Development and Compatibility of Magnesium-Matrix Fuel Plates Clad with 6061 Aluminum Alloy

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Abstract

Aluminum is a commonly used matrix for research reactor fuel plates. Reaction at the higher range of reactor fuel operating temperatures between the fuel and its Al matrix may reduce the irradiation stability of the fuel. To further understand the contribution of this reaction to irradiation stability, experiments were performed to develop a nonreacting matrix. The work focused on magnesium, which is an excellent nonreacting matrix candidate with a neutron absorption coefficient similar to that of Al. To avoid the formation of a liquid Al/Mg phase, the roll-bonding process was improved to achieve acceptable bonding at 415°C. After these methods were developed, fuel plates were produced with two fuels: uranium-2 wt.% molybdenum, and U-10 wt.% Mo with two matrices (Al and Mg). A reaction between the Mg and the 6061 Al cladding occurred during processing at 415°C. To minimize the extent of this reaction, we developed methods to roll-bond the fuel plates at 275°C. No reaction zone was observed in fuel plates processed at 275°C. With this method, fuel plates with an Mg matrix will be fabricated and included in the next irradiation matrix for the RERTR high-density fuel development program.

Introduction

Aluminum is the matrix most commonly used for research reactor fuel plates. Using Al as the matrix allows processing at 500°C because the minimum liquid formation temperature is 640°C in the U/Al binary system[1] (Fig. 1). However, the U/Al

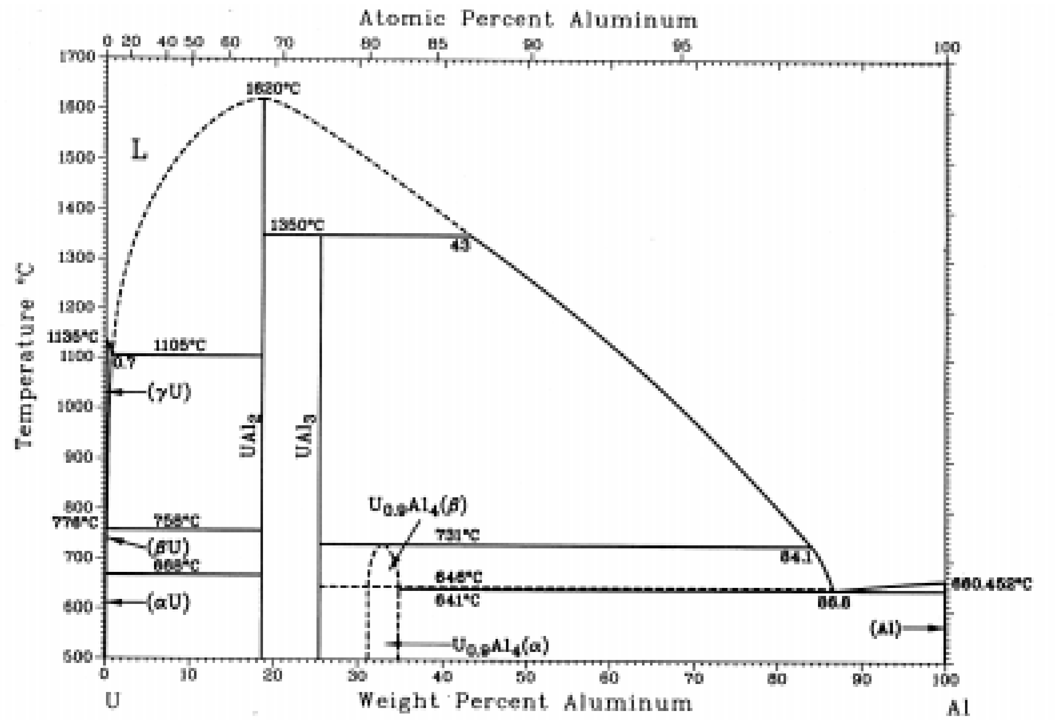

Fig. 1. U-Al phase diagram.

