

A SPECTRAL EFFECT ON PHASE EVOLUTION IN NEUTRON-IRRADIATED ALUMINUM?*

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ABSTRACT

It is proposed that the spatial distribution of transmutation-produced silicon precipitates formed in aluminum by capture of thermal neutrons is systematically modified by the fast neutron portion of the spectrum, and the tensile strength is altered accordingly. The concept is used to rationalize some previously erratic tensile data.

THE THESIS

Many water-cooled research reactors contain aluminum alloy components. The aluminum atoms capture thermal neutrons and are transformed to silicon by the reactions $^{27}\text{Al}(n,\gamma)^{28}\text{Al} \rightarrow ^{28}\text{Si} + \beta^-$. The cross section has a value of 230mb for 2200m/s ($E=0.025\text{eV}$) neutrons. These reactors operate at water temperatures between 45° and 100°C, which correspond to homologous temperatures of 0.34 to 0.4 T_m for aluminum, at which vacancies are mobile. The solubility of silicon is negligible and it forms precipitate particles¹. In high flux reactors it is not uncommon to find reactor components with neutron-generated silicon concentrations of 5 wt% or more. At such levels the silicon precipitate will dominate the radiation-induced increases in strength. The strength will depend not only on the volume fraction of silicon but also on its spatial distribution. A finely-divided phase will cause more strengthening than the same volume fraction in a coarse distribution.

This raises the possibility of a spectral effect in which the thermal fluence will control the volume fraction of silicon phase, and some other portion of the spectrum may determine the size and concentration of the precipitate particles. It is well-known that excess point defects produced by atomic displacements can raise diffusion rates and cause considerable phase instability and redistribution of solutes^{2,3}, which can result in accelerated Ostwald ripening of a precipitate phase⁴. In aluminum, some point defects are created by atomic recoils from the (n, γ) reactions but the cross section is small, $\approx 3\text{b}$, compared with a value of $\approx 1200\text{b}$ for displacements by fast neutrons with energies $> 0.1\text{Mev}$. Fast neutrons may make an additional contribution to precipitate ripening via cascade dissolution, which will release silicon atoms into the point defect streams and may be more destructive of smaller particles than larger ones. It would not be unreasonable, then, to expect that the microstructure and corresponding tensile strength of irradiated aluminum should be sensitive to the neutron spectrum, as expressed through the ratio of thermal-to-fast flux, Φ_{th}/Φ_f . A soft spectrum, i.e. a high ratio of Φ_{th}/Φ_f , will induce less displacements per unit silicon atom and should retard the ripening process, resulting in a finer distribution of the silicon phase and more strengthening; and vice versa.

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THE EVIDENCE

The evidence for the spectral effect stems from the tensile strength data of Weeks et al.^{5,6} for 6061-T6 aluminum alloy. This alloy has a nominal composition of Al-1.0Mg-0.6Si and in its T6 condition is quenched and tempered to produce a microstructure of fine, needle-like Mg₂Si precipitates. Weeks' test specimens were cut from reactor components irradiated for long periods in the heavy-water-cooled High Flux Beam Reactor (HFBR) at Brookhaven National Laboratory. The reactor coolant inlet and outlet temperatures were 55° and 65°C, respectively. The data are displayed in Fig. 1, together with a set of data obtained^{7,8} for exposures in the light-water-cooled High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory, with inlet and outlet temperatures of 44°C and 67°C. The data are plotted against thermal fluence in the upper part of the figure and against fast fluence in the lower part. In both parts, the HFIR data are a smoothly increasing function of fluence, whereas the HFBR data show considerable scatter.

