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CURRENT STATUS OF APPR-1 FUEL ELEMENT METALLURGY

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FUEL ELEMENT METALLURGY

The development by ORNL of fuel elements for the APPR-1 has been based on the use of 304L as the cladding material with highly enriched uranium dioxide and a small quantity of boron carbide dispersed in a sintered matrix of stainless steel as the fuel bearing core. The boron carbide is introduced into the matrix as a burnable poison for controlling excess reactivity at start-up.

Powder metallurgy techniques are used in preparing the core. The core is clad by the use of roll bonding and the picture frame technique. Finished cladding thickness is .005", and finished core thickness is .020". A metallurgical bond is obtained between core and cladding and between cladding elements themselves.

The materials used in the fuel bearing core of plates for both stationary and control rod fuel elements are:

Stainless Steel Powder - type 304 with high silicon in a size less than 149 microns.

UO_2 Powder - highly enriched grade containing approximately 88% uranium in which the U-235 isotope is upgraded to product level. The oxide is prepared by the reduction of hydrated oxide-uranyl nitrate, commonly called the Geneva process. Particle size is 44-88 microns, and the oxide powder is free of clinging fines.

B_4C Powder - contains approximately 76% natural boron and is screened to less than 44 micron size.

The stainless steel, uranium dioxide and boron carbide powder for each batch are weighed separately before blending. The oxide powder is weighed inside a special box arrangement for handling enriched material. The three materials are introduced into a glass container and blended in an oblique blender. A small amount of lauryl alcohol is added near the end of blending.

The blended powder is transferred to the die cavity in a box-type enclosure on a hydraulic press. The powder is cold compacted under a pressure of 33 tons per square inch.

Ten compacts are loaded in a stainless steel sintering boat which is then introduced into a high temperature furnace and sintered for one hour at 2100° F. A dry hydrogen atmosphere with a dew point of at least -70° F is maintained during sintering. At the conclusion of the sintering operation, the compacts are visually inspected and reloaded in the original die and coined to correct for the shrinkage which occurs during sintering. Acceptable compacts are weighed and stored in a desiccator for subsequent use.

The cladding for the core is originally composed of three parts, the frame for enclosing the four lateral edges of the sintered compact and a top and bottom cover shield. The frame piece is prepared by punching a nearly square hole in plate material. The previously prepared core is inserted in this hole in the frame and the cover pieces placed over and under and welded to the frame to form a three ply billet.

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The jacketed compact or billet is preheated before rolling to a temperature of 2100° F for approximately 20 minutes in a muffle furnace purged with hydrogen. The compacts are hot reduced on a 20" diameter and 30" face mill from .390" to .040" with a total reduction of approximately 90%. The jacketed compacts are reheated for a minimum of 2 minutes between each pass and after the final pass. Hot reductions per pass are from 10 to 30%. Rolling direction is reversed after each pass. After hot rolling the scale is removed by pickling in an acid bath, and wire brushing.

The hot rolled plates are fluoroscoped to locate the outlines of the core and the plate is marked for shearing. The fluoroscope examination is also used as an inspection for gross segregation. The edges of the composite plates are then trimmed to remove excess material and squared by shearing.

The plates are then cold rolled to a finished thickness of .030". A 4 high mill with a 5" diameter work roll and 14" face is used. The reduction per pass is kept to less than 5% with total cold reduction approximately 25%. The roll faces are lubricated with oil which is removed from the finished plate by vapor degreasing.

The plates as cold rolled are wavy and arched. A flattening anneal is required to bring the plates to acceptable flatness. The plates are batch stacked, pressure is applied between two bolt down platens with alumina being used to prevent sticking during annealing. Annealing is done in an atmosphere of dry hydrogen at 2100° F for approximately two hours followed by furnace cooling.