system contains three compounds (UAl_2, UAl_3, and UAl_4) that are thermodynamically very stable.[2] The reduction in energy level is a driving force for the formation of such UAl_x compounds in Al-matrix fuel plates. A reaction between the fuel and the Al matrix may reduce[3] or increase[4] (in low loaded plates) the irradiation stability of the fuel. To further understand the contribution of this reaction to irradiation stability, we conducted experiments to develop a nonreacting matrix. Any such reaction may be minimized or eliminated if the matrix material forms no U-X compounds. An excellent candidate that meets these requirements and has a neutron absorption coefficient similar to that of Al is magnesium. The U/Mg system,[1] which is presented in Fig. 2, contains no compounds. However, the introduction of Mg requires a review of the Mg/Al system[1] (Fig. 3). To prevent possible formation of a liquid phase, fuel plates with an Mg matrix must be rolled at <437°C.

This poses a problem because conventional roll-bonding and blister-testing processes require a temperature of 500°C.[5]

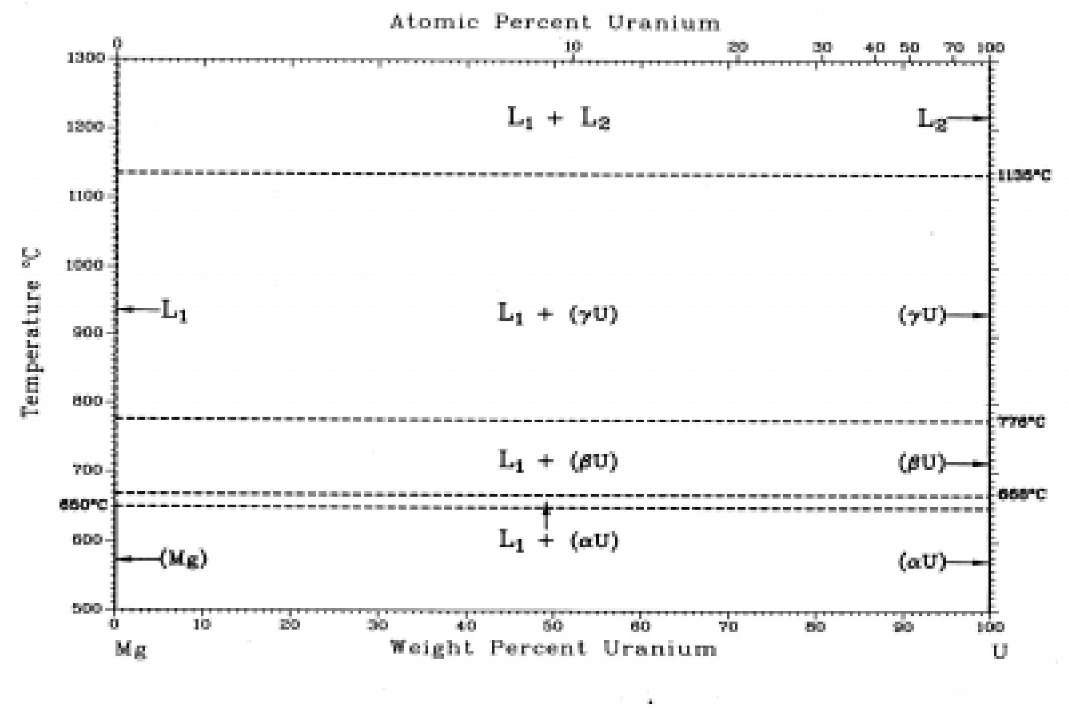

Fig. 2. U-Mg phase diagram.

