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STATUS OF APPR-1 FUEL ELEMENTS
TO: J. E. Cunningham
FROM: R. J. Beaver

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INTRA-LABORATORY CORRESPONDENCE

OAK RIDGE NATIONAL LABORATORY

To: Listed Distribution

Subject: Minutes of Meeting to Clarify Status of APFR-1 Fuel Elements

The subject meeting was held at ORNL on July 18, 1956. The object of the meeting was to:

1. Describe a serious problem associated with the manufacture of APFR-1 fuel components.
2. Present recent pertinent experimental data.
3. Recommend corrective measures.
4. Plan future course of action.

Those present were:

A. L. Boch, Y-12
Larry Schaffer, Y-12 (Army)

R. J. Beaver, ORNL
J. E. Cunningham, ORNL
R. C. Waugh, ORNL
W. R. Burt, Jr., ORNL

J. F. Haines, ALCO
R. D. Robertson, ALCO
L. Meem, ALCO
J. G. Gallagher, ALCO
E. C. Edgar, ALCO (ORNL)

J. B. Lampert, AFB, Wash.
W. Hepburn, AFB, Wash.
D. Campbell, AFB, Wash.

J. F. Smith, AEC, SOO
H. W. Behrman, AEC, OROO
A. W. Larson, AEC, OROO

I. Introduction.

The difficulties encountered during the manufacturing of the APFR-1 fuel elements were reviewed. The fabrication heat treatments were listed and are as follows:

1. Hot rolling fuel plates at 1125°C.
2. Annealing hot rolled fuel plates for 2 minutes at 1125°C and air quenching.
3. Flatten annealing fuel plates in a stack of approximately 40 plates by

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slowly heating from 300 to 1125°C; holding at this temperature for 3 minutes and furnace cooling to 300°C.

4. Brazing the fuel element by slowly heating to 1130°C; holding at this temperature for 7 minutes and furnace cooling to 300°C.

It was pointed out that although six depleted fuel elements were manufactured prior to the enriched fuel elements and were satisfactory in all respects, considerable difficulty was experienced with braze metal flow across the cladding of the fuel sections in subsequent enriched units. Later, several units were also rejected for poor plate spacings. Early in June when it became evident that it was imperative that this situation be corrected, production of these fuel elements was stopped and an investigation made to isolate the cause of the braze runout. It became evident that the fuel plates were being mis-handled after they were flattened by annealing and prior to assembling into a fuel unit. Fabrication prior to the time was carried out by personnel in the development group. Because of the accelerated schedule, there was insufficient time to conduct a dry run of depleted fuel elements using the production crew. It was also apparent that an oxide film remained on the plates after they were annealed in the hydrogen atmosphere. In addition, there was evidence that the alumina placed on the surfaces of the fuel plates to prevent self-welding during annealing was not removed sufficiently in the hot water scrub. It was felt that these two factors prevented the flow of braze metal around the groove joint and encouraged flow across the clad of the fuel plate.

The procedure was modified so that the plates were flattened by annealing after they were machined and radiographed. After washing the alumina from the plates in hot water and prior to assembling into the fuel unit, the plates were dipped for approximately 2 minutes in a 15% nitric-5% hydrofluoric aqueous solution. It is to be emphasized that type 304L SS is not subject to excessive intergranular attack in this acid. Due to a lack of insufficient time caused by the July 15 delivery date, a thorough investigation of the effects of these modifications was not made. On July 13, two batches of plates which had been annealed for two and three cycles were acid cleaned. During this cleaning, it was observed that those plates which had been annealed three times showed severe intergranular attack by the acid. Because of the enormous grain size of the stainless steel cladding after heat treatment, subsequent removal of grains by the acid exposed the fuel core. In fact, actual grains of steel were removed during this treatment. The plates which were annealed twice were examined and showed some intergranular attack, but not nearly as severe as the attack on the plates which had been annealed three times. It appeared evident that apparently the 304L cladding material was being carburized, thus placing it in a position of being susceptible to intergranular attack by the acid. Investigation was immediately initiated to isolate the reasons for the observed phenomenon and to circumvent the difficulties. A review of all past APFR-1 manufacturing was made, and it was revealed that in manufacturing the first nineteen fuel elements, acid cleaning was done at random on the fuel plates prior to brazing. No record was maintained on this procedure. Metallographic inspection to determine the extent of intergranular attack by the acid on a fuel plate in a brazed APFR-1 fuel unit, rejected for out-of-tolerance plate spacings, revealed severe intergranular attack to within a $\frac{1}{2}$ mil of the fuel core. This condition severely impairs the reliability of such fuel elements for reactor operation.

