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**AEC Research and
Development Report**

**Knolls
Atomic Power
Laboratory**

Development of Ferritic Stainless Steel—UO₂ Dispersion Fuel Elements

**W. K. Barney
W. E. Ray
H. G. Sowman**

January 30, 1958

**Operated for the
United States Atomic
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UC-25, Metallurgy and Ceramics
(TID-4500, 13th ed, Rev.)

DEVELOPMENT OF FERRITIC STAINLESS STEEL - UO_2
DISPERSION FUEL ELEMENTS

W. K. Barney, W. E. Ray, H. G. Sowman

January 30, 1958

C. E. Lacy
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ABSTRACT

A preliminary definition of a fabrication process for ferritic stainless steel - UO_2 fuel clad with Type 430 stainless steel has been achieved. The procedure is an adaptation of an earlier process for preparing austenitic fuel elements, and consists of the cold-binder extrusion of a plastic powder dispersion which is fired in dry hydrogen, fitted with welded end caps, and electroplated with nickel. This core is inserted into a length of Type 430 stainless steel cladding and the assembly is bonded by hot-swaging at 800°C .

The irradiation performance of such a fuel element has been examined by means of an MTR test. The results of this test strongly indicated that the desired lowering of UO_2 particle operating temperature was realized and that ferritic stainless steel is not unduly susceptible to irradiation damage.

DEVELOPMENT OF FERRITIC
STAINLESS STEEL - UO_2 DISPERSION
FUEL ELEMENTS

W. K. Barney, W. E. Ray, H. G. Sowman

INTRODUCTION

Ferritic stainless steel such as Type 430 have smaller thermal expansion coefficients and higher thermal conductivities than austenitic stainless steels such as Type 304. Therefore, it seems very likely that ferritic stainless steel - UO_2 fuel elements would operate in power reactor cores at lower temperatures and stress levels than elements based on austenitic compositions. The increased ductility of ferritic stainless steels in usual reactor core temperature ranges lends additional credence to the lowered stress level prediction.

This report presents progress in the development of a fabrication process for the production of ferritic fuel elements clad with Type 430 stainless steel. The process under consideration is an adaptation of the cold-binder extrusion, hot-swaging method* developed for the preparation of austenitic stainless steel fuel elements. By using a preliminary but promising fabrication procedure, elements were prepared for irradiation performance tests. The results of these MTR exposures are also presented.

PROCEDURES

Fabrication Procedure

The iron-chromium - UO_2 dispersion cores used in this work were prepared in much the same way as are cores for austenitic fuel elements. The iron powders used were of the carbonyl "L" grade, the chromium powder was prepared electrolytically, and the UO_2 was of the usual dispersion fuel type, but was not enriched. With these powders, 63 wt % iron, 12 wt % chromium, 25 wt % UO_2 cold-binder rods were prepared by (a) hand-tumbling, (b) adding Methocel Gel by hand-kneading, and (c) extruding the plastic mixture through a 0.210-in.-ID die. The rods thus obtained were subsequently fired in hydrogen to increase their density and to volatilize the Methocel binder. Two firing schedules were tried, 1100°C for 1.5 hr and 1325°C for 1.5 hr; the more appropriate diameter (0.183 to 0.190 in.) was obtained with the latter temperature and time.

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* KAPL-1557 "A Method of Fabricating Stainless Steel - UO_2 Dispersion Fuel Elements", H. G. Sowman, C. E. Lacy, and G. L. Ploetz, June 1, 1957

The cores were cut to length after firing and fitted with wrought Type 430 stainless steel end plugs. The 0.185-in.-diam end plugs were attached by induction welding. After welding, the cores were electrolytically coated with iron, chromium, or nickel.

The electroplated fuel element cores with attached end plugs were next soaked in distilled water to remove any entrapped electrolyte; dried in acetone, and held under vacuum for 1 hr. Type 430 stainless steel tubing, used as cladding for the cores, was cut to length, cleaned, and fired with the cores at 1000°C for 1 hr in dry hydrogen.

After the clean-up firing, the fuel components were hand-assembled. The clearance between core and cladding at this point was 0.002 to 0.010 in. and averaged about 0.008 in. The assembled samples were next sufficiently reduced in diameter at one end to bring the cladding into contact with the end plug. A cold-drawing operation followed (through a 0.217-in.-diam die) which brought the entire inside surface of the cladding tube into close contact with the core and end plugs. After cold-drawing the excess lengths of cladding were cut from the elements; the components were closed at both ends by heliarc welding. After attaching a 3-ft length of stainless steel handling rod to one end of each sample, the preparations for hot-swaging were complete.