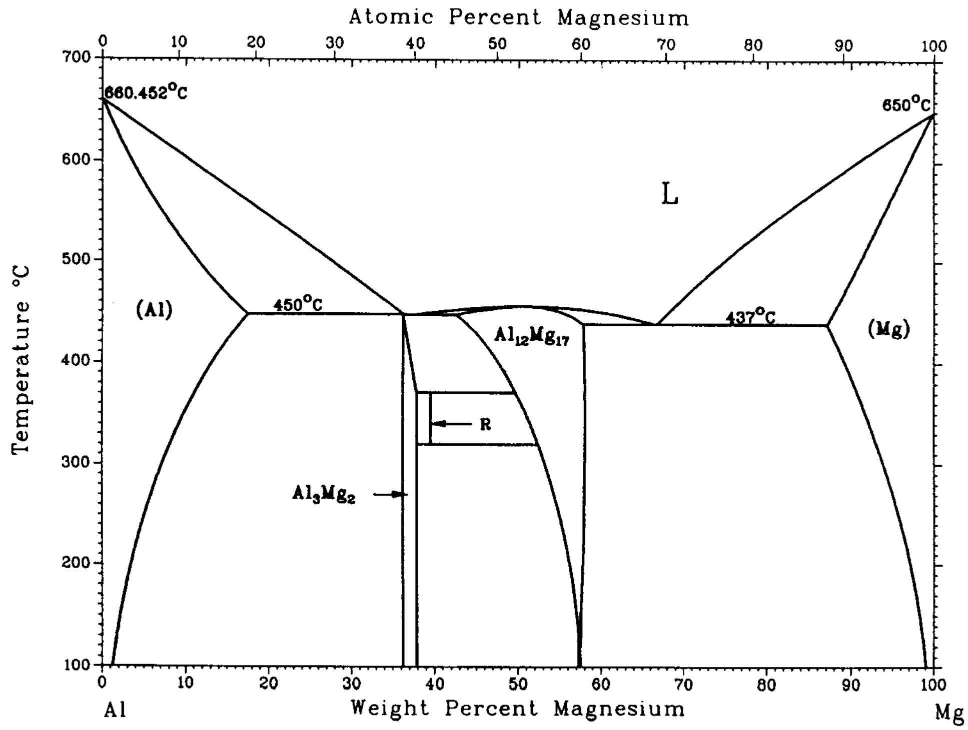

Fig. 3. Al-Mg phase diagram.

Improvements to the roll-bonding process to achieve acceptable bonding at 415°C were the first goal of the study. After the improved methods were developed, two fuels (U-2 wt.% Mo and U-10 wt.% Mo) were studied to determine if a reaction occurs between them and an Mg matrix.

Equipment and Experimental Procedures

Test plates of 6061 Al (0.40-0.8 Si;0.7 max. Fe; 0.15-0.40 Cu; 0.15 max. Mn; 0.8-1.2 Mg; 0.04-0.35 Cr; 0.25 max. Zn; 0.15 max. Ti; 0.05 max. others; balance Al) were rolled at 415°C and then annealed for 1 h at 415°C. After the development of acceptably bonded test plates, 5 x 15.2 cm (2 x 6 in.) fuel plates were fabricated to the specifications shown in Fig. 4. The fuels, matrixes and cladding used in the plates are listed in Table 1. Two fuels were studied; depleted uranium (DU)-2 wt.% Mo powder purchased from Nuclear Metals, USA; and DU-10 wt.% Mo powder received from the Korean Atomic Energy Research Institute (KAERI). Both fuels were spherical. After processing, one Mg-matrix plate was sectioned to determine the baseline extent of reaction. The remaining plates were then heat treated at 400°C for various times. Samples were sectioned and examined by optical metallography, scanning electron microscopy (SEM), and energy-dispersive analysis by X-ray (EDX) to determine any reaction zones.

Results and Discussion

Roll Bonding Tests at 415°C

After a test plate made by the conventional Argonne National Laboratory method (but with the rolling temperature reduced from 500°C to 415°C) showed nonbonds, a different rolling schedule, similar to that used by the Indonesian National Atomic Energy Agency (BATAN), was tried. The new schedule gives excellent bonding results after rolling at only 415°C, which is sufficiently below 437°C to allow testing with Mg.

