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Response of aluminum and its alloys to exposure in the High Flux Isotope Reactor*

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Pure aluminum and some aluminum alloys were irradiated to very high neutron fluences in the cooling water at 328 K in the high flux region of HFIR. Displacement levels of 270 dpa and transmutation-produced silicon levels of 7.15 wt % were reached. Damage microstructures consisted of dislocations, cavities, and precipitates which caused substantial strengthening and associated loss in ductility. Formation of cavities and related swelling were considerably reduced by alloying elements and by the presence of fine Mg_2Si precipitate.

INTRODUCTION

1. The High Flux Isotope Reactor (HFIR) at Oak Ridge is a compact, light-water cooled and moderated, flux-trap reactor operated at 100 MWt. Its internal components are built mainly from aluminum alloys. This paper describes the response of aluminum surveillance specimens to radiation in HFIR.

MATERIALS AND EXPERIMENTAL CONDITIONS

2. The surveillance specimens were made from: (a) A very high purity aluminum of 99.9999% (6–9) grade, used as the reference material; (b) a less-pure (4–9) grade; (c) a commercial purity (99% or 2–9) aluminum, known as 1100 alloy, whose major impurities are Fe and Si; (d) a solid solution hardened alloy, 5052, whose major additives are 2.2 wt % Mg and 0.2 Cr; (e) a precipitation-hardened alloy, 6061, containing 1 wt % Mg and 0.6 Si. The first four of these materials were in fully annealed ("O") conditions. The 6061 alloy was solution-annealed, quenched and tempered to produce a finely-dispersed precipitate of Mg_2Si ("T6" condition). Test specimens were in the form of right cylinders of minimum dimensions 3 mm diam × 38 mm long for the 6-9 Al, and button-headed tensile specimens with gage sections of 3 mm diam × 25 mm length for the other materials.

and an effective displacement threshold energy of 25 eV, these fluences correspond to damage levels of 1.5×10^{-25} dpa per unit of fast neutron fluence, with a maximum level of ~270 dpa.

4. The major transmutation products from reactions of fast neutrons with aluminum atoms are hydrogen (up to 500 appm) and helium (up to 85 appm). With thermal neutrons, aluminum has a large cross section for production of silicon by (n,γ) reactions; as much as 6.9 atomic % (7.15 wt %) was created. Since silicon is essentially insoluble in aluminum, the transmutant silicon is incorporated as precipitate particles.

5. Following irradiation, the specimens were subjected to transmission electron microscopy (TEM) examination, density measurements and tensile testing.

RESULTS AND DISCUSSION

6. The irradiated specimens were coated with a hydrated oxide scale. However, there was no evidence of excessive corrosion. The specimens were descaled in a chemical polishing solution to prevent the scale from interfering with bulk density measurements.

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3. All specimens were irradiated in the target region of HFIR in contact with flowing coolant water at an ambient temperature of 328 K for periods up to 3½ y. The neutron spectrum at the central target region is primarily thermal with a peak flux of ~2.3×10^{19} n/m^2/s (2200 m/s), but with a substantial fast (>0.1 MeV) component of ~1.4×10^{19} n/m^2/s. There is very little variation in fuel cycle-averaged fluxes. The duty cycle is high, ~91%. Surveillance specimens were exposed to a range of fluences reaching as high as 1.8×10^{27} fast n/m^2. Using the conversion data of Gabriel et al. (ref. 1)

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TEM observations

7. The damage microstructure consisted of dislocations, cavities and precipitates of silicon, as shown in Fig. 1. Dislocations and cavities were the most obvious features in the purer materials, and were evident even at the lowest fluences; they grew with increasing fluence. Each cavity was associated with a silicon precipitate, but there were more precipitates than cavities. In the less-pure, commercial grade 1100-0 Al there were less cavities and they tended to be segregated near inclusions and alongside grain boundaries (Fig. 1b). At the higher fluences, the cavities were coated with silicon (ref. 2). In the 6061-T6 and 5052-0 alloys the occurrence of cavities was significantly curtailed, the

purer aluminums but is displaced to higher fluences by an order of magnitude. The swelling in these three materials contains negligible contribution from the silicon precipitate; swelling is derived primarily from cavities. The reduced rate of swelling above 1% swelling may be caused by a decrease in cavity growth rate as the cavities assume increasing importance as point defect sinks and recombination sites (ref. 5). The shift of swelling to higher fluences in the 1100-O alloy is attributed to dissolved impurity elements which trap point defects and promote more recombination in the matrix. This reduces the nucleation rate of cavities and extends the incubation period for measurable swelling.

