ANL/ET/CP - 97559

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

by

Thomas C. Wiencek, I. G. Prokofiev and D. J. McGann
Argonne National Laboratory
Argonne, IL USA

October 1998

To be presented at the
1998 International Meeting on
Reduced Enrichment for Research and Test Reactors
October 18-23, 1998
São Paulo, Brazil

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

DISCLAIMER

Abstract

Aluminum (Al) is a commonly used matrix for research reactor fuel plates. It has been found that a reaction between the fuel and the aluminum matrix may reduce or increase the irradiation stability of the fuel. To further understand the contribution of the reaction to the irradiation stability, experiments to develop a non-reacting matrix were performed. The work focused on magnesium (Mg), which is an excellent non-reacting matrix candidate and has a neutron absorption coefficient similar to Al. To avoid the formation of a liquid Al/Mg phase, improvements were made to the roll bonding process to achieve acceptable bonding at 415°C. After these methods were developed, fuel plates were produced with two fuels, uranium (U) -2 w/o molybdenum (Mo) and U-10-w/o Mo with two matrices, Al and Mg. A reaction between the magnesium and the 6061 Al cladding was discovered to take place during the processing at 415°C. To minimize the amount of reaction, methods were successfully developed to roll bond the fuel plates at 275°C. No reaction zone was observed in fuel plates processed at 275°C. Using this method, fuel plates with a Mg matrix are planned to 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 aluminum as the matrix allows processing at 500°C since the minimum liquid formation temperature is 640°C in the U/Al binary system[1] (Fig. 1). However, the U/Al system contains three compounds UAl_2, UAl_3 and UAl_4 which are thermodynamically very stable[2]. The reduction in energy level is a driving force for the formation of UAl_x type compounds in aluminum matrix fuel plates. It has been found that a reaction between the fuel and the aluminum matrix may reduce[3] or increase[4] the irradiation stability of the fuel. To further understand the contribution of the reaction to the irradiation stability, experiments to develop a non-reacting matrix were performed. Any reaction may be minimized or eliminated if a matrix material is developed which forms no U/X compounds. An excellent candidate which meets these requirements and has a neutron absorption coefficient similar to Al is Mg. The U/Mg system[1], which is presented in Figure 2, contains no compounds. However, the introduction of magnesium requires a review of the Mg/Al system[1] (fig. 3). In order to prevent possible formation of a liquid phase, fuel plates with a magnesium matrix must be rolled at <437°C. This posed a problem since the current roll bonding and blister testing processes require a temperature of 500°C.[5]

Improvements to the roll bonding process to achieve acceptable bonding at 415°C were the first goal of the study. After these methods were developed, two fuels, U-2 w/o Mo and U-10-w/o Mo were studied to determine if there is a reaction between these fuels 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 aluminum) were rolled at 415°C and were given an anneal for 1 hour at 415°C. After the development of an acceptable procedure, 5 x 15.2 cm (2 x 6 in.) fuel plates were fabricated to the specifications shown in Fig. 4. The fuels, matrix and cladding used in the plates is given in Table 1. Two fuels were studied; depleted uranium (DU) 2 w/o Mo powder purchased from Nuclear Metals, USA, and the DU 10 w/o powder received from the Korea Atomic Energy Research Institute (KAERI). Both fuels were spherical. After Processing, one Mg matrix plate was sectioned to determine the baseline amount of reaction. The remaining plates were the 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 (EDAX) for any reaction zones.

Table 1. List of Fuels Fabricated into Test Plates

Test Plate Number	Fuel	Loading Volume %	Matrix	Cladding*
1	U- 2 w/o Mo	25	Magnesium	6061 Al
2	U- 2 w/o Mo	25	Magnesium	6061 Al
3	U- 2 w/o Mo	25	Magnesium	6061 Al
4	U- 2 w/o Mo	25	Aluminum	6061 Al
5	U- 2 w/o Mo	25	Aluminum	6061 Al
6	U- 2 w/o Mo	25	Aluminum	6061 Al
7	U- 10 w/o Mo	25	Magnesium	6061 Al
8	U- 10 w/o Mo	25	Aluminum	6061 Al

* The 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 aluminum

Results and Discussion

Roll Bonding Tests at 415°C

After a test plate using the conventional Argonne National Laboratory method with the rolling temperature reduced from 500°C to 415°C showed non-bonds, the Indonesian National Atomic Energy Agency (BATAN) was contacted to examine their rolling schedule. It was received, reviewed and looked quite promising. It gives excellent bonding results after rolling at only 415°C which is sufficiently lower than 437°C to allow testing with magnesium. This is a confidential schedule and we were requested by BATAN to disclose only minimal information about it. For our experiments, it was modified slightly because some of the processing is at 500°C but this change was not expected to affect the bonding.

