MONOLITHIC FUEL PLATES DIFFUSION BONDED BY ELECTROCONSOLIDATION® PROCESS TECHNOLOGY*

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ABSTRACT

Diffusion bonding and other solid-state joining processes have been proposed and demonstrated for the fabrication of monolithic U-Mo fuel plates. An innovative new process, named Electroconsolidation® Process Technology, has been studied for the diffusion bonding of monolithic fuel plates. The main advantage of this process over conventional diffusion bonding is the minimization of time-at-temperature due to the rapid heating inherent to the Electroconsolidation® Process. Bonding results of surrogate fuel plates made with stainless steel and depleted uranium monolithic fuel plates are presented. One or more fuel plates fabricated by Electroconsolidation are planned to be tested in upcoming irradiations.

Introduction

The next generation of highly loaded fuel for research reactors with low power and high neutron flux will use recently developed advanced materials such as U-Mo alloys that have high uranium density, outstanding strength, radiation resistance, and stability at low irradiation temperatures. Experiments with foil type and monolithic fuel in Al cladding have shown promising results [1-3]. Diffusion bonding and other solid-state joining processes have been proposed and demonstrated for fabrication of monolithic U-Mo plates [4].

The established industrial methods for pressure-assisted bonding are hot pressing (HP) and hot isostatic pressing (HIP). HP is limited to very simple joints. Parts with complex joints can be made by gas pressure sintering by the HIP process, but HIP autoclaves are expensive, heavy, and difficult to operate. HIP processing is also slow. In addition, the fuel plates to be densified by HIP must be pre-sintered, welded, or coated to seal the porosity so that the gas can exert pressure on the part. HIP has been used to make prototype fuel plates, but it is very expensive. Fabrication by friction stir welding requires extensive machining and surface preparation after bonding.

In contrast to HIP, heating by the Electroconsolidation® Process is very rapid and requires short cycle times. Much higher temperatures are readily achieved than possible with HIP. Expensive pressure vessels are not needed.

Experimental

Sample Preparation

Using neutronic, thermodynamic, and thermochemical calculations, Idaho National Laboratory determined the compositions of plates and the processing temperatures. All sample cleaning and preparation before Electroconsolidation were done at Argonne National Laboratory using standard fuel fabrication procedures [5].

Experimental plates were placed in a graphite die cell for treatment in the Electroconsolidation equipment at up to 600ºC for times as long as 2 hours. An initial low heating rate was used to prevent rapid out-gassing of the graphite powder chamber. The first set of control samples was fabricated by a low-pressure sintering approach (<25% of capacity). This provides a measurement to gauge the experimental results and correlations with thermodynamic predictions. Subsequently, a second set of samples will be fabricated under higher electrical current and higher pressure-assistance (Electroconsolidation) conditions. It will be also possible to roughly estimate reaction/melting temperatures.

Electroconsolidation

Electroconsolidation is one of several methods of pressure-assisted densification referred to as "soft tooling" or psuedo-isostatic pressing, in which a part to be densified is surrounded by an inert particulate medium that serves as a pressure-transmitting agent. The part and the particulate medium are contained within a die, and movement of one or more rams that are in contact with the particulate medium exerts pressure for densification. The movement of the rams and the degree of compaction of the medium are generally controlled by a hydraulic press as

illustrated in Figure 1. Electroconsolidation differs from other soft-tooling methods in the way the part is heated to the sintering temperature. Instead of heating the die chamber and having the heat transfer from the die wall to the part to be densified, an electrically conductive material such as graphite is selected as the pressure-transmitting medium. Current is made to flow through the medium, which resistively heats, thereby transferring heat directly to the part by conduction and radiation. Resistive heating provides rapid temperature rise, which enables rapid cycle times for Electroconsolidation. It has temperature capability to above 3000ºC. It also has the ability to diffusion bond all interfaces of clad fuel plates and provides more rapid cooling than HIP for the assembly in one cycle. The method has been described previously [6, 7].

