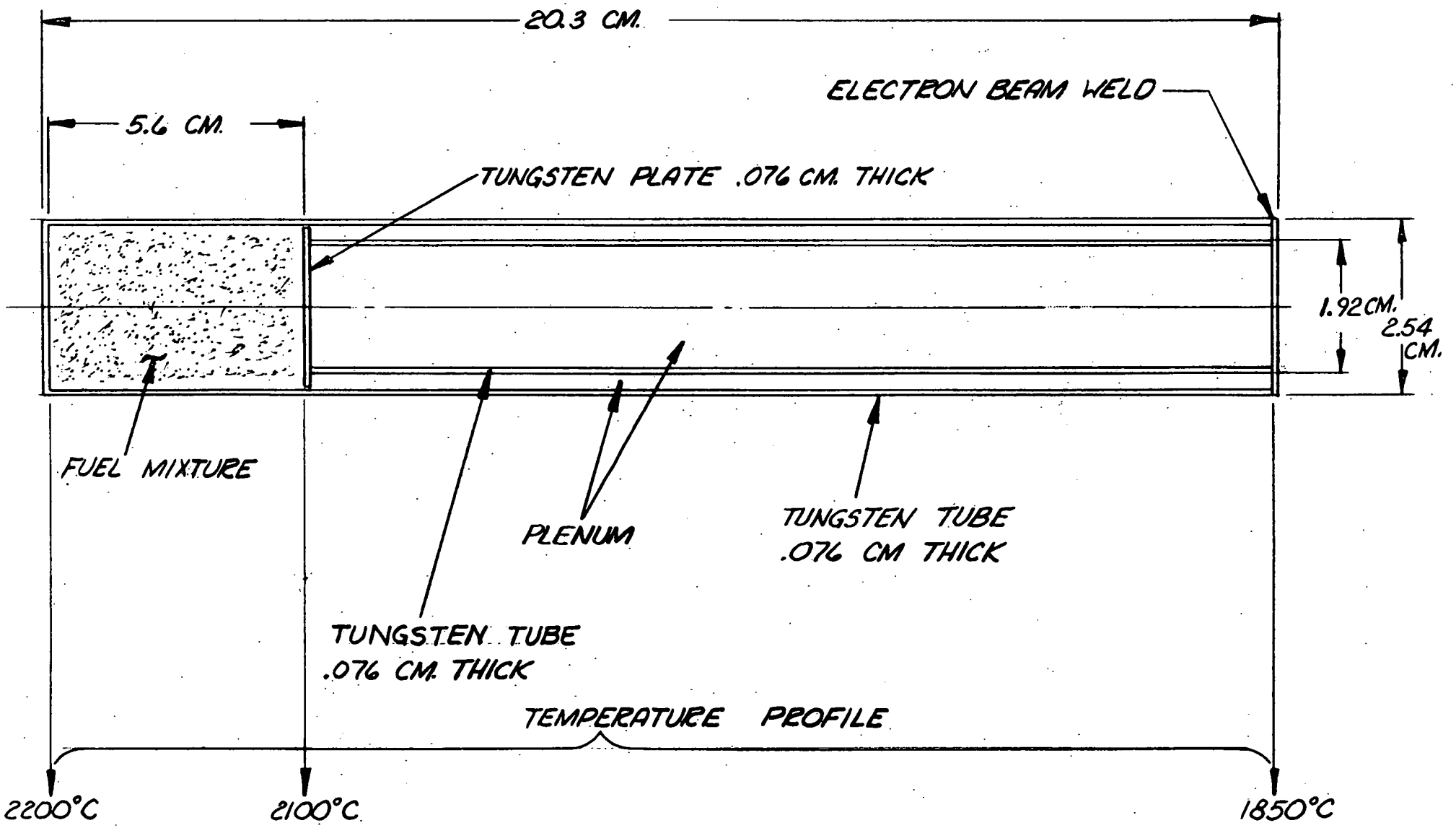


FIG. 2 TEST ELEMENT CROSS SECTION & TEMPERATURE PROFILE

V. THERMAL GRADIENT STUDIES

Investigations of the potential for formation of volatile boron species was emphasized in the previous section. Although thermodynamic calculations have shown that the formation of volatile boron species in a closed system is improbable, it was important to determine the type of boron migration which would occur if such volatile species were formed. As a result of the large ratio of radial to axial thermal gradient in an operating fuel element, condensation of any volatile phases should occur preferentially in the radial direction. However, the potential for axial movement must be assessed in view of the gross consequences to fuel element performance resulting from such movement.

