ANL/ET/CP - 94634

# Study of Diffusion Bond Development in 6061 Aluminum and Its Relationship to Future High Density Fuels Fabrication*

by

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September 1997

To be presented at the 1997 International Meeting on Reduced Enrichment for Research and Test Reactors, Oct. 5-10, 1997, Jackson Hole, WY

*Work supported by the U.S. Department of Energy, Office of Nonproliferation and National Security, under Contract W-31-109-Eng-38.

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## ABSTRACT

Powder metallurgy dispersions of uranium alloys and silicides in an aluminum matrix have been developed by the RERTR program as a new generation of proliferation-resistant fuels. Testing is done with miniplate-type fuel plates to simulate standard fuel with cladding and matrix in plate-type configurations. In order to seal the dispersion fuel plates, a diffusion bond must exist between the aluminum coverplates surrounding the fuel meat. Four different variations in the standard method for roll-bonding 6061 aluminum were studied. They included mechanical cleaning, addition of a getter material, modifications to the standard chemical etching, and welding methods. Aluminum test pieces were subjected to a 'bend' test after each rolling pass. Results, based on 400 samples, indicate that at least a 70% reduction in thickness is required to produce a diffusion bond using the standard rollbonding method versus a 60% reduction using the Type II method in which the assembly was welded 100% and contained open 9mm holes at frame corners.

## INTRODUCTION

In March 1996, after a 6-year moratorium during which there was no RERTR fuel development, the U.S. Department of Energy resumed a program to develop advanced LEU fuels with the objective of improving options and performance of research reactors undergoing conversions.[1] As a result of this continuation after such a long hiatus, initial efforts concentrated on the production of microplates, machined to dimensions of approximately 76 x 22 x 1.3 mm (3.0 x 0.875 x 0.050 in.) containing dispersion fuels with different types of uranium compounds and alloys. The path of the experimental and fabrication activity was a series of rolling experiments with plates of 6061 aluminum (1.0Mg - 0.6Si - 0.3Cu - 0.2Cr, balance Al) to determine the amount of reduction required to roll bond this alloy.

We had two goals for these experiments:

- to confirm that the smaller amount of reduction scheduled for microplates is sufficient to achieve bonding
- to study the roll-bonding process which will aid in the production of future microplates which will contain a magnesium matrix.

The plates were hot-rolled, blister-annealed, sheared into bond samples, and tested.[2]

Promising methods are discussed in this paper that may lead to engineering or tailoring the composition and microstructure of aluminum cladding alloys to provide optimized diffusion bonding. When planning future activities in the development of new fuel elements, it is necessary to use the published data base and especially to incorporate new results from future experimental studies closely focused on RERTR requirements. Subsequent feasibility studies based on these new experimental results will help discourage the use of highly-enriched uranium (HEU) in a new generation of test reactors.

## EQUIPMENT and EXPERIMENTAL PROCEDURES

Prototype fuel plates were produced by hot and cold rolling assemblies consisting of 6061 covers and frames with cavities. Equipment utilized included a 100 x 406 mm (9 x 16 in.) rolling mill with flat rolls, resistance furnace with air environment, and rolling temperature of 500°C.

The first irradiation matrix plan for the high density fuel microplates included a decrease in the total amount of reduction from the standard ~88% to ~80%. Although it is believed that this small change will not affect the quality of bonds, it was considered prudent to prove that the plates would be acceptably bonded. Additionally, a better understanding of the bonding process will be required to produce some of the plates planned for the subsequent irradiations.

The matrix of the processing methods is given in Table I and consists of 6061 aluminum test plates sized approximately 75 x 150 x 5 mm (3 x 6 x 0.187 in.) containing no compacts using four different types of assembly methods. The reference for comparison was the standard assembly method consisting of chemically cleaning the 6061 aluminum with Oakite 160 ( a commercial NaOH-based etchant) and then welding the assembly such that the corners and the center of the rear are left open.

Type II was the same as the standard method except that the assembly was welded 100% and 9mm (0.35 in.) holes near the four corners were left empty in the frame. It is believed that these holes collected residual gas and prevent blisters from forming in the fuel plate area. Type III was the same as Type II except that the inner bonding surfaces were wire brushed before assembly. Type IV was also the same as Type II except that the holes in the frame were filled with zirconium turnings. These turnings acted as a getter for any residual oxygen. Individual assemblies were given from one to four 25% reduction in thickness passes at 500°C except for the Type IV which consisted only of the two test plates.