The first test assembly consisted of a 6061 aluminum frame with two empty cavities located near the trailing end and two 6061 aluminum cover plates. The assembly was cleaned and given a total of 82% reduction in thickness at 415°C. After hot rolling, it was given a one hour blister anneal at 415°C. Two small blisters were observed over the intentionally placed cavities in the trailing end of the fuel plate which indicated 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 was taken which confirmed that there was > 50% grain growth across the interface and is shown in Fig. 5. With the bonding confirmed, test plates with a Mg matrix were successfully roll bonded at 415°C. The assembly consisted of a 6061 aluminum frame with two compacts having 25 volume percentage tungsten powder surrogate fuel in a magnesium matrix and two 6061 aluminum 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 one hour blister anneal at 415°C and bend testing.

After proving the feasibility of the concept, a matrix (see Table 1) of 8 compatibility plates was processed by the method described above. Once again, metallurgical bonding was confirmed by the presence of blisters over the empty cavities and bend testing. Following rolling at 415°C and the 1 hr blister test, a U- 2 w/o Mo magnesium matrix plate was sectioned in the as-fabricated condition. It had been hoped that with the lower rolling temperature any reaction between the clad and the matrix would be minimal. However, as Fig. 6 shows, a significant reaction had taken place. EDAX analysis on a SEM showed the layer nearest the magnesium matrix to be $Al_{12}Mg_{17}$ and the layer nearest the clad to be Al_3Mg_2. Since compatibility was planned to be tested at 400°C, this reaction would invalidate the results because one of the assumptions made for calculating the volume changes is that any possible reaction is zero or statistically small enough to ignore due to 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, it was decided to continue the compatibility studies without volumetric measurements. After a sufficient amount of 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 400 hrs. at 400°C the U/Mg matrix plates were sectioned. In order to reduce the amount of time required to produce a reaction between the U-10 w/o Mo Al matrix plate, the temperature was increased and after 100 additional hours at 500°C this plate was sectioned. Results of the experiment have indicated that there is 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 type compound. U-10 Mo had a minimal amount of reaction. These results are in agreement with previous KAERI studies.[7]

Roll Bonding Tests at 275°C

In an effort to minimize the reaction between the cladding and the Mg, the rolling temperature was reduced to 275°C and significant changes were made to the BATAN 415°C rolling schedule. An assembly consisting of a 6061 aluminum frame and two cover plates was rolled which contained four compacts, two compacts having 25 volume percentage tungsten powder surrogate fuel in a magnesium matrix and two 100% Mg compacts. After blister testing at 275°C, bonding was confirmed by blister formation and bend testing. Fig. 7 shows a cross section of the surrogate fuel zone. No reaction is seen between the cladding and the matrix and was also confirmed by EDAX.

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

Summary and Conclusions

Fuel plates with a magnesium matrix were developed in an effort to minimize any reaction between the fuel and the matrix. It was found that processing at 415°C caused significant reaction between the 6061 Al cladding and the magnesium matrix. Methods were developed which achieved 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 275°C. It is planned that magnesium matrix fuel plates will be included in the next group of micro-plates to be irradiated.

