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Mechanical properties and microstructure of neutron irradiated cold worked Al-6063 alloy

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Abstract

The impact of neutron irradiation on the mechanical properties and fracture morphology of cold worked Al-6063 were studied, using scanning and transmission electron microscopy, and tensile measurements. Specimens (50 mm long and 6 mm wide gauge sections) were punched out from an Al-6063 23% cold worked tubes, which had been exposed to prolonged neutron irradiation of up to 4.5×10^{25} thermal neutrons/m^2 ($E < 0.625$ eV). The temperature ranged between 41 and 52°C. The tensile specimens were then tensioned till fracture in an Instron tensiometer with strain rate of 2×10^{-3} s^{-1}. The uniform elongation and the ultimate tensile strength increase as functions of fluence. Metallographic examination and fractography reveal a decrease in the local area reduction of the final fracture necking. This reduction is accompanied with a morphology transition from ductile transgranular shear rupture to a combination of transgranular shear with intergranular dimpled rupture. The intergranular rupture area increases with fluence. No voids could be observed up to the maximum fluence. The dislocation density of cold worked Al decreases with the thermal neutron fluence. Prolonged annealing of unirradiated cold worked Al-6063 at 52°C revealed similar results. It thus appears that under our irradiation conditions the temperature during irradiation is the major factor influencing the mechanical properties and the microstructure during irradiation. © 1998 Elsevier Science B.V.

1. Introduction

Aluminum alloys exhibit good corrosion resistance as well as very low capture cross section of fast and thermal neutrons thus, they are extensively used as structural materials in the nuclear industry. In the past, the choice of a particular alloy was based on the properties of the unirradiated material [1]. Naturally, structural materials in the reactor core are exposed to large fluxes of fast and thermal neutrons, resulting in microstructural changes. Thus, mechanical properties changes take place during service. Numerous studies have been performed concerning the irradiation impact on aluminum alloy properties [1–19]. Irradiation usually induces degradation in mechanical properties (particularly increase of ultimate tensile strength and reduction of elongation), and swelling. On the other hand, irradiation of cold-worked Al-A5 causes its annealing, and as a result the ultimate tensile strength and elongation increase with the neutron fluence [17]. Some studies indicate that annealing of cold worked Al-1100 [15] and Al-6061 [1] may occur during irradiation. However, the impact of irradiation of cold worked aluminum alloys on the mechanical properties is not well established, especially for alloys containing Mg, which undergo precipitation hardening. The present work is aimed at investigating the post-irradiation properties of cold worked Al-6063.

2. Experimental

Al-6063 23% cold worked parts were exposed to prolonged irradiation of neutron fluences of up to 4.5×10^{25} thermal neutrons/m^2 ($E < 0.625$ eV). The temperature ranged between 41 and 52°C. For mechanical examination, tensile specimens were punched out from the tubes: 50 mm long and 6 mm wide gauge sections. According to our experience (transmission electron microscopy of the

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Table 1
Mechanical properties of cold worked Al-6063 as a function of the thermal neutron fluence

Fluence (10^{24} n m^{-2})	Stress		Elongation		Final area reduction (%)
	σ_Y (MPa)	σ_{UTS} (MPa)	uniform (%)	total (%)	
0.0	189	239	7.9	11.5	73
1.5	222	291	9.6	11.0	50
4.7	222	282	11.2	13.5	45
8.8	234	289	10.6	13.4	47
11.5	224	277	9.7	11.2	45
14.9	240	301	12.3	13.5	43
19.0	235	294	11.3	14.1	35
21.5	236	267	10.5	12.8	32
33.5	235	309	10.0	12.5	30
45	255	314	10.0	12.5	25

punched samples), the sample fabrication did not introduce a significant work hardening. The tensile specimens were then tensioned till fracture in an Instron tensiometer at a strain rate of 2×10^{-3} s^{-1}. Fractured surfaces were examined with a scanning electron microscope (Phillips SEM type 505). Parts of the aluminum were thinned for transmission electron microscopy. First, thinning to about 150 μm was performed chemically with a 1 M aqueous solution of NaOH. Then discs of 3 mm were punched out of the specimen, and further thinned by electropolishing using 20% perchloric acid in methanol at −20°C, at a voltage of 20–30 V.

Structural materials in a reactor core are influenced not only by the fast and thermal neutron interaction but undergo a prolonged low temperature annealing (52°C in our case). To study the low temperature annealing a set of

Table 2
Mechanical properties of cold worked Al-6063 as a function of the aging duration at 52°C

Aging duration (month)	Stress		Elongation		Final area reduction (%)
	σ_Y (MPa)	σ_{UTS} (MPa)	uniform (%)	total (%)	
0	189	238	7.9	11.9	73
1	189	240	9.8	13.2	72
2	202	258	10.6	13.2	73
6	222	282	11.1	12.8	50
8	229	289	11.1	12.9	51
13	234	293	11.1	12.85	45
15	228	287	11.9	14.2	42
18	245	304	11.8	12.5	37
22	242	300	10.8	12.5	37
24	243	301	11.0	12.2	35
27	247	306	11.1	12.7	30
30	245	302	10.9	11.3	27

Al-6063 23% cold worked samples were annealed at 52°C for approximately 2.5 yr and more. The samples were examined in the same way as the irradiated ones.