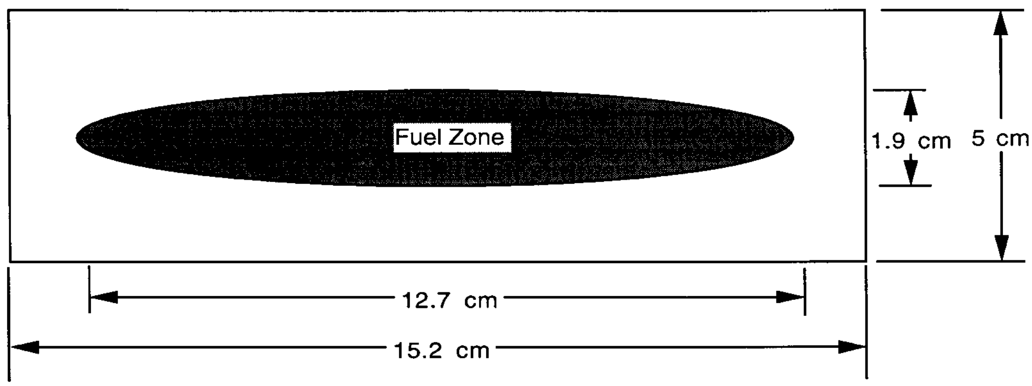

Fig. 4. Design of fuel plate for bonding and compatibility study experiments.

Table 1. Fuels Fabricated into Test Plates

Test Plate Number	Fuel	Loading Volume %	Matrix	Cladding[a]
1	U-2 wt.% Mo	25	Magnesium	6061 Al
2	U-2 wt.% Mo	25	Magnesium	6061 Al
3	U-2 wt.% Mo	25	Magnesium	6061 Al
4	U-2 wt.% Mo	25	Aluminum	6061 Al
5	U-2 wt.% Mo	25	Aluminum	6061 Al
6	U-2 wt.% Mo	25	Aluminum	6061 Al
7	U-10 wt.% Mo	25	Magnesium	6061 Al
8	U-10 wt.% Mo	25	Aluminum	6061 Al

[a]Composition of 6061 Al alloy is 0.40-0.8 Si;0.7 max Fe; 0.15-0.40 Cu; 0.15 max Mn; 0.8-1.2 Mg; 0.04-0.35 Cr; 0.25 max Zn; 0.15 max Ti; 0.05 max others; balance Al.

The first test assembly consisted of a 6061 Al frame with two empty cavities located near the trailing end, and two 6061 Al cover plates. The assembly was cleaned and given a total of 82% reduction in thickness at 415°C. After hot rolling, the assembly was given a 1-h blister annealling at 415°C. Two small blisters were observed over the intentional cavities, indicating that bonding had occurred over the rest of the plate. Bend-test samples were taken and also indicated that the assembly was bonded. As a final confirmation of bonding, a metallographic examination of the interface confirmed >50% grain growth across the interface (Fig. 5). With the bonding confirmed, test plates with an Mg matrix were then successfully roll-bonded at 415°C. The assembly consisted of a 6061 Al frame with two compacts having 25 vol.% tungsten powder surrogate fuel in an Mg matrix and two 6061 Al cover plates. Two empty cavities were again located near the trailing end. Bonding was confirmed by the observation of blisters over the empty cavities after a 1-h blister annealling at 415°C and bend-testing.

After proving the feasibility of the concept, we processed a matrix (see Table 1) of eight compatibility plates by the method described above. Once again, metallurgical bonding was confirmed by the presence of blisters over the empty cavities and by bend-testing. Following rolling at 415°C and the 1-h blister test, a U-2 wt.% Mo-Mg-matrix plate was sectioned in as-fabricated condition. We had anticipated that with the lower rolling temperature, any reaction between the cladding and the matrix would be minimal. However, as Fig. 6 shows, a significant reaction had occurred. EDX analysis on an SEM showed the layer

