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# MIXING OF AL INTO URANIUM SILICIDES REACTOR FUELS

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## ABSTRACT

SEM observations have shown that irradiation induced interaction of the aluminum cladding with uranium silicide reactor fuels strongly affects both fission gas and fuel swelling behaviors during fuel burn-up. We have used ion beam mixing, by 1.5 MeV Kr, to study this phenomena. RBS and the  $^{27}\text{Al}(p, \gamma)^{28}\text{Si}$  resonance nuclear reaction were used to measure radiation induced mixing of Al into  $\text{U}_3\text{Si}$  and  $\text{U}_3\text{Si}_2$  after irradiation at  $300^\circ\text{C}$ .

Initially U mixes into the Al layer and Al mixes into the  $\text{U}_3\text{Si}$ . At a low doses, the Al layer is converted into  $\text{UAl}_4$  type compound while near the interface the phase  $\text{U}(\text{Al}_{0.93}\text{Si}_{1.07})_3$  grows. Under irradiation, Al diffuses out of the  $\text{UAl}_4$  surface layer, and the lower density ternary, which is stable under irradiation, is the final product. Al mixing into  $\text{U}_3\text{Si}_2$  is slower than in  $\text{U}_3\text{Si}$ , but after high dose irradiation the Al concentration extends much farther into the bulk. In both systems Al mixing and diffusion is controlled by phase formation and growth. The Al mixing rates into the two alloys are similar to that of Al into pure uranium where similar aluminide phases are formed.

## INTRODUCTION

High density reactor fuels are required for either high power density or low uranium enrichment applications. In such applications, particles of the fuel,  $\text{U}_3\text{Si}$  and  $\text{U}_3\text{Si}_2$ , and the cladding, Al, are mixed together and roll-bonded together between Al plates. During use of the fuel plates in reactor, the uranium fissions into several ionized fission fragments that produce tracks of damage that may penetrate the interface and result in irradiation induced mixing of the fuel particles and their Al cladding.

The mixing of fuel and cladding is important to the fuel plate swelling behavior [1,2]. Fuel burn-up results in fission products which are approximately 13% Xe and Kr. These inert elements are chemically insoluble in all materials including the fuels. The inert gases precipitate into bubbles that grow by absorbing vacancies and gas atoms. Within the interior of the fuel particles, the diffusion of fission gas to bubbles, becomes the dominant source of fuel swelling and very large bubbles are found on the interior of the fuel particles[3]. However, no visible fission gas bubbles are observed in the mixed layer at the fuel cladding interface. At high fuel particle density, fuel failure can occur when the Al is locally

consumed by mixing into a fuel particle. This motivates a study of the details of the irradiation induced mixing of Al into these materials.

A complicating factor is that  $U_3Si$  and  $U_3Si_2$  are amorphized by neutrons or 1.5 MeV Kr ions after an irradiation dose of 0.3 dpa at temperatures below 280°C [4,5,6]. Amorphous materials under irradiation undergo plastic flow. In this situation the amorphous  $U_3Si$  and  $U_3Si_2$  have little or no strength and catastrophic swelling will occur [1,2].

## EXPERIMENTAL

Uranium silicide specimens,  $U_3Si$  and  $U_3Si_2$ , were produced from an alloys of depleted uranium and silicon that were arc melted and heat treated at 1075 K for 72 hours. The  $U_3Si$  melt was made Si rich to avoid the presence of free uranium. This resulted in about 10% volume fraction of  $U_3Si_2$  as a second phase precipitate. The  $U_3Si_2$  material was single phase. The starting ingots were sliced into wafers that were then polished flat with 0.5 mm  $Al_2O_3$  grit. The polished wafers were mounted on copper blocks using Sauereisen. The surfaces were polished and coated at room temperature with 200 to 300 nm of Al by electron beam evaporation.

Specimens were irradiated at the 2 MV tandem National Electrostatics ion accelerator facility at Argonne National Laboratory. Irradiations were performed with 1.5 MeV Kr ions at 300°C up to doses of  $10^{17}/cm^2$  for the  $U_3Si_2$  specimens and  $6.0 \times 10^{16}/cm^2$  for the  $U_3Si$  specimens. The peak in the Kr stopping occurred at 250 nm in the specimens of both alloys with the damage extending to more than 500 nm. The average damage production in the two uranium silicides was 6.0 and 4.8 dpa/( $10^{15}$  ion/ $cm^2$ ) respectively and in the ternary 4.8 dpa/( $10^{15}$  ion/ $cm^2$ ).