3. Result

3.1. Mechanical properties of irradiated specimens

Irradiation of cold worked Al-6063 caused mechanical properties changes depending on the thermal neutron fluence, as seen in Table 1. Typical stress/strain curves of unirradiated, as well as irradiated Al-6063 for different neutron fluences are presented in Fig. 1. The impact of

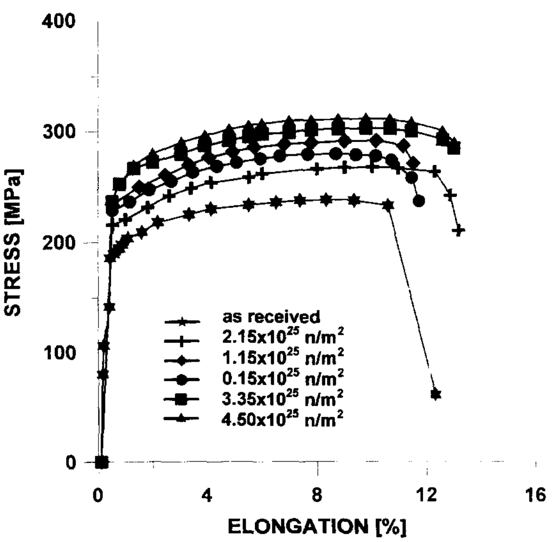


Fig. 1. Stress–strain curves of unirradiated Al-6063 samples and of samples irradiated to different thermal neutron fluences.

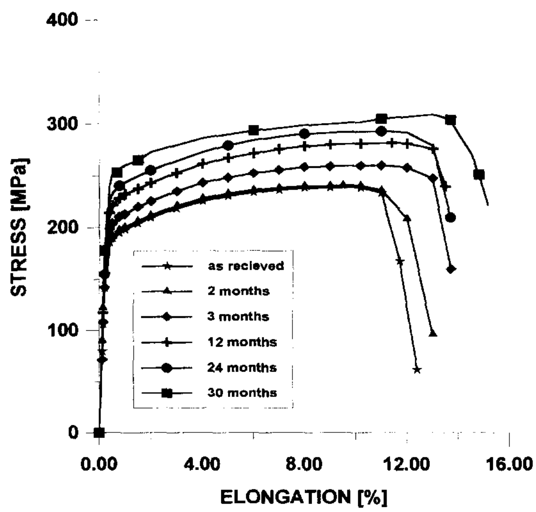


Fig. 2. Stress–strain curves of Al-6063 samples as a function of annealing duration at 52°C.