The fuel plates are then refluoroscoped to locate the boundaries of the core section. The core is positioned within dimensional limits specified in drawings D9-13-2007, D9-13-2006, A9-13-2010 and A9-13-2009, by means of a template placed over the fuel plate. The outline of the finished fuel plate is marked for removal of excess inactive stainless steel. Finished specified fuel plate size is obtained by shearing and batch machining to drawings D9-13-2006, 2007, A9-13-2010 and A9-13-2009. Machining lubricants are removed by vapor degreasing.

Side plates with longitudinal grooves running the length of the plate are gang milled to drawings D9-13-2011, 2014, to provide the recesses for the side edges of the fuel plates. Combs are also machined, primarily for use as supports for the ends of the assembled plates during the brazing operation.

The 18 fuel plates (16 in the case of control rod elements), 2 side plates and 2 combs are assembled for brazing. Coast Metals NP brazing alloy, containing 50 Ni, 27-Fe, 11-Si, 8-Nb, 4-P nominally, is used. A stainless steel brazing jig supports the assembly in loading and brazing. A pair of side plates and one end comb are initially positioned in the jig. The first fuel plate is then introduced into position. The jig is rotated in two directions to 45° from the horizontal so that brazing alloy powder can be loaded in the flat position and cemented with Micro-braze cement. A special technique is used to insure the location of the brazing alloy to prevent excessive spreading onto the fuel plate during brazing. This procedure is repeated for each plate until all plates have been assembled.

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The stainless steel brazing jig with the fuel element loaded is then put into a muffle type brazing furnace. Helium is used as a purging gas at low temperatures and pure dry hydrogen at high temperatures. Brazing temperature is 2150° F and the unit is held at temperature for 5 minutes. Cooling rate is governed by allowable distortion. Hydrogen is replaced by helium at a temperature of approximately 1400° F.

Fuel plate location and flatness are held to plus or minus 10% of plate spacing except for the two outside plates. Tolerance on these plates as measured down from side plate edges is plus or minus 5%.

End adapters are then welded to the ends of the stationary elements after facing the ends. The assembly is then machined to final dimensions per drawing D9-13-1003. On the control rod fuel elements, a handle is welded to the top end of the side plate before assembly and brazing. After brazing, a pin is inserted through and welded to the handles. Assembly is per drawing D9-13-1004. The fuel element assembly is checked for squareness and lengthwise bowing by insertion in a box in which the inside dimensions are approximately 0.031" over the nominal fuel element external dimensions.

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SPECIFICATIONS

Specifications for the manufacture of fuel elements and absorber sections are being prepared in cooperation with Oak Ridge National Laboratory personnel and are presently in final revision. Fuel element loadings are as follows:

Stationary Fuel Elements - per element		
Fuel content - gr. U-235	515.16	+.38% -.97%
Enrichment - Product level		±0.1%
Boron 10 - gr.	0.483	+1.75% -4.94%
Control Rod Fuel Elements - per element		
Fuel content - gr. U-235	417.76	+.38% -.97%
Enrichment		±0.1%
Boron 10 - gr.	0.392	+1.75% -4.94%

Tolerances are determined by accuracies obtainable in analysis, weighing and handling.

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FUEL ELEMENT IRRADIATION TESTS

Three test programs were undertaken to investigate the ability of APPR-1 type fuel elements to perform satisfactorily.

One of these tests, described in detail in CF 55-4-163, consisted of exposure of encapsulated small sized specimens in the MTR. Test variations in these samples included type of matrix material, type of oxide, initial oxide particle size, oxide and poison contents, burn-up and clad-core-clad proportions. Examination of eight of these small sized samples has been made to date by the Solid States Division of ORNL. Five of these samples received approximately 20% burn-up and the other three approximately 50% burn-up. On all eight samples no cracks, distortion, blistering, or other observable defects were observed.

