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REPORT NO. 2

FLAT PLATE BONDED FUEL ELEMENTS

Period Aug. 11 - Oct. 10, 1953

Technical Division

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Explosives Department - Atomic Energy Division
Wilmington, Delaware

MASTER

[redacted]

UNLIMITED

Report No. 2

FLAT PLATE BONDED FUEL ELEMENTS

Period Aug. 11 - Oct. 10, 1953

SUMMARY

Attention has continued to be concentrated on routes employing either wrought uranium or powder metallurgy product for the making of flat plate fuel elements of approximately 0.180" uranium metal core thickness bonded to either ribbed or ribless aluminum sheaths. Intermediate goals of the program are to have elements 18 inches long for MTR irradiation tests this fall and to make sufficient advance in the overall program in 1954 so that we can provide an initial reactor charge of 15-foot long fuels as early as possible in 1955. The development of a satisfactory process tube for retaining an assembly of several fuel elements is also required.

Uranium of satisfactory quality for fabrication into fuel elements appears to have been produced by the August high alpha rolling at Superior Steel, and it seems likely from the electroplating results that the metal can be employed for electroplating and bonding without such surface preparation as vapor blasting, grinding, or machining.

Nickel applied by electroplating to provide a thickness of not greater than 0.5 mil appears to be a satisfactory bonding layer system, and hot pressing is the preferred method for forming the bond, with step-pressing the method being given priority to make long fuel elements.

An initial lot of 14 powder metallurgy flats was received from Sylvania. By agreement with Sylvania these units will be used for destructive and non-destructive tests. An additional 20 pieces will be supplied for the MTR irradiation program.

Difficulty in obtaining aluminum components, both sheaths and process tubes, remains a bottleneck in the development program and specifically has delayed work on the wrought metal samples for MTR tests.

URANIUM COMPONENTS

At the end of the preceding report period a rolling of plate stock was carried out at the Superior Steel Corporation. Initial work with this product indicates that it possesses properties satisfactory for fabrication into fuel elements. The grain size is fine and uniform. The as-rolled surfaces are good and free of visible defects that might lead to unsatisfactory behavior on irradiation.

The metal surface preparation experimental program has been directed toward determining the most economical process for making finish-dimension pieces satisfactory for fuel element cores. Specific attention has been given to the suitability of as-rolled stock, etched but not otherwise surface treated, and of liquid-honed stock as bases on which to apply nickel electroplates. Attention has also been given to beta treatment conditions to provide a dimensionally stable structure on irradiation, with suitable handling to ensure minimum warping or other overall change in shape during the beta treatment step. The greater amount of satisfactory experience in reactors with beta treated stock makes this the first choice for irradiation tests, but portions of the metal are being retained for fabrication of the as-alpha-rolled metal into other irradiation test elements.

Tests of the acceptability of the economically preferred as-rolled surfaces given no treatment other than etching have shown them to be as satisfactory for nickel electroplating or for hot-dip treatment as liquid-honed surfaces, which are second choice from economic considerations.

Through arrangements made by NYOO, 52 plates (20" long x $3\frac{1}{2}$" wide and approximately 0.180" thick) from two billets of the August Superior Steel rolling were beta treated at Superior Steel. These plates, suspended freely in a vertical position on hooks attached to a beam, were beta treated several at a time by lowering into a lithium-potassium carbonate bath. They were held in the bath at 1350°F for two minutes. On removal from the bath they were cooled in the air in the vertical position. Minimum change in dimensions and minimum warpage occurred as a result of this treatment. Plate thickness increased by approximately 0.3 mil with an average increase in width of between 17 and 25 mils and a decrease in length of approximately 200 mils. Average camber was around 20 mils with a maximum bow after treatment of about 26 mils. The thickness range of the pieces after treatment was from 174 to 189 mils, but these dimensions represent, as stated above, increases of only 0.3 mil from the pre-beta treatment thickness. Metal was shipped to the Savannah River Laboratory (SRL) for nickel electroplating and cladding into MTR irradiation test plates. In view of the favorable results with the beta treatment procedure, NYOO was requested to arrange for similar beta treatment of most of the remaining 180-mil range stock from Superior Steel's August rolling.

This initial stock was chosen from billets having chemical composition and other properties in the center of the commercial metal production range. Request is being made to NYOO to arrange for another closely controlled, high-alpha rolling by Superior, using stock that will represent the limits of chemical composition and of other characteristics expected for 1954-55 production metal. In contrast to the first rolling this may include not only virgin metal but also recast scrap and possibly reworked uranium.