A series of tests has been attempted in which volatile boron species were purposely generated by operating vibratory compacted UO_2-B_4C fuel rods with an axial tungsten-rhenium heater at a fuel centerline temperature of 2500°C for periods up to thirty hours under conditions in which the gaseous products were removed from the reaction chamber (a thermodynamic open system). Helium was continually bubbled through the test chamber in contrast to previous tests in which the system was evacuated and back-filled with inert gas (referred to as a "semi-closed" system). A ratio of radial to axial thermal gradient of 250 was selected for these tests on the basis that this represented a more severe condition (lower potential for radial migration) than that encountered in typical water reactor fuel elements.

Two rods, described previously,[6] and containing UO_2-0.05 w/o B_4C compacted around a tungsten-25% rhenium heater, were tested in the thermal

gradient test apparatus under conditions described above. On the basis of satisfactory performance of Mylar wrapped elements (in helium and in solid metal environments) during previous tests, it was believed that the desired axial thermal gradient could be introduced by using different thicknesses of Mylar film wrapped around the stainless steel cladding. In this series of tests, with a water coolant and with the Mylar film introduced as an insulator between the coolant and the element, the desired ratio of thermal gradient could not be achieved. Erratic temperatures were being recorded along the axis of the element, indicating only intermittent effective insulation.

After two unsuccessful test runs with Mylar wrapped elements, this approach was abandoned in favor of the configuration shown in Figure 3, in which variations in cladding thickness are used to induce the desired radial to axial thermal gradient ratio. Figure 4 is a view of the test element, , after notching, prior to compaction. The notch was introduced in the element wall to minimize final machining after compacting the element. Calculations of power input requirements and radial to axial gradients are given in the Appendix and the ability of this particular design to provide the desired test conditions are demonstrated.

One of the newly designed elements has been compacted and tested with the desired radial to axial thermal gradient of 250, at a fuel centerline temperature of 2500°C and for a period of thirty hours. This element has been impregnated with "Loctite"* and sections are being analyzed for boron by the carbonate fusion-quinalizarin method. Both axial and radial boron distributions will be obtained. Visual observations of some of the sections, already obtained, have

* Previously shown to be boron-free.