References

1. T. B. Massalski, ed., Binary Phase Diagrams, 2nd Ed. (American Society for Metals, Metals park, OH, 1990)

2. P. Chiotti, et. al, The Chemical thermodynamics of Actinide Elements and Compounds, International Atomic Energy agency, Vienna, 1981

3. G. L. Hofman, J. Rest, J. L. Snelgrove, T. C. Wiencek, and S. Koster van Groos, Aluminum-U_3Si_2 Interdiffusion and It's Implications for the Performance of Highly Loaded Fuel Operating at Higher Temperatures and Fission Rates, Proceedings of 1996 RERTR International Conference, pp 94-110, Korean Atomic Energy Research Institute, 1996

4. G. L. Hofman, J. Rest, and J. L. Snelgrove, Irradiation Behavior of Uranium Oxide - Aluminum Dispersion Fuel, Proceedings of 1996 RERTR International Conference, pp 202-230, Korean Atomic Energy Research Institute, 1996

5. T. C. Wiencek, Summary Report on Fuel development and Miniplate Fabrication for the RERTR Program, 1978-1990, Argonne National Laboratory Report ANL/RERTR/TM-15 (1995)

6. T. C. Wiencek, R. F. Domagala, and H. R. Thresh, Thermal Compatibility Studies of Unirradiated Uranium Silicide Dispersed in ALuminum, Nuclear Technology, 71, 1985

7. Ki Hwan Kim, Don Bae Lee, Chang Kyu Kim, G. L. Hofman, and Kyung Wook Paik, Thermal Compatibility of Centrifugally Atomized U-Mo Powders with Aluminum in a Dispersion Fuel, pp 111-117, Nuclear Engineering and Design, 178, 1997