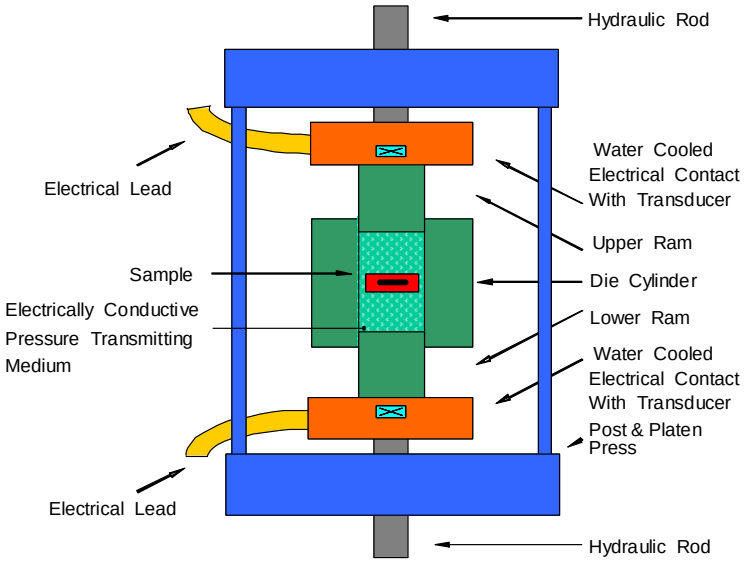

Figure 1. Diagram of the Electroconsolidation equipment and die cell.

Processing Procedure

The sample was wrapped in graphite foil and immersed in the graphite particulate medium. The foil acts as a safety precaution to contain any possible molten Al that might escape during an off-normal condition and to isolate the assembly from the graphite medium. The temperature was controlled by thermocouples installed below the sample. A pressure of 3 ksi (21 MPa) was applied prior to the heating. The temperature was increased at a nominal rate up to the set point temperature of $T_{bonding}$ +/-5ºC. The temperature and pressure required to initiate bonding depend on processing time and the interface composition. To avoid oxidation of the graphite die cell at these temperatures, it was enclosed in a flexible bellows filled with flowing nitrogen gas.

The heat treatment was followed by rapid cooling of ~150ºC /minute for 3 minutes then tapering off to room temperature. The applied pressure and temperature need to be carefully controlled to obtain maximum possible densification of the samples yet avoid stress cracking and deformation of the plate and to slow the quenching effect by reducing the thermal conduction between graphite particles.

X-ray Imaging System

The x-ray inspection system (designed and constructed by Bio Imaging Research, Inc., Lincolnshire, Illinois) enables real-time imaging and is capable of resolving dimensions to 0.07 mm (0.003 in.) during the densification process. A 160-kV, 2-ma x-ray generator (tube) and image intensifier are attached to the hydraulic press frame, as shown in Figure 2. A clear image can be displayed on the monitor every 10 seconds.

The x-ray imaging system serves as a useful control during processing and allows the real-time observation of melting-point indicators and monitoring of the densification process during Electroonsolidation.

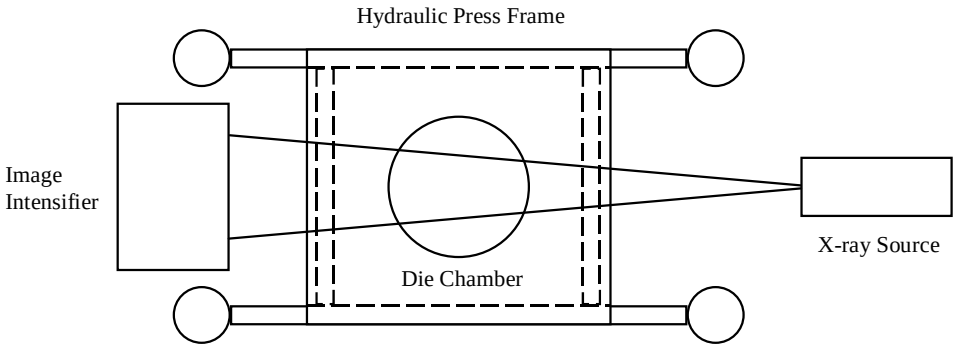

Figure 2. Top view of the press and x-ray system showing the cone angle of the x-ray passing through the graphite die chamber.