Considerable effect of oxide particle size on hardness and ductility of the stainless matrix material has been found. On the 20% burn-up samples, bend tests show fairly extensive core break-up in the oxide particle size range of 7 to 11 microns. Minute core cracking was observed with oxide particle size of 31 to 44 microns. 53 to 62 micron particles showed no evidence of cracking on bending. In general, samples containing oxide sizes under 31 microns showed fairly extensive break-up while samples with larger particle size showed only minute cracking in the range of 31 - 44 microns, and no cracking with larger sizes. On the samples with 50% burn-up, there is not yet enough evidence to establish a pattern. Complete evaluation must await examination of all irradiated samples.

Hardness measurements also indicate the effect of oxide particle size on hardness of matrix material under irradiation, as illustrated in the following table:

Oxide Particle Size (Microns)	Hardness VHP - 2 KG Load	
	Before Irradiation	After Irradiation
7 - 11	204	475
31 - 44	184	420
88 - 105	178	371

Although these results are not strictly conclusive because of the limited number examined, the data indicates that particle sizes under 31 microns have considerably more radiation-damage effect upon the matrix material than larger particle sizes.

Another irradiation test, described in detail in CF 55-6-39, consisted of exposure in the MTR of a full sized fuel element adapted to MTR size requirements. This full sized element received an estimated burn-up of 25 to 40%. Telescopic examination of the element as a unit, and of four plates removed from the element, showed no defects except a possible crack in a small section of a brazed joint. Metallurgical examination of the section containing this questionable area revealed a surface irregularity rather than a crack. A reddish-brown film outlining the core section was evident on all fuel plates.

The third irradiation test, described in CF 54-11-13 and CF 55-1-113, was of a full sized element in the STR core at ARCO. However, this element has received only a small portion of its proposed burn-up at the time of its removal from the core some time ago. Corrosion resistance under STR operating conditions

has been reported as excellent.

In brief, all APPR-1 irradiation tests to date indicate that satisfactory performance can be expected from APPR-1 fuel elements.

CONTROL RODS

It is presently planned to use Boron 10 as the absorber material in the APPR control rods, dispersed in an iron matrix and clad with 304L stainless steel. The method of fabrication is in general similar to that used in the fabrication of the fuel bearing plates, with some variations due to the difference in materials and end requirements. The picture frame technique is again used as are powder metallurgy techniques for the preparation of the Boron 10 and iron compact. Finished thickness of the absorber plate is .187" including .032" cladding. Four plates are welded by the heliarc method into a hollow box which is used in order to take advantage of the thermalizing effect of the water in the center of the box on any fast neutrons which pass through the sides of the box. Pertinent drawings are D9-13-2017 and 1002.

Absorber section loading is as follows:

Per element

Boron 10 - gr. 56.4 $\pm$ 2% (tentative)

ABSORBER IRRADIATION TESTS

The absorber plates were originally specified in ORNL-1613 as dispersions of B_4C in a copper matrix and clad with 304L stainless steel. Development work at ORNL on absorber plates was continued on this combination of materials. It soon became evident, however, that copper cored stainless clad plates could not be fabricated satisfactorily due to the differences in properties of copper and stainless steel at hot rolling temperatures. A wide range of temperatures was tried, but in all cases it appeared that copper was flowing between the stainless frame elements themselves, resulting in poor bonding. Welding of frame elements of the plates was considered, but was rejected because it was doubtful that a good seal could be obtained consistently.

A review of potential absorber and matrix materials was then instituted. Other compounds of boron were unattractive because of the larger weight percent amounts that would be required due to the smaller weight percent of boron in these compounds. Stainless steel, nickel, and nickel alloys had to be eliminated as matrix materials because of evidence of reaction with the boron compounds occurring at hot rolling temperatures. Boron 10 appeared to be a desirable absorbing element because of the much smaller amount of total poison required per absorber plate. At this time, however, boron 10 was apparently in rather short supply and obtainable from only one source. Further investigation revealed that boron 10 in the quantities required for APPR-1 could be obtained from ORNL.