II. Results of Recent Experimental Studies.

Studies were conducted to resolve the problem of apparent carburization and sensitization of the APFR-1 fuel plates. These experiments were designed to thoroughly investigate whether or not the present combination of a 304B SS matrix in the core, with its .07 w/o carbon was resulting in a higher carbon content in the 304L cladding after the various heat treatments required in the manufacturing process. Additional experiments were made to determine whether or not this effect could be corrected with substitute materials. Combinations studied in this investigation were:

<u>SS Core Material</u>	<u>SS Cladding Material</u>
(1) 304B	304L
(2) 347	304L
(3) 304B	347
(4) 347	347

The results are summarized as follows:

1. The carbon content of the 304L stainless clad was increased sufficiently by diffusion of carbon from the 304B stainless core to permit sensitization to occur. Carbon analyses of the clad indicated that after this diffusion the cladding stainless steel was no longer a 304L type, but a 304 type stainless steel.
2. The present 304B-304L material was sensitized after the specified process heat treatments, failed the Strauss Test, and would be susceptible to intergranular attack in the 15% HNO₃-5% HF aqueous solution.
3. Substitution of 347 stainless in full or in part for the 304 SS components decreased the susceptibility for sensitization. The all-347 stainless combination was not sensitized, passed the Strauss Test, and would therefore not be subject to intergranular attack in the 15% HNO₃-5% HF aqueous solution.

III. Corrosion Evaluation.

The subject of corrosion of sensitized SS in 500°F pressurized water system was reviewed. Results of testing at ANL revealed no appreciable corrosion, and their conclusion (available in ANL-5354) was: "Sensitized 300 series steel were not susceptible to intergranular attack in water at 500°F." It was pointed out that this data was based on a maximum of 2000 hours of testing. It was revealed that prior to inserting the APFR test component in the STR, ORNL provided WAPD with a depleted prototype four-plate element for corrosion testing at Bettis Field in their dynamic water loop. This fuel element contained essentially the same materials combination and was fabricated in the same manner as the

APFR-1 fuel elements, with the exception that the fuel plates were not cleaned in acid prior to assembling for brazing and were brazed in a graphite jig instead of the SS jig used in APFR-1 fuel element manufacture. The test conditions were as follows:

1. Water purity:	24 cc of hydrogen per kiloliter
10 cc of nitrogen per kiloliter
oxygen undetected
2. Water velocity:	38.2 ft/sec
3. Temperature:	500°F
4. Time in dynamic water:	6245 hrs

The results of this test were as follows:

1. This fuel element was returned to ORNL by WAPD and two plates were removed for examination. A carbon analyses was made of the cladding material in one plate, and the results revealed that the carbon content was .076 w/o carbon. In other words, although the initial cladding material was 304L, sufficient carbon contamination in the clad evidently had occurred during manufacturing to increase the carbon content from a maximum of .03 w/o carbon to the .076 w/o carbon.
2. Transverse sections were sheared from the other plate. The exposed edges of one of these sections were sealed by heliarc welding and the section Strauss tested. Other sections were metallographically examined to determine the effect of corrosion by the elevated temperature water in WAPD's loop. The section which was Strauss tested failed. Metallographic examination of the specimens which were not Strauss tested revealed negligible corrosion by the water environment in the WAPD test. This supported their statement that there were negligible weight changes on the fuel element after this test.
3. The information obtained revealed that the cladding material in the WAPD test unit was a 304 SS in the sensitized condition and subject to failure in a Strauss test.

