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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 U_3Si and U_3Si_2 after irradiation at 300°C .

Initially U mixes into the Al layer and Al mixes into the U_3Si . At a low doses, the Al layer is converted into UAl_4 type compound while near the interface the phase $\text{U}(\text{Al}_{.93}\text{Si}_{.07})_3$ grows. Under irradiation, Al diffuses out of the UAl_4 surface layer, and the lower density ternary, which is stable under irradiation, is the final product. Al mixing into U_3Si_2 is slower than in U_3Si , 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, U_3Si and U_3Si_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 U_3Si and in **Figure 4** for U_3Si_2 . After 10^{16} Xe/cm² the phase $\text{U}_{25}\text{Al}_{70}\text{Si}_5$ has formed in U_3Si . A similar composition is found in U_3Si_2 but the mixed layer is not as uniform, and the inward diffusion of Al into U_3Si_2 is much greater.

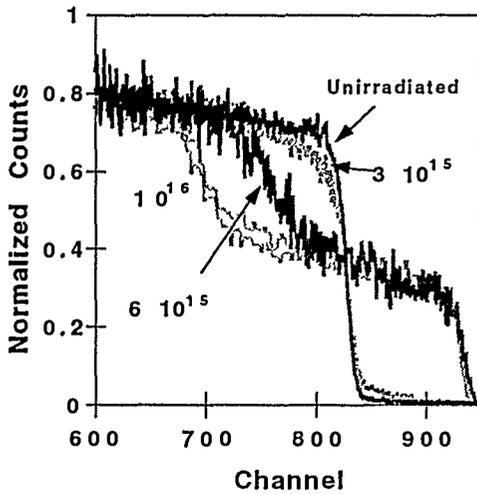


Figure 1 RBS spectra from Al/ U_3Si irradiated by 1.5 MeV Kr ions at 300°C. Irradiation doses: 0.3, 0.6 and 1.0×10^{16} Kr/cm²

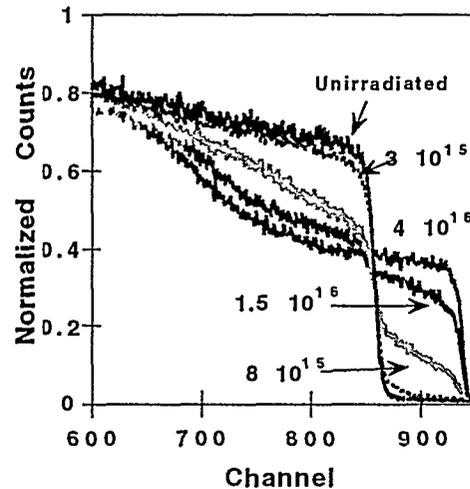


Figure 2 RBS spectra from Al/ U_3Si_2 irradiated by 1.5 MeV Kr ions at 300°C. Irradiation doses: 0.6, 1.0 and 3.0×10^{16} Kr/cm²

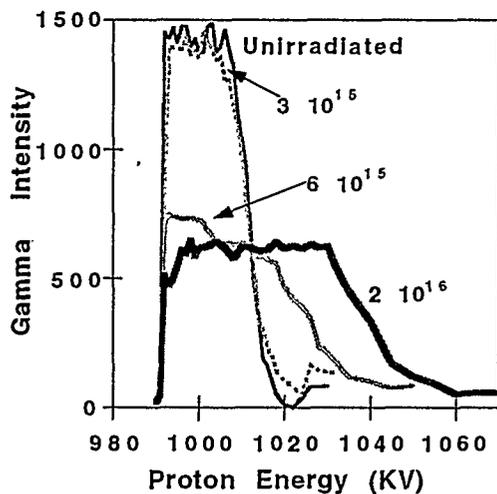


Figure 3 $^{27}\text{Al}(p, \gamma)$ intensities from Al/ U_3Si after 1.5 MeV Kr irradiation at 300°C to doses of: 0.3, 0.6 and 1.0×10^{16} Kr/cm²

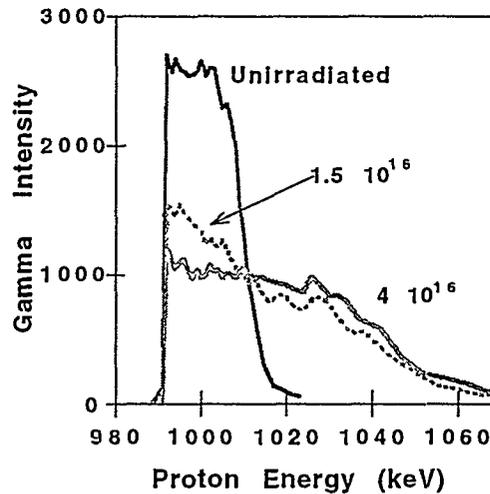


Figure 4 $^{27}\text{Al}(p, \gamma)$ intensities from Al/ U_3Si_2 after 1.5 MeV Kr irradiation at 300°C to doses of: 0.6, 1.0 and 3.0×10^{16} Kr/cm²

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Schematics of deconvolutions of the RBS and p-γ spectra for irradiated U₃Si 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 U₃Si 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 UAl₄ forms and grows at a rapid rate. As Al diffuses into the U₃Si, a phase of the type UAl₃ forms. As the irradiation continues, Al diffuses out of the UAl₄ phase and the UAl₃ type phase extends to the surface.