ALUMINUM COMPONENTS

Except for the provision by Alcoa to Sylvania of MTR sheaths for powder metallurgy cores, little progress has been made on the aluminum component program. Fabrication of unribbed aluminum sheaths for the wrought-metal-core MTR test has been prevented by a series of technical difficulties experienced by Alcoa in both extrusion and drawing operations. These have included tearing and wrinkling of the thin (30 mil) sections of the sheath adjacent to the heavier edges and failure to fill out the shape to the specified width.

Sixteen 2-foot sections have been shipped to the Savannah River Laboratory. These were recognized by Alcoa as under width with somewhat poor surfaces due to irregular "ribs and grooves", uneven wall thickness, and distortion from the desired rectangular cross section, but shipment was made at AED request in order to have some sheaths available for the cladding program. They will be used in press cladding of nickel plated cores for MTR tests with the hope that the defects will be minimized in the pressing operation.

No progress has been reported by Alcoa in the ribbed sheath design or in the several process tube designs ("B" process tube, a 1-piece extrusion with 2 diaphragms, to be used with ribbed sheath tubing; "C" process tube, a 2-piece design consisting of a square tube with corrugated edges, to hold ribless sheaths and to be drawn down tightly on the sheaths after assembly; "D" process tube, a 2-piece design consisting of a square and a surrounding round). In view of the difficulties and delays experienced with the aluminum sheaths and process tubes and with the necessary dies for producing these items, effort has been made to find other fabricators for at least some of the designs. Preferred fabricators appear to be Revere Copper and Brass and the Harvey Machine Co., Inc. Discussions have been held with Revere, and Revere personnel have expressed a belief in their ability to fabricate aluminum components and an interest in doing so.

Ribbed fuel element sheaths have always been preferred in view of the apparent advantages the ribs would provide, particularly with respect to controlled separation of the fuel elements in the process tubes. The non-ribbed designs, although less desirable, have been followed on account of the forseeable difficulties ribbed sheaths would cause in the bonding and the testing of fuel elements. The difficulties experienced by aluminum fabricators to date in making non-ribbed thin-walled designs have caused them to express concern as to greater difficulty in providing ribbed sheaths. In view of these facts, attention has continued, but with more emphasis, on the application of ribs to the otherwise completed fuel element. Optional methods of providing the ribs are by rolling (Metals and Controls Corp. - M&C) or by ultrasonic bonding (Aeroprojects, Inc.).

The M&C joined ribs of 2S aluminum to 2S aluminum sheet by rolling with a grooved roll at 800 to 1000°F. This technique produced a strong bond that is expected to have satisfactory corrosion resistance. The pieces are under corrosion test. Facilities were not available in the first test to provide ribs of the required size and shape. Additional tests are being made joining ribs of the required dimensions to aluminum sheet. The rolling of ribs onto otherwise completed fuel elements is being programmed on the assumption that rolling conditions will not damage the bond or adversely affect the metal structure.

Work has been continued by Aeroprojects on the ultrasonic soldering of solid ribs to aluminum sheet. A series of solder compositions have been used, such as 85% tin-15% zinc, 95% zinc-5% aluminum, 99% tin-1% magnesium. In corrosion tests with water close to the boiling point widely varying results have been obtained with each solder system, with some excellent showings and some poor ones. It is suspected that the poor results are not due to the corrosion resistance of the solders but to the incompleteness of the wetting and soldering. Solid ribs of modified design and others with the rib edges covered with sprayed aluminum are under test. Definite improvement in duplicability of results was being obtained at the end of the report period. In view of the favorable indications with rolled-on ribs, ultrasonic soldered ribs are considered as of second priority but as warranting continued effort for the time being, due to the need to follow up any reasonable lead on means of providing ribbed fuel elements.

BONDING

Research on bonding continues to concentrate on (1) bonding systems and (2) processes for employing the systems.

Systems that continued to receive attention during this period are nickel, copper, and chromium by electrolytic deposition; AlSi by hot-dip; and chromium by non-electrolytic methods. Processes in order of priority are hot pressing, including step-pressing; roll cladding without deformation of core; rolling with simultaneous deformation of core and sheath but using conditions that would avoid adverse uranium structure; ultrasonic soldering; and extrusion.