It was concluded that in view of the recent results of corrosion testing sensitized 304 SS at ANL, and the additional testing by WAPD on the APFR-1 prototype test element, sensitized 304 SS has satisfactory corrosion resistance in 500°F water.

IV. Status of the APFR-1 Fuel Elements.

The manufacturing status of the fuel elements was reviewed and are listed herewith:

Fuel Elements 1-19 - possibly contained fuel plates which had been cleaned in the 15% nitric--5% hydrofluoric aqueous solution prior to assembling into a fuel unit. Since no record was maintained of the cleaning of these plates, it was recommended that these fuel elements be rejected insofar as use in the reactor is concerned.

Fuel Elements 20,21,22,23,24,25,34,36,37,38 - are considered rejects insofar as the reactor is concerned because they contained acid cleaned plates.

Fuel Elements 28,29,30,31,33,35,39 - are considered rejects for the reactor because of excessive flow of braze metal across the cladding of the fuel core.

Among the above listed fuel elements, numbers 3R,16,19,23,37 and 38 contain plate spacings which are out of tolerance and are considered rejects for this additional reason.

The only stationary fuel elements which are considered satisfactory for the reactor are 26,27 and 32. All seven control rod fuel elements which were manufactured are considered suitable for the reactor.

All of the above fuel elements are considered satisfactory for the critical experiments to be conducted in ALCO Products' critical facility.

The fuel plate rejections were reviewed, and it was revealed that 14.8% of the 1132 stationary fuel plates manufactured were rejected. Of 180 control rod fuel plates manufactured, 22.2% were rejected.

V. Conclusions and Recommendations.

Sufficient experimental data were obtained during the short period available to gain a clearer understanding of our problem. When these data are evaluated in the light of the abundance of pertinent information available in the project and open literature, it is possible to conclude the following:

1. Carbon is migrating from the core to the cladding material during fabrication of the 304 SS-enriched fuel plates. The carbon gradient, thermal treatment and possibly the silicon content in the core material are playing a role in this diffusion process. The extent of carburization is such that low carbon (3 pts. max.) starting material for the clad is essentially being converted to a regular 304 grade stainless with a maximum of 8 points carbon. An increase in carbon content should be avoided if possible because it increases the susceptibility of stainless to intergranular attack by corrosive media.
2. Sensitization, or precipitation and concentration of chromium carbide at the grain boundaries, occurs during the flattening anneal operation. The rate of cooling after annealing, or other subsequent thermal treatment called for in the manufacturing process, is not rapid enough to keep the carbides in solution, and thereby maintain a single-phase

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homogeneous structure. Thus, the second requirement for intergranular type of attack is met by depleting the area adjacent to the grain boundaries of chromium. Chromium is the main ingredient responsible for the resistance of SS to corrosion. Sensitized fuel plates will not pass a modified Strauss Test.

