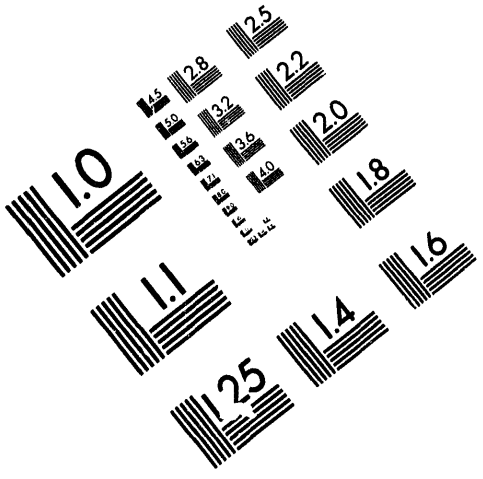

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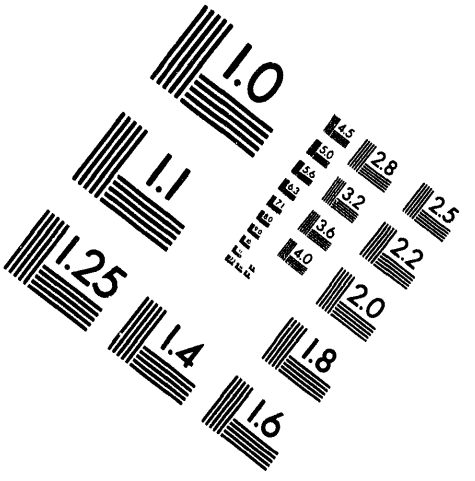