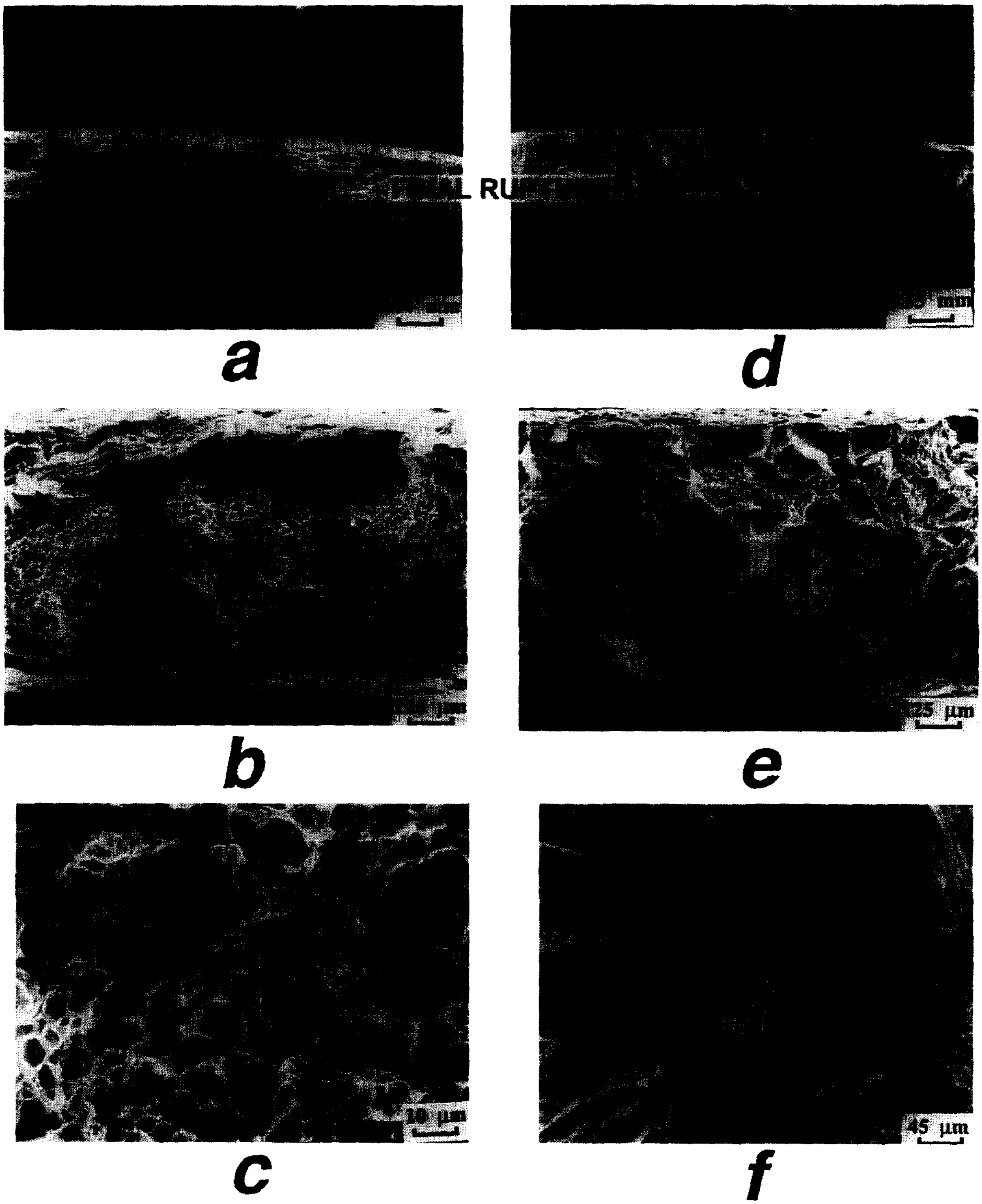


Fig. 3. Secondary electron images illustrating the microstructure of 23% cold worked Al-6063 before irradiation (a)–(c), and after irradiation to 1.2×10^{25} thermal neutrons/m^2 (d)–(f).

neutron irradiation on the mechanical properties is summarized in Table 1. As seen from Fig. 1 and Table 1, the uniform elongation, the yield strength (σ_Y), and the ultimate tensile strength (σ_{UTS}), are increasing functions of the neutron fluence, while the final area reduction of the fracture is a decreasing function of the fluence. It should be mentioned that many of the samples fractured outside the extensiometer. Therefore, the uncertainty in the total elongation is quite large. Generally the total elongation seems to be mostly unchanged, with only a slight tendency for increase with the neutron fluence.

The mechanical properties of samples which were subjected to a 2.5 yr annealing at 52°C without irradiation show similar mechanical properties change as occurred during irradiation, as seen in Table 2 and Fig. 2.

3.2. Fractography

The impact of irradiation on the fracture morphology and on the final area reduction of the fractured specimen irradiated to 1.2×10^{25} thermal neutrons/m^2 is illustrated in Fig. 3. Two major changes occurred due to irradiation: reduction of the final area fracture, and fracture morphology. There is an increase of the final fracture thickness (i.e. decrease of the area reduction of the final rupture) with increasing thermal neutron fluence. For example, the final area reduction for unirradiated samples is about 80%. Irradiation to fluences of 1.0×10^{25} and 2.7×10^{25} thermal neutrons/m^2 reduces the local area reduction to 42% and 32%, respectively. An improved view, at a higher magnification, of the irradiation impact on the fracture morphology is illustrated in Fig. 4. The fracture of the unirradiated samples exhibits a transgranular dimpled shear rupture microstructure (Fig. 4(a)), characteristic of a ductile fracture. After irradiation, the fracture morphology changes to a mixture of transgranular dimpled shear rupture, and intergranular dimpled rupture that appears as large facets. The fraction of the intergranular rupture areas increases with the thermal neutron fluence. Similar behavior of fracture morphology was observed for specimens annealed at 52°C for long periods of time, as illustrated in Fig. 5. A larger magnification of the facets observed in both cases reveals a dimpled structure as seen in Fig. 6. The dimple size is by an order of magnitude smaller than the one revealed in the shear transgranular rupture.