3. Fuel plates in the sensitized condition corrode rapidly on exposure to 15% HNO_3 -5% HF water solution. The type of attack is selective and inter-crystalline in nature which renders the plates unsuitable for reactor service.
4. Sensitized austenitic stainless steels (300-type) are not susceptible to intergranular attack in water at 500°F. The corrosion behavior of the test element exposed for 6200 hrs in the Bettis Test Loop clearly demonstrated this point. Therefore, sensitized 304 stainless fuel plates are satisfactory for reactor service provided they have not been pickled in HF solution while in the sensitized condition.
5. The Strauss Test is a poor criteria for evaluating the suitability of fuel plates for service in 500°F demineralized water. It does, however, afford an excellent means to determine whether or not the fuel plates have been sensitized. Because of the importance of integrity, carburization and sensitization of the fuel plates should be avoided.
6. This objectionable phenomena of carburization and sensitization of the fuel plates can be greatly reduced by the employment of substitute materials in the make-up of the composite fuel plate. Of the material combinations studied, 347 SS is unquestionably the best structural and cladding material for the proposed application. This particular grade is normally not affected by heat treatment because the carbides are stabilized by the addition of niobium. A strong preference should be given to the 347 stainless clad frame and powder core material combination in the make-up of future components.
7. Other material combinations studied (which are less desirable from the viewpoint of reliability but acceptable) are rated in order of preference as follows:
 - a. 347 clad frame and 304B powdered core material.
 - b. 304L clad frame and 347 modified powder core material.
 - c. 304L clad frame and 304B powdered core material.
8. The burnout poison boron can probably be incorporated into the fuel component in the form of either B_4C or ZrB_2 . The results favor the use of the B_4C compound slightly.

The fuel element is the central and most important single component in the heterogeneous reactor system. It is the source where the potential energy of

fission is converted to heat as well as the site where useful neutrons are born. In the design and construction of reactor fuel elements at ORNL, a premium has always been placed on absolute integrity. It is with this philosophy in mind that the following recommendations are made.

1. Employ the initial loading 39 stationary fuel elements, scheduled for delivery on July 30, 1956, for service in the critical facility only.
2. Employ the initial loading of 7 fuel sections and 7 absorber sections for the control assembly (also scheduled for delivery on July 30, 1956) for service in either the critical facility or APPR-1.
3. Void the present order calling for 19 spare, stationary fuel elements and construct a new core loading of 48 stationary (38 regular plus 10 spares) 347 grade stainless steel fuel elements for service in APPR-1. Tentative date for shipment of the new loading to the reactor site is December 30, 1956. Such action would allow a loading consisting of 38 stationary (347) fuel elements and 7 complete (304) control assemblies to be loaded into the reactor early in December of 1956.
4. Issue a change order calling for 7 spare fuel and 7 spare absorber sections for the control rod assemblies to be constructed of 347 rather than 304 grade stainless steel. Tentative date for shipment of these components to the reactor or critical facility site is March 1, 1957. These control rod pieces could be employed in either the critical facility of APPR-1 core - depending on the reactor start-up date. In any event, complete loadings for both facilities would be available shortly after March 1, 1957.
5. Employ natural boron in the form of B_4C for the burn-out poison in the make-up of new stationary fuel components.

VI. Discussion.

After lunch, the meeting was opened for discussion. It was impossible to record all the pertinent discussion during the three and one-half hour period. Below are given the salient points of the discussion on which decisions were made and concurrence given:

1. The seriousness of braze metal flow over the clad of the fuel core was evaluated. The results of a recent investigation revealed that of approximately 75 samples sectioned from APPR enriched unit No. 17, the maximum depth of penetration of metal into the clad was 0.75 mils. Although these results strongly indicate that this effect of braze metal flow is not serious, the later flow has recently been confined by the use of Colmonoy green stopoff applied along the edge of the preplaced braze metal on each plate. This stopoff can be effectively removed by bristle brushing in hot water. It was therefore considered advisable at this time to prevent the braze metal flow rather than to accept it.