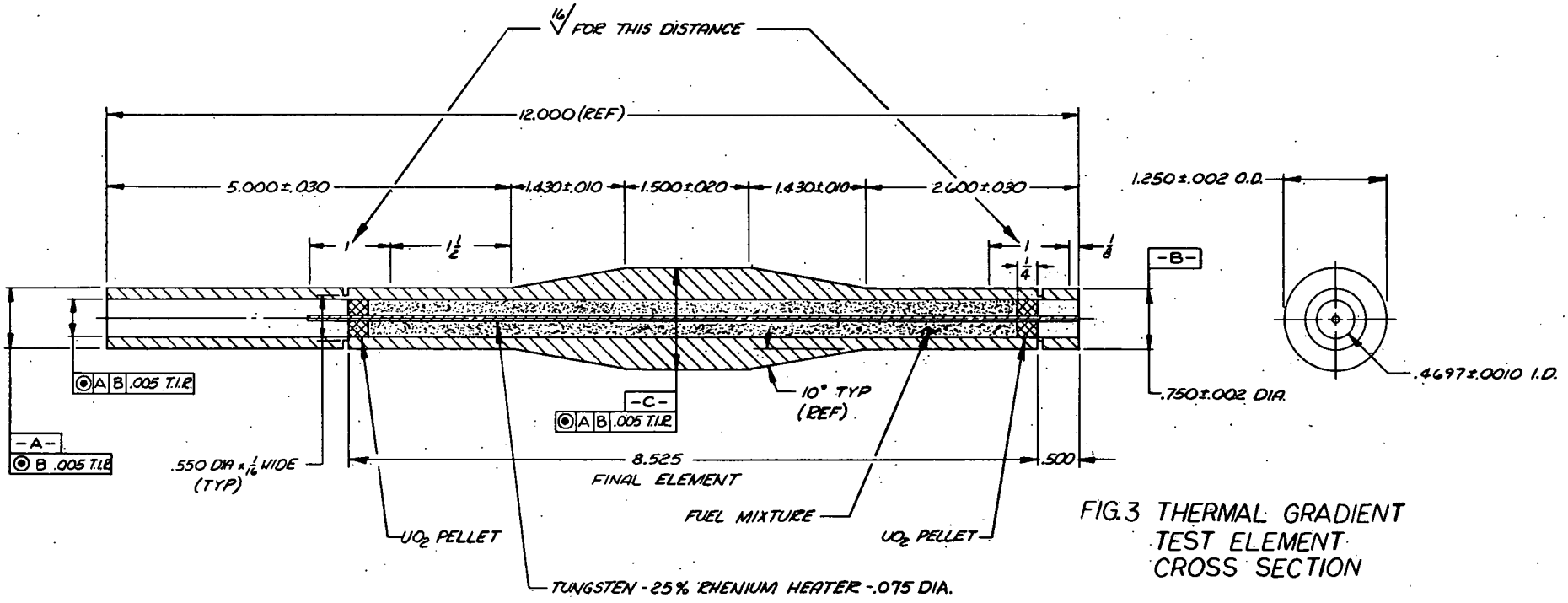


FIG. 3 THERMAL GRADIENT TEST ELEMENT CROSS SECTION

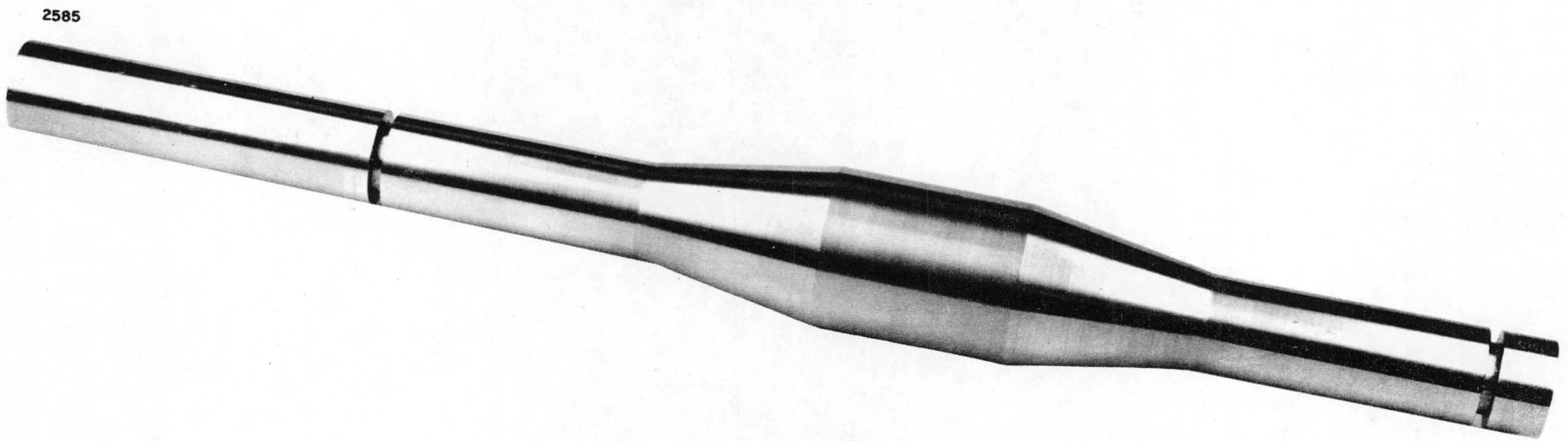


Figure 4 - THERMAL GRADIENT TEST ELEMENT

indicated that the desired reaction has probably occurred. Thus, volatile boron species have probably been formed. Therefore, this test should provide results which will establish the mode of boron migration.

VI. LOOP TESTING OF DEFECTED ELEMENTS

The objective of this task is to compare the behavior of rods containing UO_2 and B_4C with those containing only UO_2 under defect clad conditions. In order to evaluate the effect of density on the potential B_4C washout and dissolution when compared with UO_2, a set of six rods, three containing pure UO_2 and three containing UO_2 + 0.05 w/o B_4C, were fabricated to a range of density from 83 to 86% of theoretical. A compilation of the density and composition of the six rods is presented in Table III.

TABLE III

Density and Composition of Defected Elements

Element No.	Composition		% Theoretical Density
	UO_2	UO_2 + 0.05 w/o B_4C	
60		X	83.3
70		X	84.6
71		X	85.9
73	X		83.7
59	X		84.8
72	X		85.9

In order to approximate conditions representative of those encountered in a large water cooled power reactor, the samples were subjected for two hours to centerline temperatures up to 2000°C. Subsequent to thermal treatment, the samples were defected and stored until such time when a loop, being presently designed under another AEC contract, will be made available for these tests.