Hot-swaging procedures will be discussed on p. 14. Pre-heatings were done in a hydrogen atmosphere furnace and a gas burner was used at the swaging machine to maintain the fuel elements at the desired temperature at the die face. A photograph of this arrangement is shown on Figure 1.

Irradiation Test Procedure

Three-inch lengths of ferritic stainless steel - enriched UO_2 fuel elements were used in MTR irradiation performance tests. These shortened, Type 430 stainless steel clad samples were provided with end plugs which were attached by brazing under helium. A 60% manganese - 40% nickel braze composition was used. The completed samples were then assembled, together with austenitic stainless steel fuel elements as control specimens, into irradiation test capsules.

The desired MTR test space could not be obtained; therefore, an alternate position was used which gave estimated element surface temperatures between 400 and 600°C for the 2300-hr test exposure. At the end of this abbreviated test period, the five-capsule slug was mistaken for another slug and discharged. This error was not apparent until the capsules arrived at KAPL. The slug had been cut by MTR for cask shipment and could not be re-inserted in MTR. Thirty-two per cent burnup was desired so that a meaningful comparison could be made between ferritic

FIGURE 1 Swaging Apparatus for Final Sizing of Fuel Elements

KS-8484
Unclassified

and austenitic samples but only 11% was reached. Because of the low per cent burnup actually realized, the value of this comparison was largely lost. Therefore, only one capsule (the one receiving the highest burnup) was examined and the remainder scrapped. Metallography was performed on the ferritic element to determine effect of irradiation on the microstructure.

RESULTS AND DISCUSSION

Fabrication Process Development

Initial attempts to fabricate ferritic stainless steel - UO_2 fuel dispersions clad in Type 430 stainless steel followed fabrication conditions developed for austenitic stainless steel fuel elements. The steps in fabricating the assembled components were:

1. Cold-swage sufficiently to bring the cladding into contact with the core and end plugs.
2. Make welded closures on both ends.
3. Swage through 0.210, 0.194, and 0.185-in.-ID dies at 900 to 1100°C.
4. Anneal 3 hr at 1300°C in hydrogen to homogenize the core matrix and promote core-to-cladding bonding.
5. Hot-swage through 0.176-in.-ID die at 900 to 1100°C.
6. Centerless grind to final size.

The results of this treatment are shown in Figure 2. Note that the grain sizes of both core and cladding are very coarse. This structure is not acceptable since grain boundaries penetrate completely through the cladding. If intergranular corrosion began at the surface of such a fuel element, the cladding would be penetrated quickly. Notice that this fuel element shows excellent bonding of the cladding to the core; in fact, the presence of the nickel bonding layer initially present is barely noticeable.

To achieve smaller grain sizes than those produced in the initial hot-swaging attempts, reduction of processing temperatures was indicated. Therefore, in the next fabrication attempt the same sequence of hot-swaging passes was used, but the processing temperature was lowered into the range of 800 to 850°C. The temperature of the bonding anneal was also decreased from 1300 to 1000°C. After swaging, the elements were annealed 3 min at 850°C. The upper section of Figure 3 shows the microstructural results of this treatment. One-half of the cladding is coarse-grained, the remainder is small-grained. It was found that this non-uniformity was produced during the post-swaging anneal. That temperature was high enough and of sufficient duration to produce not

FIGURE 2 Ferritic Stainless Steel - UO_2 Fuel Element Swaged
at 900 to 1100°C with an Intermediate Anneal at 1300°C for
3 Hours

Transverse Section Etched in Mixed Acids

20X

KS-20579
Unclassified

Longitudinal Section

20X

Transverse Section

20X

FIGURE 3 Ferritic Stainless Steel - UO_2 Fuel Element Swaged at 800 to 850°C with an Intermediate Anneal at 1000°C for 3 Hours

KS-20580
Unclassified

only desirable recrystallization but also undesirable grain growth. By reducing the final anneal to 750°C for 10 min it was found that grain growth was always reduced and sometimes eliminated. Annealing for 5 to 10 min at 600°C was found to be a more reliable grain refining treatment. The lower section of Figure 3 shows the transverse structure of the same fuel element. Note that many small voids are apparent in the bonding layer in the transverse section; insufficient diffusion of the nickel bonding layer into the core and cladding has occurred.

To obtain a satisfactorily small and uniform cladding grain size and at the same time increase the extent and uniformity of nickel bonding layer diffusion, a number of modifications of the basic fabrication procedure was examined. Of the modifications tried, the following produced the most favorable results.

1. Swage at 950 to 1000°C through 0.210, 0.194, and 0.185-in.-ID dies,
2. Anneal 3 hr at 1000°C in hydrogen,
3. Swage through 0.176-in.-ID die at 800°C, and
4. Recrystallize anneal 10 min at 600 to 700°C.