10. Similar considerations apply to the more highly alloyed 6061-T6 and 5052-O materials. These two alloys also have an additional factor that further depresses swelling — they contain high concentrations of fine Mg_2Si precipitates that foster point defect recombination. Consequently, cavity formation is relatively difficult in these alloys, and much of their measured swelling at high fluences is caused by the silicon precipitate. Their swelling rate exponents, however, are about 1.4 to 1.5, reflecting the influence of cavities. The swelling curve for the 5052-O alloy falls below the dashed line for the contribution from silicon because in this alloy the silicon is precipitated as Mg_2Si from a solid solution of Mg in Al. Although Mg_2Si is less dense than elemental silicon, its formation entails withdrawal of magnesium from solid solution with a consequent increase in the density of the matrix. The net effect is that the density changes in the 5052 alloy due to radiation-induced precipitation are smaller than in the other materials.

Tensile properties

11. Tensile tests at moderately slow strain rate were made at 323 and 423 K, with the results shown in Figs. 3 and 4. Data points surfaces in this alloy displayed facetting akin to intergranular failure.

12. The pronounced irradiation strengthening displayed by these alloys is compatible with hardening by the radiation-induced dislocations, cavities, and precipitates, as described quantitatively for 1100 alloy (ref. 8). The formation of fine Mg_2Si precipitate during irradiation gives additional strength to the 5052-O alloy. The overall losses in ductility are neither precipitous nor intolerable. They result largely from reduction of plastic deformation by the hardening. At the highest fluence at 323 K, and at the higher test temperature there was further ductility loss seemingly associated with intergranular failure. The latter may be exacerbated by helium or, in the case of the 1100-O alloy, by tearing along the near-grain boundary cavities.

CONCLUSIONS

13. Aluminum and several of its alloys exposed to very high neutron fluences in the cooling water of HFIR at 328 K display some commendable radiation responses:

- Large quantities of insoluble, transmutation-produced silicon are accommodated.
- Corrosion resistance remains satisfactory.
- Resistance to cavity formation and related swelling increases markedly with alloy content and is especially high in alloys containing fine Mg_2Si precipitate.
- The dislocations, cavities and precipitates created during irradiation cause considerable increases in tensile strength and associated losses in ductility.
- There is no overt tendency for brittle failure at the reactor coolant temperature; even at the highest fluences there is significant ductility. However, a temperature excursion to 423 K enters a region of low uniform ductility where overstressing must be avoided.

are omitted for clarity; greater details on most of the tests are available elsewhere (refs. 4,6,7). At both temperatures, flow stresses and tensile strengths were progressively and considerably, increased by irradiation. The rate of strengthening was decidedly greatest in the 5052-O alloy. As strengths increased, ductilities were decreased to some plateaux values. Total elongations at 323 K were reduced to 8 to 15% at fluences $>10^{26}$ n/m^2, but the fracture modes remained transgranular with considerable necking; at the highest fluences the 1100-O alloy exhibited a further drop in ductility concurrent with an apparent intergranular fracture. Uniform elongations were reduced to 3–5% for the 4–9 Al and 1100-O alloy at fluences $>2 \times 10^{25}$ n/m^2, and to about 4% for the 5052-O and 6061-T6 alloys at fluences $>10^{26}$ n/m^2. At the higher test temperature of 423 K the total elongations in the irradiated materials were reduced to <5% despite the high ductilities in the unirradiated conditions at this temperature. Uniform elongations in the 5052-O alloy were reduced to <1%; at the very highest fluence the fracture

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predominant features being dislocations and precipitates. In the 6061-T6 alloy (Fig. 1c), the original Mg_2Si precipitates were retained and were supplemented by a precipitate of silicon. In the 5052-0 alloy (Fig. 1d) a precipitate of silicon did not form at the lower fluences but, instead, the radiation-induced silicon reacted with dissolved magnesium and formed a finely-divided precipitate of Mg_2Si (refs. 3,4), at least up to a fast fluence of 3×10^{26} n/m^2 at which the dissolved magnesium should be fully depleted. Presumably excess silicon precipitated at higher fluences, but it was not discernible. In this 5052-0 alloy there were very few cavities (Fig. 1d).

Density changes

8. Densities were measured by Archimedes' displacement, and were lower for irradiated specimens than for unirradiated controls. This swelling, expressed as change in density, $(\rho_o - \rho_i)/\rho_i$ %, where the subscripts o and i denote the unirradiated and irradiated conditions, is plotted as a function of fast neutron fluence in Fig. 2. These data include swelling from the silicon precipitate which is less dense than aluminum. This contribution is indicated by the dashed line in Fig. 2.

