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EFFECTS OF PREHEAT TIME IN COMPOUND LAYER FORMATION

By

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EFFECTS OF PREHEAT TIME IN COMPOUND LAYER FORMATION

Introduction

Hanford fuel elements are made by brazing uranium cores in aluminum cans. The interface, or junction, between the AlSi braze and uranium core known as the compound layer, is composed of an intermetallic complex. The properties of this compound layer, brittleness, ductility, toughness, are considered to have a pronounced influence on the performance of fuel elements in the reactors. This report presents the results of initial efforts to identify the compound layers and establish some of the process conditions and variables which influence the properties of the compound layer, in particular, the lead preheat time.

Summary

Compound layer formation on uranium cores in the duplex bath (AlSi floating on lead) takes place in two main steps. The first step is the formation of a silicon rich uranium-aluminum-silicon complex. This layer is very hard and brittle. As the reaction continues, the original complex is broken up and replaced by a UAl_3 -type compound which is more ductile and tougher. Bond strength was found to increase as the preheat time in the lead was increased from 8.5 to 34.0 seconds at 606 C. Increasing the reaction rate by raising the duplex bath temperature improved the bond strength. Decreasing the silicon content of the AlSi layer allowed more UAl_3 -type compound layer to form and produced better bonds.

Discussion

Hanford fuel elements are made by sealing uranium in aluminum cans. The uranium cores are bonded to the aluminum can by brazing with AlSi. The AlSi braze in the finished fuel element serves two main purposes: (1) it is a heat transfer medium between the core and the can, and (2) it acts as a diffusion barrier between the uranium and aluminum.

The core is wet in the AlSi layer of the duplex furnace after preheating in the lead layer. A complex reaction, about which little is known, takes place during wetting. The resulting intermetallic compounds, known as the compound layer, are formed at the interface between the uranium and AlSi. Variations within this compound layer have been observed which result in good and bad bonding. The purpose of this report is to show the effect of varying lead preheat time upon the resulting compound layer.

Figure 1 shows compound layers formed on the outside of I & E cores while lead preheat time was varied between 8.5 and 34.0 seconds at 606 ± 3 C with a constant AlSi time of three seconds at 597 ± 2 C. Compound layers formed as a result of 8.5 and 14 seconds lead preheat show the typical type associated with brittle bonds. The dark area, for simplicity sake, will be called primary phase. Actually this phase is a multilayered structure highly susceptible to cracking as revealed by etching with ion bombardment.¹ It was also very brittle, as determined by the "chisel test". Bond strength increased as the lead preheat

1. Bierlein, T. K. and Mastel, B., Letter to W. A. Blanton, July 24, 1958.

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time was lengthened. The photomicrographs show that there is considerably more of a light colored material present at the longer lead preheat time. This material will be called a UAl_3 -type compound since it exhibits properties typical of UAl_3 .²

As lead preheat time is lengthened, more so-called "scrambling" above the compound layer is observed.

Compound layer formation appears to take place in two main steps. The primary phase is assumed to be a silicon rich uranium-aluminum-silicon complex since uranium has a greater affinity for silicon than aluminum. Evidence of this greater affinity of silicon for uranium is shown by (1) the formation of primary aluminum grains adjacent to the compound layer and (2) the duplex $AlSi$ is depleted of silicon during canning. The secondary phase consists of forming a UAl_3 -type compound adjacent to the uranium. This formation is a result of a break in the primary phase. As the reaction continues, portions of the primary phase and primary aluminum grains are consumed. The so-called "scrambled" effect associated with tougher bonding is a result of expansion during formation of the UAl_3 -type compound.

Figure 2 shows compound layers formed on the I & E bore surface. $AlSi$ did not contact this surface until after the lead preheat because the bore was closed with lead plugs. For a given preheat time there is more of the UAl_3 -type compound on the bore surface than on the outside surface. The lack of $AlSi$ freeze out on the inner surface during submergence may have allowed a more uniform preheat which in turn could have permitted a more rapid compound layer formation.

Comparison of bond strength tests and photomicrographs of compound layers indicate that it is desirable to produce more of the UAl_3 -type compound.

A greater percentage of UAl_3 -type compound can be formed by increasing the reaction rate and by decreasing the duplex $AlSi$ silicon content. The reaction rate can be increased by raising duplex furnace temperatures. $AlSi$ silicon content reduction allows more UAl_3 -type compound to form because the increased percentage of aluminum interferes with the uranium-silicon reaction. As a result, the primary phase compound layer is thinner allowing easier penetration and formation of more UAl_3 -type compound.

A combination of higher temperatures and reduced $AlSi$ silicon content are being used to produce larger areas of UAl_3 -type compound layers.

Photomicrographs of compound layers shown in the report were prepared by using the standard polish with a two per cent chromic acid lubricant and Linde B abrasive. The samples were etched 15 seconds in standard phosphoric acid electrolytic bath at 18 to 20 volts and swabbed for two to three seconds with 0.5 per cent hydrofluoric acid.³

2. Todd, R. H., Personal Communication, June, 1958

3. Todd, R. H., Personal Communication, July, 1958

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Compound layers appearing in this report produced as a result of a certain lead preheat time, should not be considered typical production samples. The primary purpose of this experiment was to decrease core heating time by eliminating bore plugging with AlSi. Since it was possible to assemble pieces after a very short preheat time and there was a trend in bond strength, it was assumed that there would also be a trend in compound layer formation. Actually, compound layers produced by an 18 second preheat at 606 C using lead bore plugs appear to be equivalent to compound layers formed after a normal 34 second preheat at 606 C.

Additional work on compound layer formation is underway, but will not be completed for at least three months. Similar work to that reported herein with closer temperature control has been initiated. Lead preheat temperatures were varied between 600 C and 624 C with preheat times varied between ten and sixty seconds to determine compound layer formation. Another series of tests has been started in which lead preheat temperatures were varied between 600 C and 624 C at 7.5 to 10.5 per cent duplex AlSi silicon content.

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FIGURE 1
 EXTERNAL COMPOUND LAYERS LEAD PLUGGED I&E FUEL ELEMENTS
 (250x)
 Duplex Lead: Temperature $-606\text{ C} \pm 3\text{C}$ Time-as indicated
 Duplex AlSi: Temperature $-597\text{C} \pm 2\text{C}$ Time-3 seconds

FIGURE 2
INTERNAL COMPOUND LAYERS LEAD PLUGGED I&E FUEL ELEMENTS
(250x)

Duplex Lead: Temperature - $606^{\circ}\text{C} \pm 3^{\circ}\text{C}$ Time - as indicated
Duplex AlSi: Temperature - $597^{\circ}\text{C} \pm 2^{\circ}\text{C}$ Time - 3 seconds

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