The geometry and the fabrication procedures for these elements are similar to those used in previous thermal gradient tests.[2] Compaction times were adjusted to attain the desired fuel density. After subjecting the rods to thermal treatment, they were defected at two positions as shown in Figure 5. One defect consisted of a 0.5 mm diameter hole drilled through the clad. The second defect was in the form of a slot, 6.4 mm long and 0.5 mm wide, and was produced by sawing the clad with a high speed steel saw, just deep enough to break through the clad and expose the UO_2. Extreme care was exercised in subsequent handling of the specimens. The defects were covered and an end cap was welded at each end of the rod. Figure 6 shows one of the completed specimens which is being stored in polyethylene bags during the interim period between fabrication and testing.

The test loop designed under another AEC contract will permit simultaneous testing of six elements. Loop conditions can be adjusted to provide 540°F water at 950 psia and at velocities of about 760 cm/sec over the fuel sections. The scheduled test time will be a minimum of 100 hours.

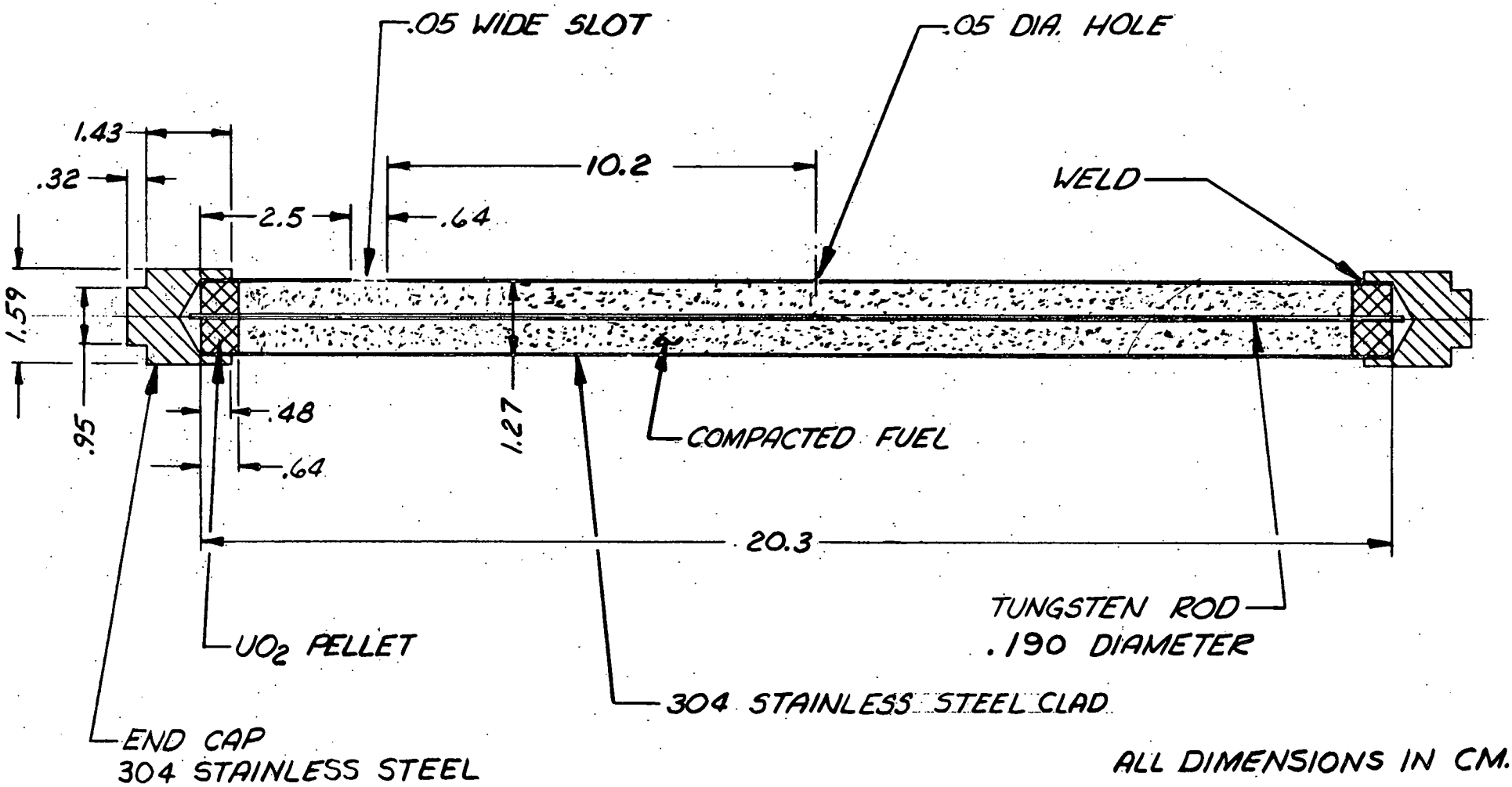


FIG. 5 CROSS SECTION OF DEFECTED ELEMENT

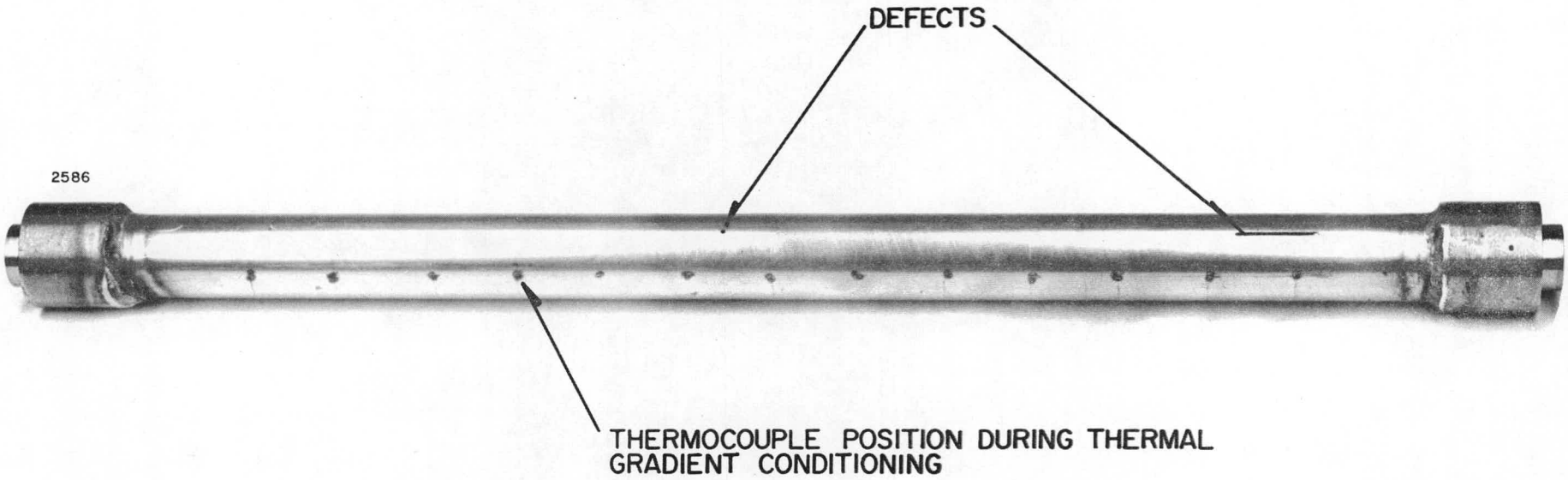


Figure 6 - DEFECTED ELEMENT

VII. IRRADIATION TESTING

An "Invitation for Bid" for an irradiation test program was transmitted during the last quarter to four installations believed to be best qualified to undertake such a program.

The specific objective of the irradiation program is to determine the extent of boron migration under typical power reactor irradiation conditions. Two options were included in the bid request.

Option A: Irradiation of full length rods in a water cooled power reactor.

Option B: Capsule irradiation of 12-inch long fuel rods in a test reactor.

It was specified that prime consideration would be given to those proposals offering an arrangement for irradiation under Option A.

The Invitation for Bid, suggested irradiation of two rods containing UO_2-0.05 w/o B_4C compacted by Combustion Engineering to 88 $\pm$1% of theoretical density. The Invitation for Bid requested that:

A. Irradiation be performed at a maximum fuel centerline temperature consistent with the requirement that the melting point of UO_2 will not be exceeded at any time during irradiation,

B. An axial gradient of approximately 10°C/cm should be maintained under Option B. This requirement was waived in the case of Option A where the axial temperature profile attained in the power reactor is acceptable.