After rolling, the plates were cut in half. One half was retained in the as-rolled condition and the other half was blister annealed. The plates were then marked and cut into nominally 12.5mm x 75mm (0.49 x 2.95 in.) strips with the longer dimension transverse to the rolling direction.

The samples were then bend tested to an approximately 1-T bend. The bend testing consisted of one 90° bend to the left; return to 0° and then a 90° bend to the right; return to 0°. This tested both of the bonded interfaces in a tensile mode. This is similar to the test used by CERCA, on full-sized fuel plate assemblies to check the bonding.

## EXPERIMENTAL RESULTS and DISCUSSION

Approximately 400 bend-testing samples from the rolling experiments with 6061 aluminum plates were examined to determine the amount of reduction required to roll bond this alloy. Table 1 lists the bend test results. No bonding was observed after the first two 25% reduction in thickness passes. Partial and good bonding was observed after the third pass. After the fourth pass, the methods were rated as follows. The best bonds appeared to be generated by fully welded samples. This method was followed by the wire brushed samples, then the gettered samples, and finally the standard method. It is encouraging that all three changes to the standard method appeared to improve the bond quality.

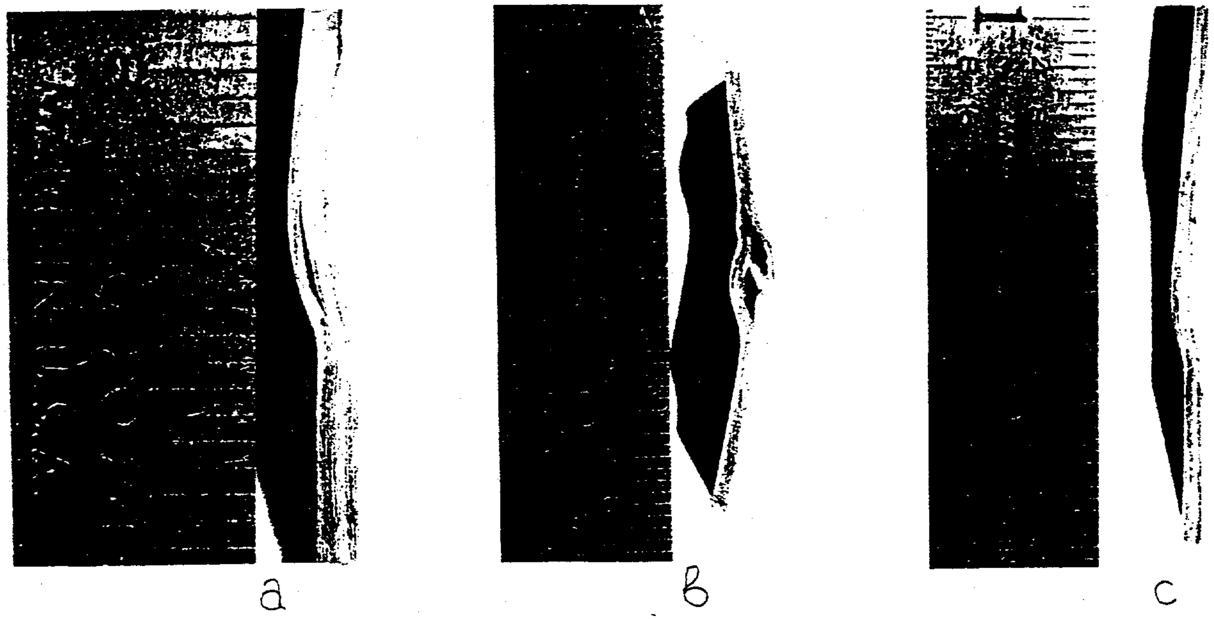

Figure 1 - a, b, and c, show typical samples after bend testing. In Fig. 1a, the bend-testing sample is shown to be partially bonded, In Fig. 1b, the bend-testing sample is shown to be not bonded. In Fig. 1c, a good bond is illustrated.