Development work was therefore initiated on using boron 10 as the absorber and iron powder as a matrix material. It was found that rolling could

be performed in only one direction due to necessity for maintaining a square and full thickness of core at the end adjacent to the fuel element. Also, since starting thickness of frame and covers is limited by CRNL press capacity, finished absorber plate thickness has been reduced to .187" in order to obtain the reduction desired for satisfactory bonding. This development work has resulted in an absorber plate which is considered excellent from the structural viewpoint.

Considerable concern, however, has developed as to the capability of boron-containing control rods to withstand irradiation. Under irradiation boron is transformed into Lithium 7 and helium. Calculations by CRNL personnel indicate that the helium pressure resulting from boron burn-up could be excessive provided the helium atoms formed diffused into a void. Unfortunately, there is no irradiation experience directly comparable to the boron 10 dispersion in an iron matrix proposed for the APPR-1. Wrought stainless steels containing boron in amounts less than that required for APPR-1 absorber plates become tremendously embrittled under irradiation. Blisters or warts have developed on an unclad irradiated 1% boron dispersion in titanium. Cracks have developed under irradiation in an unclad dispersion of boron in zirconium. However, due to differences in properties of materials, and of the finished plates, there is a very good possibility that the boron 10 dispersions in iron clad with .032" of stainless steel will not develop irregularities or defects which would bar them from giving satisfactory service. The clearance between the control rod housing and the absorber box section is sufficiently large that a gross distortion or a gross surface blistering would be necessary to cause interference with the proper functioning of the control rod assembly.

In order to attain irradiation damage experience as soon as possible, a program has been set up to make and irradiate small samples of boron dispersed in iron and clad with stainless steel. These small samples will be inserted in leaky rabbits and irradiated in the MTR in a manner similar to the Phase II APPR-1 fuel element samples. The proposed test samples and irradiation cycles are listed in the table below.

Test	MTR Cycles	Priority
B_{10} Design Loading	1	1
	2	2
	3	3
	4	4
	5	5
60% B_{10} Design Loading	3	6
	5	7
12% B_{10} Design Loading	3	8
	5	9
Low Densification Core *	3	10
High " " "	3	11
Low " " "	5	12
High " " "	5	13

*These samples have subsequently been eliminated from the test program.

This test is based upon the assumption that five MTR cycles are the equivalent of 100% burn-up and that five cycles will require approximately 15 weeks irradiation. The 60% B_{10} content samples are included because preliminary calculations indicate that this content will furnish the required blackness at the end of one core life. It is, of course, necessary to check this content in

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the APPR-1 low power tests at Schenectady. The sample containing 20% excess B_{10} is included as a possible accelerated test. The low and high densification samples are included to determine the effects of processing variables.

These thirteen samples are presently being fabricated and it is expected that they will be shipped to the MTR by June 1st. It is also planned to test a full size control rod adapted for insertion in the MTR. This full sized element is in the stage of preliminary design at the present time, and it is expected that it will be fabricated and shipped to the MTR in July.

In the event that these tests prove B_{10} to be an undesirable absorber material, consideration has been given to other materials for both immediate and long range back-up efforts. Calculations have been made by Alco which indicate that Hafnium would be a satisfactory substitute absorber material. A hollow box constructed of solid Hafnium with walls .3" - .4" thick is required. The availability of Hafnium for this purpose is being investigated by ANP.

Practically all potential absorber materials have been considered by Alco personnel and evaluated in connection with the long range back-up effort. High cross section rare earth materials appear unsatisfactory with present fabrication techniques because of the large amount of each material that would be required to attain reactivity control throughout core life equivalent to the present design using B_{10}. It is entirely possible, of course, that a new fabrication technique can be developed which would permit the incorporation of the relatively large amount of materials needed. Cadmium, possibly in the form of a cadmium silver alloy at a content of about 30%, appears to be a more likely long range back-up material at this time. Wall thicknesses again in the order

of .4" would be required. An experimental reactivity evaluation of any alternate absorber material in the APPR-1 core is required before a final decision can be made to use a material other than boron.

END

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