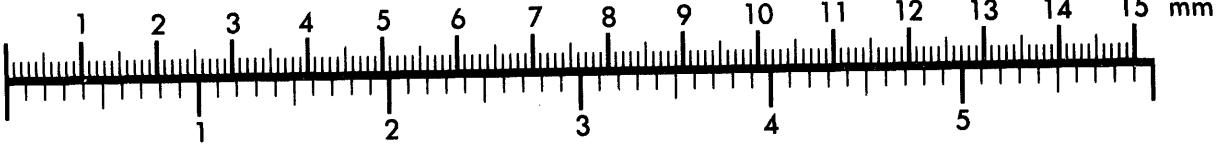

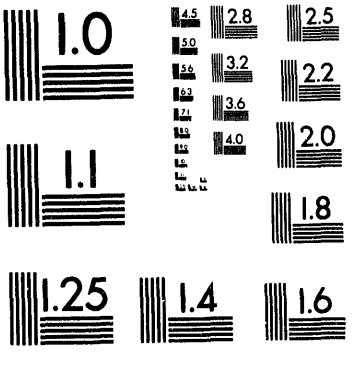

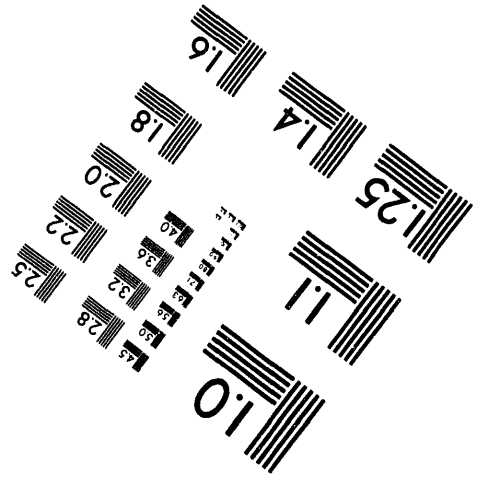

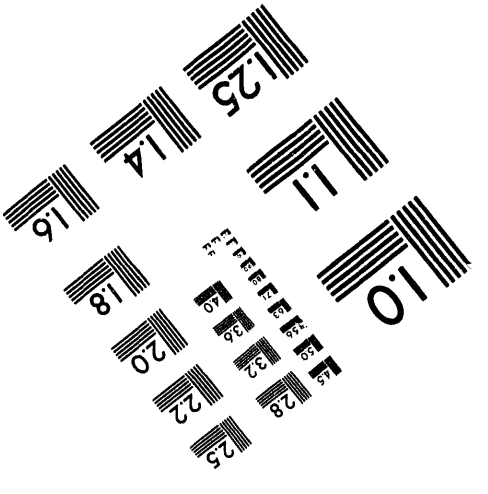

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POTENTIAL LIMITATIONS OF AL-SI BONDED FUEL ELEMENTS

W. H. Hodgson

Engineer, Process Engineering

Fuels Preparation Department

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POTENTIAL LIMITATIONS OF AL-SI BONDED FUEL ELEMENTS

INTRODUCTION

Tests in which aluminum-jacketed, Al-Si bonded uranium fuel elements were baked at various temperatures have shown there is a time-temperature relationship for Al-Si layer decomposition. For heat transfer and secondary coolant barrier considerations, the extent of bonding layer deterioration during fuel element irradiation is important. Under present reactor operating conditions, Al-Si bonded fuel elements show evidence of internal bond deterioration, and to a lesser degree, external bond deterioration following irradiation. Such evidence has aroused concern for the ability of the Al-Si bonding layer to withstand future reactor operating conditions.

Currently, several potential uranium fabrication and canning process improvements are being developed to further advance fuel element stability and performance. Optimization of process conditions based on these improvements may provide the necessary margin of safety for good bond layer integrity during irradiation. Before a decision can be made to continue improvement of the present process or convert to a new canning process, more information on the stability of the present fuel element bond is needed. The purpose of this report is to summarize data derived from past and present testing and to recommend courses of action to more fully evaluate Al-Si bond integrity under anticipated future reactor operating conditions.

SUMMARY

The Al-Si jacket-to-core bond consists of several parts. As illustrated in Figure 1, the bond consists of: (1) a homogeneous aluminum to Al-Si juncture, (2) the Al-Si layer and (3) the heterogenous Al-Si compound layer-uranium juncture. The compound layer actually consists of several zones or layers of intermetallic compounds of aluminum, silicon, and uranium. These layers are relatively brittle and if laminar, are subject to brittle fracture under relatively small tensile, shear, or compressive forces. For most fuel elements, the strength of the bond is dependent on the strength of the compound layers.

A series of baking and jacket-peel tests have been utilized to measure the relationship of bond deterioration with temperature and time. Since, under reactor operating conditions, the bond is probably in compression,[1] more realistic tests involving high temperature-pressure autoclaving and jacket-peeling have been conducted. More recently, to obtain a more quantitative measure of bond strength, a stud-pulling

[1] Personal Communication - K. Merckx. May, 1960

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technique has been developed and is currently being utilized extensively. Alternate methods of measuring heat transfer from core to jacket qualitatively and quantitatively are under development. The results of these tests and pertinent in-reactor production tests are summarized below.

Limited bake-test data have indicated fuel elements which possess a good bond prior to baking suffer little or no deterioration in over 50 hours at temperatures up to 250 C. More extensive data at temperatures in the 300-400 C range indicate the diffusion of aluminum into the uranium and formation of UAl_3 compound is acclerated with increased temperature. Results of a recent test conducted by Jacky[2] are included in Table I in the appendix. Formation of small voids in the compound layer accompanied the diffusion of the aluminum. Decreases in bond strength, possibly due to depletion of Al in the layer adjacent to the Al-Si, resulted from baking at high temperature and pressure (1000 psi and 450 C). Under these conditions, extensive diffusion of aluminum to form UAl_3 and void formation in the original compound layer were both observed.

Visual inspection of irradiated fuel elements in the reactor basins and in the Radiometallurgy Laboratory has shown that most fuel elements, which failed as a result of a hot spot, had incurred some bond separation between the Al-Si and uranium under the heated area. An example is shown in Figure 2 in the appendix. Ultrasonic testing at the C basin of irradiated fuel elements having hot-spot patterns indicated the presence of a hot spot did not necessarily indicate porosity in the bond under the hot spot. A production test specifically designed to evaluate the uranium - Al-Si bond deterioration during irradiation with bond temperatures in the range 230-250 C indicated no serious bond defects or stability problems; however, the number of pieces examined was small and would indicate only gross changes in bond quality.

Examination of four enriched (1.47 percent U-235) self-supported fuel elements irradiated to 850 MWD/T exposure at 75, 86, 88 and 110 KW/ft. average specific powers was recently completed in the Radiometallurgy Laboratory. Extensive bond separation, 50-90 percent, was evident on the I.D. of all cores, as shown in Figure 3, whereas broken bonds extended 30 to 50 percent of the distance around the O.D. of the cores. The actual cause of the bond separation may not have been irradiation damage since impact during discharge or unusually rough handling are possible causes. Metallographic investigation of the unbroken bond as shown in Figure 4 indicated adequate UAl_3 formation and good compound layer integrity. The broken bonds bore evidence of less extensive UAl_3 formation. Figure 5 is included to show the estimated relationship of fuel element bond temperature to fuel element specific power.