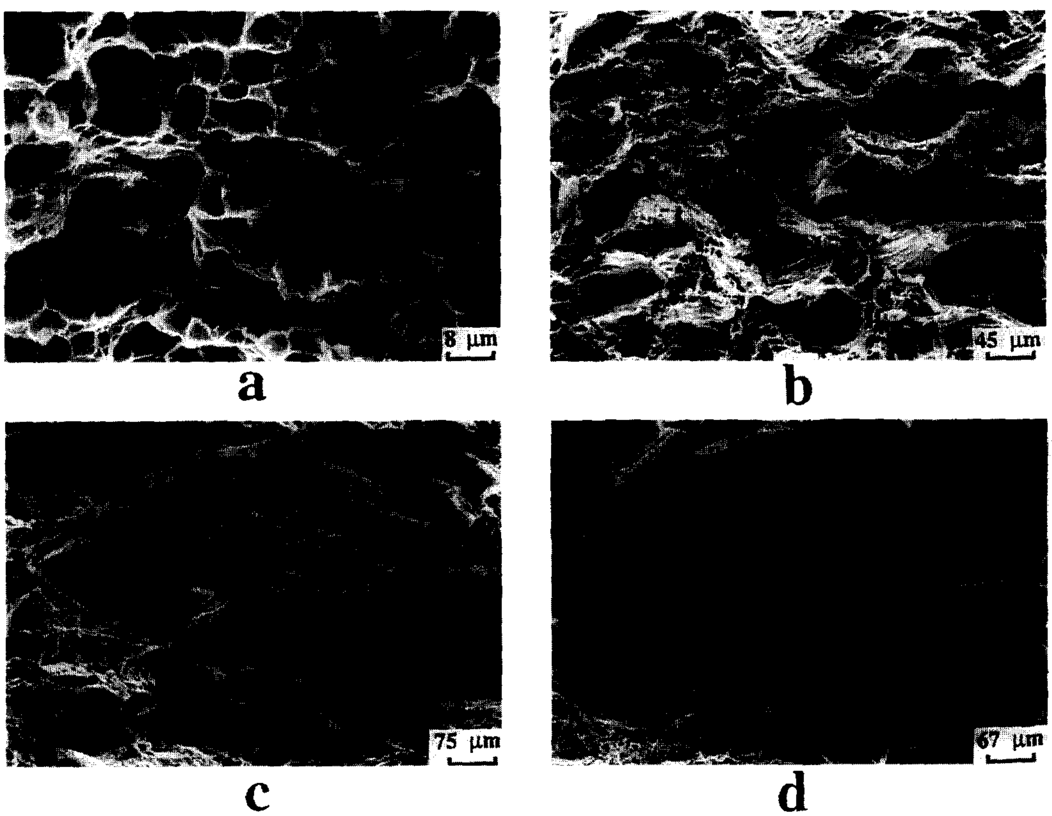


Fig. 4. Morphology of fractured surfaces for different thermal neutron fluence; (a) unirradiated sample, (b) irradiated to 1×10^{25} thermal neutrons/m^2, (c) irradiated to 2.5×10^{25} thermal neutrons/m^2, (d) irradiated to 4.5×10^{25} thermal neutrons/m^2.