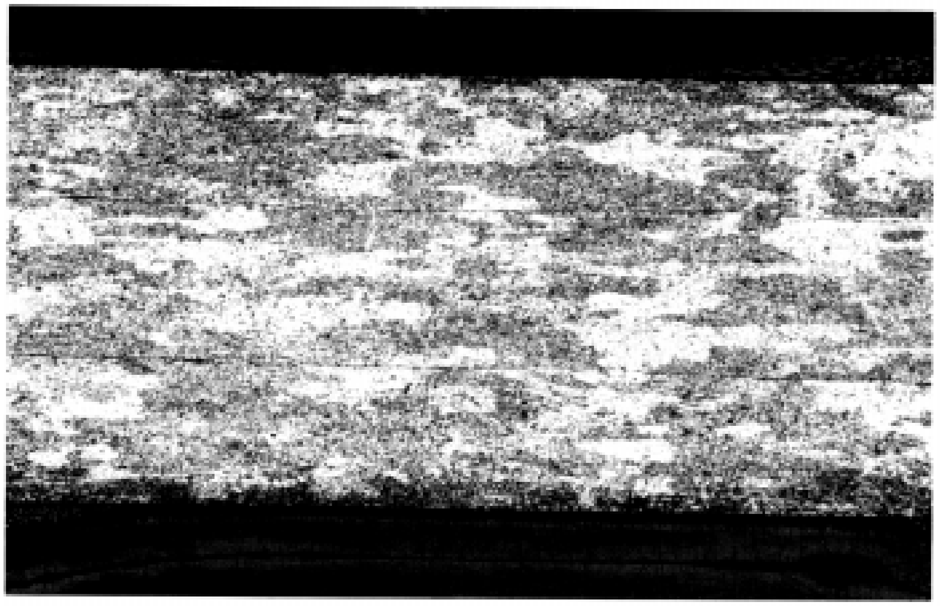

Fig. 5. Bonding at 415°C confirmed by grain growth across interfaces; ≈ 50 X.

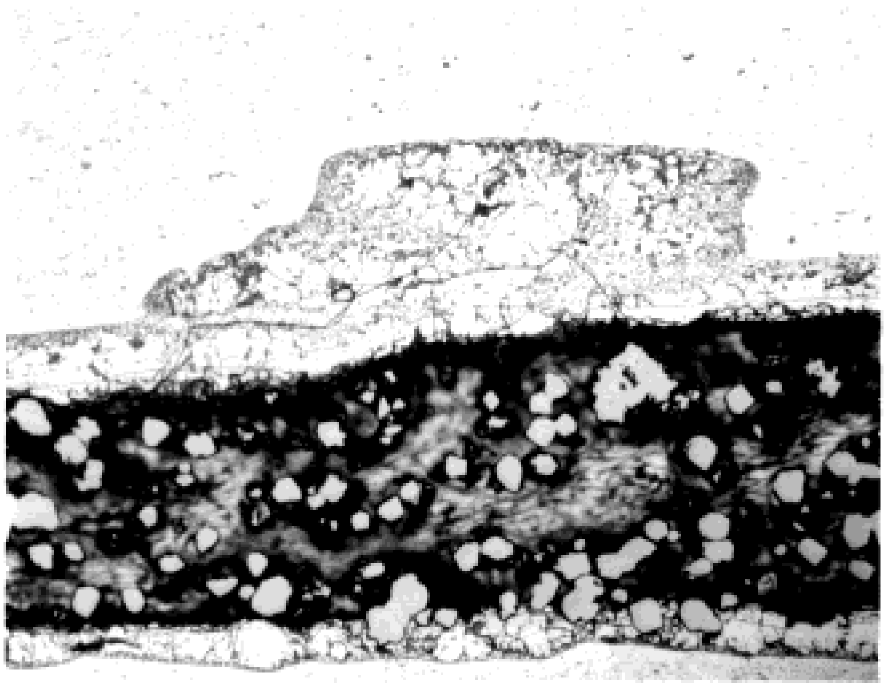

Fig. 6. Diffusion zones for 6061-Al-Mg-matrix processed at 415°C; ≈ 200 X.

nearest the Mg matrix to be $Al_{12}Mg_{17}$, and the layer nearest the cladding to be Al_3Mg_2. Since compatibility was to be tested at 400°C, this reaction would invalidate the results because one assumption made for calculating the volume changes is that any possible reaction is zero or statistically small enough to ignore because of the lower amount of cladding/matrix interface available for reaction. This assumption of near-zero reaction had been found to be true for Al-matrix fuel plates.[6]

After the reaction was found, we decided to continue the compatibility studies without volumetric measurements. After sufficient time, the center of the fuel zone was examined for a reaction product. It was hoped that the reaction zone between the cladding and the matrix would grow to become an effective barrier to cladding diffusion between the matrix and fuel. After 420 h at 400°C, the U/Mg matrix plates and the U-2 wt.% Mo/Al matrix plate were sectioned. To reduce the time required to produce a reaction between the U-10 wt.% Mo-Al-matrix plate, the temperature was increased after 184 h at 400°C and, after an additional 236 h at 500°C, this plate was sectioned. Results of the experiment indicate very little or no reaction between these fuels and the Mg matrix. U-2 Mo was found to react with an Al matrix and formed a UAl_3 compound. U-10 Mo underwent only minimal reaction at 400°C but also formed UAl_3 after heat treatment at 500°C. These results are in agreement with previous KAERI studies.[7]

