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REPORT NO: 7
FLAT BONDED FUEL ELEMENTS
Period June 11 - August 10, 1954
Technical Division

DELETED
VERSION

SR/H--856
DPW-54-19-7A
11-29-88

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Report No. 7

FLAT BONDED FUEL ELEMENTS

Period June 11, 1954 - August 10, 1954

SUMMARY

Improvements in rolling, beta transformation, and machining were made and provided higher yields of uranium flats suitable for bonding.

Several unsuccessful attempts were made by one aluminum fabricator to obtain sheaths with wall thicknesses approaching 20 mils in place of the current 30-mil thickness. The difficulties experienced were not considered insurmountable, and, in view of the desirability of having less aluminum in the fuel element system, experiments are continuing. Die and aluminum ingot quality improvements are being made.

Corrosion tests have continued on the uranium-nickel-aluminum system. Water temperatures of 95° and 100°C have been employed. The sheaths and bond layers have been perforated with holes varying from 15 to 60 mils in diameter. Results have been highly erratic, indicating some uncontrolled factor in the experiments. This may be a uranium hydride layer under the nickel.

Three plate elements with alpha-rolled and beta-treated cores were discharged from the MTR after satisfactorily withstanding irradiation at a mean exposure level of approximately 1500 MWD per ton. There was no detectable change in the length of the plates and no warp. Widths increased by an average of 6 mils with about 3 mils of this caused by tiny surface bumps.

Three plates with cores prepared by high-alpha rolling but not beta-treated showed prohibitive changes in dimensions after a mean exposure of approximately 580 MWD per ton in the MTR. Increase in length of the plates ranged from $3/4$ to $1-1/8$ inch, the thickness decreased by as much as 15 mils, and the width decreased by as much as 47 mils. Based on previous experience, the results indicate that control of rolling conditions was not adequate to provide an alpha-rolled structure, previously obtained with rods, that would undergo irradiation without dimensional instability.

I. URANIUM COMPONENTS

A. Metal Supply

From the 31 slabs rolled at Superior Steel on February 22, the final portion, consisting of 48 sections, 14' X 3-1/2", was beta-transformed satisfactorily at Atlas Steels Ltd., using again the conditions which had given good results on the earlier beta-transformation at Atlas. One modification of conditions largely eliminated a yield-reducing defect of the previous beta-transformation. This defect was the overall distortion at the lower end of the plate; it was overcome by attaching a weight to the bottom of the plate.

On July 28, a meeting was held at Superior Steel for the purpose of setting up arrangements to obtain the continuing supply of uranium plate, ready for electroplating, which will be required for the remainder of the plate development experimental program. Arrangements were discussed by representatives of Superior, OROO, SROO, and AED, of the steps necessary to convert unconditioned slabs through to plates ready for electroplating. This meeting resulted in tentative agreements whereby Superior would condition the slabs, roll the strips, and after beta-transformation at Atlas Steels, finish-machine the wrought core plate to specified dimensions. Implementation of this arrangement is not expected until after work on the next experimental lot of metal has been completed.

Detailed planning resulted in better controlled rolling conditions at Superior on August 3, where 50 slabs were hot-rolled to strips nominally 0.183" thick. For this rolling the edges of the slabs, as well as the flat surfaces, were conditioned by planing off the surface defects. Operating conditions provided a better salt cover during rolling and reduced the amount of water splashed from the bearings onto the metal surfaces. Because of the necessity for using in this rolling left-over slabs from another program and of somewhat less than the desired quality, the yield of satisfactory plates may not be much better than obtained on the previous rolling. Slabs were narrower and shorter than previously rolled slabs and resulted in much smaller allowances for edge-machining, scrap, and end cropping.

The conditions employed resulted in a definite improvement in flatness and camber over previous rollings, and by using closely-controlled rolling times and temperature the quality of the strips, except for the chemical and physical defects due to the slabs, was much more uniform than previously obtained.

About 60% of the strips came within the specification limits of $0.183" \pm 0.003"$ directly on rolling. Approximately 40% of the strips were over size in thickness. However, on pickling, Superior found that all but three strips were brought down to a satisfactory thickness. These three strips are to be cold rolled to tolerance and used to test the effect of limited cold rolling as respects structure and satisfactoriness of the metal on irradiation. The strips meeting dimensional specifications on rolling were cropped and sheared into sections 15' 6" long varying from 3" to 3-1/2" in width and then sent to Atlas Steels for vertical beta-transformation. The strips brought to thickness tolerance, either by pickling or by cold rolling, are also being sent to Atlas Steels as a second lot for beta-transformation.

