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REPORT OF MEETING ON THE ANSI BONDING
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REPORT OF MEETING ON THE AlSi BONDING LAYER

By

R. H. Todd

Metallography Laboratories Operation
 REACTOR AND FUELS RESEARCH AND DEVELOPMENT OPERATION

August 4, 1958

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By DK Hanson, 12-20-93

Verified By Jerri Mally 1-5-94

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HANFORD ATOMIC PRODUCTS OPERATION
 RICHLAND, WASHINGTON

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REPORT OF MEETING ON THE AlSi BONDING LAYER

PURPOSE

This meeting was held between members of HLO, FPD, and IPD with the purpose of acquainting all interested parties with the present problems being encountered in the canning process. A short discussion of previous canning methods and their problems was presented as background information.

DETAILS

The search for a bonding agent to bond the aluminum can to the uranium core covered such materials as AlSi, lead, tin, aluminum, and copper. Much of this early work was guided by the University of Chicago. The Grasselli (du Pont) Laboratory developed the triple-dip process where the uranium was first heated in a copper-tin bath, rinsed in a tin bath, then canned in an AlSi bath with AlSi as the bonding agent. It appeared at this time that the best AlSi composition to use was a little toward the aluminum side of the eutectic composition of 12.5 percent silicon.

Hanford fuel element (slug) production has gone through four eras. The first of these was the "Whiz-bang" which lasted from April to September of 1944. The second era used the triple dip process. Gamma extruded uranium was used in all processes until early 1948, and alpha rolled, heat treated uranium has been used since that date. The third era came in with the lead-dip or duplex process in March 1954. This process had been tried with the gamma extruded uranium for a short time in 1947 but suffered from excessive distortion of the uranium cores. With the more stable alpha-rolled, heat treated uranium the lead dip process has been successful.

The fourth era can be termed the expansion era. In this period many different types of product have been asked for. These involve new geometries, new materials and longer life under higher power and higher temperatures.

It has been accepted as reasonable that the present out-of-reactor non-destructive tests give fair assurance of accepting only those fuel elements which will give good reactor performance. It must be admitted however that a number of potential failures do pass the testing line and are sent to the reactors. On this basis research has been directed toward making a fuel element which will pass the present non-destructive tests and give good reactor performance.

Since the available non-destructive tests do not give a complete picture of fuel element quality, a number of destructive tests are used which give a little more information. One of these is the chisel test, used to test bond strength. This test consists of removing the can from the uranium core with a hammer and chisel. As long as one man does the testing he can say with fair certainty that one can had a better bond than another. However the test is purely qualitative and the same man must be the one to test any series of slugs for quality.

A current problem and the one with which this meeting was primarily concerned is that of bond failure where the bond between the can and core is broken. Using the chisel test as a guide the following variables have been found in this most recent study to have an effect on the strength or quality of the bonding layer.

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1. Geometry of the holding basket.
2. Position of the uranium core in the duplex bath, whether held vertical or near horizontal.
3. Turbulence in the lead bath.
4. Lead cover above the slug during preheat.
5. AlSi cover above the slug during preheat.
6. Time allowed for preheating in the lead bath.
7. Temperature of the preheat bath.
8. Percentage of silicon in the duplex bath.
9. Geometry of the slug being processed.

The composition of the AlSi in the canning bath apparently has little if any effect on the bonding layer. Other variables such as uranium surface roughness and oxide layer, and can cleanliness are known to have an effect on quality. At this time such other variables are being held as near constant as possible while those listed are being evaluated. The effects of the above named variables seem to be as follows:

1. If the holding basket only partially encloses a slug, that part of the slug not in the basket will have a more brittle bond. If the basket encloses the full length of the slug, the bond will be more nearly uniform over its entire length.
2. If the slug lies near horizontal in the lead bath instead of vertical the bond will be more uniform over the length of the slug.
3. The degree of turbulence can be either too great or too slight to promote maximum bond strength.
4. & 5. The deeper the lead cover and the deeper the AlSi cover, the stronger will be the bond.
6. Longer time in the preheat bath (with upper limits around one minute) will promote a stronger bond.
7. Higher temperature in the preheat bath promotes a stronger bond. (This finding is contrary to that reported in Reference #1).
8. A lower percentage of silicon in the duplex bath promotes a stronger bond.
9. In general, I&E slugs and other geometries having higher surface-to-mass ratios than the normal slugs will have better bonds when canned under identical conditions.

Mr. Strand has found that most of the above variables could be varied enough to produce a significant change in the compound layer and still be within production line limits. One of the worst conditions in this respect was temperature which was allowed to fluctuate as much as 14°C. The reference point for comparison of the above variables is taken to be the average product of the present production line. The optimum condition for each of these variables is now the target of present experiments. It was noted during discussion that during most of the past history

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of Hanford, there was no complete record of any testing program set up to determine the effect of each of these variables independently. Because several variables were being changed at once, no conclusive data could be obtained. Also equipment limitations prevented the close control needed.

