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Combined Effects of Displacement Damage and High Gas Content in Aluminum

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MASTER

OAK RIDGE NATIONAL LABORATORY

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HIGH GAS CONTENT IN ALUMINUM

K. Farrell and J. T. Houston

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COMBINED EFFECTS OF DISPLACEMENT DAMAGE AND
HIGH GAS CONTENT IN ALUMINUM

K. Farrell and J. T. Houston

ABSTRACT

A solid solution alloy of 2300 appm of ^6Li isotope in aluminum was neutron irradiated at about $0.36 T_m$ in high, fast and thermal fluxes producing a damage level of 2 to 3 dpa and simultaneously inducing a gas content of about 2200 appm each of helium and tritium from burnup of ^6Li . The gases significantly increased the nucleation of structural defects but did not change the degree of swelling; cavity concentrations were increased ~ 1000 -fold, cavity sizes were decreased ~ 10 -fold and there was ~ 10 -fold increase in the concentrations of dislocations. Also, large cavities were developed on grain boundaries. The cavities were consistent with their being gas-filled bubbles. The refinement of damage structure by the gases caused a considerable increase in radiation hardening. Bend tests at 77 and 296 K revealed severe embrittlement and intergranular fracture. Comparison with data from material irradiated to produce comparable gas levels but relatively little displacement damage indicates that premature intergranular failure is much enhanced by the presence of a defect-hardened matrix. Postirradiation annealing tests showed the cavity and dislocation structures to have high resistance to annealing. Annealing also encouraged the development of a secondary population of large cavities believed to be associated with migration and precipitation of tritium.

INTRODUCTION

The question "What will happen to damage structures and to mechanical and physical properties of metals when significant displacement damage and high gas generation rates (and levels) occur simultaneously?" is especially pertinent to the conditions envisaged for the first wall of a controlled thermonuclear reactor (CTR) operating on a D-T reaction. Here the irradiation temperatures will be in the range for void formation, and the generation rates for helium and hydrogen from (n, α) and (n,p) reactions will be much higher than those in existing fission reactors.¹ Moreover, further quantities of these gases will be introduced from the plasma and from the lithium breeder blanket. Concurrently there will be high displacement damage rates. There are no precedents for these conditions and no neutron facilities in which the effects of these conditions can be determined explicitly. We have therefore done a simulation experiment to examine some of the effects of displacement damage and high gas generation rates in a simple material, high purity aluminum.

EXPERIMENTAL CONDITIONS

High purity aluminum is not likely to be used to build a CTR but it does have certain advantages for our simulation experiment. In the first place it readily develops radiation damage structures consisting of the usual loops, dislocations, voids and transmutation products at low and easily attainable neutron fluences.²⁻⁵ Second, it has a very high solid solubility for lithium. We can therefore utilize the large cross-section for the ${}^6\text{Li}(n,\alpha)$ reaction with thermal neutrons to produce significant quantities of helium and the hydrogen isotope tritium. And these gases will be created uniformly throughout the test specimens.

Our test material was a dilute alloy of high purity aluminum and 2300 appm (0.052 wt %) of ${}^6\text{Li}$ isotope (added to the melt as a pellet of 98% ${}^6\text{Li}$ + 2% ${}^7\text{Li}$). The composition of the alloy was analyzed by mass spectroscopy for the ${}^6\text{Li}$ isotope. This alloy, together with unalloyed base aluminum (designated 6-9 Al for six nines purity) was processed to

rods about 50 mm long by 3 mm diam which were annealed for 1 h at 873 K in air then furnace-cooled (FC) or water-quenched (WQ). (Actually we found such big differences in behavior between the 6-9 Al and the Al-Li alloy after irradiation that we did not pursue any effects of prior heat treatment.) The rods were irradiated in the hydraulic facility of the Oak Ridge High Flux Isotope Reactor (HFIR) either in direct contact with the cooling water at about 328 K ($0.35 T_m$) or packed in aluminum powder in a sealed capsule in which the temperature during irradiation was estimated to be about 348 K ($0.37 T_m$).