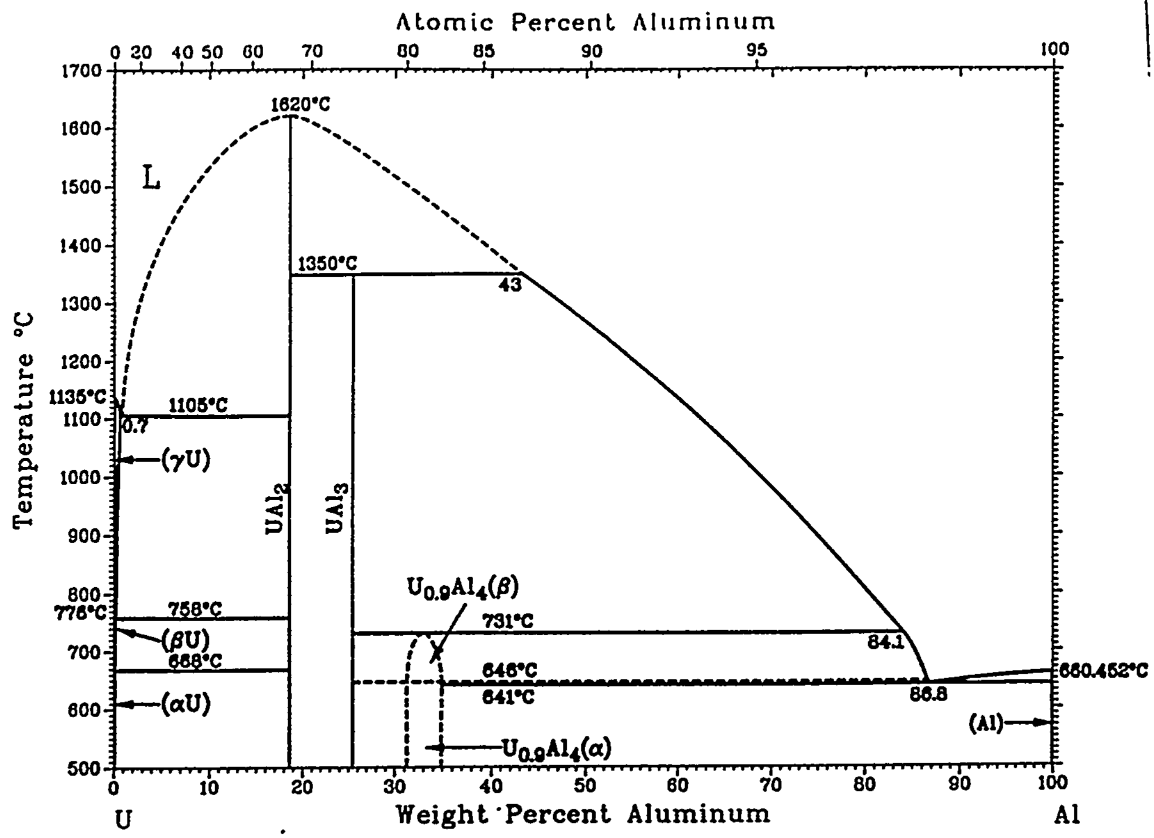

Fig 1. U-Al phase diagram

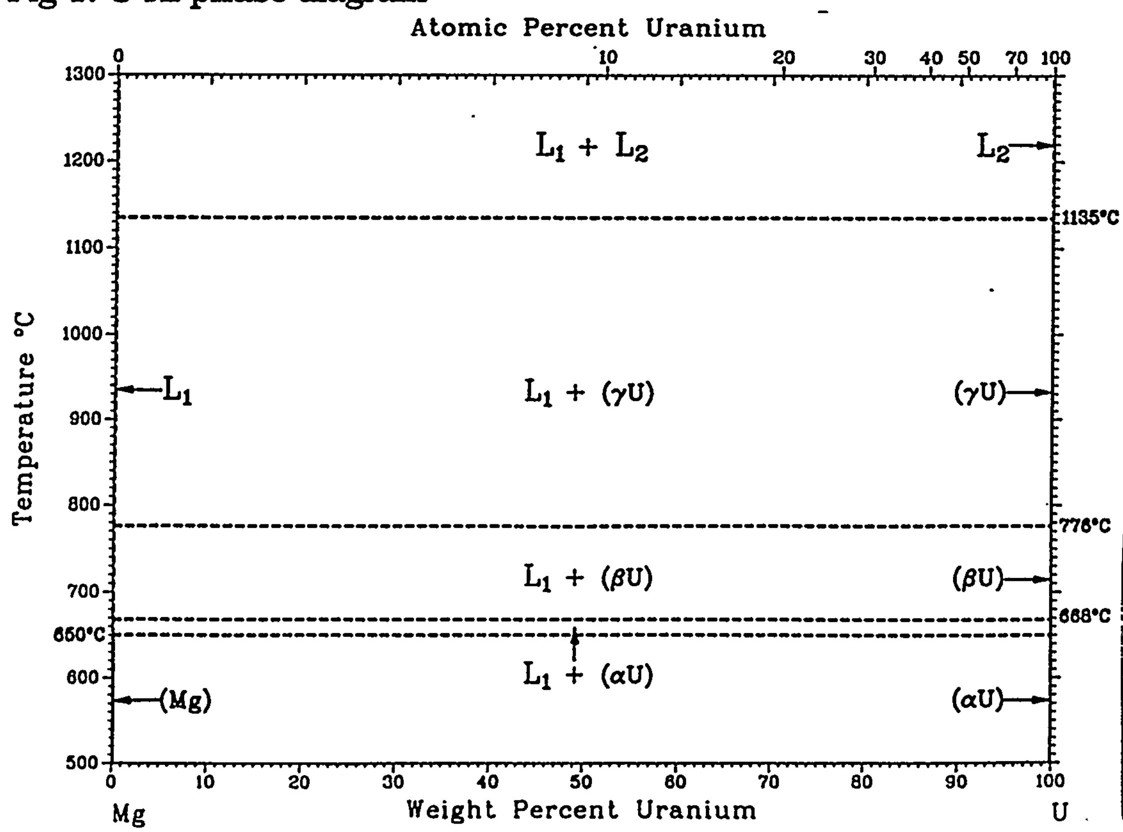

Fig 2. U-Mg phase diagram

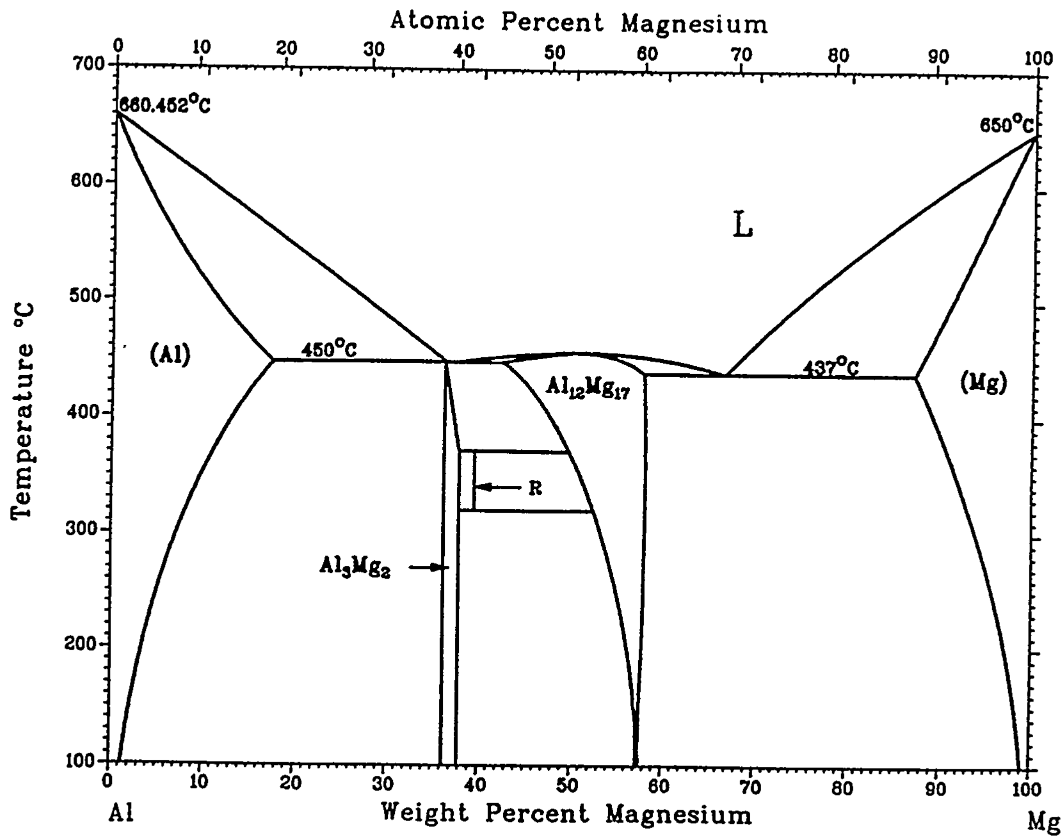

Fig 3. Al-Mg phase diagram