Uranium distributions were determined by RBS, using standard curve fitting techniques, with 1.5 to 1.8 MeV He ions. Al concentration profiles were directly measured by resonance nuclear reaction of  $^{27}Al(p, \gamma)^{28}Si$  which occurs at a proton energy of 991.9 keV with a FWHM of 100 eV.

## RESULTS

RBS spectra taken from  $U_3Si$  and  $U_3Si_2$  specimens after 1.5 MeV Kr ion irradiations to different doses and at a temperature of 300°C are plotted in **Figure 1** and **Figure 2** respectively. Because of the large mass differences, these RBS spectra are only sensitive to back scattering from uranium atoms. No changes were detected during annealing at 300°C before irradiation. RBS spectra in **Figure 1** and **Figure 2** indicate that the mixing rate is higher in  $U_3Si$  than that in  $U_3Si_2$ . The depth dependence of the Al concentrations were independently determined by use of the

resonance nuclear reaction of  $^{27}\text{Al}(p, \gamma)^{28}\text{Si}$ . The gamma intensities are plotted in **Figure 3** for  $\text{U}_3\text{Si}$  and in **Figure 4** for  $\text{U}_3\text{Si}_2$ . After  $10^{16}$  Xe/cm<sup>2</sup> the phase  $\text{U}_{25}\text{Al}_{70}\text{Si}_5$  has formed in  $\text{U}_3\text{Si}$ . A similar composition is found in  $\text{U}_3\text{Si}_2$  but the mixed layer is not as uniform, and the inward diffusion of Al into  $\text{U}_3\text{Si}_2$  is much greater.

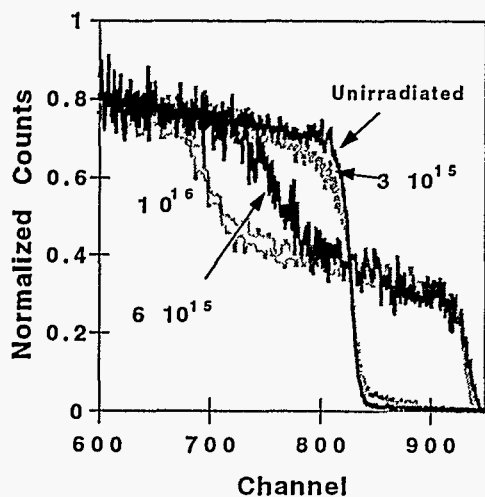


Figure 1 RBS spectra from Al/ $\text{U}_3\text{Si}$  irradiated by 1.5 MeV Kr ions at 300°C. Irradiation doses: 0.3, 0.6 and  $1.0 \times 10^{16}$  Kr/cm<sup>2</sup>

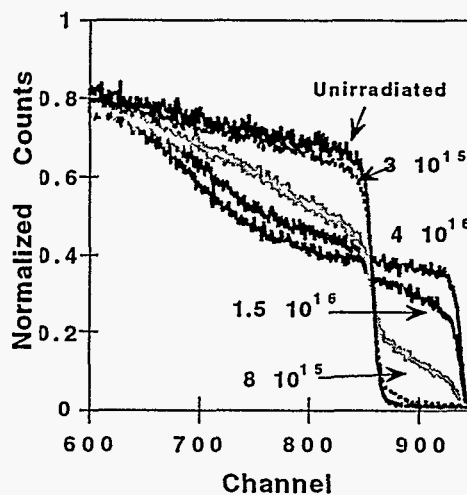


Figure 2 RBS spectra from Al/ $\text{U}_3\text{Si}_2$  irradiated by 1.5 MeV Kr ions at 300°C. Irradiation doses: 0.6, 1.0 and  $3.0 \times 10^{16}$  Kr/cm<sup>2</sup>