Nickel by electrolytic deposition is the preferred system and is the only system being given attention for the MTR plates. Nickel appears to be deposited controllably and uniformly within a range of 0.2 mil for the preferred maximum thickness of 1/2 mil. Methods for determining thickness in this range to 0.1 mil have been developed. Battelle Memorial Institute (BMI) finds that a thickness of 0.2 mil is sufficient to stop diffusion of uranium at 300°C for at least 100 hours. Assuming that bonding conditions can be worked out to maintain most of the nickel as such, and this appears likely, a goal of 0.3 mil thickness has been set for future attainment. In general, nickel coats have behaved satisfactorily as bonding layers, but there have been some cases in which for an undetermined cause the nickel adheres to the aluminum and fails to adhere to the uranium. Full scale nickel plating of 18-inch length uranium cores required for MTR test pieces has begun at the SRL.

The nickel-copper system will be considered only in case bonding experiments on a larger scale show that this is necessary to provide a greater reproducibility than can be obtained with nickel as indicated in some small scale experiments at the BMI. Hot dip bonding using aluminum-silicon is being suspended because of the process difficulties evident in this process even with short lengths of flat plates and the likelihood of much greater problems with long lengths. Several advantages of chromium over nickel, chiefly the lower loss of neutrons by parasitic capture for equal thicknesses of chromium and nickel, caused continued attention to be given to this system, employing electroplating methods at the BMI and non-electrolytic methods at National Research Corporation (NRC). In small scale tests the electroplated chromium has been found to bond satisfactorily although requiring somewhat more severe conditions than are necessary for nickel. Interest in the non-electrolytic chromium process has been maintained because of the information from NRC as to speed and stated simplicity of the process and its stated provision of a dense corrosion-resistant coating. The bonding qualities of a chromium coating so provided are to be tested.

Considerable attention has been paid by BMI to relative effectiveness of various bonding layers in delaying corrosion of the uranium core. Tests have been carried out in the bonded systems by boring small holes through the aluminum and bonding layer and into the uranium core, then exposing the pieces at near-boiling temperatures. Effectiveness in delaying corrosion has been measured by determining the area of uranium corroded in a given time. It has been found that the uranium corrosion takes place at the boundary of the bonding layer rather than down into the body of the uranium itself. Although nickel has been shown to be excellent in these tests, the BMI has been concerned over the location of the corrosion at the boundary with nickel. BMI believes that an incubation period is primarily reponsible for the difference in corrosion rate with different bonding layers and has proposed a program for more thorough investigation of the way in which corrosion takes place.

Hot pressing of nickel bonds has been chosen for making the MTR plates. The 18-inch die for making the wrought metal MTR test plates is in operation at the SRL. After preliminary adjustment it is giving excellent control of temperature on all parts of the pressing. Conditions for pressing are approximately 5000 psi at 450-500°C for 10 minutes. Preliminary examination indicates that good bonds are obtained.

Initial results obtained by Sylvania in step-pressing uranium powder indicate this is an attractive method for making long fuel elements. Steps have been mainly 4 inches, but up to double that length has been used. Time at temperature has been as low as 10 seconds, with some indication that this may be too short but with 30 seconds being more than ample. Dies have been ordered to enable carrying out an investigation of step-pressing with wrought metal at the BMI and at Savannah.

Larger scale tests at M&C undertaking to produce bonds by roll cladding and with deformation of the aluminum without deformation of the uranium core did not duplicate the favorable results obtained at the BMI on pieces 1x1" in area. Confirmation of the difficulty with the larger pieces has been obtained by BMI. In 1x1" pieces nickel plated cores were satisfactorily bonded to aluminum sheaths with as low as 5% pack reduction and without uranium reduction below a pack reduction of at least 12%. Scale-up to pieces 3x1" brought a lowering of the critical per cent pack reduction without deformation of the core to approximately 3%. It appears possible that pieces with larger dimensions will result in still further lessening in the critical pack reduction without deformation of the uranium, but a series of experiments is under way to determine whether rolling conditions can be found that will raise the value with larger dimensional areas. Nothing has occurred to change the view that any considerable amount of deformation of the uranium core is undesirable. Information is lacking on the effect of very low percentages of deformation. Because of the attractiveness of the method as a means of cladding, search is being made with larger size pieces for conditions which will provide bonding with less than 5% deformation of the core.