Future experimental cycles will use very small pellets of an Al-Si or Al-Mg eutectic distributed at various locations throughout the pressure-transmitting medium. These pellets are angular in shape and placed on edge with respect to the x-ray path. This arrangement provides the best view of the sudden shape change that occurs when the well-defined eutectic temperature is attained. The observation of the melting event will allow the temperature-time combination to be determined very accurately and will warn of over-temperature that would prevent melting of the assembly. From such measurements, very accurate temperature distributions throughout the die at various times can be calculated. The pellets will be used in various experiments to determine the reproducibility of the experiments as well. For the same applied current, the resulting temperature-time plot in volume distributions will show melting to occur within a few seconds of each replicated experiment.

Various eutectic materials, stable in the presence of carbon, with a wide range of melting points will be used to determine the temperature of the pressure-transmitting medium. These indicators are small enough to be placed in the medium without interfering with the sample. The computer based analytical system was programmed to take a sequence of x-ray images at fixed time intervals and record them to provide a history of densification.

Results and Discussion

The initial experiments were performed to estimate the elongation of samples and bonding of Al plates. Results were positive, and the applicability of Electroconsolidation was tested with a monolithic plate. Figure 3 is a cross section of an Al/stainless steel surrogate plate inside a metallographic clamping device. The top has a rough surface as a result of pressing by the graphite particles. The bottom surface is a smooth replica of the polished graphite block. The block can be modified and used as a contouring press or/and embossing die for outward flanging or cooling fins.

Figure 3. Cross section of Al/stainless steel surrogate plate inside clamping device.

The sample was polished and examined with an optical microscope. As expected, a thin reaction zone between the Al and stainless steel was formed with thickness of ~ 10 microns. Figure 4A shows the middle of the Al/ stainless steel surrogate plate and Figure 4 B, the same thickness of reaction zone at the edge of the plate.

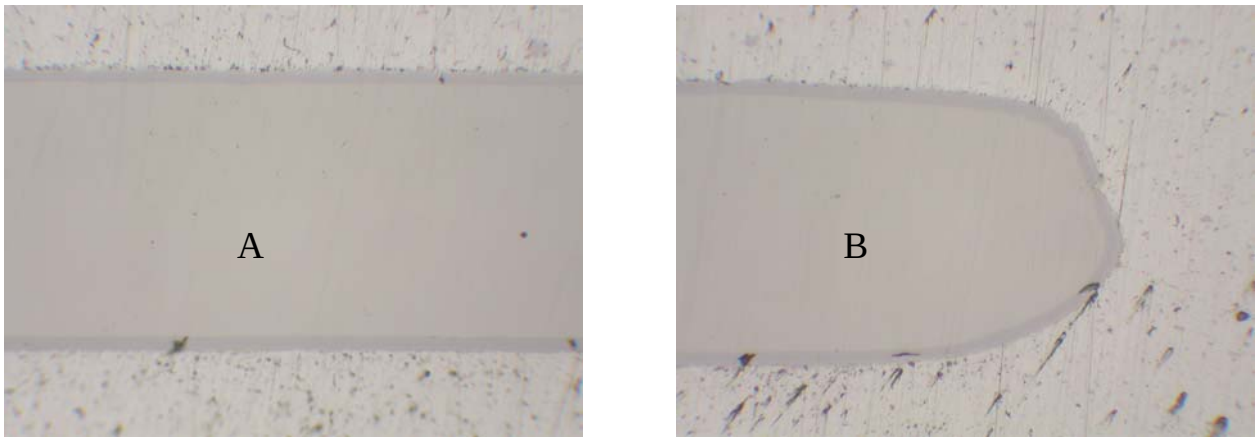

Figure 4. Cross section of Al/ stainless steel surrogate plate with reaction zone in middle (A) and at edge (B).