Since May of this year the production of fuel elements using X-8001 alloy (M 388) has been placed under new process specifications. By incorporating only two of the changes, silicon composition and temperature, the normal reject rate of 20 to 30 percent was reduced to only 3 or 4 percent. Exact information on this change can be obtained from C. A. Strand in Building 306.

At the present time tests are in progress to determine the effects of variable preheat times (from 10 to 60 seconds) at each of several temperatures (600 to 624°C) and at each of several silicon compositions, all in the duplex bath. Evaluation will be by the chisel test, undercutting corrosion test, and metallography.

Early in this year an effort was made to obtain a better etch for the compound layers present in the finished slugs. Repolishing and reetching of these samples revealed that slight variations in etching technique affected the appearance of the compound layer considerably. A procedure has now been established which produces consistent results and which, in the opinion of the metallographers, reveals as many of the compound layers as can be shown by chemical etching.

For compound identification, a specially prepared sample from standard production material was submitted to the X-ray laboratory for X-ray diffraction work. The results from this sample revealed the following compounds to be present in a stabilized and tough compound layer: UAl_3 , USi_3 , and UPb_3 . It may be worth noting here that Bud Kahle did some work on the reaction of uranium with liquid lead. (Reference #2). One of the negative results he obtained but did not report in his document was the observation that the normal chemical test, used to show the presence of lead, failed completely where lead uranium compounds were present. Thus it is now known that the chemical test will not reveal the presence of lead when that lead is combined with uranium. However, X-ray diffraction will reveal the presence of lead and show in what structure or compound it occurs. This could explain why a lead compound was found by X-ray diffraction when it was believed by many that no lead was present.

One compound has been identified by comparison to be UAl_3 . Sections of hot pressed slugs have shown diffusion of aluminum into uranium to form UAl_3 . This UAl_3 structure is so nearly identical to that of one of the compounds in an AlSi bonded fuel element that it must be the same compound in each case. Where this compound (UAl_3) occurs in the AlSi-uranium system only two others appear. These would be USi_3 and UPb_3 . The UPb_3 compound layer should be identifiable by its absence in slugs canned by the vacuum canning process. Such a sample was studied but it contained the same three compounds as a normal slug. However, it is not known if the AlSi used for canning this specimen was completely free of lead. This approach may yet reveal the identity of the compounds.

Whenever a weak or brittle bond appears, the compound layer contains little or no UAl_3 , while strong bonds invariably contain a fair amount of it. Accordingly compound identification was important to enable the investigators to know what process changes would be most likely to produce the desired results.

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After mechanical and electrolytic polishing the compound layer of a normal sample shows many cracks, especially in and parallel to the bonded layers. Further etching by ion bombardment uncovers more cracks in the bonding layer which were originally masked by flowed metal from the polishing operation. (Ref. #3). The uranium adjacent to the compound layer is fine grained; presumably, the cold work on the surface of these slugs is sufficient to cause recrystallization during the canning cycles. Large primary grains of aluminum in a layer one grain thick appear adjacent to the compound layer. These grains probably develop as a consequence of silicon depletion in the AlSi if silicon is preferentially consumed in the formation of the compound layer. These aluminum grains may also develop due to a preferential nucleation and growth at the compound layer or uranium AlSi interface. Regions of the compound layer appear to spall off and float into the AlSi. The fact that these bonds contain a high concentration of uranium has been substantiated by microradiographs. Many of the specimens show regions in the layered intermetallics in which a new feathery phase has formed at the expense of the layered compounds. An appreciable change in the density for the feathery phase must occur, since a banding of the layered structure occurs. Spalling of the layered structure into the AlSi is also shown to be associated with regions where the feathery compound forms. The fact that USi_3 has a (100) spacing of 4.09 Å, whereas that of UAl_3 is 4.15 Å might indicate that pure UAl_3 with a larger volume per U atom is the feathery phase. Accurate calculations of the density of UAl_2 , UAl_3 , USi_3 , UPb_3 , etc., should be made to ascertain theoretical density changes associated with the formation of these phases.

The intermetallic compound formation has been shown to occur in the duplex bath. When extracted from the duplex bath the tops of slugs have less adherent AlSi than the bottoms due to gravitational rundown. Insertion of such tapered pieces into cans should result in a greater washing action at the lower part of the slug than on the top provided the adherent AlSi is semi solid or completely solid. If the AlSi on the uranium is liquid, the washing action is still greatest at the bottom and least at the top. This reasoning would account for the scrambled intermetallics at the bottom of slugs. If this washing action is applied to the can the reverse situation applies. This latter situation could account for the poorer AlSi wettability observed in the lower bottom regions of a can.

Cocked or eccentric slugs have shown susceptibilities toward cracked intermetallics in the region where AlSi thickness is limited. At a point where very little AlSi was present, no bond was observed. If a bond existed it was extremely narrow and could have broken out during the polishing.

A comparison of the compounds shows the feathery layer (UAl_3) to have definite channels at right angles to the bond. The layered type has up to six bands and shows many cracks. At the uranium-compound interface mounds are present which are probably oxide impurities.