2 HW-63636, "Interim Evaluation of Nickel Plate on Aluminum Jacketed Fuel Elements". G. F. Jacky. February 8, 1960. (Secret)

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CONCLUSIONS AND RECOMMENDATIONS

Most bond layer studies conducted to date were for the expressed purpose of determining the rates and extent of uranium and Al-Si diffusion. Dissociation of the uranium - Al-Si bond under elevated temperature and pressure conditions has been of secondary interest. As a consequence, little is known of how the stability of the bond during irradiation is related to fabrication techniques. Further information is required to advance fuel element bond technology and to establish the potential effects of reactor power and exposure on bond quality. Irradiation testing must include intermediate and high-exposure irradiation of self-supported standard and optimum quality fuel elements at present and future specific powers. This test[3] may best be accomplished by careful preparation and irradiation of fuel elements of natural, 0.94 percent and 1.47 percent enrichment, having normal and optimum quality jacket-to-core bonds, utilizing all compatible process improvements known to improve bond strength, toughness, and soundness or freedom from porosity.

At the same time, an attempt should be made to improve laboratory testing to more nearly duplicate in-reactor conditions. For instance, it has been hypothesized that the elasticity of aluminum and Al-Si assures plastic deformation of the jacket in preference to bond fracture during expansion and contraction. The similarity of expansion coefficients for aluminum, Al-Si, and uranium and the differential in average temperatures of Al-Si and uranium during reactor operation cause the bond to be in compression except during shutdown. To simulate reactor power and temperature conditions, testing should be conducted in an autoclave environment. A set of temperature and pressure conditions which would approach bond temperature and possibly pressures during reactor operation would be an autoclave test with pressures in the range 100 psi to 325 psi gage (165 C 220 C). The pressure achieved in this test would probably be less than experienced in reactor, but would be more representative than unrestrained baking in an oven. This test, however, would not measure the effect of irradiation damage on the Al-Si, compound layer, or uranium. Laboratory testing should include extensive stud-pulling and metallography of carefully prepared samples (jacketed fuel elements) assembled by present and improved canning techniques withdrawn before and during autoclave tests covering an approximately 100-to-200 day exposure period. Thermal cycling of fuels during reactor shutdown and startup could be duplicated by cycling with alternate hot and cold water during the autoclave test.

3 HW-65870 A "Design of Production Test IP-344-A-FP, Determination of the Limitations of the Al-Si Process". W. H. Hodgson and M. A. Clinton. To be issued. (Secret)

DISCUSSION

For several years programs have been underway to improve the strength of the Al-Si bond obtained from the lead-dip canning process. During this period, improved uniformity of bond strength, and continuity have been achieved through numerous canning process improvements. From time to time, however, concern about the stability of the Al-Si bond at proposed reactor power and exposure goals has led to evaluation of alternate cladding techniques including: hot-press canning,[4] cable cladding,[5&6] and vacuum canning.[7] In each instance, however, improvements to the Al-Si process were either adequate to improve the Al-Si bond and yet retain the competitive position of the Al-Si process, or certain deficiencies were encountered in the alternate fuel element cladding process which were of a magnitude sufficient to preclude further evaluation without large monetary expenditures.

In view of the proposed IPD program for increases of reactor power and coolant temperatures over the next five years,[8] and with no new alternate fuel element cladding techniques under study, the necessity for initiating new studies in FY 1961 depends on the stability of the Al-Si bond under the projected conditions.

To date, a single production test has been specifically conducted to measure the stability of the Al-Si bond under irradiation.[9] All other bond quality tests have been conducted out of reactor. There has been no correlation between out-of-reactor and in-reactor tests.