9. Two effects are immediately obvious from Fig. 2: The alloys have considerably greater resistance to swelling than do the purer aluminums, and the rate of swelling is decreased above about 1% swelling. Considering the latter first, we see that the purest aluminum shows measurable swelling at quite low fluences corresponding to doses of only 0.1 dpa. Over the fluence range up to about 2×10^{25} n/m^2, at which about 1% swelling is reached, the swelling rate can be described by an expression of the form $S \alpha (\phi t)^n$, where S is the degree of swelling, ϕt is the fast fluence, and the exponent n has a value of about 1.25. When swelling exceeds about 1%, n is decreased to about 0.9. The curve for the commercial purity (2-9) 1100-0 alloy follows a similar form to the

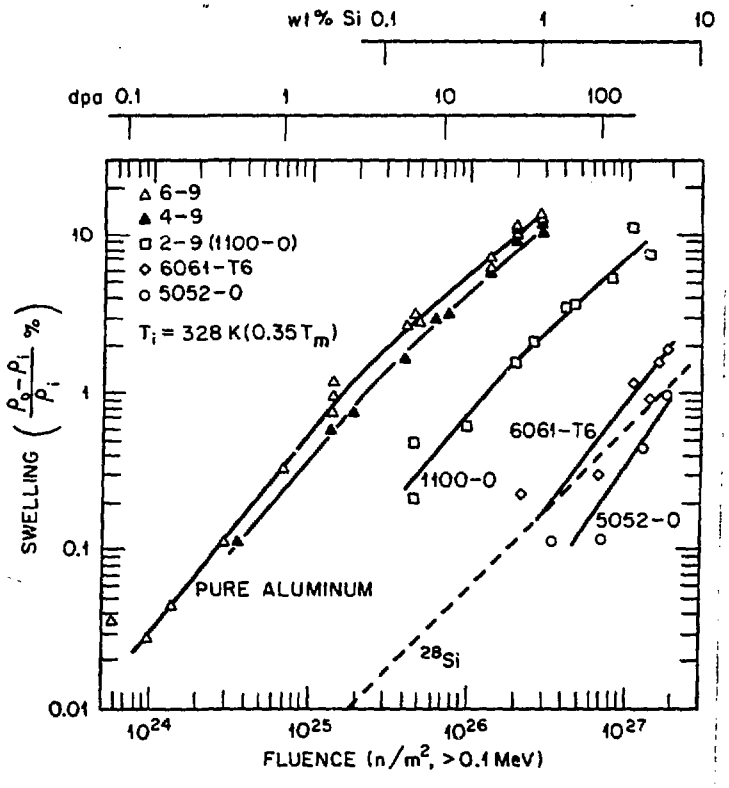

Fig. 2 Swelling versus fast fluence, as measured from decreases in densities.