The neutron spectrum in the HFIR has both high thermal and fast components, thus assuring that in the Al-Li alloy displacement damage and high gas generation rates will occur simultaneously. The conventional thermal (2200 m/s or $E < 0.0253 \text{ eV}$) flux on the specimens was about $2 \times 10^{19} \text{ n/m}^2$. The fast ($E > 0.1 \text{ MeV}$) flux was about $9 \times 10^{18} \text{ n/m}^2$, equivalent to a displacement damage rate for aluminum of 1 to $2 \times 10^{-6} \text{ dpa/s}$. The neutron fluences attained are shown in Table 1, the displacement damage levels being in the range 2 to 3 dpa. The cross-section for the ${}^6\text{Li} (n, \alpha)$ reaction with conventional thermal neutrons is $950 \times 10^{-28} \text{ m}^2$. Consequently, the Al-Li alloy suffered 95-99% burnup of ${}^6\text{Li}$, resulting in estimated final helium and tritium levels in the region of 2200 appm each. Such gas levels are of the order of those expected to be created in the first wall of a CTR during 10 years of operation. In our experiment they were attained in a period of only about 20 days. The displacement damage rate is of the same order as that expected in a CTR first wall material but the damage level is equivalent to only a few weeks of CTR damage.

OBSERVATIONS

Hardness and Density Measurements

Hardness measurements were made at room temperature on ground and polished specimens cut from the irradiated rods. A Kentron tester was used with a diamond pyramid indenter and a load of 0.5 kg. The average

Table 1. Irradiation Conditions, Hardness, and Swelling Values

	Irradiation Temperature (K)	Neutron Fluence ($\times 10^{25}$ n/m ²)		Hardness (MPa)	Swelling (%)
		E < 0.0253 eV	E > 0.1 MeV		
6-9 Al (FC)		0	0	137	
Al-0.05 Li (FC)		0	0	137	
Al-0.05 Li (WQ)		0	0	196	
6-9 Al (FC)	~328	5.3	2.2 (3 dpa)	382	
Al-0.05 Li (FC)	~328	5.3	2.2 (3 dpa)	902	
Al-0.05 Li (WQ)	~328	5.3	2.2 (3 dpa)	863	
6-9 Al (FC)	~348	3.3	1.5 (2 dpa)	245	0.64
Al-0.05 Li (FC)	~348	3.4	1.4 (2 dpa)	745	0.56/0.79

hardness values from three indentations for each data point are given in Table 1. The Al-Li alloy displayed considerably greater irradiation hardening than did the 6-9 Al.

Densities (ρ) were measured by Archimedes' displacement on just two of the rods. The degree of swelling, $S = \frac{100(\rho_u - \rho_i)}{\rho_i}$, where the subscripts u and i denote unirradiated and irradiated conditions, is about equal at 0.6% (Table 1) for the pure aluminum and for the Al-Li alloy. The two values of swelling quoted for the Al-Li alloy depend on whether we use the density of Al-Li alloy or of pure aluminum, respectively, for the unirradiated reference material. The use of the density of pure aluminum in this respect allows for the change in composition caused by burnup of ^6Li .

Embrittlement

Mechanical testing consisted of bending the rods between two pairs of hand-held pliers in air at room temperature (296 K) and in liquid nitrogen (77 K). At both temperatures the unirradiated materials and the irradiated 6-9 Al could be bent through a full circle without failure. The irradiated Al-Li alloy snapped cleanly with an audible crack and with no measurable bend angle; more force was needed to cause fracture in liquid nitrogen. At both temperatures the fracture path in the irradiated Al-Li alloy was wholly intergranular (Fig. 1a).

Examination of the fracture surfaces of the irradiated Al-Li alloy using scanning electron microscopy and, for better resolution, carbon replicas revealed the boundaries to be riddled with cavities (Fig. 1b,c,d). Fracture occurred by ductile tearing of the grain boundary areas between the cavities. The scale of this ductile tearing was smaller in specimens tested in liquid nitrogen. The shapes and concentrations of cavities varied widely from one grain boundary to another. The photographs in Fig. 1 were selected to show the range of cavitation. Measurements of the cavities indicated that they covered up to 90% of the grain boundary area on some of the fracture facets. On other facets the cavities were relatively small and they covered as little as 10% of the grain boundary area. Typically, the coverage was 50% or more. Frequently the cavities

Fig. 1. Intergranular Fracture in Al-Li Alloy Irradiated at 348 K and Bend Tested at 296 K (a,b,c) and 77 K (d).

were interconnected and it was difficult to distinguish individual cavities. Rough measurements indicated that there were at least 10^{12} cavities/m² of grain boundary. If these are assumed to be spherical then the size of cavity required to touch one another and give almost complete grain boundary coverage is just 1 μ m. This is quite compatible with the observations. These grain boundary cavities did not result from the mechanical testing. They were created during neutron irradiation.