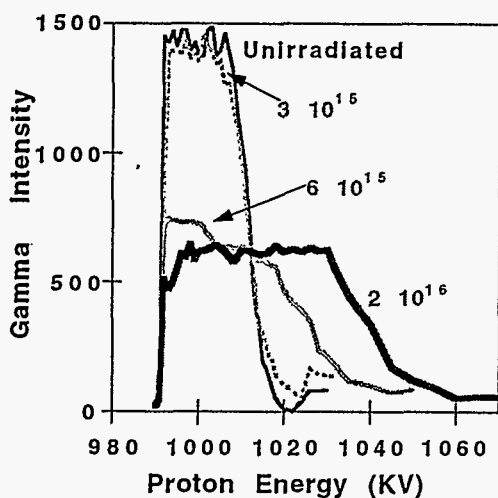


Figure 3  $^{27}\text{Al}(p, \gamma)$  intensities from Al/ $\text{U}_3\text{Si}$  after 1.5 MeV Kr irradiation at 300°C to doses of: 0.3, 0.6 and  $1.0 \times 10^{16}$  Kr/cm<sup>2</sup>

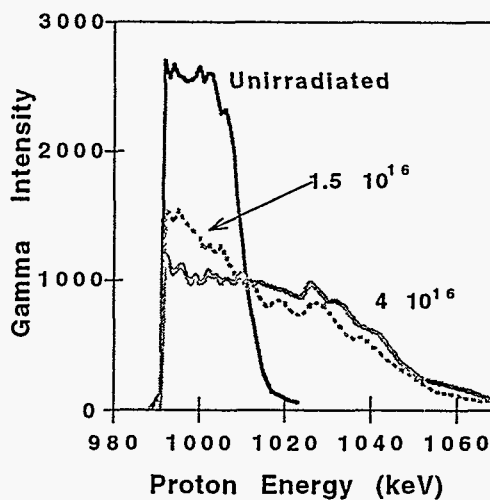


Figure 4  $^{27}\text{Al}(p, \gamma)$  intensities from Al/ $\text{U}_3\text{Si}_2$  after 1.5 MeV Kr irradiation at 300°C to doses of: 0.6, 1.0 and  $3.0 \times 10^{16}$  Kr/cm<sup>2</sup>

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Schematics of deconvolutions of the RBS and p-γ spectra for irradiated  $U_3Si$  is shown in Figure 5. Indications from SEM observation after in pile irradiation of these alloys suggested that Al is the diffusing species into both uranium alloys [1]. However after our lowest dose, the RBS measurements show that U atoms are found throughout the Al layer, and the p-γ measurements show that Al atoms are found in the  $U_3Si$  far from the initial interface. This indicates that both U and Al are mobile during irradiation at 300°C. As U atoms diffuse into the Al, a phase of the form  $UAl_4$  forms and grows at a rapid rate. As Al diffuses into the  $U_3Si$ , a phase of the type  $UAl_3$  forms. As the irradiation continues, Al diffuses out of the  $UAl_4$  phase and the  $UAl_3$  type phase extends to the surface.