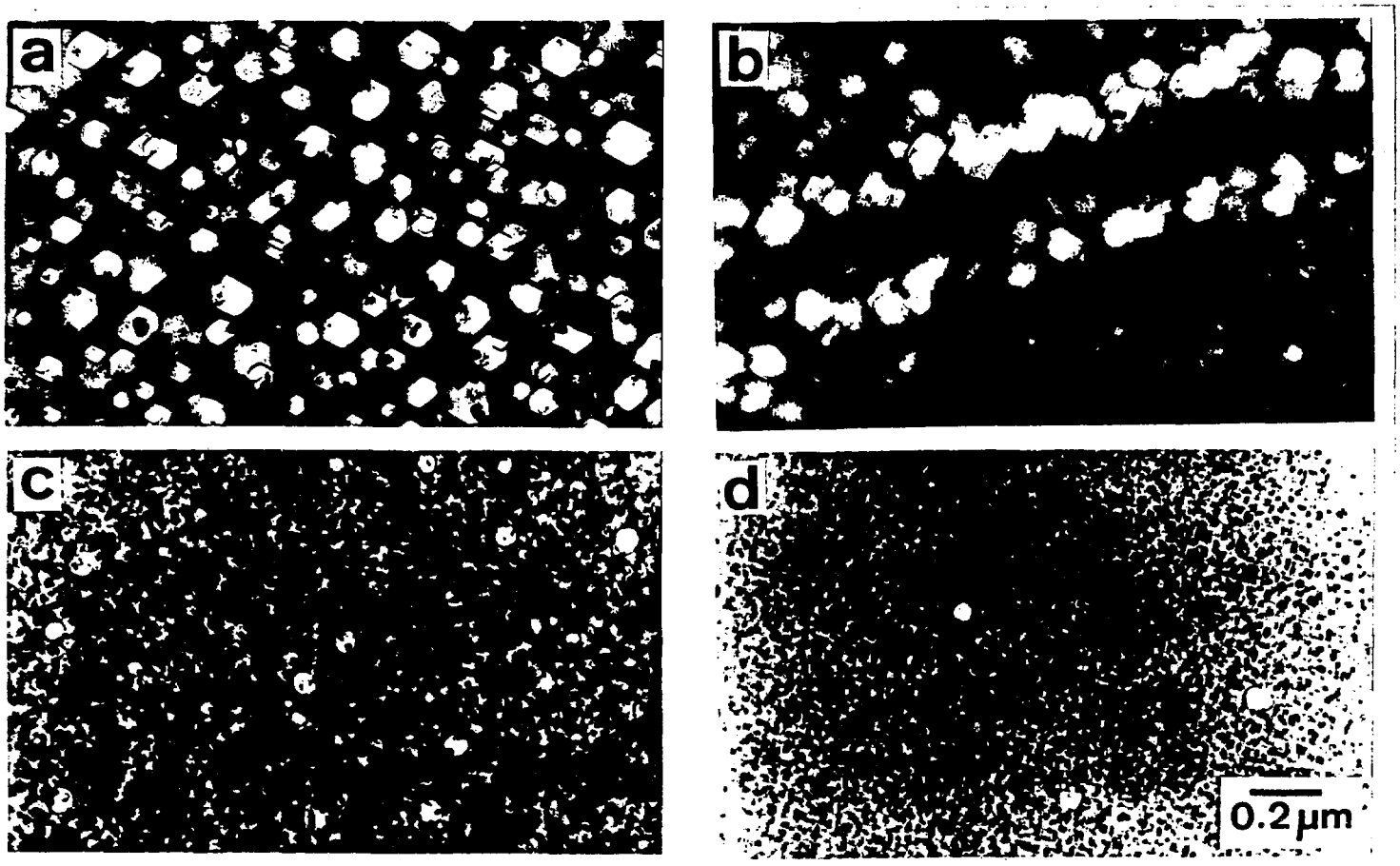

Fig. 1 Damage microstructures showing cavities and precipitates. (Dislocations are out of contrast). (a) 6-9 Al, ϕt(fast) = 2.9×10^{26} n/m^2, (b) 1100-0 Al, 2.8×10^{26} n/m^2, (c) 6061-T6 Al, 1.8×10^{27} n/m^2, (d) 5052-0 Al, 1.8×10^{27} n/m^2.

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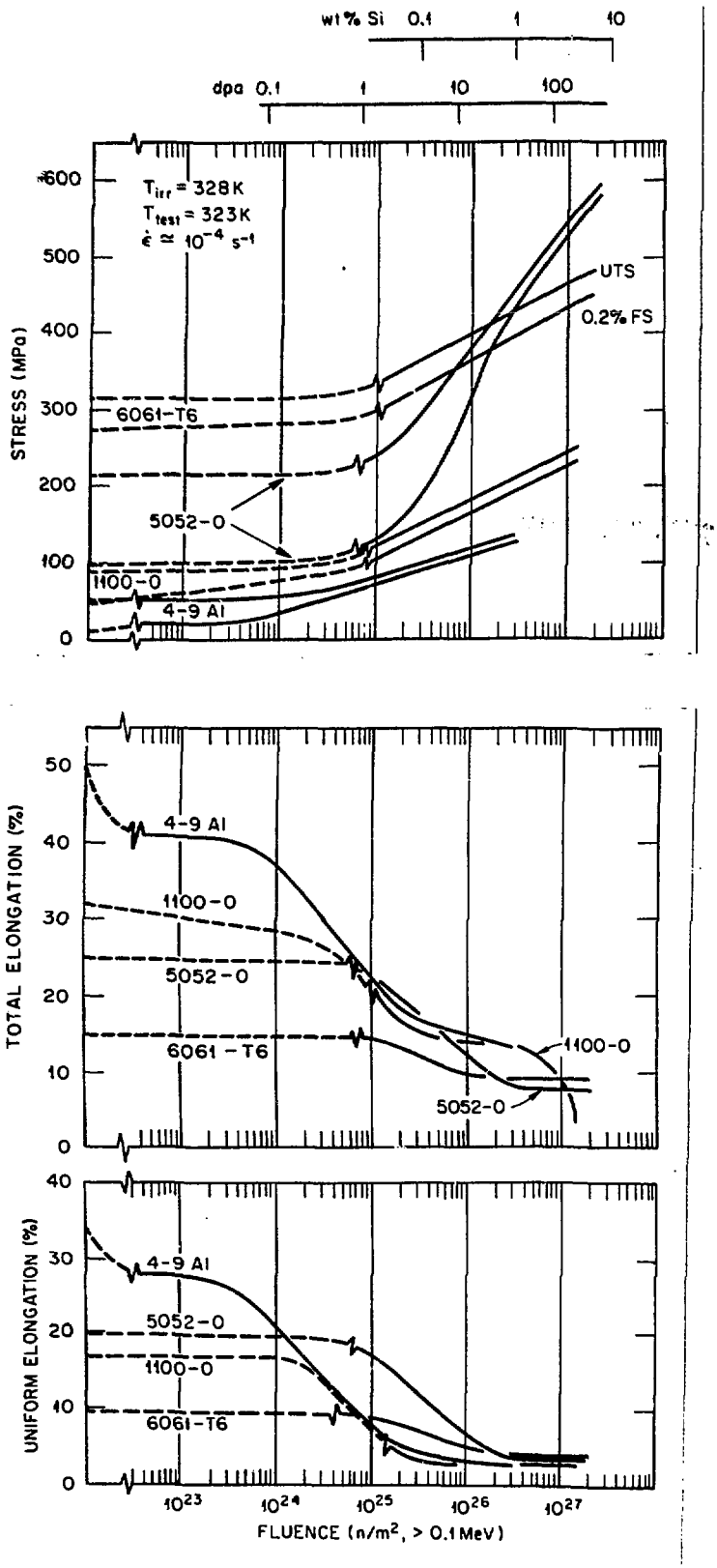