The primary purpose of the bond between the jacket and the core of a fuel element is to permit uniform heat transfer from the core to the coolant. Lack of a good bond, or only partial bonding, may lead to non-uniform heat transfer, high local jacket temperatures and possible fuel element rupture due to excessive corrosion of the jacket material. If it is assumed that a fuel element is operating

4 HW-61995, "Specifications, Laboratory Hot Press Process for "C" Size I&E Fuel Elements". J. C. Tverberg. 9/25/59. (Secret)
5 HW-56801, "Extrusion Cladding Uranium with Aluminum Using the 'Schloemann' Cable-Cladding Press-Mechanical Aspects". G. F. Jacky. 7/21/58. (Unclassified)
6 HW-56802, "Evaluation of Extrusion Clad Hanford Fuel Elements - Metallography, Corrosion and Jacket-Core Bonding". G. F. Jacky. 8/25/58. (Confidential)
7 HW-57692, "Evaluation of Vacuum-Canned and Hot Press Fuel Elements PT-IP-44-A and IP-45-A". E. A. Smith. 10/15/58. (Secret)
8 HW-62862, "Plant Improvement Program, Irradiation Processing Department". A. B. Greninger. 4/1/60. (Secret)
9 HW-58929, "Production Test IP-229-A, Evaluation of the Uranium - Al-Si Bond at High Temperature". W. K. Kratzer. 1/10/59. (Secret)

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under ideal conditions of perfect alignment and coolant flow, sound core to jacket bond, a "crud" layer temperature drop of 60 C, and a specific power of 80 KW/ft., from Figure 5 it is estimated the bond temperature will be within the 80 to 98 C range above the bulk coolant temperature. Alternate bond temperature conditions for specific powers from 40 KW/ft. to 120 KW/ft. can be estimated from the curve.

Under present reactor power and temperature conditions, misalignment and resultant high-jacket temperature which induces hot-spot formation, probably contributes to dissociated or broken compound layers under virtually all hot-spot areas. Location of unbonded areas, however, may not be limited to hot spots.[10] Production tests[11, 12] to determine the relationship of braze porosity to the incidence of hot spots on both solid and I&E elements were irradiated. A brief summary of these tests is given below.

PT IP-68-A-90 FP

Solid eight-inch fuel elements having large voids, small voids, and hot-pressed pieces having no voids were exposed to 750 MWD/T at specific powers up to 50 KW/ft. Data from the test indicated: (1) there was no significant difference between the numbers of hot spots and hot areas which occurred on the best acceptable, barely acceptable, and void-free fuel elements classified by autoradiography; (2) only four of 43 hot spots correlated with void and porosity areas shown on the autoradiographs; (3) there was a significant correlation of hot spots to warp of the same or upstream adjacent fuel elements, and (4) location of half the hot spots in one-fourth of the tubes strongly suggested misalignment or distortion was a contributing factor in causing hot spots.

PT-IP-210-A-1-FP

Enriched (0.94 percent U-235) six-inch, I&E fuel elements were classified as to bond quality by production bond testers into the categories premium, borderline (slightly better than reject), and reject, to compare incidence and severity of hot spot genera-

10 HW-56028, "Ultrasonic Bond Testing of Irradiated Fuel Elements". K. P. Bokish. May 19, 1958. (Secret)

11 HW-49559 D, "Final Report on Comparison of Void-Free Fuel Elements With Standard Production Fuel Elements, Production Test IP-68-A-90-FP". W. H. Hodgson. June 15, 1959. (Secret)

12 HW-57645 F, "Final Report on Comparison of Bond Quality with Reactor Performance of Enriched I&E Fuel Elements - Production Test IP-210-A-1-FP". W. H. Hodgson. January 15, 1960. (Secret)

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tion during irradiation. These columns were exposed to 800 MWD/T at specific powers up to 80 KW/ft. A total of 90 of the 1020 fuel elements exhibited hot spots following irradiation. Approximately 70 percent of the hot spots were associated with misalignment or warp of the elements in the tubes. The remaining hot spots indicated bond quality may be a contributing factor, but the data were not conclusive. Hot spots did not correlate with the braze voids. It is possible, however, that brittle bond or non-wet conditions which may be detected by bond testers and were not detected by autoradiography, were present in some borderline and reject pieces and resulted in severe hot spots in those categories.

PT IP-229-A[9]

This production test was conducted to determine specifically, if possible, the changes that occur in the uranium - Al-Si bond during irradiation at bond temperatures between 255 C and 285 C. Twenty-five (25) X-8001 aluminum alloy clad dip-canned depleted (0.14 percent U-235) uranium solid fuel elements, having removable supports attached to each end were irradiated to approximately 500 MWD/T goal (40 days) in high temperature water. Bond temperatures were calculated to be in the 230 C to 250 C range. Difficulty in removing the supports prevented bond testing of more than a few of the elements following irradiation. Those tested indicated no serious bond defects or deterioration.