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FUEL ELEMENTS BY POWDER METALLURGY TECHNIQUES

Early in September Sylvania provided 14 MTR-size flat plates, all of which had passed preliminary non-destructive tests made by the vendor. The majority of these, however, had one or more characteristics rendering them open to question for use in MTR test. In each case the characteristic was such that it was considered that a small amount of additional work could eliminate it and provide material with properties felt desirable for irradiation tests. A new lot of 20 samples is being prepared and is expected for delivery shortly after the end of this report period. Changes that will be made include reduction in thickness of the nickel layer from values in the range of from 1 to 3 mils down to 1/2 mil and reduction or elimination of the nickel powder placed in the original samples at the end of the uranium powder section. Nickel pressing characteristics are believed to have been the cause for the marked thinning of the aluminum sheath during pressing at the place where the sheath came into contact with the nickel powder. In some cases this resulted in reducing the sheath almost to paper thinness. In the preliminary samples graphite had been used as a parting compound in the pressing operation, and a metallurgical examination disclosed it had not been possible to remove it completely. Alumina powder or colloidal silica appear likely to serve as satisfactory parting compounds without introducing the difficulties caused by the graphite. There were some blisters in the samples, believed to have been caused by gas absorbed by the aluminum powder used at the top closure of the powdered metal MTR piece, and it appears that this can be removed and the blisters avoided. Two properties appeared that were not expected and for which the cause has not been definitely established. One was a rather marked variation in thickness occurring irregularly along the length of the piece; the second was a dimpling, in an irregular pattern, of the sheath. Inasmuch as there was some question on the quality of the aluminum employed in making the sheaths, the dimples may have been caused by local concentrations of impurities or to imperfections in the die faces. Again, these are difficulties which further experience should remove.

The Sylvania powder metallurgy pieces are being given the usual non-destructive tests at SRL. These include radiography to determine the location of the core in the sheath and characteristics of the closure, particularly at the upper end where there is nickel powder and considerable amounts of aluminum powder. Frost and ultrasonic tests are being used to determine completeness of bond. Some unbonding appears to be present but it is not too general. Ends have been welded to some of the powder metal plates without difficulty except where the closure was made by pressing the aluminum powder. In this case the weld appears to be somewhat brittle and weak, indicating need for consideration of other means of closure than the present aluminum powder pressing.

Results have been obtained from the preliminary corrosion testing of typical wrought metal versus powder metal of varying densities from 18.87 down. Under test conditions the highest density powder metallurgy product showed a corrosion rate of the same order as the wrought material, though somewhat greater. With lower densities, corrosion rate increased regularly and relatively rapidly, showing the need for maintaining high density powder compacts. (It had been thought possible that for Savannah River low density product might be

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satisfactorily used, with attendant simpler conditions for obtaining powder metallurgy product). Under test conditions powder metallurgy product with a density of 18.87 showed weight losses of 5.5 mg/cm^2/hour. Toward the lower end of the density range tested a sample of 17.61 density showed a weight loss of the order of 100 mg/cm^2/hour.

The original lot of powder metallurgy product plates is being used generally at Savannah to work out the techniques of making MTR test pieces by welding ends to them and of using the destructive and non-destructive tests, which include autoclaving. This should enable the lot of 20 pieces, agreed on to be provided by Sylvania for irradiation, to be made up into MTR pieces and tested promptly on receipt at Savannah.

TESTING

Non-destructive tests programmed for flat plates include: quantitative tests for the transformation to beta phase of alpha fabricated metal; a means of determining the thickness of electroplates, especially nickel; tests for the completeness of bonding both by ultrasonics and the frost test; determination of the sheath thickness generally and detection of thin spots in aluminum sheaths; and determination of a number of characteristics by radiography, including particularly the soundness of welds and freedom from flaws of end sections beyond the lengths of the uranium core. A non-destructive test considered desirable, but not yet formulated as to method, is a means of determining by characteristics of structure that wrought metal has been rolled under proper high alpha temperature conditions. Progress on some of these tests has been mentioned earlier in this report.

Ultrasonics proved of little value in the detection of thin spots in aluminum cladding, i.e., less than 15 mils, and attention is being given to detecting thin spots by use of auto-radiography, but the nickel bonding layer may present difficulty in the use of this method.

On flat plates tested so far, ultrasonic results were identical with those revealed by frost test. Non-bonded areas of 1/16" in diameter appear to be reliably detected.

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