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AN ANALYSIS OF HOT DIE SIZE FUEL BONDING

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Fabrication Development Unit
Fabrication Metallurgy Section
Metallurgy Department

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AN ANALYSIS OF HOT DIE SIZE FUEL BONDING

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Abstract

A critical analysis of the Al-Ni-U bond and the hot die size bonding technique is given as the result of a literature search. Metallurgical aspects of the bond and the technique of hot die sizing are commented upon in particular.

Introduction

This analysis is the result of a search and study of the literature concerning diffusion bonding of aluminum to nickel plated uranium. ⁽¹⁾ A vast amount of work has been done on this type of cladding process the world over during the past 15 years. Hot Press Canning, Step Pressing, Gas Pressure Bonding and Hot Die Sizing are the four processes that have been used to diffusion bond nickel plated uranium to aluminum cladding. Work began in the United States in the early fifties and since then work has been done in Canada, Europe, Japan, India and probably in other locations. This analysis will be restricted to the hot die sizing process and the resulting bond. Areas will be suggested where further work may be done in order to improve or insure the quality and integrity of the bond.

Discussion of HDS Process

For those not familiar with the Hot Die Sizing Process as it is currently practiced at Hanford ^(2,3) a short synopsis is in order. The hot die size cladding process begins with a uranium slug which is etched, activated and nickel plated

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with an average thickness of less than 0.5 mils (13 microns). The nickel plated core and the top end cap are loaded in an aluminum can which either has the base end cap welded in or is an impact extrusion. The aluminum components are cleaned and deoxidized prior to assembly. The assembled fuel element components are coated with lubricant on the outside aluminum surfaces and placed in a preheated furnace set near 680°C. In 12 minutes the uranium core has reached an average temperature of 550°C. During this time Ni-U diffusion has begun to bond the nickel plate to the uranium core.

Upon leaving the preheat furnace the assembly begins to cool in the air. The maximum time allowed between the furnace and the sizing press is 45 seconds. The temperature of the slug-aluminum interface just prior to bonding is controlled to 380° to 550°C by controlling the cladding temperature to 380° to 450°C (for fuels less than 1.6 inch OD).

In the sizing press the preheated slug is end loaded by two opposing rams in order to seat the end caps on the core and the cladding is sized to the core by passing the components through internal and external dies. Sizing of the cladding is a drawing operation in which the uranium acts as a moving mandrel. The clad undergoes a 25 to 65% reduction of area, producing a high strength, uniform, diffusion bond on the lateral surfaces. The mechanical action of drawing the aluminum over the core breaks up the aluminum oxide film and provides nearly ideal diffusion conditions. The aluminum end caps are not fully bonded to the core ends by the sizing step because of the lack of metal movement at the cap-core interface and the chilling effect of the rams. A separate step, end bonding, is required. The end bonding step is essentially a hot-pressing process in which inductively heated dies press the cap and core together at 560° to 580°C, for up to 3 minutes. After end bonding the core and clad are fully bonded and the fuel element is finished by the normal processes before irradiation.

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The resulting bond contains an aluminum-nickel diffusion zone of about 1 micron thickness and a nickel-uranium diffusion zone somewhat thinner. Each of the two diffusion zones normally contains two intermetallic compounds. Al_3Ni and Al_3Ni_2 appear in the Al-Ni zone, and Ni_5U and NiU_6 appear in the Ni-U zone. Although intermetallic compounds are generally brittle and poor conductors, their thickness in the hot die size bond (HDSB) has negligible influence on the strength and thermal conductivity of the bond. In fact the strength of a good HDS bond is greater than that of the aluminum clad. The bond itself is ideal with three exceptions:

1. The presence of nickel in the bond has a small effect on neutron economy. For this reason the nickel plate thickness is kept as small as possible.
2. The bond offers virtually no secondary corrosion resistance once the aluminum clad is penetrated by water.
3. The strength and continuity of the bond are lowered by exposure to (4,5,8) the reactor environment.

It is possible that one of the above deficiencies could be improved upon, but most likely at the expense of aggravating one or both of the other. Any improvements would have to be made through the hot die size bonding process itself which to date has been superbly engineered to yield a high quality bond before pile exposure. Obviously, any bond improvement that might be made would have to be justified economically, after the possibilities are demonstrated.

Summary and Conclusions

It is concluded that the secondary corrosion resistance of HDSB fuels may be improved significantly by a thicker nickel plate and/or more Ni-U diffusion. A more basic understanding of the bond behavior may be gained from knowledge of the properties of the intermetallics in the bond. Practical limits should be

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established for the in pile exposure conditions of HDSB fuels and the factors leading to bond degradation studied.