C. The first rod should be irradiated for approximately 900 to 1800 MWD/MTU and the second rod to 5000 MWD/MTU.

D. A suggested program for post-irradiation testing of the fuel element including visual and dimensional inspection, gas collection and analysis for Xe, Kr, He, CO_2, CO, radial and axial boron distributions, and metallography be outlined.

After reviewing the proposals submitted, the following conclusions were reached:

1. All proposals were technically acceptable.
2. All did not provide adequate sampling plans for boron analysis.
3. All did not provide adequate methods for boron analysis.

It was felt that items 2 and 3 could be resolved during subcontract negotiations. A subcontractor has been selected and this selection has been approved by the United States Atomic Energy Commission. The bases of selection were:

A. The proposal selected provides a technically acceptable program at the lowest cost.

B. The proposal offers an irradiation program arrangement under Option A. This option, as specified in the bid request, did receive prime consideration.

Subcontract negotiations are being initiated with Westinghouse for irradiation in Saxton.

VIII. PLANNED EFFORT DURING THE NEXT REPORTING PERIOD

A. Incremental compaction of fuel elements containing self-shielded B_4C particles will be completed.

B. Work on the irradiation test program will be initiated.

C. Compatibility tests of UO_2-B_4C in a closed thermodynamic system will be completed.

D. The analysis of the rod tested with a radial to axial thermal gradient of 250 in a thermodynamically open system will be completed.

REFERENCES

1. Zuromsky, G., Brayer, R. C., and Chernock, W. P., "Burnable Poison Additions to UO_2," USAEC Report CEND-181, Combustion Engineering, Nuclear Division, November 1963.

2. Brayer, R. C. and Chernock, W. P., "Burnable Poison Additions to UO_2," USAEC Report CEND-197, Combustion Engineering, Nuclear Division, October 1963.

3. Williamson, H. E., General Electric, San Jose, California, "The Performance of B_4C Powder in the Dresden Control Rods," in American Nuclear Society Transactions, Vol. 7, No. 1, p. 98, Philadelphia, June 15-17, 1964.

4. Robson, H. E. and Gilles, P. W., "The High Temperature Vaporization Properties of Boron Carbide and the Heat of Sublimation of Boron," Journal of Physical Chemistry, 68, No. 5: 983 (1964).

5. Hildenbrand, D. L. and Hall, W. F., "The Decomposition Pressure of Boron Carbide and the Heat of Sublimation of Boron," The Journal of Physical Chemistry, 68, No. 5: 989 (1964).

6. Brayer, R. C. and Chernock, W. P., "Burnable Poison Additions to UO_2," USAEC Report CEND-3107-210, Combustion Engineering, Nuclear Division, April 1964.

APPENDIX I

Determination of Power Input and Radial to Axial Thermal Gradient Ratio Per Unit Length for the Element Shown in Figures 2 and 7

Data - Maximum fuel center temperature = 2500°C

Clad temperature = 40°C

Total radial temperature gradient ($\Delta T_{r(t)}$) from heater OD to clad OD = 2460°C

Assumption - The conductivity values are constant with temperature, $K = K_{effective}$. K_{eff} has been selected for the temperature range of interest and the K_{eff} for the fuel includes the boundary conductance.

Calculations - The radial temperature drop in any successive concentric layer of an infinite cylinder is given by the following equation:

$$\Delta T_r = \frac{Q}{2\pi l} \quad \frac{\ln \frac{Do}{Di}}{K}$$

Where ΔT_r = radial temperature drop (°C)

Q = input power (watt)

l = length of heated zone = 19.7 cm

Do = outside diameter of the layer (cm)

Di = inside diameter of the layer (cm)

K = thermal conductivity of the material (watt/cm^2/cm/°C)

The value of $\frac{\Delta T_r}{\frac{Q}{2\pi l}}$ is calculated for each section in the thicker portion of the element and the total value for the element from the