Little is actually known of the relationship of unbonded areas to hot spots. The presence of severe corrosion in the center of a hot spot indicates good local heat transfer from the core to the jacket exterior where the aluminum corroded rapidly. The high rate of local heat transfer is suspected as a cause of local bond deterioration under the hot spot. Corrosion in the center of the hot spot is not expected to be severe if the fuel element bond under the hot spot is fractured or separated before irradiation, since maximum heat transfer would be through the bonded zone on the periphery of the separated bond.

Attempts have been made to classify the quality of the bond by various mechanical strength tests: peel testing; stud pulling; bake and stud pulling; and high-pressure autoclaving with peel testing. Concommitant and separate metallography of bonds has shown that bonds which have high tensile strength may not possess good bond toughness. In this instance, toughness means the ability to resist brittle fracture due to thermal shock or cycling. Little is actually known of the characteristics of a bond which will withstand irradiation treatment best. There appears to be a distinct difference between a strong bond as demonstrated by baking and stud pulling and a strong bond as shown by thermal cycling and metallographic inspection. The difference may be of importance to the reactor performance since during operation the

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jacket temperature is normally determined by a combination of core and coolant temperatures and is low in comparison to the core temperature.

The method of fuel element quenching following canning and the elasticity of the cladding metal, combined with the differences in thermal expansion coefficients indicate the bonding layer should be in a state of compression following quench. During reactor startup and operation, the bond should remain in compression. However, during shutdown, especially in the case of a reactor scram, the bond may be in tension. Calculation of the pressures exerted on the bond under most severe reactor condition changes and during quench following canning show the quench conditions are by far the most severe. In no case, however, will the quench or the changes occurring in-reactor approach the stress applied in stud-pulling tests.

The effects of irradiation on the physical properties of uranium are familiar. Uranium becomes brittle and more subject to fracture from thermal shock. If the same holds true for the compound layer which contains compounds of uranium, aluminum, and silicon, intermittent and extensive thermal cycling during irradiation may contribute to or ultimately cause fracture of the bond. And the possibility exists that brittle fracture of the uranium adjacent to the UAl_3 diffusion zone may occur.

To achieve conditions which would simulate reactor operation, limited testing has been performed in high-pressure autoclaves.[13] In this fashion, the compressive forces and high temperature at the bond layer during irradiation were produced. Post-autoclave bond conditions were studied, using metallography and peel testing. Areas of can-wall collapse noticed after 800 hours at ~ 450 C and 1000 psi probably resulted from poor mechanical characteristics of the thickened intermetallic compound layer (brittle and subject to failure by stress, or friable) and from the formation of diffusion voids at or near the primary aluminum in the Al-Si (Kirkendall effect). Although 450 C operation with the bond under compression for limited time (several hundred hours) may be tolerated by fuel elements, it appeared from peel tests that longer exposure may have altered the bond sufficiently to sensitize it to mechanical or thermal chock. Long-term exposure in-reactor at 300-350 C with minimum thermal cycling may be feasible.

In baking tests which were used to improve the character of chemically deposited nickel plates, the growth of UAl_3 at temperatures above 200 C was confirmed; however, since UAl_3 has been shown to be one of the more ductile compounds formed during canning, increased quantities of this compound may toughen the bond with some loss in tensile strength.

13 HW-56300, "Progress Report, High Temperature - High Pressure Bake Testing and Diffusion Studies of Aluminum Clad Fuel Elements". J. W. Gifford and R. G. Wheeler, 7/25/58. (Declassified)

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Recent use [14,15] of the stud-pulling test to evaluate the effect of silicon concentration on bond strength of standard production fuel elements indicated the bond strength on the cap end (normally quite brittle) varies inversely with the Si concentration in the range studied (8.5 to 10.5 percent Si). Thickening of the UAl_3 layer was the most significant change in the bonding layer, as determined by metallography. In spite of the improved elasticity of the bond with increased UAl_3 formation, high exposure causes deterioration of the bond. Reasons for this phenomenon are not fully understood. Examination of enriched self-supported I&E elements irradiated to 850 MWD/T at specific powers of 70, 86, 88 and 110 KW/ft. show broken bonds in from 50-100 percent of the spire compound layers and 20 to 50 percent of external compound layers. Under 300X to 750X magnification, broken bonds were observed in the compound layer. The actual cause of the broken bonds in these pieces is not known since impact in the basin following discharge or unusually rough handling are possible causes. Adjacent uranium did not appear to contain diffused aluminum, a condition which has been associated with good bonding in out-of-reactor bond studies.[16] The cause of broken bonds in work reported by Goffard[13] may have been the layering of U-Si compounds in the compound zone which was depleted by diffusion of Al into the uranium.