B. Uranium Characterization

It was reported previously that, employing the uranium plate rolled at Superior on August 3, 1953, a schedule for testing the mechanical and physical properties of uranium plate was being developed. This schedule has been completed. It consisted of tensile, impact, bend, and hardness tests on representative specimens taken from both alpha-rolled and beta-treated metal. The tests were conducted at 75°F, relatively close temperature control being maintained because of tentative information that some properties changed rather markedly with small differences in temperature.

Physical properties of the as-rolled plate showed a pronounced dependence upon specimen orientation with respect to rolling direction. Subsequent beta-treatment practically eliminated this effect. Metal uniformity, as determined from all the data, was good.

Some of the results have been given earlier, but complete results are reported below:

Alpha-Rolled Metal

Impact Tests - Transverse impact tests yielded a mean value of 1.22 ft. lbs. The longitudinal specimens had a mean value of 1.79 ft. lbs.

Bend Tests - Transverse specimens had tensile elongations of 1% and deflections of 5 to 10 degrees. Longitudinal specimens had tensile elongations of 3 to 8% and deflections of 20 to 55 degrees.

Tensile Tests - Specimens of as-rolled material showed pronounced directional properties, as illustrated below:

	<u>Transverse</u>	<u>Longitudinal</u>
Average ultimate strength (psi)	87,000	105,000
Average yield strength (0.2% offset)(psi)	74,000	42,500
Average modulus of elasticity ($\times 10^{-6}$ psi)	25.8	17.5
Percent elongation	1-3	3-14

Hardness Tests - The hardness was uniform, with an average value of R_B 95.5.

Beta-Treated Metal

Impact Tests - The mean value for the impact strength of beta-treated uranium was 3.18 ft. lbs. There was no significant difference between longitudinal and transverse specimens.

Bend Tests - For beta-treated material the tensile elongation was 3 to 6 per cent and the deflection was 20 to 40 degrees. Within experimental accuracy there was no difference between longitudinal and transverse specimens.

Tensile Tests - The directional properties which were so pronounced in the as-rolled material were practically eliminated by beta-treatment, as shown by the data below:

	<u>Transverse</u>	<u>Longitudinal</u>
Average ultimate strength (psi)	84,000	84,500
Average yield strength (psi)(0.2% offset)	42,500	41,500
Average modulus of elasticity ($\times 10^{-6}$ psi)	21.8	20.7
Percent elongation	4-9	4-9

Hardness Tests - The hardness was uniform, with an average value of R_B 92.5.

An investigation was made to determine time and temperature limits that might be employed for beta heat-treatment of uranium plates of from 2-3 inches width and somewhat less than 0.2 inch thickness. Results showed that a satisfactory beta-transformed structure was obtained with salt bath temperatures of from 700-740°C employing heating times of 2-10 minutes and allowing the metal to cool in air after transformation. Further work is under way to establish the extremes of each variable that can be tolerated based on the effect in terms of grain size, hardness, mechanical properties, surface appearance, and metal loss.

II. ALUMINUM COMPONENTS

A. Procurement of Fuel Element Sheaths and of Process Tubes

On account of the judgment by aluminum vendors that it will be possible to produce the required sheaths only if porthole extrusion is used in place of the mandrel extrusion preferred from a metal reliability standpoint, evaluation has been under way of porthole closures in extruded sheaths made respectively by ALCOA, Revere, and Harvey. The examination of ribbed sheaths, made by these vendors and having the porthole closure in thick portions of the sheath cross-sections, revealed no significant metallurgical defects. No results from simulated service tests are available nor are non-destructive test methods for the regular examination of the weld sections for such defects as inclusions or non-welds.

Several unsuccessful attempts have been made by Revere to produce sheaths with walls less than the current 30 mil thickness and preferably of the order of 20 mils. Difficulties that have been experienced are not considered insurmountable and die and aluminum ingot quality modifications are being made in further experiments to obtain the thinner-walled sheaths.