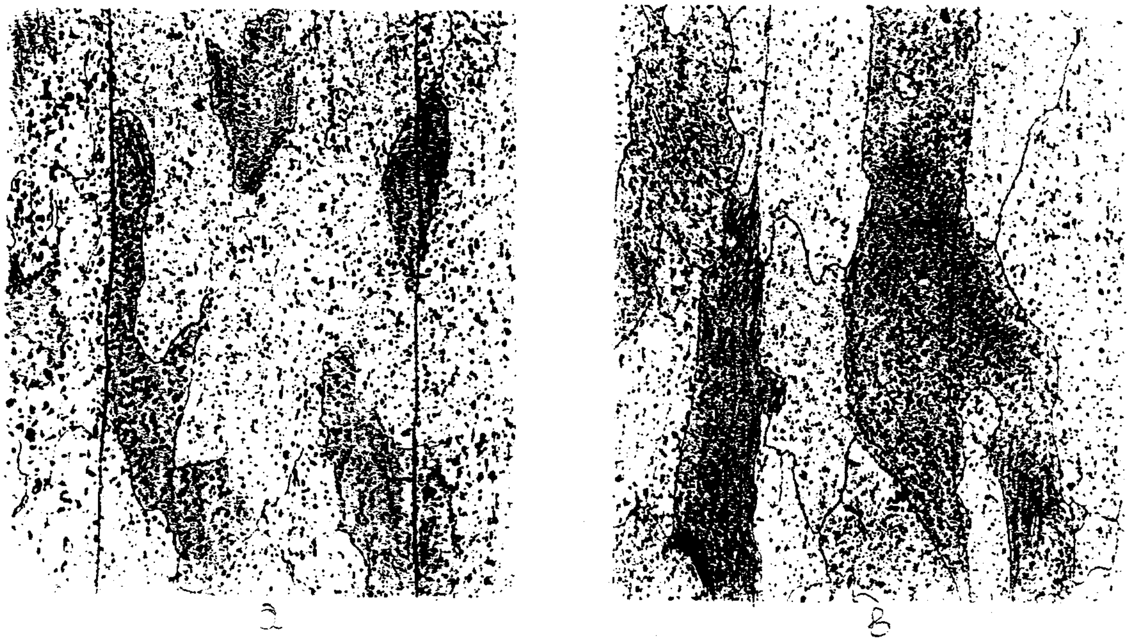

Figure2. a and b. show the microstructure zone adjoining two plates. Figure 2a shows some grain growth. Figure 2b shows more than 50% grain growth.

Table 1. Results from Roll Bonding Experimental Matrix

| Rolling Pass (25% Reduction per each pass) | Standard[a] Assembly Method | Type II[b] Assembly Method | Type III[c] Assembly Method | Type IV[d] Assembly Method |
|---|---|---|---|---|
| First | Not bonded | Not bonded | Not bonded | No sample |
| Second | Not bonded | Not bonded | Not bonded | Partially bonded |
| Third | Partially bonded | Bonded | Partially bonded | No sample |
| Fourth | Bonded<br>Fourth Best | Bonded<br>Best Quality | Bonded<br>Second Best | Bonded<br>Third Best |

[a] The standard assembly method consists of chemically cleaning the 6061 Al and then welding of the assembly such that the corners and the center of the rear are left open.

[b] The same as the standard method except that the assembly is welded 100% and 9mm holes near the four corners were left open in the frame.

[c] The same as Type II except that the inner bonding surfaces are wire brushed before assembly.

[d] The same as Type II except that the holes in the frame are filled with Zr turnings.

As mentioned earlier, one of the objectives of these experiments discussed herein was to study the roll-bonding process which will aid in the production of future microplates containing magnesium matrix. However, it is necessary first to understand a little better about the result of previous experiments using degassed Alcoa No. 718 Al powder (with 12.2 wt% Si - 0.4wt% Fe) as an alternative to the standard MD101 Al powder (99.3 wt% Al).

## Al-Si Powder Chemistry

It had originally been postulated that the reaction between Al and U would be reduced by as much as a factor of five if a eutectic Al + Si alloy replaces Al, a very beneficial possible effect.

Experimental results showed that the use of 718 Al (12.2 wt% Si - 0.4wt% Fe) as the matrix produced an increase in the fuel zone volume growth rate after annealing at 400°C. This was with a $U_3Si_2$ fuel alloy making up 32% of the matrix. The volume increase was ~20X greater compared to MD 101 Al powder (99.3 wt% Al) being used as the matrix under otherwise identical conditions. That was interpreted as an increased reaction zone compared with MD 101 Al powder and higher fabricated porosity in the 718 matrix fuel plates.[2]

Today we can formulate two causes for these observed phenomena. Previously, the release of hydrogen was suggested[2] as being a possible cause of the increased porosity and fuel zone volume. Today, we can also suggest that interfacial conditions between the matrix and the fuel alloy could have played a leading role.

Analysis of data in the literature shows a complex structure formed for eutectic alloy with 12% Si. The structure contains dendritic solution and globular forms of Si particles existing between branches of dendrites.