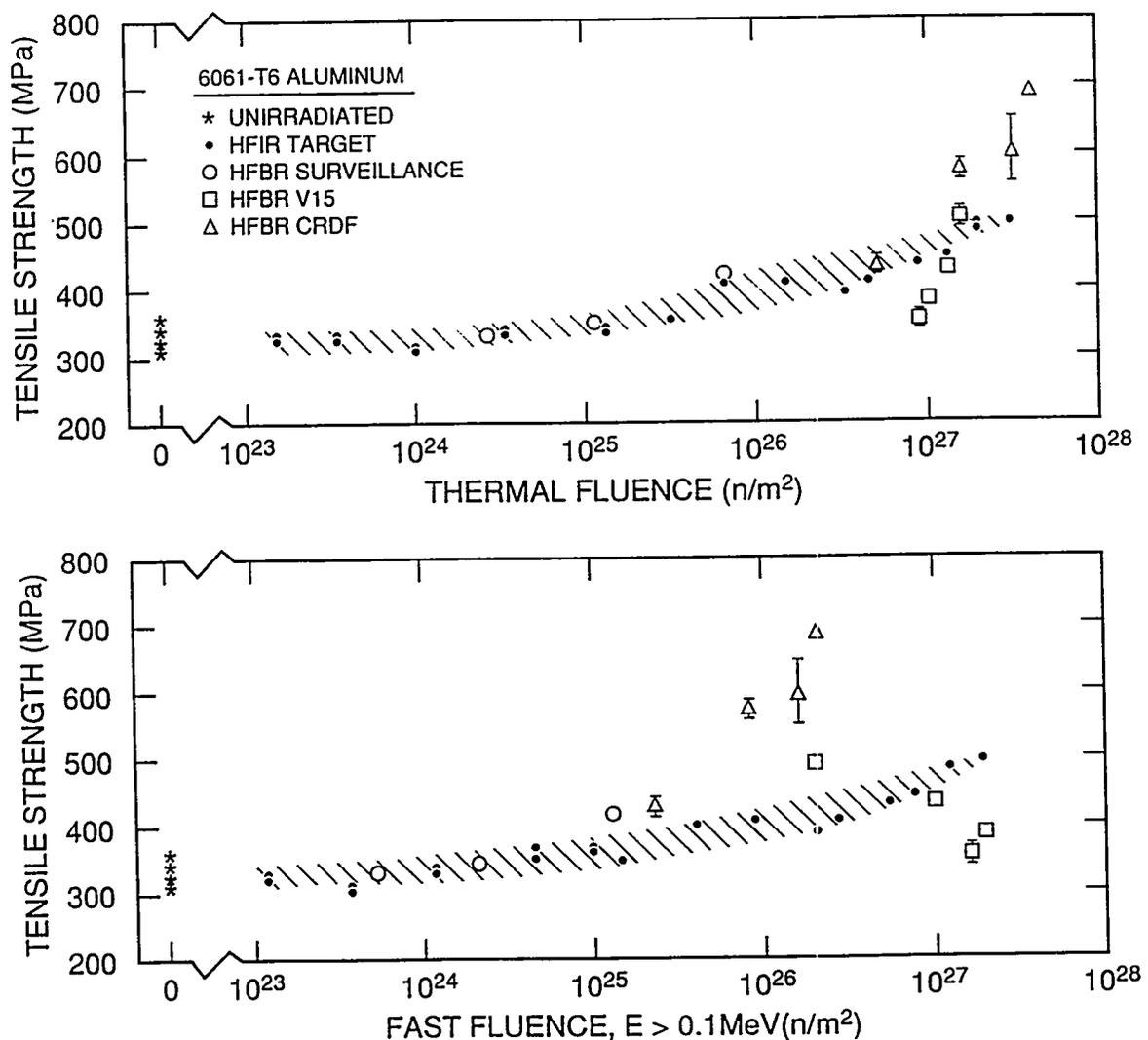


Figure 1. Fluence dependencies of tensile strengths of 6061-T6 aluminum alloy irradiated in the HFBR^{5,6} and the HFIR^{7,8}.

The picture changes when the data in Fig. 1 are assessed in terms of their Φ_{th}/Φ_f ratios. The HFIR test specimens were all irradiated in the flux trap where the fluxes are high and the spectrum is fairly hard, with a Φ_{th}/Φ_f ratio in a very narrow range of 1.7 to 2.5. Hence, from the spectral effects perspective, they should yield smooth plots against both thermal fluence and fast fluence. The HFBR materials were exposed at different locations in the reactor, some inside the core, others away from the core in the softer spectra of the heavy-water reflector. Their Φ_{th}/Φ_f ratios vary from 0.5 to 20. We should not attempt to represent these data by a single line. Rather, each datum should belong to one of a family of curves each depicting a narrow range of Φ_{th}/Φ_f ratio and each following a fluence response of similar form to the HFIR specimens, as shown in Fig. 2. Here, the HFBR curves straddle the HFIR curves and share a pattern of smoothly-varying behavior with both fast and thermal fluences. The curves merge to the unirradiated strength level at thermal fluences below $1 \times 10^{25} \text{ n/m}^2$ and fast fluences below $1 \times 10^{24} \text{ n/m}^2$, where the transmutation-produced silicon levels are less than about 0.02%.