B. End Closures for Fuel Plates

Based on earlier results favoring the hot forging technique as the method likely to produce satisfactory enclosures, investigations of this continued at ALCOA. The technique involves wrapping 3/4" wide 2S aluminum strips around the plate and upsetting under heat and pressure. ALCOA reports achievement of side and edge bonding between a 2S aluminum plug, the sheath, and the strip. Bond strengths of 5 specimens tested average 15,000 psi with a minimum of 12,000 psi. The sheath thickness was reduced 40% in the forging process. Metallographic examination showed the oxide layer had been broken up at the forged interfaces.

III. BONDING

A. Bonding Systems

Work is still being done only on nickel as the bonding layer, and the nickel is being deposited on rolled and pickled uranium primarily by electroplating. The thickness of the nickel deposits continues within the limits of 0.3 to 0.5 mil. It has been found at ERL that the amount of gas and water vapor removed from a sample degassed at 1-5 μ pressure at 525°C-550°C for 30 minutes increases in

the following order: bare uranium, uranium electroplated with nickel, and electroplated uranium that has been chemically stripped and replated with nickel. At BMI, work is in progress to identify the six compound layers (excluding the uranium core, nickel electroplate, and aluminum cover) that are recognizable metallographically in the polished cross-section of aluminum-clad, nickel-plated, hot-press bonded uranium cores; the first layer above the uranium, dark brown in color and not obviously metallic-looking, is of major interest, as it is suspected of contributing to the poor resistance of nickel electroplate to undercutting during corrosion testing. A single sample containing wrought nickel foil 3 mils thick which was employed as a bonding layer between uranium and aluminum, and bonded by pressing at [REDACTED], did not show this dark brown layer adjacent to the uranium.

Some difficulty was encountered during the report period with the nickel plating solutions. The nickel plate is darker than previously obtained and contains a thin coating of black sludge that can be wiped off easily. It is felt that this sludge may be the result of impurities in the salts used to make up the plating solutions. Purification of the bath was undertaken by operating it at low current densities in order to try to plate out unidentified impurities. Following this, the quality of the nickel plate appeared to be better. Some suspicion is also attached to the introduction of calcium and magnesium through the use of tap water for making up the baths, since experiments with distilled water show a much lower quantity of deposit and grayish rather than black color in what deposit there is.

The differences, noted in the last report, that were found between BMI and SRL results in corrosion testing assemblies, in which holes were drilled through the aluminum sheaths and nickel-plate bonding layers into the uranium cores may now have been explained. Initially, SRL had corrosion-tested samples in water contained in a cell that was sealed to the surface of the specimen; the assembly was maintained 95°C by means of an air bath. Under these conditions, there was no evidence of undercutting with some samples up to 22 days (528 hours). BMI has corrosion-tested samples under these conditions, and found no undercutting up to 35 days (840 hours). The size of the drilled hole in this test does not seem to be important. When on the other hand similarly drilled samples are totally immersed in water at 100°C, undercutting proceeds quite rapidly; at both SRL and BMI, a series of samples failed between 168 and 300 hours, while two are not appreciably undercut after 36 days (864 hours). Under these conditions the size of the drilled hole does make a significant difference;

the rate of corrosion diminishes markedly on going from 60 to 30 to 14 mil holes. The corrosion resistance seems to be a function not only of the method of testing and of the method of fabrication, but also of factors in each not yet understood. The sample mentioned in the preceding paragraph, made up with a wrought nickel foil bond 3 mils thick, has shown no indication of undercutting after 336 hours in water at 100°C. It shows a minimum of the brown layer noted above and tentatively identified metallographically as uranium hydride. The presence of this compound resulting from hydrogen in the metal from manufacturing or introduced during cladding and not removed in bonding is a plausible explanation of rather erratic corrosion results.

In a continuation of the study of the strength of bond between aluminum and nickel-plated uranium pressed at 510°C, 6000 psi, BMI has found that the bond strength is independent of time at temperature in the hot pressing time range from 0.5 to 5 minutes. Tensile tests made to determine bond strength by means of studs welded to the sheath over the bond showed a range of 10 to 13 X 10³ psi for this time range. The nickel-to-nickel bonds obtained by hot pressing nickel-plated aluminum to nickel-plated aluminum under the above conditions had a strength approximately 60% that of the uranium-nickel-aluminum bonds. Metallographic examination and chemical spot tests on the fractures of stud-welded tensile specimens of nickel-electroplated, aluminum-clad uranium bonded at 550°-575°C, 8000 psi, showed that in 28 out of 30 cases the fracture had occurred at the uranium-nickel interface.