The metalworking forces during the sizing step, as a function of process variables, were investigated and are discussed. A compilation of diffusion data from the literature is made.

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Nickel Plate

Nickel electroplating uranium has been studied extensively. A review of the current state of the art is given by Lundquist, Braun, and Stromatt.⁽⁶⁾

The biggest deficiency of the electroplates used in HDSB is the porosity of the plate, which is inherent in such thin nickel electroplates on uranium. Each pore provides an entryway for water once the aluminum clad is penetrated. Essentially pore-free electroplates with very good corrosion resistance may be produced at thicknesses approaching 3 mils⁽⁷⁾ but they are not economical from a neutron standpoint.

The only obvious possibilities for reducing plate porosity lie in other plating processes such as carbonyl nickel, and vacuum vapor deposition but neither of these processes are economically competitive with electroplating.

Secondary Corrosion Resistance

The principal disadvantage of the HDSB fuel element is its lack of a secondary corrosion barrier. Apparently, neither the intermetallic layers nor the nickel plate are sufficiently thick or continuous to act as a barrier. Generally, the nickel plate is kept as thin as possible for neutron economy and the intermetallic diffusion thicknesses are kept down in order to limit the thickness of the brittle intermetallic, Al_3Ni_2 .

However, the HDSB process does offer the flexibility of controlling the Ni-U diffusion during preheat separate from the Al-Ni diffusion during sizing. The Ni-U layers may be thickened without increasing the thickness of the brittle

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Al-Ni compound, Al_3Ni_2 . There is every possibility that increased Ni-U diffusion will form a sufficiently thick and continuous layer of Ni-U intermetallics to provide appreciable resistance to water penetration. Increased diffusion could also "heal" the porosity in the nickel plate. A slightly thicker nickel plate would naturally be required to accommodate the increased Ni-U diffusion.

To date this possibility has not been pursued. It does have enough potential to be investigated. A study of the secondary corrosion behavior of fuels manufactured with increased Ni-U diffusion and/or increased nickel plate thickness should be carried out. Also, a study of the corrosion characteristics (corrosion rates and EMF's) of the individual intermetallics in the bond would provide helpful basic information. Thermal conductivity data should also be developed for each intermetallic in order to assess the effect of increased thicknesses on the heat transfer of the bond. The possession of the basic data on the intermetallics may also uncover more possibilities for improving overall bond quality.

In Reactor Bond Degradation

(8)

Data from Savannah River show that the bond strength of nickel bonded fuel elements (Mark VB fuel) decreases during irradiation by as much as 75%. Also, gaps are formed between the uranium core and the bond which cover up to 65% of the bond area. The bulk of the bond degradation occurred linearly with exposure to 1300 MWD/T. The mechanism of bond deterioration is not now understood fully and the only way of predicting bond degradation is through irradiation tests.

Bond degradation could be due to irradiation effects on the intermetallics in the bond or fuel bumping (local uranium distortion) or a combination of both. Certainly fuel bumping can aggravate any poor bond condition.

The causes of bond degradation should be studied to ensure that the HDS bond will perform satisfactorily at higher exposures, temperatures, and heat transfer rates. Fuel elements operating at higher temperatures and exposures can be

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irradiated and the bond quality evaluated subsequently. One objective of such tests should be to establish the upper service limits for HDSB fuels.

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APPENDIX

Comparison of Diffusion Rates - Published Results

Metalworking Forces Generated During the HDSB
Sizing Step

Comparison of Diffusion Rates - Published Results

There has been a substantial amount of work done in the Al-Ni and Ni-U diffusion systems. Most experimenters have found that the second order proportionality between diffusion zone thickness (strictly speaking, intermetallic phase thickness) and time is valid, as well as the Arrhenius equation:

$$x^2 = Kt$$

Second order parabolic relation where:

X=diffusion zone thickness

K=penetration constant

t=time

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$$K = K_0 e^{-\frac{Q}{RT}}$$

Arrhenius equation describing temperature dependence of penetration constant:

K=penetration constant at temperature, T

K₀=penetration constant at temperature T = ∞

derived from plot of 1/T vs log K

Q="activation energy", a constant

R=universal gas constant

T=temperature degrees Kelvin

The actual values of K₀ and Q published by different authors are not in strict agreement, especially in the Al-Ni system, where there is substantial disagreement.