The transverse structure of an element after step 3 is shown in Figure 4A. Note the absence of voids in the bonding layer as well as the extremely coarse grain size. After the recrystallization anneal (step 4), the structure is modified to the one shown in Figure 4B. The grain size of this structure is satisfactory; however, it is apparent that nickel diffusion is still not extensive.

Since nickel is an austenite stabilizer, it seems likely that elements in which nickel electroplates are used might have partially austenitic zones between core and cladding which would reduce heat transfer performance. Consequently, some work was done to investigate the feasibility of iron and chromium bond layers. Since the core and cladding components are rich in these two elements, the driving force of a large concentration gradient, which is operative with nickel bond layers, is greatly reduced in elements with iron- or chromium-plated cores. The results of the work were not promising, presumably because of this reduction in concentration gradients. A further cause of difficulty in the case of chromium-plated cores may have been the presence of Cr_2O_3 - contaminated surfaces. Chromium oxide is reduced only with difficulty during firing at 1000°C in completely dry hydrogen. Oxide layers of this type would drastically inhibit solid-state diffusion, leading to spotty bonding of the type observed.

A. Annealed 3 Hours at 1000°C in H₂, Reswaged at 800°C

B. Annealed 10 Minutes at 650°C in H₂ after 800°C Swaging

FIGURE 4 Ferritic Stainless Steel Fuel Element Swaged at 950°C

KS-20581
Unclassified

Early work with austenitic elements indicated that satisfactory bonds could be obtained in the absence of an electroplated bond layer. Trials with unplated ferritic stainless steel elements were unsuccessful. Apparently the necessarily reduced working and annealing temperatures are too low to permit extensive self-diffusion of ferrite containing chromium.

Irradiation Performance

Typical unirradiated and irradiated UO_2 particles are depicted in Figures 5 and 6. On the basis of preirradiation data, it can be safely assumed that there is little, if any, difference between the UO_2 particles used in these ferritic elements and the UO_2 employed in preparing austenitic samples previously tested. There is little evidence of the spherical pores and sintering effects usually noted in irradiated UO_2 particles irradiated in austenitic matrices.*

Irradiation-induced porosity is most affected by per cent burnup, test temperature, and time. In austenitic elements, complete sintering and pore formation has been observed at burnups much lower than 11%. Further, the ferritic elements under discussion were in the MTR as long as were most of the austenitic samples tested. It is deduced on these bases that the lesser degree of porosity and sintering noted in the ferritic elements is attributable to lower UO_2 operating temperatures. This, of course, assumes no error in gross surface temperature calculations. Past experience indicates this assumption to be permissible.

Visual examination and dimensional measurement of the irradiated ferritic fuel elements lead to the conclusion that no gross effects were produced by the test. No irradiation damage of the ferritic stainless steel cladding or matrix alloys was observed other than that experienced by the layer of matrix surrounding each UO_2 particle. The matrix material surrounding each UO_2 particle is damaged by fission product recoil. Therefore, the existence of the layer is a result of the nature of the dispersed particle and cannot be eliminated by changing the composition of the matrix.

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*KAPL-1688, "Irradiation of UO_2 - Stainless Steel Dispersion Type Fuel Elements", W. K. Barney and W. E. Seymour, September 10, 1957
KAPL-1836, "Metallography of Irradiated UO_2 Containing Fuel Elements", W. K. Barney and B. D. Wemple, January 15, 1958

SUMMARY AND CONCLUSIONS

Preliminary fabrication development work on ferritic stainless steel - UO_2 fuel elements with bonded Type 430 stainless steel cladding indicates that such elements can be satisfactorily prepared by a cold-binder extrusion, hot swaging procedure. The work demonstrated the necessity of using an intermediate bonding layer of electroplated metal between the core and cladding. This layer should preferably be of nickel, since the evidence available indicates that suitable extensive iron or chromium diffusion will not result during hot-swaging operations at 800°C . The final annealing operation must be carried out between 600 and 700°C , since higher temperatures result in grain coarsening.

The irradiation data obtained to date on this fuel element are confined to metallographic results obtained from specimens irradiated to 11% burnup. This information is favorable and indicates that the UO_2 particles operate at lower temperatures in a ferritic stainless steel matrix than in austenitic matrices. No irradiation damage of the ferritic stainless steel itself could be found.

As-Polished

500X

Etched

250X

FIGURE 5 Unirradiated UO_2 Particles in Ferritic Stainless Steel

KS-20582
Unclassified

Etched

500X

Coarse Grain Size of Cladding is the Result of Fabrication Practice. Etched

100X

FIGURE 6 Irradiated UO_2 Particles in Ferritic Stainless Steel

KS-20583
Unclassified