Another possible explanation for fracture of the UAl_3 rich compound layers mentioned by Goffard is related to the stability of thin brittle films. It is known, for instance, that thin brittle glue films withstand fracture under more severe conditions than thicker films. Since Al-Si - Uranium compound layers are brittle in relation to the aluminum, uranium and Al-Si layers thickness may be a determining factor in the incidence of fracture. If, for instance, during exposure to long-term high-temperature conditions, the UAl_3 layer and total compound layer become thicker, the susceptibility to brittle fracture increases, notwithstanding the relatively better ductility of the thicker UAl_3 layer.

Since there is no out-of-reactor test which will demonstrate the ability of the Al-Si bonding layer to withstand future irradiation conditions, a small-scale irradiation test under closely controlled conditions appears to offer the best answer to the question of Al-Si limitations. Such a test would involve closely monitored irradiation of standard and carefully prepared optimum quality fuel elements of varied enrichment levels to intermediate and high-exposure goals, with complete characterization of the condition of the bonding layer before and after irradiation. The optimum quality fuel elements would be fabricated by the standard process, incorporating, however, process improvements to

14 Personal Communication - C. A. Strand.
15 Undocumented, "Bond Strength Evaluation of the Brittle Bond Problem in Production Fuel Elements". J. C. Tverberg. 11/10/58. (Confidential)
16 Personal Communication - C. A. Strand.

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achieve the following:[17]

a. Improved component cleaning and vibration of the components in the canning baths to facilitate wetting and reduce braze porosity.

b. Utilization of optimized canning assembly process for maximum braze integrity and strength.

c. Isolation of the effects of sources of hydrogen pickup in canning baths for tighter braze porosity control.

d. Utilization of pressurized quenching of assembled fuel elements to minimize braze porosity.

e. Modification of ultrasonic bond testing equipment to improve reproducibility, including sensitivity to non-wets and fractured or brittle bonds.

The optimum quality fuel elements would have bonding layers representative of the quality expected to be produced in CY-1962 - 1965.

Additional basic investigation of the nature and cause of bond fracture and the conditions necessary to produce a bond which withstands extreme reactor operating conditions is needed since little is known of the mechanism of failure of the bond. Application of the stud-pulling technique to determine strength of intact bonds on irradiated cores may indicate relative effects of bond quality fabrication improvements and various reactor power, temperature and exposure conditions. Further tests are also warranted in autoclaves at high temperature and pressure or through the use of other devices to duplicate in-reactor conditions insofar as feasible.

W. H. Hodgson

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Process Engineering

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17 "Review of Al-Si Process Limitations and Potential Alternate Processes", W. A. Blanton. 12/2/59. (Confidential-Undocumented)

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14 Strand, C. A. Personal Communication

15 Tverberg, J. C. Bond Strength Evaluation of the Brittle Bond Problem in Production Fuel Elements. Undocumented. November 10, 1958 (Confidential)