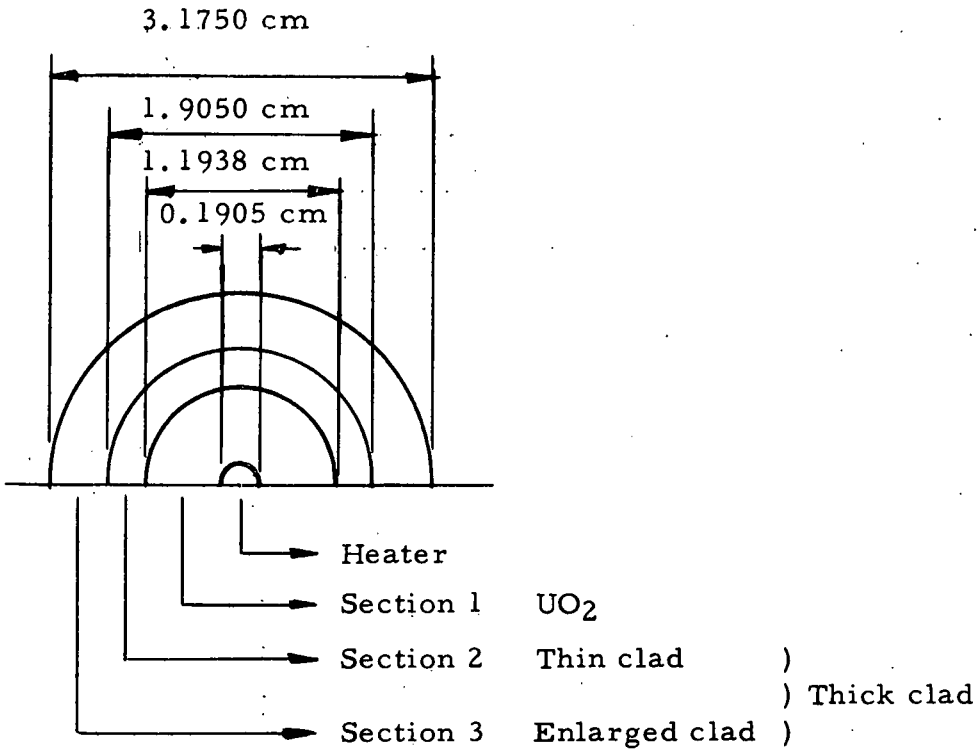


FIGURE 7.

Thermal Gradient Test Element Radial Cross Section

heater outside surface to the clad surface is calculated by summation.

Section	Material	Do (cm)	Di (cm)	$\frac{Do}{Di}$	$\ln=\frac{Do}{Di}$	K (watt/cm^2/cm/^{o}C)	$\frac{\Delta T_r}{\frac{Q}{2\pi l}} = \frac{\ln \frac{Do}{Di}}{K}$
1	UO_2	1.1938	0.1905	6.267	1.8353	0.018	101.95
2	304 SS	1.9050	1.1938	1.596	0.4675	0.173	2.70
3	304 SS	3.1750	1.9050	1.667	0.5110	0.173	2.95
Σ1-2-3							107.60

Then, if $\frac{\Delta T_{r(t)}}{\frac{Q}{2\pi l}} = 107.60$

$$Q = \Delta T_{r(t)} \frac{2\pi l}{107.60} \sim 2840 \text{ watts}$$

Input power = 2840 watts

The radial temperature drop across the fuel (section 1) is:

$$\Delta T_{r(1)} = \Delta T_{r(t)} \times \frac{101.95}{107.60}$$

and the radial temperature drop per unit length across the fuel is:

$$\Delta T_{r(1)} \times \frac{2}{D_{o(1)} - D_{i(1)}} = \Delta T_{r(t)} \times \frac{101.95}{107.60} \times \frac{2}{D_{o(1)} - D_{i(1)}}$$

The radial temperature drop across section 3 (cladding protrusion) is:

$$\Delta T_{r(3)} = \Delta T_{r(t)} \times \frac{2.95}{107.60}$$

The axial temperature drop from section 2 OD to section 3 OD per unit length is:

$$\Delta T_{r(3)} \times \frac{1}{\text{length of taper (cm)}} = \Delta T_{r(t)} \times \frac{2.95}{107.60} \times \frac{1}{3.63}$$

Assuming that the axial temperature profile of the center of the element reproduces the temperature profile of the inner clad, the ratio of radial thermal gradient and axial thermal gradient in the fuel per unit length is:

$$R = \frac{\Delta T_{r(t)} \times \frac{101.95}{107.60} \times \frac{2}{D_{o(1)} - D_{i(1)}}}{\Delta T_{r(t)} \times \frac{2.95}{107.60} \times \frac{1}{3.63}}$$

$$R = \frac{101.95}{2.95} \times \frac{3.63 \times 2}{(1.1938 - 0.1905)} = \sim 250$$

$\underline{R \sim 250}$