B. Step-Press Bonding - Savannah River Laboratory

Ultrasonic testing of plates clad by step-pressing in 3-inch steps for [REDACTED] indicate complete bonding of the cores. Under SRL mechanical pressing conditions of [REDACTED] psi, stud-weld bond strengths between 52S aluminum end plugs and 2S aluminum sheath ran from 9 to 21 X 10³ psi on the flat surfaces, but edge bonding was poor; consequently the end-closure problem has not yet been solved.

Satisfactory results were obtained in the horizontal assembly of 14-foot uranium plates and ribbed sheaths, by clamping the sheath to a table and pulling the uranium through the tube. In order to press-clad flat plates with ribbed sheaths, grooved die inserts were installed on the forge press die; with these, several MTR-size uranium plates were successfully press-clad with ribbed sheaths at [REDACTED]. Satisfactory bonding was obtained except for some discontinuous lines about 1/16" wide

beneath the ribs. Difficulty was initially encountered in removing the clad ribbed plates from the die after pressing, because the plates became wedged between the fixed side of the die and the rib grooves; this was remedied by grinding a taper on the fixed side of the die, but the resulting larger die opening caused formation of a heavy flash at the edge of the plate with consequent poor edge bonding. The die is being redesigned to eliminate these defects.

C. Pneumatic Bonding - Engineering Research Laboratory

Metallurgical examination of small (1" X 6" X 0.2") plates pneumatically press-clad at the ERL showed that they were well bonded. No corrosion tests, similar to those made on mechanically press bonded elements have yet been made. They are desirable as metallurgical examination has not always correlated with corrosion results. Corrosion tests are being deferred until pneumatically-bonded MTR-size pieces are available. This is because of previous evidence at BMI indicating the unreliability of judging the satisfactoriness of the corrosion resistance of pieces of MTR size by the use of small ("half-dollar" sized) test pieces.

The uranium for the pneumatic program was nickel-electroplated at SRL and was pneumatically clad at ERL with and without prior degassing, at temperatures from [REDACTED] and at pressures of [REDACTED] psi for time periods of [REDACTED]. The end plugs were coated with about 0.08 mil of nickel, as bonding layer, by an evaporation technique. Bond strengths of 8000 to 19,000 psi were obtained between the core and sheath, and a strength of 20,000 psi between the end plugs and sheath. Metallographic examination of a polished cross-section from a specimen pressed under these conditions shows 7 distinct layers (including the core, nickel plate, and sheath); the intermetallic compound layers are thin, and have not yet been identified. The pneumatic chamber and holders for plates of MTR size have been completed.

IV. DEVELOPMENT OF TESTING METHODS

Further refinements were found to be required in the ultrasonic non-bond tester in order to increase the sensitivity and reliability, and to decrease the pickup of transients.

Attention has been given to the development of a method of chemical spot tests for determining impurities, such as iron, copper and uranium, on the external surfaces of the aluminum sheaths of completed fuel elements. Such chemical inspection, to study impurities in a large area of cladding, has been desired as a means of assuring optimum corrosion resistance of the sheath and as a supplement to autoclave testing. The initial experiments for achieving a simple method for testing a large area have been more successful than had been hoped. The method involves the deposition of a film containing an electrolyte and a "developer" or indicator to give a characteristic color for each of the impurities it was desired to detect. The following procedure was used:

- (a) The surface was degreased, then coated with a film by dipping into a solution of carboxymethylcellulose containing potassium nitrate (the electrolyte) potassium ferrocyanide (the indicator), talc and titanium dioxide (opacifiers). The opacifiers served primarily to lessen the transparency of the film and to increase the contrast of any color developed by the ferrocyanide. Gelatin was used originally as the film-forming agent, but the "CMC" was found more stable, more adherent, and more easily applied.
- (b) The film was dried, either with hot air or by vacuum.
- (c) The specimen was then immersed in a bath of potassium nitrate solution and connected to the anode of a dry cell. It was allowed to remain in the bath until the coloring developed.
- (d) The specimen was then dried at room temperature and examined. Iron inclusions were blue, copper inclusions were red-brown, and uranium at discontinuities was brown.

The results, although of a preliminary nature, are definitely encouraging but indicate the need for considerably more work before wide application will be possible.