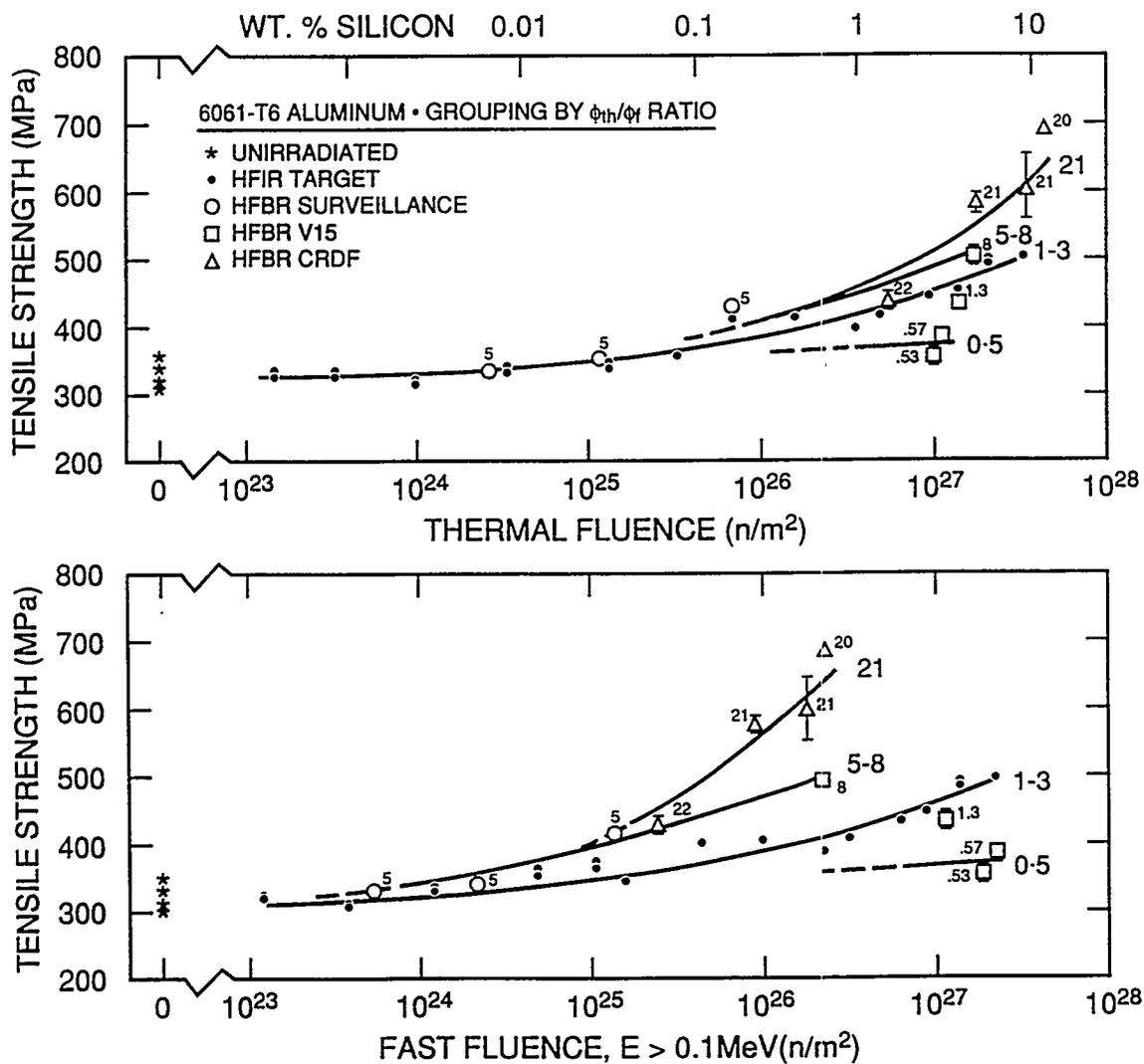


Figure 2. Data from Fig. 1 assessed by Φ_{th}/Φ_f ratio.

At a common thermal neutron fluence, i.e. at equal levels of transmutation-produced silicon, those HFBR specimens with larger Φ_{th}/Φ_f ratios than the HFIR specimens are stronger than the HFIR specimens, and those with smaller ratios are weaker. In this scenario, a HFBR material with a larger Φ_{th}/Φ_f ratio than the HFIR specimens should have a finer distribution of silicon precipitates than the corresponding HFIR specimen.