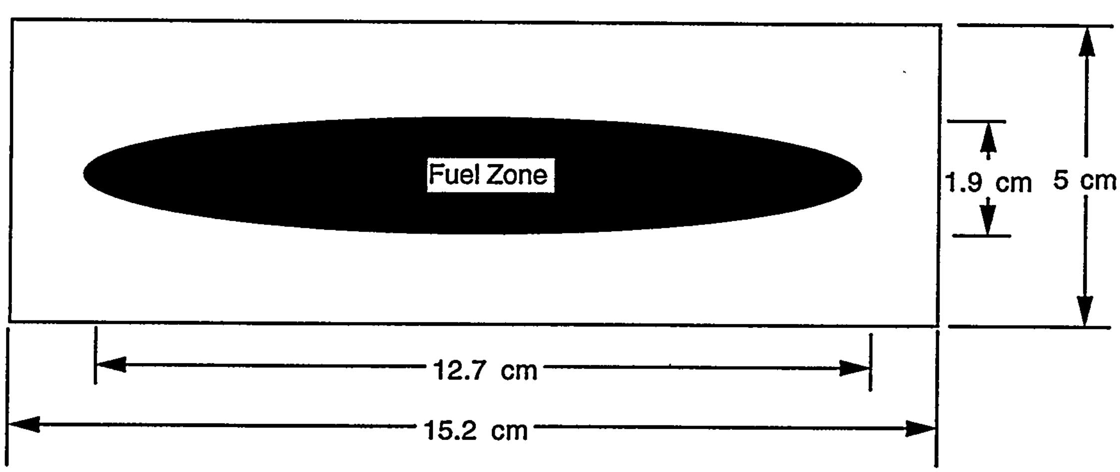

Fig 4. Design of fuel plate for bonding and compatibility study experiments

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

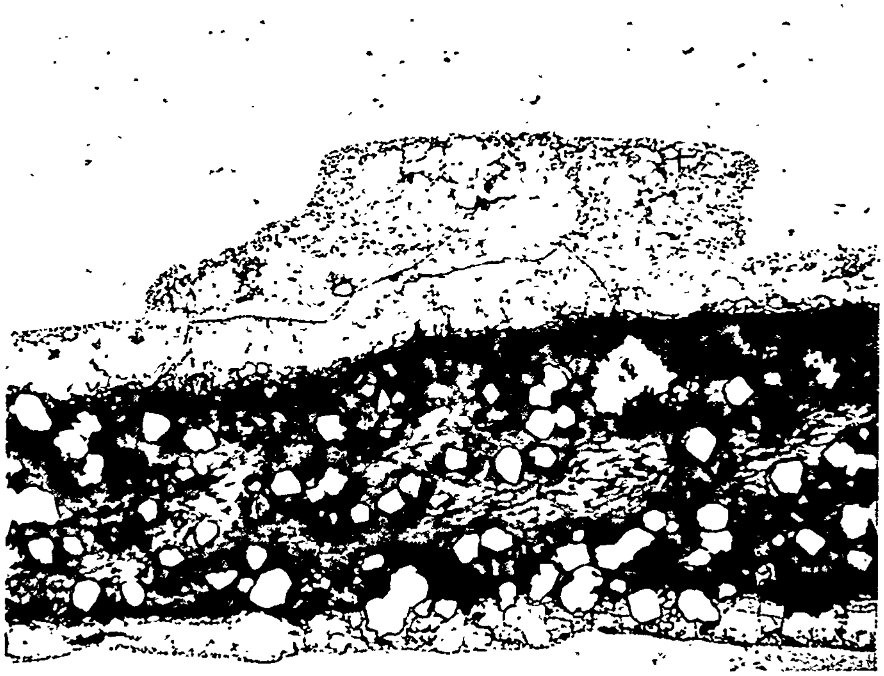