16 Strand, C. A. Personal Communication

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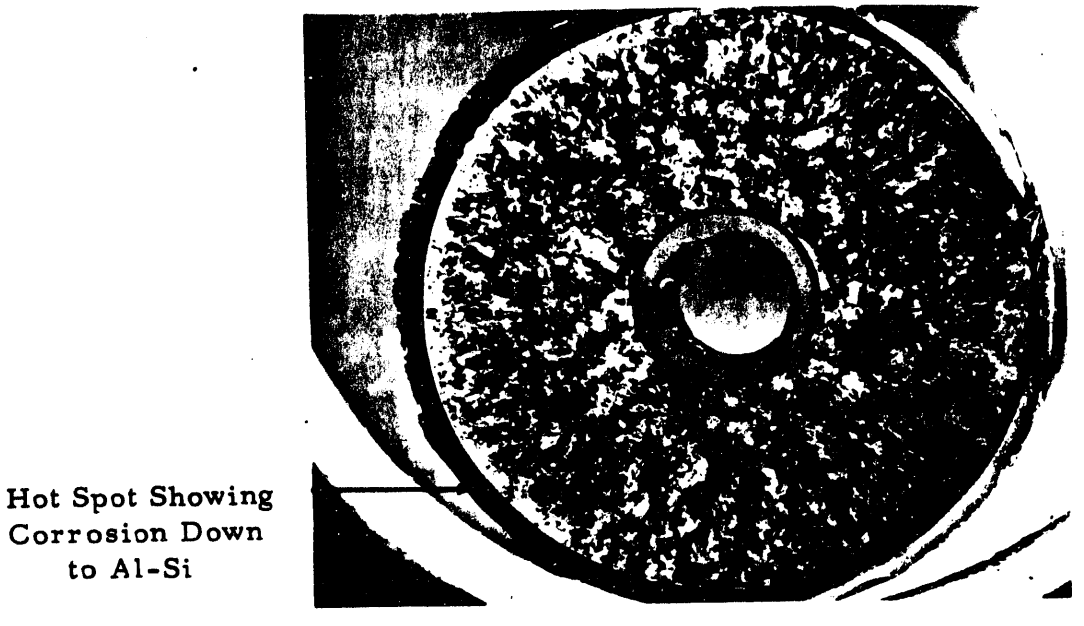
TABLE I[2]

POUNDS FORCE REQUIRED TO PULL 1/4" DIAMETER STUD

Bake Temp., °C	Time Hr.	Max.	Min.	Median	Average	Elements	Total No. Studs.	
No Bake	--	1150	300(2)	1000	860	10	30	Group I(1)
300° C	3	1150	400	1000	910	4	12	
	6	1150	250(3)	875	820	4	24	
	12	950	200	500	530	4	24	
	25	800	200	300	350	4	24	
	50	700	200	300	330	4	12	
350° C	3	1050	450	630	670	4	12	
	6	1100	300	550	580	4	12	
	12	750	150	430	450	4	12	
400° C	1/2	1300	350	950	860	4	24	
	1	1200	350	800	790	4	36	
	3	850	400	600	630	4	12	
	5	975	90	450	500	4	24	
	22	500	200	350	340	4	12	
No Bake	--	1050	300(4)	700	680	10	30	GROUP II(1)
300° C	3	1050	200(5)	730	680	10	30	

1 Elements in a single group canned on same day
2 Three studs at < 400 lbs.
3 Two studs at < 400 lbs.
4 Six studs at < 400 lbs.
5 One stud at <400 lbs.

Fig. 1. Photomicrograph of Al-Si bonding layer showing can wall, Al-Si, compound layers and uranium.

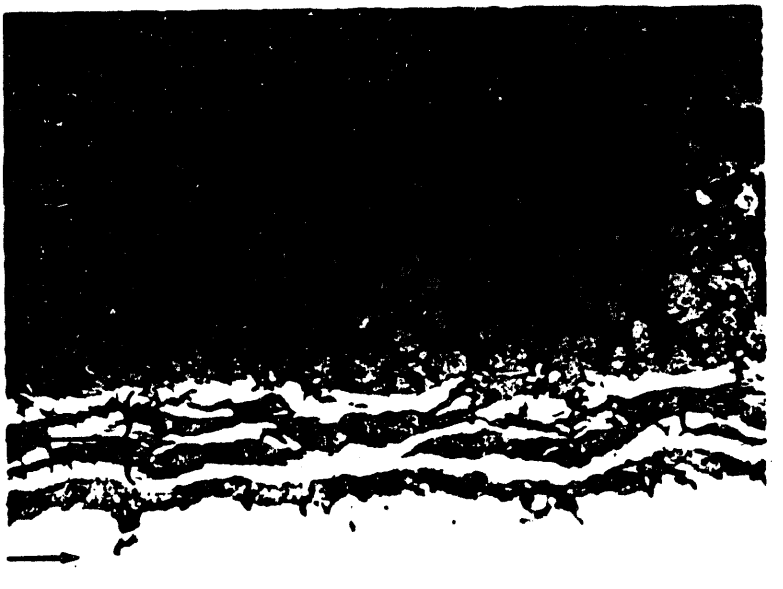


Cathodic Etch 2.3X

Fig. 2. Incipient rupture showing broken bond under the hot spot extending 5/6th of the distance around the periphery.

Polished 250X

Fig. 3. Example of broken bond found adjacent to the internal coolant channel of the I & E fuel elements.