There are difficulties in comparing the published data due to the various methods results are reported. Results can be reported as:

1. D₁=chemical diffusion coefficient for one metal in a phase;
from Ficks law.
2. D=interpenetration coefficient for two interdiffusing phases:

$$D = \left[\frac{N_2}{N} D_1 + \frac{N_1}{N} D_2 \right] \quad \text{where } D_1 = D_{O1} e^{-\frac{Q_1}{RT}}$$

and $\frac{N_1}{N}$ = partial molar fraction of component, 1. ⁽⁹⁾

3. K_1 = penetration coefficient for growth of one $X_1^2 = K_1 t$ intermetallic phase in one direction.

4. K_{1j} = penetration coefficient for growth of one intermetallic phase in two directions

$$X_{1j}^2 = K_{1j} t$$

$$\text{since } X_{1j} = X_1 + X_j \text{ then } K_{1j} = (\sqrt{K_1} + \sqrt{K_j})^2$$

5. K = penetration coefficient for growth of total width of intermetallic phases during diffusion.

$$X^2 = Kt$$

$$\text{since } X = X_{1j} + X_{k1} + X_{mn} + \dots$$

$$K = (\sqrt{K_{1j}} + \sqrt{K_{k1}} + \sqrt{K_{mn}} + \dots)^2$$

Relationships between D and K are difficult to arrive at (see Reference 9).

K is related directly to diffusion zone widths and is reported in a majority of the studies of Al-Ni and Ni-U diffusion. For this reason diffusion data were compared by reducing all reducible data to a value of K . The square root of K vs $1/T$ was then plotted in Figures 1 and 2. (The square root of K is proportional to X when t is held constant. Also, measurement errors involved in X are not amplified as badly when $\sqrt{K}$ is plotted.)

The effect of pressure on diffusion is shown in the plots of Storcheims (10,11,12) data, i.e., pressure decreases both U-Ni interpenetration and Al-Ni interpenetration. A pressure of 26 tsi was found to completely inhibit formation of the Al-Ni intermetallics γ (NiAl_3) and β (Ni_2Al_3). A pressure of 20 tsi will inhibit formation of the brittle γ ⁽¹³⁾ phase. Since the pressures generated during hot die sizing are on the order of 6 tsi or lower, the effects of pressure on

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FIGURE 1
 $\sqrt{K_{Ni-U}}$ vs $1/T \times 10^3$

FIGURE 2
 $\sqrt{K_{\text{Al-Ni}}} \text{ vs } 1/T$

diffusion are negligible. Storcheim also found that increasing temperature significantly increased the γ/β ratio between 500°C and 625°C. Decreasing pressure from 20 to 11 tsi significantly increased the γ/β ratio.

The rate of growth of Al-Ni layers during the initial stages is not resolved. (14,15) Diffusion proceeds at a rate faster than parabolic (volume diffusion) (15) until a thickness of about 40 microns is reached. Konishi fitted a linear rate law to his data for the initial stage of growth of Al_3Ni_2 .

As can be seen in Figure 1 there can be a factor of 3 difference in the U-Ni diffusion zone widths predicted by separate investigators. Likewise there can be a factor of 5 difference for Al-Ni diffusion. It is worth noting that all the investigators used rolled nickel as one component of the diffusion couples with the exception of the French who used electrolytically deposited nickel. Storcheim's measurements were made of the maximum penetrations which represent optimum diffusion conditions, while other investigators measured the average intermetallic widths.

All things considered the optimum conditions for HDS bonding appear to be the cleanest surfaces and combined with the lowest diffusion temperature (lowers γ/β ratio).

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Metalworking Forces Generated During the HDSB Sizing Step

The HDSB sizing operation is a drawing process in which an aluminum tube is drawn down upon the nickel plated slug which acts as a moving mandrel. The forces generated during sizing are of interest in understanding the bonding process and in estimating the effect of pressure upon diffusion rates.

(22)

The following analysis is based on the discussion by Dieter concerning tube drawing with a moving mandrel. A schematic of the draw canning operation is Figure 3, and in Figure 4 the stresses acting on an element of the aluminum cladding tube, as it is drawn through the die are shown. Both figures depict the external clad only. The geometry and forces acting on the internal clad during sizing are similar. The following analysis is equally valid for either the external or internal clad.

A basic assumption of the analysis is that plane-strain conditions exist, and, therefore, the distortion energy yield criterion applies:

$$\sigma_x + p = 1.15 \sigma_0 = \sigma'_0$$

where:

σ_0 = uniaxial flow stress of the aluminum

σ'_0 = flow stress under conditions of plane-strain

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The above equation establishes a direct relationship between the longitudinal stress on the aluminum tube wall and the transverse compressive stress acting between the aluminum tube and the nickel-plated uranium core.