Microstructures

Disks were cut from untested, irradiated rods and were thinned for transmission electron microscopy. The grain size was large, 0.6 mm, and the grain boundaries in the Al-Li alloy tended to be gouged out during electropolishing. The few boundaries that we did manage to retain in thinned areas of the Al-Li alloy are considered to be the better boundaries (i.e., those with the smallest cavities). Two of these are shown in Fig. 2. On such boundaries the cavity sizes ranged from about 15 to 400 nm diam, with concentrations of about 10^{13} /m² of grain boundary area.

There were many tiny cavities within the grains in the Al-Li alloy. These are barely visible in Fig. 2 but can be seen more clearly in Figs. 3b and 4b. Also evident in Fig. 3b is a defect-free zone on each side of the grain boundary. Examination of several grain boundaries indicated that the width of this denuded zone was about equal to the diameter of the largest cavity visible on the boundary.

In the irradiated 6-9 Al there were no cavities on grain boundaries (Fig. 3a) and the denuded regions at the grain boundaries were a factor of 3 or more wider than those in the Al-Li alloy. The cavities within the grains were very much larger and of much lower concentrations than the cavities within the grains of the irradiated Al-Li alloy. Precipitate particles of silicon, a solid transmutation product of aluminum,⁶ were present on grain boundaries and on cavities in the 6-9 Al but none was observed in the Al-Li alloy.

Fig. 2. Large Cavities on Grain Boundaries in Al-Li Alloy Irradiated at 348 K.

Fig. 3. Differences in Grain Boundary Denuded Regions and Matrix Cavities After Irradiation at 348 K. (a) Pure aluminum. (b) Al-Li alloy.

Dislocation concentrations were higher in the Al-Li alloy and were arranged in a uniform configuration as shown in Fig. 4a. There was no discernible association of the dislocations with the numerous, small matrix cavities that can be seen more clearly in Fig. 4b. Quantitative analysis of the microstructures of the irradiated specimens are presented in Table 2. The measured cavity concentrations in the Al-Li alloy are believed to be somewhat low because of significant overlapping of cavity images and because there probably were cavities present whose sizes fell below our resolution limit of about 3 nm.

Annealing Experiments

The microstructures in the irradiated Al-Li alloy were found to have much greater resistance to annealing than those in the irradiated 6-9 Al. Disks about 1 mm thick were cut from each material and were annealed for 1 h in air at temperatures of 573, 673, and 773 K. During 1 h at 573 K all cavities and dislocations completely disappeared in the 6-9 Al and the silicon precipitate coarsened. In the Al-Li alloy the dislocation structure and the small cavities appeared to remain substantially unaffected by the anneal except for a small but obvious growth of some cavities like strings of beads along the dislocations. However, two new features appeared — coarse silicon particles and a uniform distribution of large, secondary cavities within the grains, as shown in Fig. 5 upper (compare with Fig. 4a). These annealing cavities were frequently associated with the coarse silicon particles and they had a wide variety of shapes ranging from rod-like to plate-like, all with crystallographic facets. Their sizes ranged up to 200 nm or so. The formation of these secondary cavities did not involve dissolution of the original, very small cavities in their immediate vicinity (Fig. 5, lower).

At 673 K the dislocation structures and the small matrix cavities were still present in the Al-Li alloy. Growth of cavities on dislocations was no longer obvious but a slight coarsening of the matrix cavities was evident. The large annealing cavities were larger (up to 1 μm) and fewer than those at 573 K, and they were more equiaxed in shape. At 773 K the irradiated Al-Li specimens tended to disintegrate and thinned foils could not be obtained.

Fig. 4. Dislocations and Small Matrix Cavities in Al-Li Alloy Irradiated at 348 K.

Table 2. Microstructural Data

Material	Irradiation Conditions	Cavities/m ³	Cavity Diameter (nm)	Loops/m ³	Loop Diameter (nm)	Dislocations (m/m ³)
6-9 Al	348 K, 2 dpa	8.7×10^{19}	52	6.7×10^{18}	~40	1.3×10^{13}
Al-0.05 Li	348 K, 2 dpa	$>7 \times 10^{22}$	~4	$>8 \times 10^{18}$	~50	$>6 \times 10^{13}$
Al-0.05 Li	328 K, 3 dpa	2×10^{23}	<4	5×10^{19}	~20	2×10^{14}

Fig. 5. Large Matrix Cavities Developed in Al-Li Alloy During Postirradiation Annealing for 1 h at 573 K. Black particles are coarsened precipitates of transmutation-produced silicon. Lower photograph shows at high magnification tiny cavities in vicinity of one of the anneal-induced cavities.