Comparison of microstructural data for the highest fluence HFIR specimens and the high fluence, high Φ_{th}/Φ_f ratio HFBR specimens at silicon levels of 7 to 9% agree with this interpretation. Details of the microstructural examinations are described elsewhere^{8,9}. The most essential observation for the present purposes is seen in Fig.3, where it is clear that the dark, spherical objects, which are silicon-rich particles, are much smaller and more numerous in the HFBR specimen. Other microstructural changes involved the nearly-complete re-resolution of the original, fine, acicular precipitate of Mg_2Si phase in both cases. Flakes of a silicon-rich phase were formed on grain boundaries, and there seemed to be more of them in the HFIR material. Zones on each side of grain boundaries were denuded of the silicon-rich phase, the zones being wider for the HFIR materials. Voids were present in the HFIR material and were coated with silicon; voids were absent in the HFBR materials. The silicon-rich precipitates were amorphous. Intermetallic inclusions, which were originally crystalline, became amorphous during irradiation.

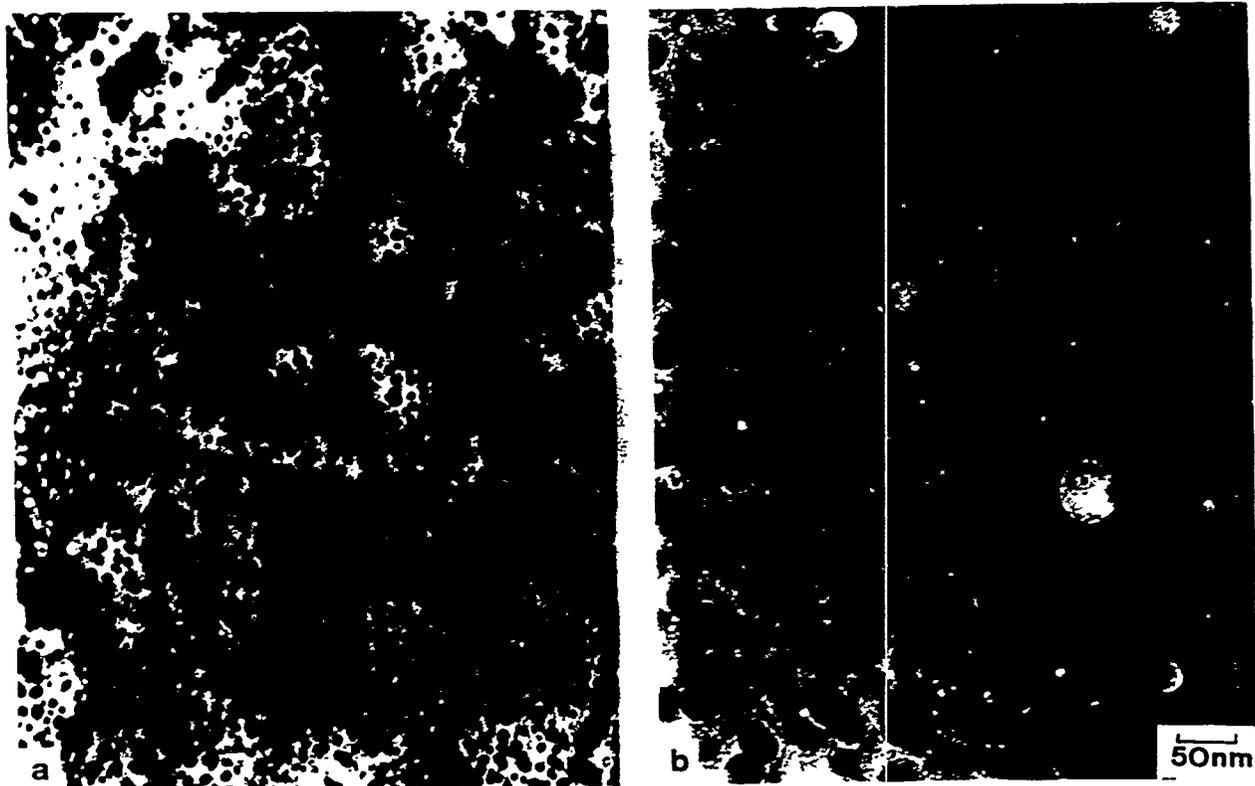


Figure 3. Microstructures of 6061-T6 aluminum alloy after high fluence irradiation in (left) the HFBR at $\Phi_{th}/\Phi_f = 20$, and (right) the HFIR at $\Phi_{th}/\Phi_f = 1.7$.