Etched 250X

Fig. 4. Example of unbroken bond found on external periphery of I & E fuel elements.

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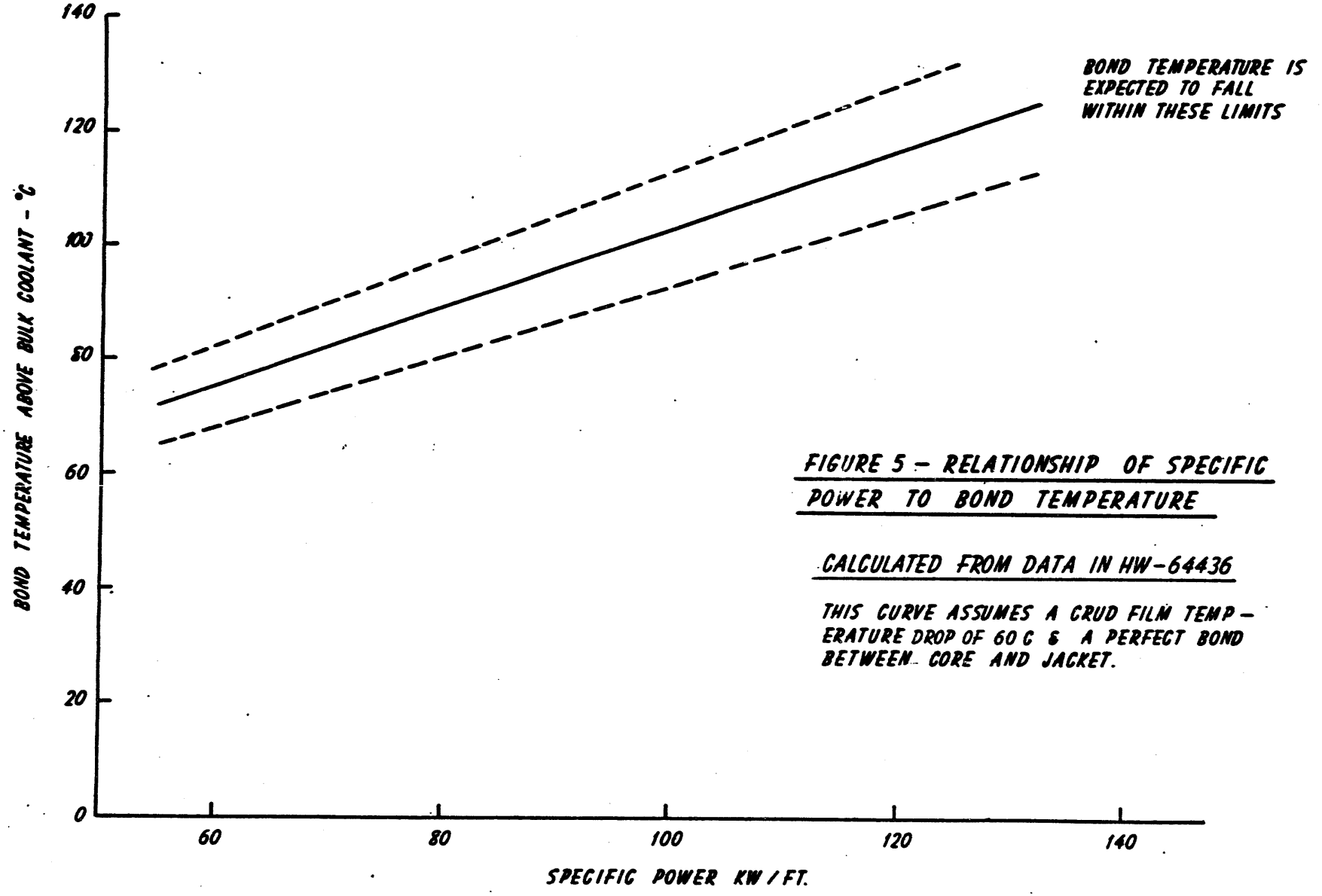

FIGURE 5 – RELATIONSHIP OF SPECIFIC POWER TO BOND TEMPERATURE

CALCULATED FROM DATA IN HW-64436

THIS CURVE ASSUMES A CRUD FILM TEMPERATURE DROP OF 60 C & A PERFECT BOND BETWEEN CORE AND JACKET.

DATE
FILMED
8/17/94

END