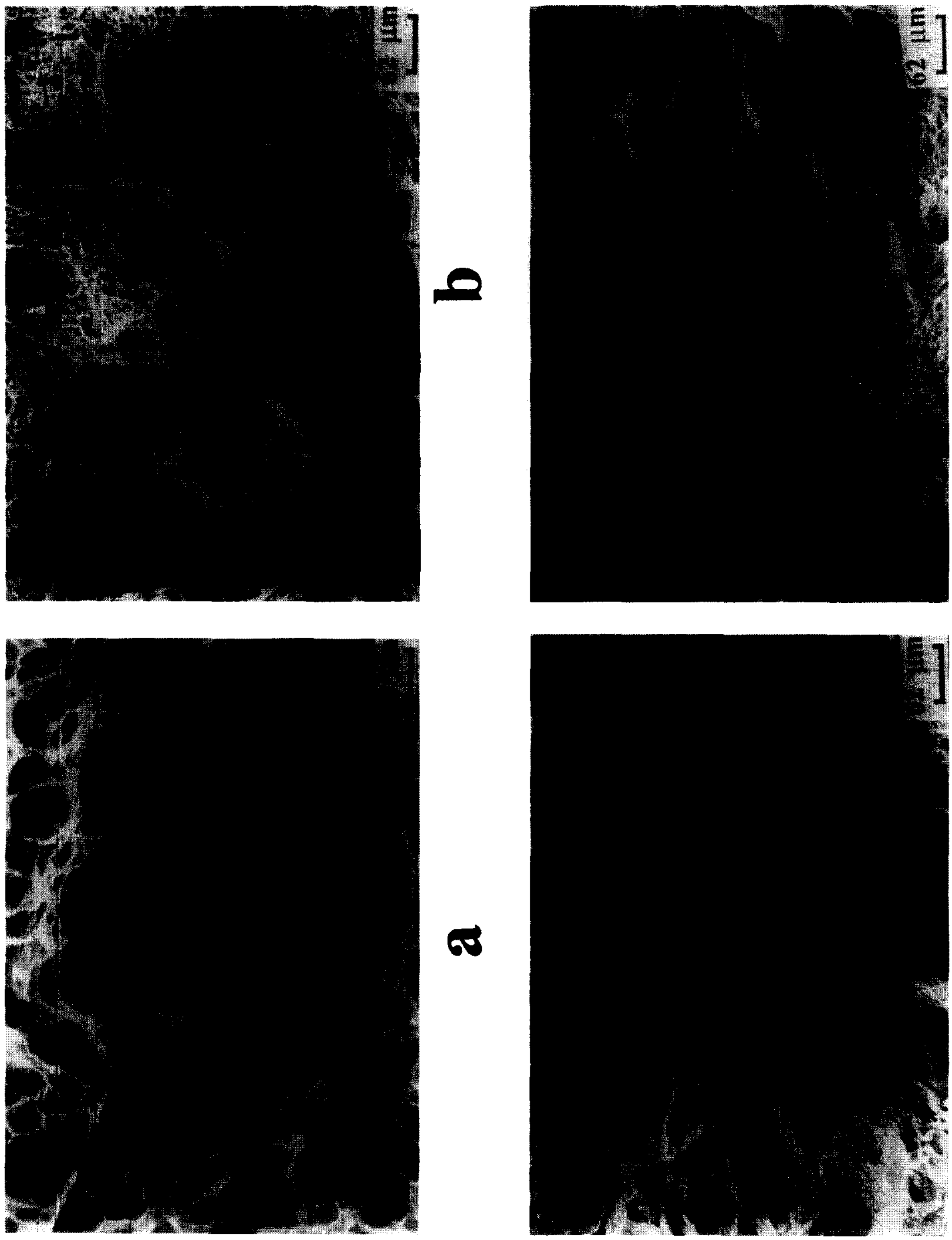
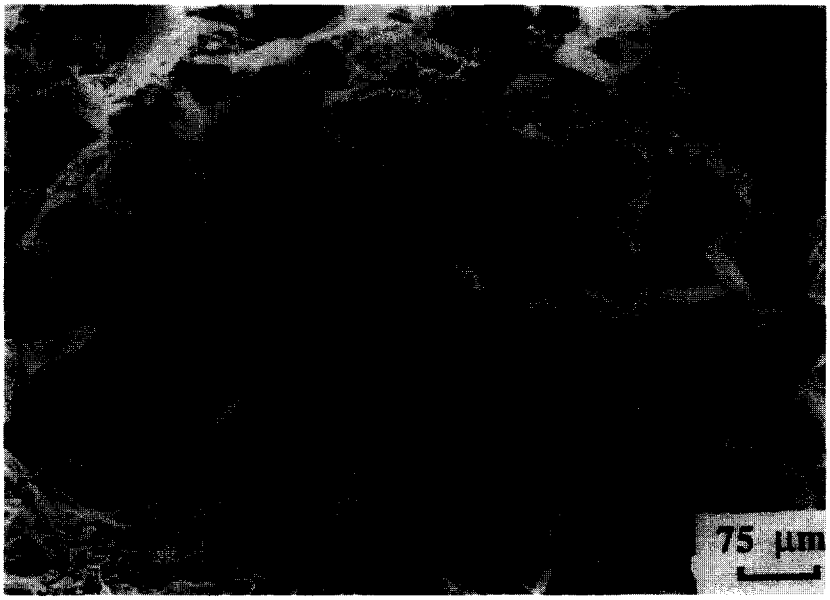


Fig. 5. Morphology of fractured surfaces for different annealing duration at 52°C of 23% cold worked Al6063: (a) before annealing, (b) annealed for 6 months, (c) annealed for 15 months, (d) annealed for 30 months.

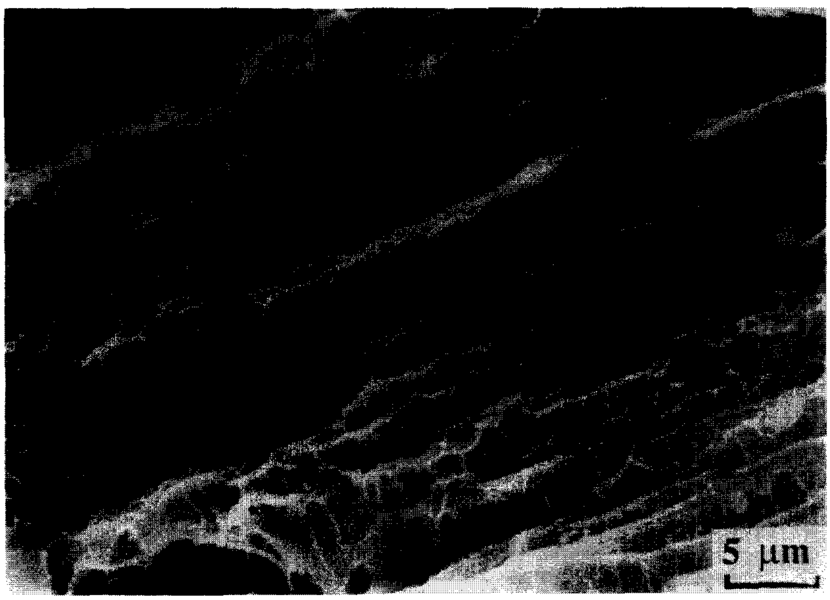
3.3. Irradiation impact on the internal microstructure