Roll Bonding Tests at 275°C

To minimize the reaction between the cladding and the Mg, we reduced the rolling temperature to 275°C and significantly changed the 415°C rolling schedule. An assembly consisting of a 6061 Al frame and two cover plates was rolled; the assembly contained four compacts: two with 25 vol.% W powder surrogate fuel in an Mg matrix, and two consisting of 100% Mg. After blister-testing at 275°C, we confirmed bonding by observing blister formation and by bend-testing. Figure 7 is a cross section of the surrogate fuel zone. No reaction is seen between the cladding and the matrix, and this was confirmed by EDX.

An additional benefit of reduced working temperature is the lowering of the tendency of phase transformations (γ decomposition) of particularly high U alloys during fabrication.

Any possible improvement in the irradiation properties of plates processed at 275°C could not be practically tested by compatibility studies because the diffusion rates at 275°C are very low. The effect of an Mg matrix can be tested only in-pile, and we are currently planning to include Mg-matrix fuel plates in the next microplate irradiation matrix.

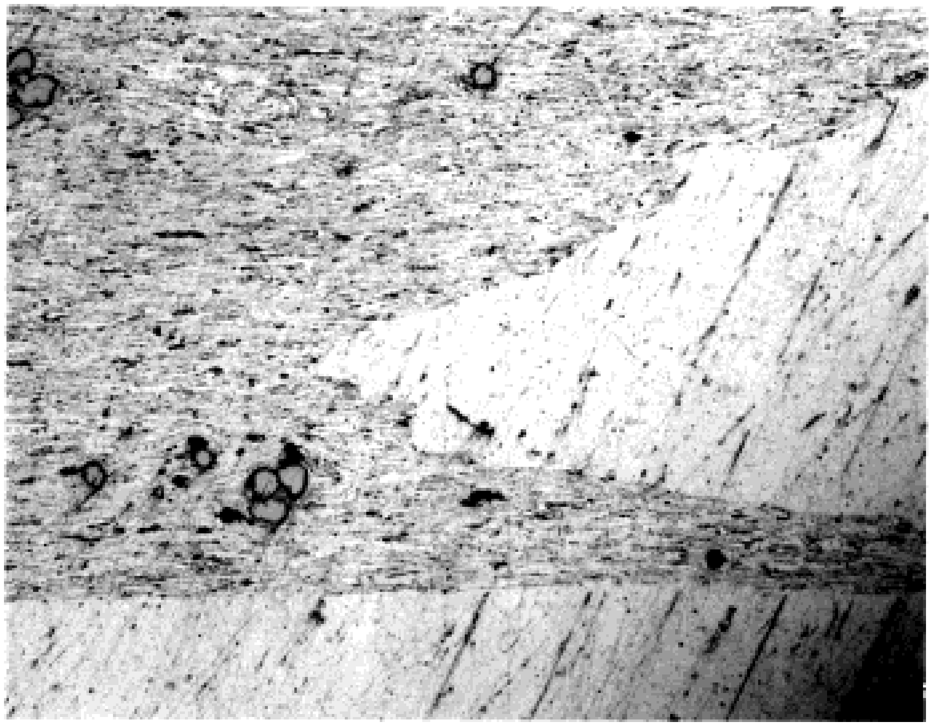

Fig. 7. Interface between 6061-Al-clad Mg-matrix processed at 275°C; ≈ 200 X.

Summary and Conclusions

Fuel plates with a magnesium matrix were developed in an effort to minimize reaction between the fuel and the matrix. We found that processing at 415°C caused significant reaction between the 6061 aluminum cladding and the Mg matrix. Methods were developed to achieve metallurgical bonding of the cladding by processing the fuel plates at 275°C. No evidence of any reaction was seen in the fuel plates rolled at that temperature. We plan to include Mg-matrix fuel plates in the next group of microplates to be irradiated.

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