The following experiment would be of interest: Prepare a slug using silicon rich AlSi in the duplex bath in order that primary silicon will solidify first on cooling, rather than aluminum. Moreover, the metallographic appearance of the compound layer might very likely be altered considerably and permit by comparison, a better interpretation of the bonding in aluminum rich AlSi. It is recommended that micro x-ray diffraction studies be conducted on the intermetallics formed in canned slugs. Since the examples of the microradiographs show their utility, the possible use of radioisotopes of lead, silicon and aluminum should be investigated to determine their applicability in identifying the intermetallics which form.

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It is also possible to identify the constituents by a micro-spark, spectrographic method. Such a unit is being constructed for Pete Anderson, 3706 Building.

The examination of the AlSi bond in irradiated slugs by Radiometallurgy has been quite limited, since most interest to date has been in the condition of the uranium cores and aluminum cladding. (Reference #4).

Cursory observations of bond layers from several fuel elements irradiated from 200 to 2000 MWD/T have been made; however, the data obtained are insufficient to draw any conclusions concerning the effects of irradiation on bond deterioration. Both low and high exposure fuel elements have exhibited some pronounced unbonded areas. However, the bond in other areas of the same transverse sections appeared to be perfectly sound. The observed separations generally occurred within the compound layer, and were nearly always wide enough to be easily seen at a magnification of 250X, and in some cases with no magnification. Compound layer cracking has not been seen, possibly because observations have been restricted to the as polished condition. Very small cracks in the compound layers would very probably be masked over by flowed metal, and etching would be required to make them visible.

Evidence of bond deterioration as a result of overheating was found in a recently examined fuel element. The slug was unruptured; however, a large "hot spot" pattern was observed on the jacket. Underneath the hot spot a large unbonded area with a maximum separation of 10 mils between the U and AlSi was found. There was no evidence of a compound layer throughout most of the unbonded section. Spheroidization of the Si in the AlSi indicated that overheating occurred before the jacket became unbonded.

It was agreed that Radiometallurgy should not undertake any extensive investigative programs in connection with the bonding problem at the present time. Until more conclusive information is obtained through current investigations of unirradiated material, it would be quite futile for Radiometallurgy to undertake anything more than routine observation of bonding layers in slugs normally submitted for radiometallurgical examination.

SUMMARY AND CONCLUSIONS

Present investigations are aimed at producing the best possible fuel element as judged by destructive and non-destructive tests in the unirradiated state. Equipment has been recently modified (and is still being improved) to provide the closest possible control over all process variables. Tests are being run to evaluate the effect of all variables which have been recognized as having a definite effect on fuel element compound layers. Variables such as bath impurities, e.g., iron, chromium, and nickel, should be considered.

It is recognized that very little is known about the ternary system of aluminum, silicon, and uranium, and the quality of a fuel element depends in large part on the bonding layer which is made up from these elements during canning. Lead is also present in the bonding layer and its exact role is not known. The ternary and quaternary phase diagrams for these four metals should be thoroughly investigated. However, at present there are no plans for such an investigation to proceed. It may be necessary to identify each chemical element and follow it through the canning process as it changes from element to final compound before we can hope to understand

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the bonding mechanism. With this knowledge it should be possible to produce the optimum compound layer. Some of the testing procedures which could be utilized to good advantage in such investigations are metallography, microradiography, electron microscopy, micro x-ray diffraction, micro-spark spectrography, and micro-beam X-ray fluorescence.

The program has advanced far enough to show that fuel elements with compound layers of consistent uniformity can be produced. It is the opinion of the author that the characteristics of that compound layer which gives the best out-of-reactor performance will be recognizable in metallographic sections.

Where possible, the Radiometallurgy Laboratory should make examinations of compound layers in such a fashion that they can be correlated with examinations of unirradiated material. Until this correlation is made, the value of the other experiments will lie only in the production of fuel elements of consistent quality, not necessarily the best quality. Routine examination of irradiated elements should provide information which will indicate how well any characteristic compound layer will stand up under actual reactor exposure. All personnel who are interested in this problem should be made information addressees for this information.

In-reactor performance of a fuel element may be entirely different from out-of-reactor performance of that same element. Only when a correlation is made of in-reactor vs. out-of-reactor performance will it be possible to set up specifications for optimum fuel elements.

The following individuals attended and contributed to this meeting:

T. K. Bierlein
W. A. Blanton
R. R. Bloomstrand
S. M. Gill
W. J. Gruber
W. H. Hodgson
B. Mastel
C. A. Strand
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REFERENCES

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2. HW-54628 (confidential) "The Diffusion Layer Formed by Molten Lead Reaction with Uranium," V. E. Kahle and A. L. Bement, January 27, 1958.
3. Letter to W. A. Blanton from T. K. Bierlein and B. Mastel, July 24, 1958.
4. Letter to R. H. Todd from D. L. Zimmerman, August 1, 1958.

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