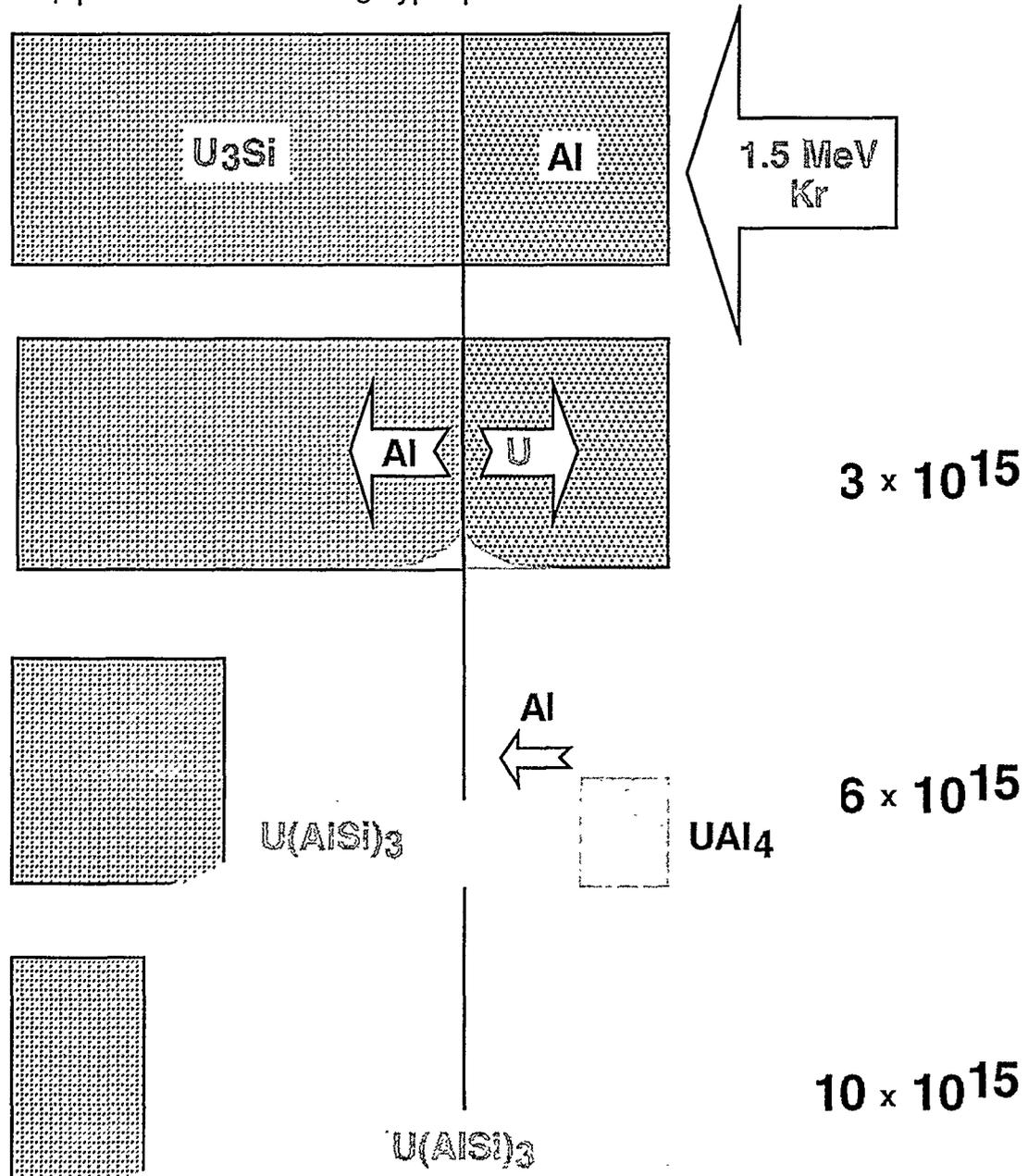


Figure 5 Schematic of Mixing of an Al layer on U₃Si at 300°C. Ion doses are indicated next to each section of the figure.

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 - γ measurements indicate the formation of a uniform mixed region with composition of $U_{25}Al_{70}Si_5$. 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 $U_{25}Al_{70}Si_5$. Al diffusion out of the UAl_4 surface layer, through the growing $U_{25}Al_{70}Si_5$ layer controls mixing at 300°C. After the UAl_4 layer is consumed, the $U_{25}Al_{70}Si_5$ 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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