DISCUSSION

It is clear that major changes in microstructural defects and in mechanical behavior occur in aluminum when large quantities of tritium and helium are created during displacement damage. These changes are more a matter of difference in the scale or degree of damage than of radical alteration in the nature of the damage. The same neutron-induced structural features (e.g., dislocations, cavities, and precipitates) are involved but there are many more of them when they are created in the presence of the gases. The gases evidently enhance nucleation of the structural defects but do not change their form. The cavities differ in the sense that those in the 6-9 Al are probably empty and can anneal readily whereas those in the Al-Li alloy are undoubtedly gas-filled and are relatively resistant to annealing. The amount of gas required to stabilize the cavities in the 6-9 Al as equilibrium gas bubbles is estimated to be 1.5×10^3 appm (taking a surface energy value of 1.2 J/m^2 for aluminum⁷ at 348 K, and using Van der Waals' constant for helium gas⁸ of $1.91 \times 10^{-29} \text{ m}^3/\text{atom}$ and assuming this constant applies to hydrogen, too). The estimated gas levels in the 6-9 Al from $\text{Al}(n,p)$ and $\text{Al}(n,\alpha)$ reactions are only 4 appm H and 0.7 appm He. In the Al-Li alloy, however, the presence of cavities on the grain boundaries is strong testimony for gas bubbles. Also, the amount of gas present is quite enough to stabilize the cavities; gas levels of 2200 appm each of ^4He and ^3He can support a population of 4 nm diam bubbles of $1.8 \times 10^{23}/\text{m}^3$, which is of the order of the cavity concentrations measured in the Al-Li alloy. These calculations are essentially unaffected by the loss of gas to the large grain boundary bubbles. Even in the extreme case of complete grain boundary coverage with 1 μm bubbles the amount of gas involved at the boundaries is only 30 appm, a negligible quantity with respect to the total gas content.

Despite the large differences in gas content between the 6-9 Al and the Al-Li alloy, and irrespective of whether the cavities are gas-filled or not, both materials swell to the same degree, about 0.6%. The gases greatly increase the concentrations of cavities but do not increase the level of swelling. Presumably at the high cavity concentrations in the

Al-Li alloy the cavities have become the predominant sinks for both vacancies and self-interstitials thus minimizing growth of cavities and controlling the swelling. The distributions of cavities differ in the two materials. There are no grain boundary cavities in the 6-9 Al. Those in the Al-Li alloy are massive compared with the cavities within the grains, but these large cavities do not dominate the swelling. The concentration, N , of large cavities on the grain boundaries in the Al-Li alloy was in the range 10^{12} to $10^{13}/\text{m}^2$. The average diameters of such cavities required to completely cover the grain boundaries are the center-to-center bubble spacings on the boundaries (i.e., $N^{-1/2}$ or 10^{-6} to 3.2×10^{-7} m, respectively. Swelling from these cavities is given by $\frac{2N\pi d^3}{6\ell}$ where ℓ is the grain diameter and $\frac{2}{\ell}$ is the grain boundary area per unit volume. In these materials ℓ is 5.8×10^{-4} m, and the maximum swelling from grain boundary bubbles will lie in the range 0.18 to 0.06%. The balance of the measured swelling, 0.5 to 0.6%, must be caused by the small cavities within the grains. The measured concentration of these small cavities is roughly $1 \times 10^{23}/\text{m}^3$ and their size is about 4 nm diam (i.e., about 0.33% swelling). This is a reasonably good agreement considering our belief that we have undercounted the small cavities. In fact, as we showed earlier, the concentration of 4 nm diam gas bubbles calculated from the known gas level is $1.8 \times 10^{23}/\text{m}^3$, which will give 0.59% swelling.