The size of the Si particles are related to the temperature of the melted solution. After quenching from a temperature of 650-700°C, these Si particles have a size of 0.010-0.040 µm, inversely dependent on the concentration of Si in Al. The larger particles are associated with the least Si-enriched Al. The concentration of Si particles in the eutectic varies also as a function of the Si in Al and can range from approximately $10^3$ - $10^4$ in a $\mu m^3$ volume. For noneutectic conditions and a higher concentration of Si, for instance 20% Si, it may be $6 \times 10^5$. For noneutectic conditions and a lower concentration of Si, for instance, 6%Si, it may be $4 \times 10^3$ Si particles. Equilibrium solid-solution between Si particles and solution of Si in aluminum is temperature and time dependent. It can be seen from standard Si/Al phase diagrams, with temperatures from ambient to 300°C, the solid-solution Si concentration $< 0.1$ %. As the temperature increases to 400°C, the Si concentration increases to 0.3%.

As the temperature increases, for example from ambient to 400°C, the solid-solution of Si in Al will have a deficit of Si. In other words and for this particular situation, Al will devour Si either from the globular particles or can react with $U_3Si_2$.

It is necessary to make additional special experiments about the solubility, reactivity, and diffusivity of this system under powder (45-150 µm particle size) conditions. Previous work[3] was done under solid-solution conditions; varying interfacial grain boundaries and morphologies of powder particles will possibly produce quite different results including different rate-controlling mechanisms. Experimentally determining any differences between uranium compound powder reaction kinetics and thermodynamics and the system in Al-Si solid-solution may be quite revealing. The large surface area associated with powder reactions can accumulate more gaseous impurities but may also have more nonequilibrium energy which can accelerate reactions at the surface

Sokursky et al [4] wrote about previous experience in using Al-Si alloys with Si concentrations ranging from 10.5-11.5 %; this is below the eutectic value. Wiencek et al [2] discussed their experience in working with 718 Al (containing 12.2 wt% Si) and apparently above the eutectic value, variously referenced, but ~12wt% Si. Small differences in concentration near the eutectoid point can play an important role for this reaction.

A possible problem with using Al-Si alloys is the lack of knowledge that we currently have about their mechanical properties (i.e. plasticity as an example) is their intrinsic limitations with plasticity. It is necessary to make a note of the mechanical properties of Al-Si alloy. Earlier researchers [5,6] reported that irradiation appeared to not have a big influence on the elongation of the Al-7%Si alloy. However their interpretation is not clear because plasticity had a level of approximately 2%. Because of heat transfer problems, such alloys exhibiting little elongation should not be used with uranium compounds which have the potential of a few per cent swelling and different thermal expansion coefficients.

## Magnesium

Magnesium is an appealing prospective alternative to Al-Si as the matrix material because (1) it is commonly believed to have less reactivity (greater compatibility) with uranium alloys and silicide fuels, (2) low cross-section capture area for thermal neutrons, and (3) ready commercial availability.

Previously, magnesium matrix plates rolled at 400°C using the standard method did not bond[2] and some changes in the standard procedures will be necessary to successfully fabricate these plates. It is not clear why the Al base 6061 clad did not bond to each other with hot rollbonding at temperatures of 385°C and 415°C when containing Mg matrix compacts. Our results in the present study show that there may be significant room for improvement in the roll bonding techniques and roll bonding of magnesium matrix samples may be achievable.

Applications of magnesium for use as the fuel zone matrix can be developed by two directions.

One direction is by improving the process of cold rolling. Chinese researchers[7] developed an innovative cold rolling technique for $U_3Si_2$-Al dispersion fuel plates with high uranium loading. Although never applied to Mg-matrix fuel plates, they developed a special regime of rolling through which cold rolling for developed fuel elements or plates for 7 gm/cc equivalent uranium loading was achieved. Their early work needs further development in many respects and needs to be tested with Mg-matrix fuel plates. For cold-rolling, it is probably preferable to use a Mg-containing modification to 6061 Al alloy. From an elemental perspective, alloys in the systems of Al-Mg-Zr or Al-Mg-Si or Al-Mg-Ni may be good first candidates to test in this modified regime for coldrolling and annealing. For cold-rolling, larger concentrations (~1%) of Ni and Fe can increase resistance to both nonuniform cold deformation and embrittlement. The influence of these alloying elements (Ni and Fe) can be attributed to a different mechanism for microplastic deformation when compared to Zr, Mo, or Mn as alloying elements. Ni and Fe can substitute into an intermetallic phase and not initiate embrittlement or failure. By slightly changing the composition of the basic 6061 Al alloy, the properties will be dramatically altered.