V. FLAT FUEL ELEMENTS BY POWDER METALLURGY TECHNIQUES

At Sylvania Electric Products, Inc., studies designed to supply information for use in preparing 14-foot long bonded elements were completed early in the report period using 4-foot long "preforms". Temperatures of 600°C and pressures of 9-11 tons per square inch were established as optimum for satisfactory bonding. Successful bonds were obtained by hot pressing with an initial 14-inch long step (the full working area of the die) held for 10 minutes, followed thereafter by pressing with 12-inch long increments held for 5 minutes. Additional facilities required for the 14-foot long elements were completed. These consisted of a powder loading unit, 16 feet long, horizontal furnace designed to minimize temperature gradients from end to center of the furnace and an exit guide to the mill used to roll "preforms" in order to minimize camber introduced through rolling.

Several 12-to 13-foot elements were fabricated by the "preform" technique using the following procedure. Uranium powder was loaded in an argon atmosphere into carbon steel sheaths. After evacuation at 450°C, the assemblies were preheated at 600°C and rolled in four passes to about 95% of full uranium density. The pieces were reheated between passes. Following this densification, the steel sheaths were removed and the "preforms" ground and trimmed prior to nickel plating. The nickel-plated cores were fitted into end plugs and slipped into 2S aluminum sheaths. Following evacuation at 450°C, the assemblies were step-pressed using the optimum conditions described above. Employing the above bonding technique, the total length of the element was press-bonded in about two hours with pre-evacuation of the assembly requiring about four hours. The initial elements so prepared showed too great a variation in density and contour to be considered usable. Increased pulverizing of the uranium was employed to raise the density, and four additional elements were fabricated. Non-destructive tests showed the complete elements to be bonded. Destructive tests on these pieces are under way.

VI. FLAT ELEMENTS PRODUCED FROM WROUGHT URANIUM CORES

A. Fabrication of As-rolled Plates

Work on numerous details involved in firmly establishing the process for the assembly of mechanically step-press bonded wrought metal plates has been carried on, but no fundamental changes have been made in the process during the current report period. Vertical assembly of the electroplated cores into ribbed or unribbed aluminum sheaths with approximately 1/32-inch clearance appears satisfactory.

The most favorable end closure results have been obtained by forging. Good bonds result by step pressing at 8,000 psi and 1-3 minutes at 550°C, although lower values of temperature and time may be possible. Attention has been given to the facilities required to make a substantial number of 14-foot long elements for irradiation tests.

B. MTR Irradiation of Wrought Uranium Elements

Three plate elements with alpha-rolled and beta-treated cores which had then been mechanically press-bonded were discharged from the MTR after satisfactorily withstanding irradiation to a mean exposure level of approximately 1500 MWD per ton. There was a probable variation of 3 to 4-fold in exposure over the length of the plates. There was no detectable change in the length of the plates and there was no warp. The widths decreased by about 1/10% (approximately 3 mils). There were increases in thickness averaging about 6 mils but varying from 2-11 mils. The variation in thickness showed no apparent relationship to the degree of burnup. Part of the increase in thickness is caused by tiny surface bumps, averaging about 3 mils in height and uniformly covering the plates. There was no evidence of corrosion or other indication of defects developing which might lead to fuel element failure.

Three plates with cores prepared by high-alpha rolling but not beta-treated showed prohibitive changes in dimensions after a mean exposure of approximately 580 MWD per ton. Increase in length of the plates ranged from 3/4" to 1-1/8", while the thickness decreased as much as 15 mils and the width decreased as much as 47 mils. The plate nearest the core of the reactor was warped, bent, and twisted. This plate was so badly distorted that it was prevented from touching an adjacent fuel plate only by a ribbed spacer of aluminum. None of the plates exhibited noteworthy changes of surface or evidence of corrosion. The fact that the aluminum sheaths remained intact, in spite of the severe distortions that were encountered, is encouraging. Based on previous experience, the results indicate that the control of rolling conditions was not adequate to provide an alpha-rolled structure, previously obtained in other shapes, that would undergo irradiation without change in dimensions.

In view of the unfavorable results with the alpha-rolled plates, work was suspended on the preparation for MTR irradiation of plates made by alpha rolling followed by a 5% cold rolling. Work is continuing on fabrication and testing of alpha-rolled, plus beta-treated, plates which have been subjected to a 5% cold rolling following beta treatment. Information on the irradiation behavior of material so treated is desired because of the expectation that in the fabrication of any considerable number of plates, some treatment, similar in its effect on structure to a small amount of cold rolling, will be involved.

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