The irradiation of cold worked Al-6063 causes two main microstructural changes, as illustrated in the transmission electron images in Fig. 7.

There is a decrease in dislocation density with thermal neutron fluence. The microstructure of the unirradiated specimen (Fig. 7(b)) exhibited tangled dislocations, and any individual dislocation could not be distinguished from the rest. After irradiation, the dislocation density de-



a



b

Fig. 6. Secondary electron images illustrating the faceted nature of fracture at two magnifications of irradiated to 2.5×10^{25} thermal neutrons/m^2.

creases, and individual dislocations could be resolved. However, the dislocation density of the irradiated material is higher than in Al-6063 alloy annealed for 4 h at 500°C (Fig. 7(a)). In addition there is an increase in the Mg_2Si precipitation with irradiation fluence as evidenced by the increase in material strength. However, no individual pre-

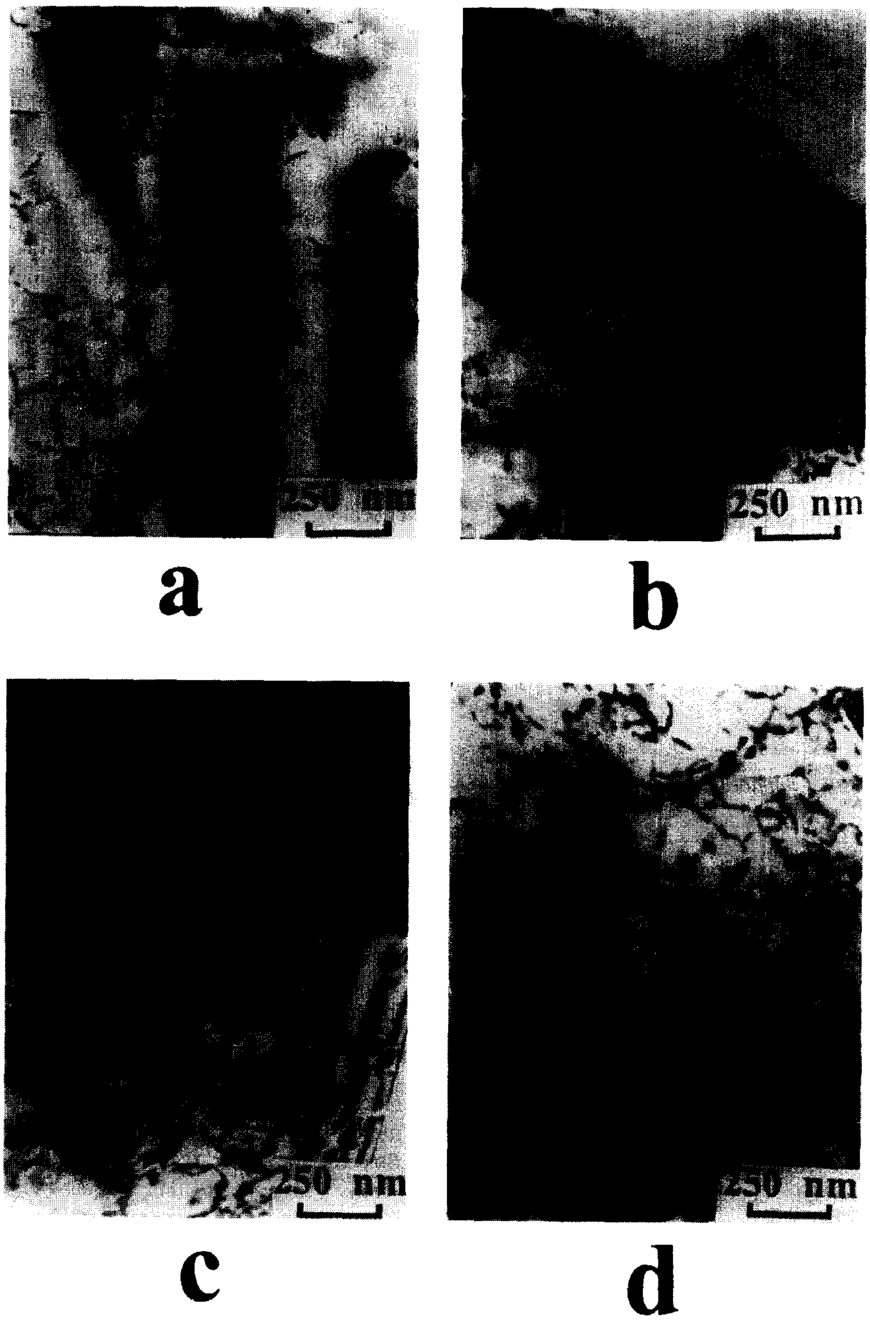


Fig. 7. Transmission electron images of 23% cold worked Al-6063; (a) annealed to 500°C for 4 h, (b) 23% cold worked unirradiated samples, (c) irradiated to 2.0×10^{25} thermal neutrons/m^2, (d) irradiated to 4.5×10^{25} thermal neutrons/m^2.

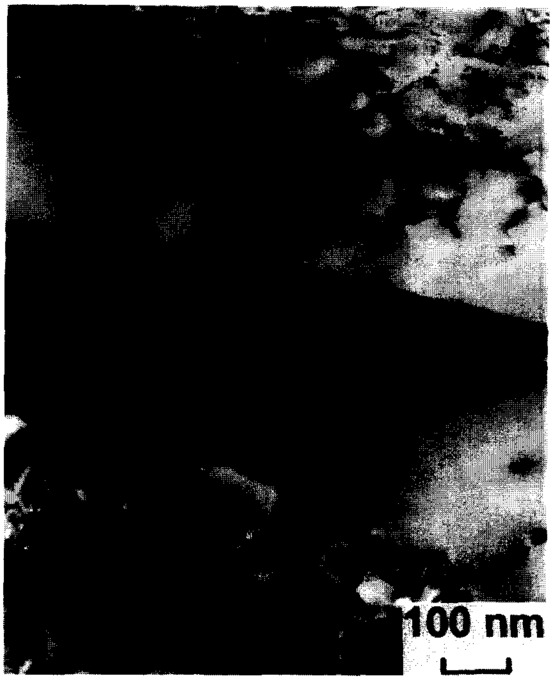


Fig. 8. Transmission electron image illustrating the onset of PFZ formation at grain boundaries.

cipitates could be observed. Irradiation also induces the beginning of the formation of precipitation free zones (PFZ) along both sides of the grain boundaries as demonstrated in Fig. 8. No voids could be observed up to the maximum thermal neutron fluxes used.

4. Discussion

4.1. Neutron irradiation impact on the mechanical properties

Under our irradiation conditions, neutron irradiation of cold worked Al-6063 caused increase in yield stress, as well as increase in ultimate tensile strength and elongation. The same behavior was observed after irradiation of cold-worked Al-A5 [17]. Generally, neutron irradiation induces changes in precipitation, dislocation density, and void formation, which are strengthening mechanisms, and thus