The tensile strength or flow stress of an irradiated metal containing several species of structural defects is described⁹ by an equation of the form

$$\sigma_i = \sigma_u + [(\Delta\sigma_{\text{dis}})^2 + (\Delta\sigma_{\text{cav}})^2 + (\Delta\sigma_{\text{other}})^2 + \dots]^{1/2}, \quad (1)$$

where σ_u is the strength of the unirradiated metal, and $\Delta\sigma_{\text{dis}}$ is the additional strengthening from a dispersion of radiation-induced dislocations, cavities or other defects that impede dislocation movement. Similarly for hardness,

$$H_i = H_u + [(\Delta H_{\text{dis}})^2 + (\Delta H_{\text{cav}})^2 + (\Delta H_{\text{other}})^2]^{1/2}. \quad (2)$$

The hardening increment from grown-in dislocation is given by the expression $\Delta H_{\text{dis}} = 3 G b \lambda^{1/2}$, where the factor 3 converts flow stresses to hardness values,¹⁰ G is the shear modulus (2.65×10^4 MPa for Al), b is the Burgers vector (0.286 nm), and λ is the concentration of grown-in dislocations on the slip plane. The contribution from dislocation loops¹¹ is given by $\Delta H_{\text{d}} = 6 G b (N d)^{1/2}$, where N is the concentration of loops per unit volume of aluminum, and d is their mean diameter; in the present work the hardening component from loops is almost negligible. Cavities make a significant contribution¹² via the equation $\Delta H_{\text{cav}} = 6 G b (N d)^{1/2}$. Using these equations and the quantitative microstructural data from Table 2 we have calculated the hardness values given in column 5 of Table 3. The calculated values seriously exceed the measured hardnesses at the higher hardness values. This, we believe, is due to the use of the ΔH_{cav} equation which may overestimate ΔH_{cav} . We get a different result if we consider the cavities to act as hard particles opposing dislocation motion, for which we can derive⁹ the expression

$$\Delta H_{\text{cav}} = \frac{3 \times 1.04}{\pi} G b (N d)^{1/2} \cdot \ln \left(\frac{d}{4b} \right) .$$

This reduces the calculated hardness values to those shown in column 6 of Table 3. Note that the two sets of calculated hardnesses now embrace the measured values. Presumably we could play around with hardening equations until we obtained a satisfactory correlation but that is not the primary purpose of this exercise. Rather, we wish to draw attention to the theoretical hardening expressions, all of which indicate that the increase in hardness caused by microstructural defects is an inverse function of the spacing between the defects. Hardness should thus increase with the concentrations of defects which, of course, is qualitatively what we see in our materials. In short, the irradiated Al-Li alloy is much harder than the irradiated pure aluminum because of the finer scale of the damage structure in the Al-Li alloy.

Table 3. Hardness Correlation

Material	Irradiation Temperature (K)	H_u (MPa)	H_i (Meas.) (MPa)	H_i (Calc.) (MPa)	
				Cavities	Hard Particles
6-9 Al	348	137	245	265	235
Al-0.05 Li	348	137	745	912	363
Al-0.05 Li	328	196	863	1520	618

The brittle behavior of the irradiated Al-Li alloy can be related to the irradiation hardening and to the structural modifications induced at grain boundaries. Grain boundaries are distorted lattice regions that are known to be good sources of vacancies and also are sinks for vacancies, self-interstitials and impurities. During irradiation the grain boundaries absorb impurities and irradiation-induced point defects from regions immediately adjacent to, and on each side of, the boundary, leaving clearly visible zones denuded of defects. Such zones are soft compared with the defect-hardened matrix. Gases entering the grain boundaries develop into large bubbles with the aid of readily available vacancies in the boundaries. We have made simple calculations of the amount of gas required to form the observed concentrations and sizes of grain boundary gas bubbles in the irradiated Al-Li alloy and find that adequate quantities of gas can be drawn from the denuded regions alone. These bubbles are built-in holes which can tear open under appropriate shear strains. Shear strains are concentrated at grain boundaries because the boundaries are natural barriers to transmission of slip, and because of the denuded zones at the boundaries. During mechanical testing the hard matrix can support a higher load than the softer denuded zones which must therefore deform prior to the matrix. Plastic strain becomes localized in the grain boundary regions. The grain boundaries, already weakened by the presence of gas bubbles, become overloaded and tear open. The result is highly intense local deformation at grain boundaries with little or no measurable bulk strain, and with intergranular fracture. Similar fracture is avoided in the irradiated 6-9 Al because the matrix is relatively soft, the grain boundary denuded zones are wider, and the grain boundaries are free of irradiation-induced cavities.