σ_x reaches a maximum near the die exit and is expressed by:

$$\sigma_x = \sigma'_0 \frac{1 + B'}{B'} \left[1 + \left(\frac{hf}{ho} \right)^{B'} \right]$$

where:

ho = original wall thickness of the aluminum tube

hf = final wall thickness of the aluminum tube

B' = a parameter

FIGURE 3

Simplified Draw Canning Operation; External Clad Only

- σX = Axial Stress
 P = Die Pressure
 f_1 = Coefficient of Friction Along Die-Tube Interface
 f_2 = Coefficient of Friction Along Tube-Mandrel Interface
 α = One-Half Apex Angle of Die

FIGURE 4

Stresses Acting on an Element of Aluminum Tubing During Draw Canning

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$$B' = \frac{f_1 - f_2}{\tan \alpha}$$

Since values of the coefficients of friction, f_1 and f_2 , are unknown the values for B' were chosen and calculations of σ_x made for various values of B' . Values of B' were set at $0 \leq B' \leq -1$ by the following reasoning:

1. $B' \geq -1$. If $B' < -1$, σ_x is compressive which is a condition of instability leading to tube damage or tool breakage.
2. $B' < 0$. If we assume $f_1 \leq f_2$ then $B \leq 0$. This assumption is based on the fact that f_1 is for lubricated surfaces and that f_2 is for the surfaces being bonded.

Table I.

σ_x As A Function of B' and hf/ho or R.A.

B'	<u>hf/ho</u>		
	.73	.55	.37
	<u>R.A.</u>		
	<u>27%</u>	<u>45%</u>	<u>63%</u>
-0.1	.29 σ'_0	.55 σ'_0	.94 σ'_0
-0.5	.17 σ'_0	.35 σ'_0	.64 σ'_0
-0.9	.036 σ'_0	.079 σ'_0	.16 σ'_0

The magnitude of the flow stress, σ_0 , varies with strain rate (or ram speed and die angle) and wall reduction (strain hardening). The strain rate increases slowly from the die entrance to a maximum at the die exit. The true strain rate is given by:

$$\dot{\epsilon} = \tan \alpha \times R.S. \times \frac{hf}{h^2}$$

R.S. = ram speed

At the die exit:

$$\dot{\epsilon} = \tan \alpha \times R.S. \times \frac{1}{h_f}$$

<u>Ram Speed</u>	<u>$\dot{\epsilon}$ (Die Exit)</u>
40 in/min	164 in/in/min
65 "	266 "
90 "	369 "
250 "	1020 "

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Data for the true flow stress of X8001 alloy aluminum as a function of temperature and strain rate are not available. These data for 1100 alloy aluminum are given by Hockett⁽²³⁾ and are plotted in Figure 6. Note that the data plotted in Figure 6 for 4½% R.A. (reduction of area) are taken from tensile tests on X8001 alloy, Figure 5. There seems to be good agreement between the data for the two alloys. The flow stress values for X8001 and 1100 aluminum alloys should then be similar. For this reason Hockett's data was used to obtain flow stress, σ_0 .

The results of the calculations based on the available data are given in Figure 7. σ_x and P are plotted for reductions of area of 27%, 45% and 63%. The variation of stress with ram speed is plotted only for σ_x at 63% R.A. Study of the curves yields the following general, but significant, conclusions:

1. Increasing R.A. increases the tensile stress, σ_x .
2. In general, as R.A. increases the compressive stress, p, decreases.

It is concluded from this that the better bonding at higher reductions of area is related solely to an enhanced smearing action at the Al-Ni interface which breaks up oxides. It is not due to a higher compressive force.

3. As the lubrication of the aluminum can improves (f_1 decreases and B' decreases), p increases and σ_x decreases.
4. Ram speed has a negligible influence on metal working stresses. Slower ram speeds have been found to result in increased chilling effects on the fuel ends, however.

5. Compressive stresses range from 0 to 6 tsi, which is too low to have a significant slowing effect on the growth of the brittle γ phase (Al_3Ni_2).
6. It is granted that higher or lower can wall temperatures will vary the flow stress, σ_0 , and σ_x and p will vary proportionally.

It is well to interject a word of caution regarding the accuracy of the results plotted in Figure 7. These equations were developed by Sachs and Espey⁽²⁴⁾, and were verified in experiments where the coefficients of friction (f_1 and f_2) were nearly equal at the tube-die and tube-mandrel interfaces. This is not the case in the HDS drawing operation because the tube-mandrel (Ni-plated U) interface is under sticking conditions and the tube-die interface is well lubricated.

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FIGURE 5
Tensile Tests of X8001-F at 400 °C

FIGURE 6
Uniaxial Flow Stress vs True Strain Rate at 400 °C

FIGURE 7
Drawing Stresses at Die Exit as a Function of B' and R. A. and Ram Speed

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