DISCUSSION

Prima facie, the above evidence seems to uphold the thesis of a spectrum effect. The higher tensile strengths and the finer spatial distribution of the silicon precipitates in the HFBR materials obey the predictions for the effects of higher Φ_{th}/Φ_f ratio. While this is encouraging, it is not proof of a spectral effect. Other explanations can be envisioned. The most obvious one is that the differences in tensile strengths and microstructural features described above might be symptomatic of a lower irradiation temperature for the HFBR materials. This explanation would be compatible with nuclear heating loads, which were less than 8 W/g in the HFBR but were about 35 W/g in the HFIR trap. Unfortunately, irradiation temperatures were not measured, and the issue can not be resolved with only the present data. It is noted, however, that the HFIR experiments were engineered to specifically accommodate the high heat load by ensuring sufficient flow of cooling water over the specimens, and there were no other signs that the goal temperature was not attained.

Temperature aside, the Φ_{th}/Φ_f ratio is not the only variable in the HFIR and HFBR exposure conditions. There are differences in exposure times, fast and thermal fluxes, and fast fluences. For the microstructural comparison specimens, the exposure time for the HFIR specimens was 3.5y, versus 20y for the HFBR material. The thermal flux was 2.7×10^{19} n/m²/s for the HFIR and 6.4×10^{18} n/m²/s for the HFBR. The fast flux was 1.6×10^{19} n/m²/s for the HFIR and 3.2×10^{17} n/m²/s for the HFBR. And the fast fluences were 1.8×10^{27} n/m² and 2.0×10^{26} n/m², respectively. The exposure times are only approximate because they include outages for refueling, etc. Nevertheless, they serve to show that time is not a prime determinant in the observed ripening; ripening was greater in the HFIR specimens, which had the shorter exposure time.

In principle, the degree of nucleation of the silicon precipitates might be expected to affect the final precipitate concentrations. Presumably, nucleation will be governed by the instantaneous supersaturation of silicon in solid solution, which will vary with thermal flux. In which case, the HFIR specimens, with their five-fold higher thermal flux should yield the higher concentration of particles. On the contrary, the HFBR materials displayed the greater concentrations. This discrepancy can not be attributed to the somewhat higher silicon levels in the HFBR materials; nucleation must have ceased long before the final silicon levels were reached. We must conclude that either the supposed connection of nucleation with thermal fluence is wrong or that precipitate ripening has masked the actual nucleation behavior.

Differences in fast neutron flux and fast fluence are the remaining contenders for the cause of the microstructural differences. The presence of the silicon-coated voids in the HFIR specimens testifies to the effectiveness of the 12 times higher fast fluence in promoting movement of vacancies and silicon atoms, and makes a strong case for particle ripening controlled by fast neutron dose. But the large factor of 50 higher fast flux for the HFIR specimens offers an alternative route. An increase in dose rate may promote precipitate ripening by allowing more point defect transport. For the case when loss of point defects is dominated by bulk recombination, the steady-state vacancy concentration varies as the square root of the flux¹⁰, and a rough guide to the relative point defect diffusion currents is given by;

$$\frac{\text{Diffusion current}_{(HFIR)}}{\text{Diffusion current}_{(HFBR)}} \propto \frac{t_{(HFIR)} \sqrt{\Phi_{(HFIR)}}}{t_{(HFBR)} \sqrt{\Phi_{(HFBR)}}} = \frac{3.5 \sqrt{50}}{20 \sqrt{1}} = 1.23 ,$$

where, for simplicity, time is in years and the fluxes are ratioed to the HFBR value.

For the case in which most of the point defects are lost to sinks, the appropriate expression¹⁰ is

$$\frac{\text{Diffusion current}_{(\text{HFIR})}}{\text{Diffusion current}_{(\text{HFBR})}} \propto \frac{t_{(\text{HFIR})} \cdot \Phi_{(\text{HFIR})}}{t_{(\text{HFBR})} \cdot \Phi_{(\text{HFBR})}} = \frac{3.5 \times 50}{20 \times 1} = 8.75 .$$

Thus, for both of these cases, the higher dose rate in the HFIR irradiations may have increased the level of atomic transport. This offers the prospect of a dose rate effect within a spectral effect, and adds another element of complexity to the situation.

If these data truly represent a spectral effect, not an inadvertent temperature effect, definitive spectral experiments under controlled conditions of temperature and dose rate are needed to resolve the issues raised herein.

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