The annealing response of the Al-Li alloy deserves comment. The fine matrix cavities were remarkably stable to annealing even despite the strong evidence of vacancy migration. This migration can be seen in the growth of the silicon precipitates and in the development of the secondary population of large cavities during annealing. In previous work on cavity annealing in neutron irradiated aluminum^{5,13} of low gas content, as well as as in the present work on 6-9 Al, it was found that growth of silicon precipitates was concomitant with cavity shrinkage or growth, implying a

coupled migration of vacancies and silicon atoms. Also, when large cavities grew, it was always at the expense of their immediate smaller neighbors by a ripening process. This is not so in the Al-Li alloy. Here the small matrix cavities in the vicinity of the anneal-induced large cavities are not dissolved during the anneal. They may shrink a little but they do not disappear. The difference is presumed to be due to the effects of the ^4He and ^3H . We suspect that in the Al-Li alloy the small cavities retain their inert helium during annealing but release tritium which migrates and precipitates to form the large, annealing cavities. Hydrogen has a very low solubility in aluminum and it readily forms bubbles during proton bombardment;¹⁴ moreover, these hydrogen bubbles coarsen during postbombardment anneals at 573-773 K and eventually disperse, although they may cause extensive grain boundary cracking in the interim. This picture fits nicely with our observations on annealing of the irradiated Al-Li alloy. We certainly had significant tritium migration even at room temperature. Every time we opened our lead storage can to retrieve an irradiated Al-Li specimen we contaminated the walls of our glove box with tritium. Also, when we returned to some thinned electron microscopy foils to reexamine grain boundaries we found that the boundaries had cracked open during storage at room temperature. The most logical explanation of this is that tritium precipitated into the large grain boundary cavities and caused them to burst through the foil surfaces.

These observations and deductions raise the question of the role of tritium in the development of cavities and damage structure. Unfortunately, the irradiated metals contain at least equal quantities of helium and hydrogen (or tritium), and helium is a noted promoter of cavities.^{4,15} We cannot readily distinguish a separate role of hydrogen but if hydrogen bubbles can form easily in aluminum during annealing at temperatures above about 370 K we see no reason why they should not develop during irradiation at 328 and 348 K, particularly at grain boundaries. And the grain boundary cavities in the Al-Li alloy are very large. Perhaps they do contain more tritium than helium. The tritium can migrate much more readily than the helium, and its low solubility would force it to precipitate at any free surface. It may also be involved in nucleation of the small matrix

cavities. Evidence in the literature indicates that hydrogen increases the concentration of cavities in neutron irradiated aluminum⁴ and in electron-irradiated PE16 alloy,¹⁶ 316 stainless steel¹⁷ and ferritic steels.¹⁸ Until recently hydrogen was not seriously considered an element of radiation damage in most reactor constructional materials, except zirconium, because it was assumed to escape. The writing is now on the wall. In CTR materials the hydrogen generation rates will be many times greater than those of helium, and possible effects of hydrogen should not be overlooked.

It is obvious from this work that when high gas contents and significant neutron displacement damage occur simultaneously in aluminum the resulting effects on damage structures and on mechanical properties are much greater than if the displacement damage occurs alone. It is not clear whether the gas and the displacement damage act synergistically (i.e., whether their combined effects are greater than the sum of their independent effects) since we have not measured the effects of gases alone. Other experimenters¹⁹⁻²² have studied Al-Li alloys irradiated in predominantly thermal reactors where the neutron spectrum contained relatively minor portions of fast neutrons and hence induced no significant build-up of displacement damage. In these early experiments the major microstructural change was the formation of gas bubbles on grain boundaries and on preexisting dislocations. These bubbles caused some swelling and hardening and loss in ductility but usually considerable ductility was retained and the fracture path remained transgranular except at elevated test temperatures above about 470 K where intergranular fractures occurred and ductility values fell to very low levels at high gas contents. Some of these changes are illustrated by the data in Table 4 for irradiations done at 348 K to give gas levels similar to those in our Al-Li alloy. Note, in Table 4, that with increasing neutron fluence (and gas content) the average size of gas bubbles increases and the average concentration of bubbles decreases markedly. At the gas level of interest to the present work (4480 appm) the concentrations and sizes of bubbles within the grains are quite different from those in our irradiated Al-Li alloy; they are, in fact, almost identical to those of the cavities in the