affects the mechanical properties. In our studies, no voids could be observed up to the maximum thermal neutron fluence used. Rather, the irradiation caused a decrease in the dislocation density, indicating partial recovery, which increases elongation and decreases the ultimate tensile strength. At the same time, irradiation induced Mg_2Si precipitation in the Al-6063. It is a well known phenomenon that in Al–Si–Mg alloys, like the Al-6063, one begins to observe Mg_2Si precipitation only at full aging condition (T6) [20]. Therefore, it is not surprising that no precipitates could be observed. However, their influence on the mechanical properties was observed as evidenced by increase in the yield stress and ultimate tensile strength. The presence of precipitates is also evidenced by the beginning of formation of precipitation free zones (PFZ) near the grain boundary, as demonstrated in Fig. 8.

The ultimate tensile strength and elongation results from superposition of all effects mentioned above. It turns out that the positive contribution of the recovery to the elongation is dominant, and the dominant contribution to the increase in ultimate tensile strength is caused by precipitation hardening. Therefore, there is a net increase in elongation, along with the ultimate tensile strength dislocation recovery. Thus, irradiation induces increase in ultimate tensile strength which is accompanied by a decrease in elongation [21].

Theoretically, the contribution of each strengthening mechanism to the change in ultimate tensile strength may be calculated [14], as a function of the precipitation density, average precipitation size, dislocation density, and void size and density. In the case of Al-6063 alloy, these parameters are difficult to estimate. Only the general trend may be pointed out. A special technique needs to be developed, which will allow an exact count of dislocation and precipitation. This subject is under study in our laboratory.

Our results agree well with studies indicating that annealing of cold worked Al-1100 [15] and Al-6061 [1] may take place during irradiation.