Fig. 3 Tensile properties at 323 K.

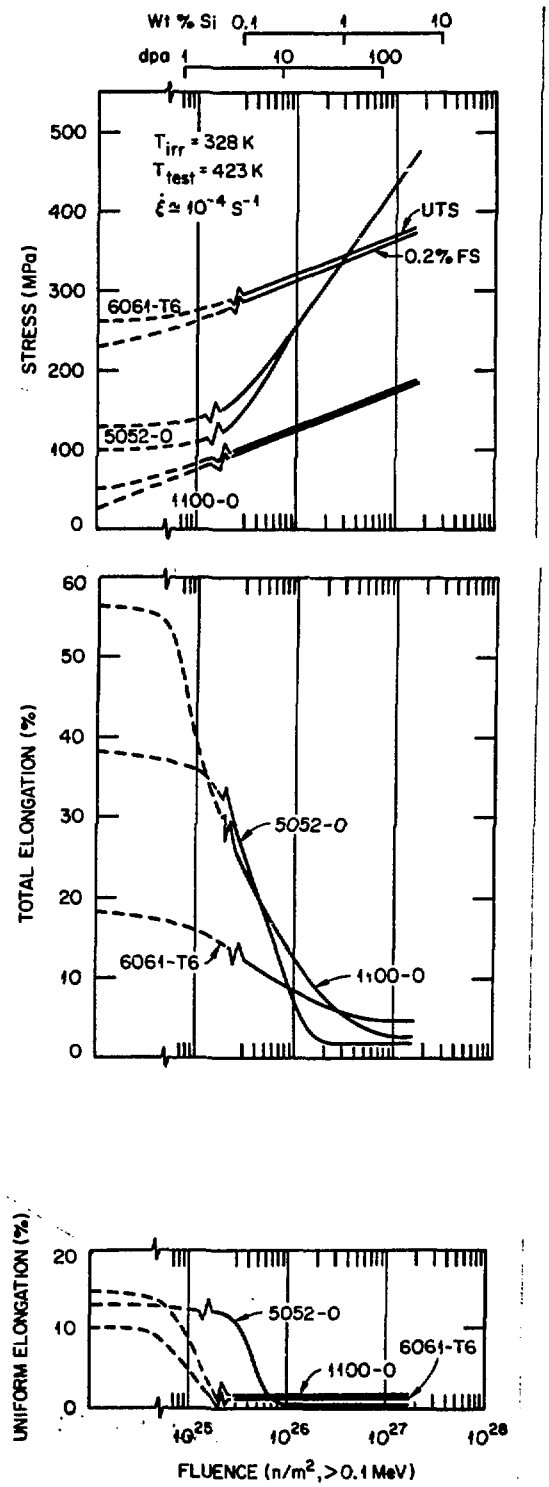

Fig. 4 Tensile properties at 423 K.