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2. The subject of whether or not the current materials combination should be changed was discussed. ORNL pointed out that sufficient material was available in any case, although there was an excess of 347 material. There was sufficient 304L material available to manufacture more than 30 fuel elements. In the meantime, additional 304L material could be ordered.
3. ORNL advised that there were approximately 75-100 spare plates plus 130 sintered cores in stock if it was decided to continue with the present materials combination and pointed out that if the 347 combination was desired, it would not be possible to initiate manufacturing until August 27.
4. B_4C is preferred for the burnout poison because of lack of the behavior of ZrB_2 under radiation, and such a substitution would require a change in specifications.
5. Advantages of an all-347 stainless materials combination were:
 - a. more flexibility in fabrication,
 - b. finer grain size than 304,
 - c. homogeneous single-phase structure in finished component,
 - d. stabilized stainless steel not subject to sensitization.
6. It was pointed out by APB that at the present time there was a disadvantage in converting to an all-347 stainless steel materials combination because it had not been demonstrated that type 347 SS fuel plates could be fabricated and fuel assemblies brazed. ORNL did not feel that any difficult problems would be encountered, although it was apparent that limited development would be necessary to ascertain that the same fabrication procedures for the 304L materials combination could be substituted for the 347 system. It was estimated that fabrication compatibility could be demonstrated by August 27, 1956.
7. The subject of a "crud" problem occurring because of the addition of type 347 SS in a reactor containing type 304 SS was discussed. ORNL did not feel that galvanic corrosion would occur because 304 and 347 grades are identical structurally (austenitic) and chemically (iron base). With the exception of the 1% addition of niobium, the chemical composition of 347 SS is very nearly that of the 304 grade.
8. ALCO Products proposed a slight delay in initiating fabrication of the new loading in the event a materials change was desirable from results of the critical experiment. This holdup would cause further delay, and it was not certain that data from critical testing could be analyzed on September 7 in order to make the required change in specifications. It was decided if critical tests indicated a need for a change in specifications, the situation would be evaluated at the time.

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9. A review of manufacturing schedules revealed that there was a better opportunity that ORNL could deliver 35 stationary fuel elements on December 1, if the present materials combination was unchanged. However, it was pointed out by ORNL that this was merely a target date and some uncertainty of meeting this date existed because of the uncertainty in completing SPERT B development and manufacturing on schedule, which ORNL was already committed to do. It was also pointed out that there was a possibility that additional orders may have been received by the AEC of which ORNL was not aware, and which could possibly have priority over the APFR. Therefore, ORNL could not definitely commit themselves to a specific December date. However, it was recommended that a request to fabricate an additional loading of APFR fuel elements be immediately forwarded to the Oak Ridge Operations Office for approval.
10. The APB representatives felt that they would prefer to use the present materials combination because it appeared more likely that such fuel elements could be delivered to Ft. Belvoir sooner than if substitute combinations were used. They emphasized the importance of getting the reactor operating at the earliest possible date.
11. It was finally agreed that pending approval, ORNL would furnish 35 stationary fuel elements to Ft. Belvoir by December 1, 1956, and 9 additional stationary fuel units, 9 control rod fuel units, and 8 additional absorber sections by January 30, 1957. These fuel elements would be manufactured with the same materials combination as in the first loading. The manufacturing procedures and specifications would be essentially the same with the exception that no fuel plates would be cleaned in a solution containing HF acid after the flatten annealing or any subsequent operations. The first loading manufactured at ORNL would be shipped to Schenectady on June 30, by air. Since three of these fuel elements were considered acceptable for the reactor, these elements would be later shipped to Ft. Belvoir by December 1, along with the 7 control rod fuel elements and absorber sections so that all of the necessary components would be at the reactor on that date.

A point not brought out in the meeting is that sensitized 304 SS fuel elements are subject to some rusting under certain conditions. If the fuel elements are exposed for long periods of time in moist air with an unlimited supply of free oxygen, rusting will probably occur due to depletion of chromium at the grain boundaries. In the event the elements are not employed in the reactor, they should be oiled for storage as a precautionary measure.

12. AEC, Schenectady Operations Office are to request the changes to be made in the program, provide a fuel allocation, and furnish additional financing. ORNL estimated that they were \$40,000 overspent under the present financial arrangement, and requested that they be provided with an additional \$140,000 to manufacture the components listed under 11 above.

J. Beaver
J. Beaver

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