4.2. The impact of prolonged aging on the mechanical properties

It appears that neutron irradiation and prolonged aging at low temperatures have similar effects on the mechanical properties (compare Fig. 1 and Table 1 to Fig. 2 and Table 2). Therefore, it is reasonable to assume that under our irradiation conditions, annealing and aging processes are the dominant factors influencing the microstructural and mechanical properties due to the temperature field during irradiation. However, it should be noted that during irradiation, Si is also formed from Al by the Al(n, γ)Si reaction. The presence of Si might increase the strengthening during irradiation by increasing the precipitation of Mg_2Si or of pure Si in the Al.

Since aging and annealing of the Al parts inside the reactor depend on the irradiation duration, the neutron flux might be a very important factor. A large thermal neutron flux (of the order of 10^{18}–10^{19} thermal neutrons/m^2) means a large Si creation rate, as well as a shorter exposure to the aging temperature. There appears to be an incubation period of 2 to 3 months before the mechanical properties changes induced by annealing could be observed (see Table 2). Thus, in the case of low fluxes (of the order of 10^{16}–10^{17} thermal neutrons/m^2), like in our case, the aging process during irradiation would be the major factor influencing the Al mechanical properties. In contrast, in a high flux isotope reactor (HFIR), where the irradiation duration required to obtain fluences of the order of 10^{25}–10^{26} thermal neutrons/m^2 is of the order of several months, irradiation effects like atom displacements, dislocation and voids formation, become dominant.

4.3. Impact of precipitation free zone on the fracture morphology

On first sight, there appears to be a conflict between two irradiation effects on the mechanical properties: the increase of uniform elongation with thermal neutron fluence which indicates an increase in ductility, and simultaneously a decrease in local area reduction with an increase in density of intergranular facets, which indicates a reduction of elongation.

It is well known that Al–Mg–Si alloy, like Al-6063, undergoes a sequence of precipitation: G.P. zones → θ'' → θ' → θ. The G.P. zones are actually regions locally Mg enriched, which then transform to a coherent θ'' precipitation which consisted of metastable Mg_2Al precipitates. However, it is difficult to detect these precipitates in Al–Mg–Si [20]. Only after a fully aged condition (T6) is reached, the precipitates become semi-coherent (θ' precipitate) when they become visible. Usually, in this process a precipitation-free zone (PFZ) around the grain boundary is created. There are two main reasons for creating a PFZ adjacent to a grain boundary:

(1) Mg diffusion into the grain boundary and the formation of Mg_2Si precipitation at the grain boundary.

(2) Enhanced vacancy diffusion to the grain boundary. Therefore, a denuded vacancy zone is created. In a region of small vacancy concentration it is hard to cause precipitation.

We have found evidence that PFZ begins to form during irradiation, as seen in Fig. 8. These PFZ regions are responsible for creation of the dimpled intergranular fracture, as well as the increase in the homogeneous elongation. Due to precipitation, the grains become stronger, therefore a higher stress is required for necking. Increase of the ultimate tensile stress, as was found by mechanical properties examination, would cause an increase in the uniform elongation. On the other hand, the PFZs are weaker than the grains; Therefore, at a certain stress they will rupture throughout the PFZ, but not necessarily at the grain boundary. As may be seen in a well developed PFZ, in an Al-6063 alloy at T6 aging conditions, needle precipitation protrudes into the PFZ, and is the source of the dimpled nature of the fracture. Actually, under fully aged conditions (T6), the entire cross section structure is intergranularly dimpled [22]. Therefore, it is hard to say that the presence of transgranular dimpled structure is typical of a brittle fracture. On the other hand, the local area reduction decreases due to ductility losses in the grains at the necking regions.

The creation of PFZ zones might be the reason why Alexander [23] found large crack propagation in spite of only minor changes in the fracture toughness. A similar explanation of the fracture morphology was given by Sturcken [13].

5. Summary and conclusions

The irradiation of cold-worked Al-6063 in a thermal reactor yields the following effects.

(1) Radiation induced recovery of cold worked samples, as evidenced by a decrease in dislocation density and an increase in elongation, as a function of the thermal neutron fluence.

(2) No voids could be observed up to a fluence of 4.5×10^{25} thermal neutrons/m^2 ($E < 0.625$ eV).

(3) A decrease in local area reduction at rupture as a function of fluence was observed.

(4) The major factor influencing the microstructure and mechanical properties during irradiation is prolonged thermal annealing caused by the temperature field generated during irradiation.

(5) Annealing of cold worked Al-6063 at 52°C for long periods of time revealed similar results, as observed for the impact of irradiated material. In both cases, the fracture morphology changes from a transgranular shear rupture to a mixture of transgranular dimpled shear rupture and intergranular dimpled rupture. The fraction of the intergranular rupture area increases as a function of the thermal neutron fluence.

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