Fig 6. Diffusion Zones for 6061 aluminum clad magnesium matrix processed at 415°C; ≈ 200 X

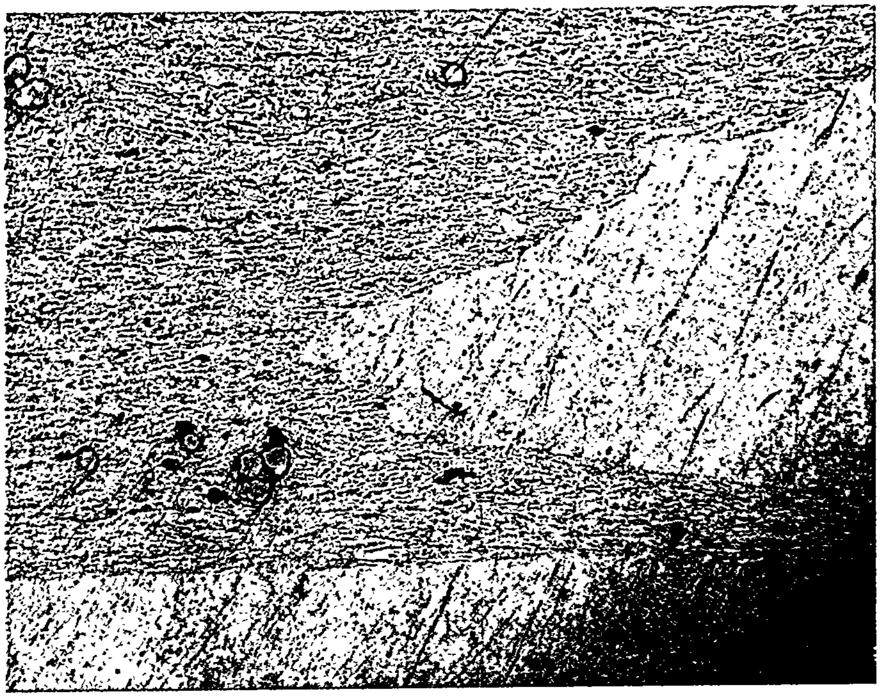

Fig 7. Interface between 6061 aluminum clad magnesium matrix processed at 275°C; ≈ 200 X