The second way to proceed to be able to utilize Mg for use as the fuel zone matrix is to first develop a special aluminum alloy powder with high concentrations of Mg. Researchers[8] at the Baikov Institute of Metallurgy in Moscow have developed Al-Mg alloys with much higher Mg concentrations. The ultimate tensile strength increases as a function of increasing Mg content, and ranges from 200MPa to 400MPa. The engineering strain is approximately 0.20 (for melting and quenching conditions). Plasticity of the Al-Mg alloys remained approximately constant in the range of Mg<15%; there was no data for Al-Mg alloys with greater than 15%. However, it is known that the compound $Al_5Mg_8$ (>15% Mg) can play a role in initiating microcracking in condition of deformation.

Although from the preceding mechanical properties information, Mg-enriched Al alloys look very appealing, the behavior of Al-Mg-Si alloys under irradiation conditions need more study to determine their stability. The addition of small quantities (<0.5%) of the alloying elements, Zr and Mo, have the potential of increasing the resistance of grains to recrystallization and growth at high temperatures due to the formation of intermetallics which control migration of boundaries but, since both Mo and Zr are used in U alloys, it is not clear if there would be a problem with diffusion during hotrolling or future irradiation conditions.

Methods have been developed[8] for minimizing the size of intermetallic inclusions in various alloys, such as Al-Mg; one such method is based on using a $K_2ZrF_6$ salt for melting. Their methodology can be further examined to begin to formulate standards for the size of intermetallic particles in alloys for nuclear fuel plates.

Our experiments of rollbonding indicate that the condition of the surface of the fuel plates is a key factor in achieving good quality bonding (Table 1). It appears reasonable that similarly the surface condition and surface micro-relief will have a main role in bonding Al-base alloys at different temperatures when using cold-rolling techniques. At lower temperatures, the surface micro-relief will be coarser and more pronounced compared to conditions at higher temperatures, such as at 300°C. Micrographs indicate that there is extensive variability in surface micro-relief associated with the type and concentration of alloying elements. By controlling conditions of cold-rolling, alloying, and annealing, and carefully controlling surface micro-relief and constraining grain size increase as a function of recrystallization, it should be possible to create good quality bonding using Mg-containing powders.

### Implications

This and the previous experience of the ANL team as well as that of other fabricators have all demonstrated the potential of roll-bonding technology for fabrication of fuel elements of different types. Application of roll-bonding was demonstrated by other researchers[7,9] and industrial companies in the conversion of reactors to use new low-enrichment materials as a substitute for previous fuels.

In parallel with the process of replacing old fuel elements and cladding in previously-constructed test reactors, it is necessary to develop new fuel elements for future test reactors which today are under conceptual or engineering design study. It is also essential to conduct concurrent studies on candidate cladding materials, such as low activation stainless steel, zirconium, vanadium, and other alloys.

For the conceptual design of test reactors and facilities, it is necessary to have data concerning the fuel and whether it will meet the heat transfer, burnup level, and burnup rate requirement. This data base can be generated as part of the base technology development and during the testing of the design prototype.

Vatulin[10] demonstrated that it is important that the geometry (plates or rods) of the fuel element components used for future assemblies of future reactor design can have a governing effect on mechanical properties such as swelling. Our experience with fabricating plates demonstrates that it is possible to regulate the parameters that are important for future design.

Nonproliferation activities such as the RERTR program will be effective in preventing the use of highly enriched uranium for new applications if the current work will result in the development of fuels for future reactors also. Thus new generations of nuclear reactors will not need to rely upon a foundation of higher-enriched uranium but will have a strong technical and engineering foundation provided for the use of LEU.

## SUMMARY and CONCLUSIONS

Results show the possible potential decrease in plastic deformation for bending while maintaining hermetic joints and good microstructure. Aspects of these results can possibly be extrapolated for use with fuel plates containing a magnesium matrix. Nonproliferation activities such as the RERTR program will be effective in preventing the use of highly enriched uranium for new applications if the current work will result in the development of appropriate new fuels for future reactors also. The creation of fabrication resources in ANL-W and the renewal of the fabrication and processing group in ANL-E will give the possibility to use unique technical resources for developing new processes and materials for the RERTR program.

We are glad to be cooperative partners for the future path:

- develop process making powder with different size and composition
- develop technology for compacting, isostatic pressing, rolling, welding, and heat treatment, new materials and fuel elements
- fabricate equipment and samples for reactor irradiation.
- participate in feasibility studies of new designs of fuel elements for facilities and test reactors and neutron sources.

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