The microstructure of the boundary between the two Al parts of the plate was studied after deep etching. Grain growth across the interface was observed, but excessive grain growth did not develop because the two Al parts of the upper and lower cladding plates had different grain sizes. Figures 5 and 6 represent the typical microstructure of the boundary in different experiments.

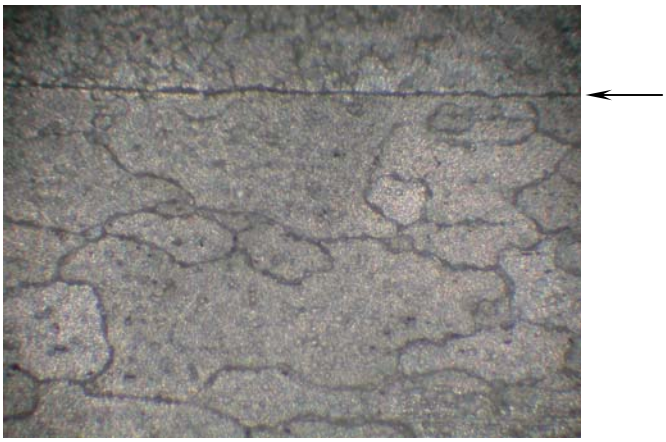

Figure 5. Cross section of Al/Al interface of stainless steel surrogate plate outside reaction zone.

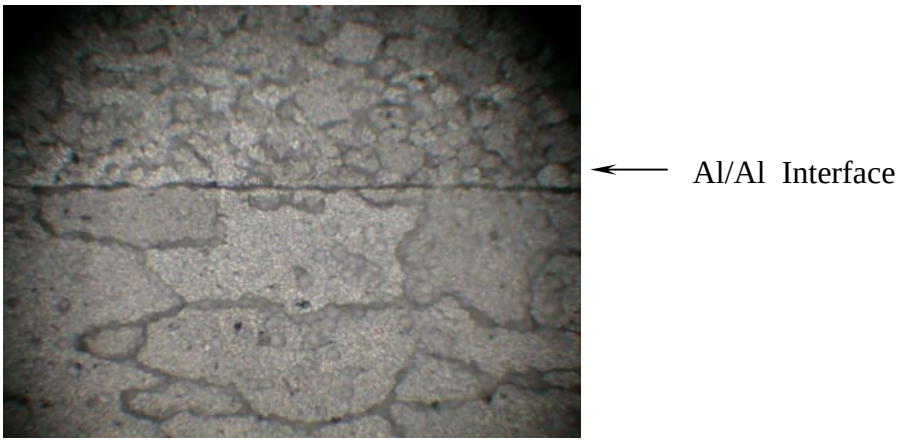

Figure 6. Typical microstructure of boundary between two Al parts of plate with grain growth across the interface of two plates of different grain size.

Grain growth at the interface was observed in the heat-affected welding zone also (Figure 7). The process of growing grains was connected with changing of the boundary profile.

Figure 7. Typical microstructure of boundary between two Al parts in heat-affected welding zone.

Conclusions

These preliminary results have demonstrated that the Electroconsolidation® Process is a promising candidate to produce diffusion-bonded monolithic fuel plates with contoured and/or complex surfaces. The main advantage of this process over conventional diffusion bonding is the minimization of time-at-temperature due to the rapid heating inherent to the process. This is important for uranium alloys with metastable structures such as U-10 Mo. Bonding results of surrogate fuel plates made with stainless steel and depleted uranium monolithic fuel plates demonstrated the important role of aluminum alloy selection with small grain size to obtain the required cross-boundary grain growth. One or more fuel plates fabricated by electroconsolidation are planned to be included in upcoming irradiations.

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