Table 4. Microstructural Data and Tensile Properties of Al-⁶Li Alloy
(0.3% Li Enriched 96% in ⁶Li) Irradiated at 348 K in a
Predominantly Thermal Reactor
(Extracted from References 22a and 22b)

Properties	Thermal Fluence, n/m ²				
	0	6×10 ²²	1.2×10 ²³	1×10 ²⁴	2×10 ²⁴
Estimated ^a gas concentration, appm	0	150	280	2350	4480
Size of grain boundary bubbles, nm	0	20+	Occasionally up to 1 μm	20+	250+
Width of grain boundary denuded regions, nm	0	0	0	400	750
Bubble density in matrix, per m ³	0	?	2×10 ²²	1×10 ²¹	8×10 ¹⁹
Average diameter of matrix bubbles, nm	0	~2	~5	13	50
Yield stress, MPa					
at 77 K	22	33	45	46	(b)
at 296 K	14	23	33	36	(b)
at 523 K	8	19	21	18	(b)
Ultimate tensile stress, MPa					
at 77 K	124	134	143	128	(b)
at 296 K	68	93	79	69	(b)
at 523 K	17	30	30	22	(b)
Elongation, %					
at 77 K	23	33	26	20	(b)
at 296 K	26	25	13	22	(b)
at 523 K	69	10	8	1	(b)
Matrix Swelling, ^c %	0	?	0.13	0.11	0.52

^aA gas concentration of 150 appm implies 75 appm ³H and 75 appm ⁴He.

^bSpecimens contained surface cracks and fissures. Not tensile tested.

^cMatrix swelling is calculated from the bubble density, N , and average bubble size, d , and is given by $100(V_b/1-V_b)$, where V_b is $N\pi d^3/6$, the volume of bubbles per m³ of swollen material.

6-9 Al. Consequently, the degree of radiation-induced hardening is expected to be small. Indeed an extrapolation of the ultimate tensile strength from the lower fluence UTS values at room temperature (296 K) indicates no more than about 69 MPa, or a maximum hardness of about 207 MPa which, again, compares more favorably with our irradiated 6-9 Al than with our Al-Li alloy. A similar extrapolation in elongation values would imply retention of significant ductility at 77 and 296 K, again more in keeping with our irradiated pure aluminum than with our irradiated Al-Li alloy. The only points of similarity with our Al-Li alloy observations are the grain boundary bubbles and the degree of swelling.

This comparison strongly suggests a synergistic effect of gases and displacement damage on microstructure and embrittlement. But there is one little flaw in the argument; the data in Table 4 were obtained in a reactor whose thermal neutron flux was undoubtedly much lower (by at least a factor of 10) than that in HFIR. So the data not only represent relative absence of displacement damage but also a lower gas generation rate. We cannot, therefore, conclude that a synergistic effect prevails in our observations. Certainly, however, the combined effects of high gas level and displacement damage are very much greater than the separate effects of displacement damage or high gas level. Another important point illustrated by this comparison is that although gas bubbles on grain boundaries will cause some loss in ductility at low temperatures (Table 4) superimposition of a hardened matrix brings about severe embrittlement (Fig. 1). Indeed, if such hardening, and a weakening of grain boundaries, occurs under CTR conditions then embrittlement and premature fracture, not swelling, is likely to be the Achilles' heel of CTR materials.

CONCLUSIONS

In aluminum neutron irradiated at moderate temperatures ($\sim 0.36 T_m$) the presence of about 2200 appm each of helium and tritium created concurrently with significant displacement damage causes: (1) No change in overall swelling. (2) Considerable refinement of damage structures without radical change in the type of damage; cavity concentrations are increased 1000-fold, cavity sizes are decreased 10-fold, dislocation concentrations are increased about 10-fold. (3) Development of large cavities on grain boundaries. (4) A narrowing of defect-free regions adjacent to grain boundaries. (5) A large increase in irradiation hardening. (6) Severe embrittlement during mechanical testing associated with intergranular fracture even at very low temperatures.

The combined effects of high gas level and displacement damage are much greater than the effects of displacement damage alone, but it is not clear if there is a synergistic effect. Gases enhance nucleation of defect structures, and at high gas levels the cavities are gas-filled. This refinement of damage structure increases the irradiation-hardening. A combination of a hardened matrix and weakened grain boundary regions leads to severe embrittlement and intergranular fracture. Postirradiation annealing causes the development of a secondary population of large cavities believed to be associated with tritium migration and precipitation. Migration of tritium is evident even at room temperature.

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