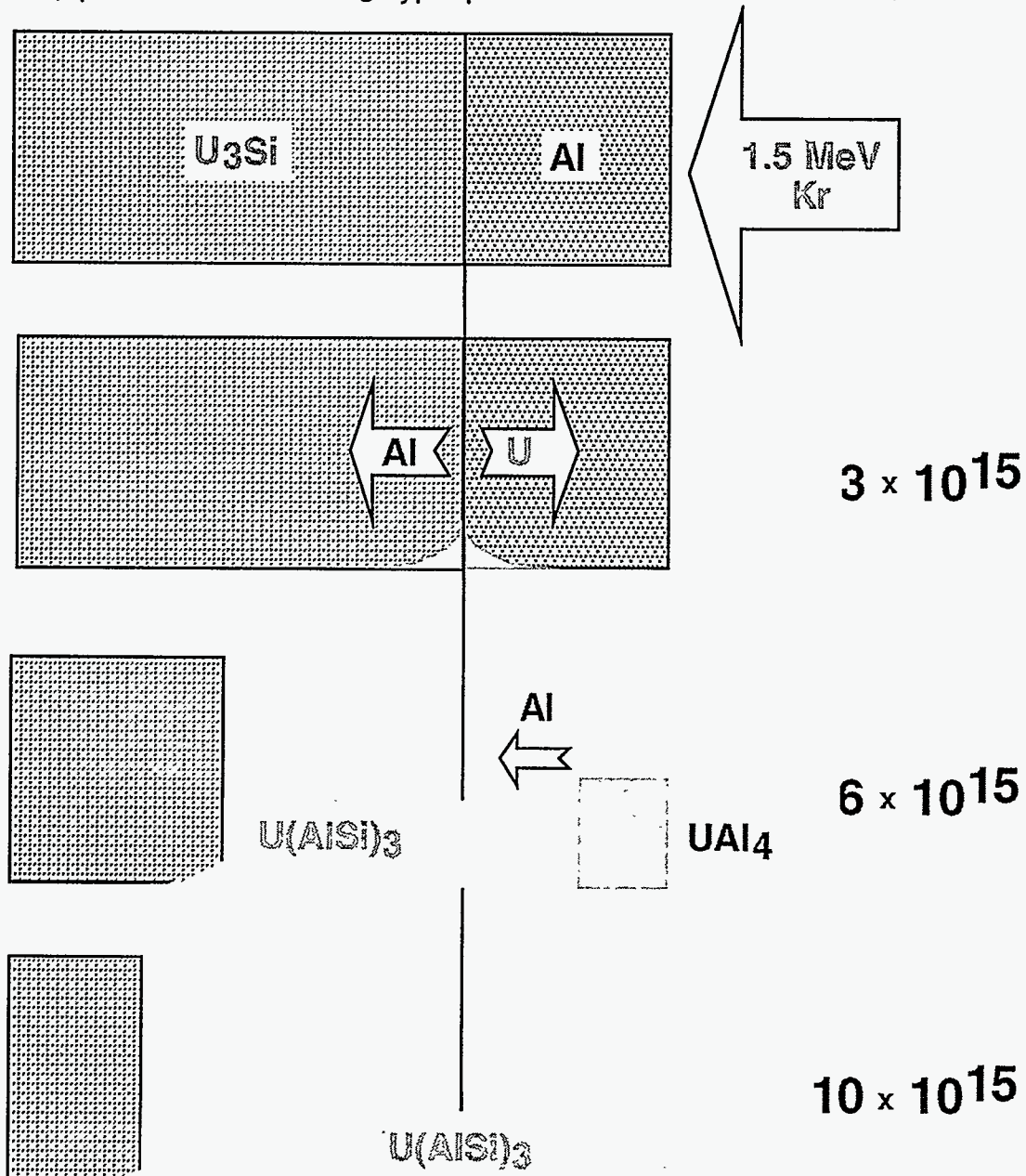


Figure 5 Schematic of Mixing of an Al layer on  $U_3Si$  at 300°C. Ion doses are indicated next to each section of the figure.

The growth rate of the stable  $U(Al_{.93}Si_{.07})_3$  phase is controlled by Al diffusion out of the  $UAl_4$  type phase and through the existing  $U(Al_{.93}Si_{.07})_3$  layer. The RBS and p- $\gamma$  measurements indicate the formation of a uniform mixed region with composition of  $U_{25}Al_{70}Si_{15}$ . This phase is found after melting  $U_3Si$  and Al [7]. Once this phase formed, its composition was unchanged by additional irradiation. Mixing of Al into  $U_3Si_2$  follows the same sequences.

Phase formation also controls the mixing of Al into pure U [8]. The mixing in this system was rapid, and during Xe irradiation the mixed region evolved from  $UAl_4$  to  $UAl_3$  to  $UAl_2$  as Al diffused out of the mixed layer. Mixing with Ar ions resulted in the formation of  $UAl_4$  without the observation of intermediate phases. At 450K, the phase layer grew at a rate of  $10^{-9} \text{ \AA}^2\text{cm}^2$  and, the mixing was too rapid to allow determination of diffusion coefficients. This rate is similar to that of Al mixing with  $U_3Si$ , and Al mixing into  $U_3Si$  is about twice as fast as into  $U_3Si_2$ .

### **SUMMARY**

The mixing between Al and uranium silicides is controlled by the diffusions of Al and U atoms leading to phase formation. U diffusion into the initial Al over layer results in formation of a  $UAl_4$  surface layer, while Al diffusion into  $U_3Si$  results in  $U_{25}Al_{70}Si_{15}$ . Al diffusion out of the  $UAl_4$  surface layer, through the growing  $U_{25}Al_{70}Si_{15}$  layer controls mixing at 300°C. After the  $UAl_4$  layer is consumed, the  $U_{25}Al_{70}Si_{15}$  mixed layer is stable during continued irradiation. The mixing rate of Al into  $U_3Si$  is about twice as fast as into  $U_3Si_2$  and is similar to that of Al mixing into U by Ar ions at same temperature.

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