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EFFECTS OF HIGH THERMAL NEUTRON FLUENCES ON TYPE 6061 ALUMINUM

REFERENCE: Weeks, J. R.; Czajkowski, C. J.; and Farrell, K.; "Effects of High Thermal Neutron Fluences on Type 6061 Aluminum," Effects of Radiation on Materials: 16th. International Symposium, ASTM STP 1175, Arvind S. Kumar, David S. Gelles, and Randy K. Nanstad, Editors, American Society for Testing and Materials, Philadelphia, 1993.

ABSTRACT: The control rod drive follower tubes of the High Flux Beam Reactor are constructed from precipitation-hardened 6061-T6 aluminum alloy and they operate in the high thermal neutron flux regions of the core. It is shown that large thermal neutron fluences up to -4×10^{23} n/cm² at 333K cause large increases in tensile strength and relatively modest decreases in tensile elongation while significantly reducing the notch impact toughness at room temperature. These changes are attributed to the development of a fine distribution of precipitates of amorphous silicon of which about 8% is produced radiogenically. A proposed role of thermal-to-fast flux ratio is discussed.

KEYWORDS: 6061-T6 alloy; Thermal neutrons; Tensile strength; Notch impact properties; Ductility; Fracture mode; Radiogenic silicon; Amorphous precipitates; Thermal-to-fast flux ratio.

In a previous publication [1] the nature of the flux distribution in the heavy water moderated High Flux Beam Reactor (HFBR) was discussed in detail. The reactor is built from 6061-T6 aluminum alloy (Al-1Mg-0.6Si) which derives its good strength from fine precipitates of Mg₂Si phase. Some of the major constituents that are exposed in the high flux regions of the core at a temperature of approximately 333°K are the eight control rod drive follower (CRDF) tubes. Since reactor startup in 1965 a surveillance program has been in place to monitor the effects of neutron exposure on removable components, including the CRDF tubes. Mechanical properties results obtained from the first three of the CRDF tubes, removed prior to 1984, were given in the earlier paper [1]. In the spring of 1989, CRDF A-2 was removed for examination. The "area of interest" on this tube (the portion exposed to the peak neutron fluxes) had received calculated fluences of 4.2×10^{23} neutrons/cm² (thermal) and 2.0×10^{22} neutron/cm² (E>0.1 MeV). The following tests were run on material from this CRDF tube:

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- tensile tests for elongation and tensile strength,
- miniature Charpy impact tests,
- notched tensile tests,
- scanning electron microscopy of the fracture surfaces,
- transmission electron microscopy of specimens from the Charpy end-pieces, and
- chemical analyses of specimens for silicon, to verify the calculated thermal fluences.

The purpose of this paper is to present the results of these tests, compare them with the other published data, and discuss what these observations tell us about the mechanisms of the effects of neutron radiation on type 6061-T6 aluminum.

EXPERIMENTAL METHODS

The CRDF tube is 3.25 cm OD with a wall thickness of 4.06 mm. The length of the high fluence region is about 35 cm. Figures 1 and 2 give the dimensions of the miniature tensile and impact specimens machined from this CRDF tube in the BNL hot cell. Tensile and impact specimens were also cut from areas of the tube exposed primarily in the reflector region above the core, where the fluxes are not well known, but are believed to be similar in thermal but much lower in fast ($E > 0.1$ MeV) neutrons than the area of interest adjacent to the core. To approximate a notch-tensile test, unfractured "Charpy" specimens, like those shown in Figure 2, were pulled on the tensile machine.

FIG. 1--Tensile test specimens cut from CRDF tube.

Dimensions In Millimeters

FIG. 2--Miniature impact specimens cut from CRDF tube.

Tension testing was performed in the hot cell using a remotely operated MTS machine calibrated against NIST standards in the hot cell by the manufacturer. Impact testing was performed in the hot cell by a Tinius Olsen impact tester (designed for testing plastics and adapted for aluminum alloys). Impact properties of the unirradiated aluminum alloys tested were measured on standard specimens by an independent testing laboratory using ASTM E399 as modified for aluminum alloys by ASTM B645 and B646. Fractographs were made of the fracture surfaces of the tensile, impact, and notched tensile specimens. Tensile and impact data were compared with those of unirradiated 6061-T6.

Selected areas from end pieces of the Charpy and tensile specimens were cut into wafers at BNL and sent to Oak Ridge National Laboratory (ORNL) for transmission electron microscopy (TEM). TEM examinations were done in a Philips CM12 microscope operating at 120 kv. Selected areas were examined by energy dispersive X-ray spectroscopy analysis. Wet chemical analyses for silicon were also performed at ORNL using one half of a broken impact or tensile specimen.

RESULTS

Mechanical Tests and Chemical Analyses Data

The data obtained from the mechanical tests, are given in Tables 1 and 2.

TABLE 1--Tensile test results on specimens cut from CRDF A-2.

Test Temperature = 23°C (296 K)		
Specimen	Tensile Strength MPa	Percent Elongation
<u>Unnotched bar (Fig. 1)</u>		
Unirradiated [1]	330 ± 4	10.3 ± 0.5
Irradiated (Outside Area of Interest)	641	8.5
Irradiated (Area of Interest)		
1	684	7.4
2	666	7.1
3	694	7.9
4	686	6.4
Average	682.4 ± 10.3	7.2 ± 0.75
<u>Notched bar (Fig. 2)</u>		<u>No. of Specimens</u>
Unirradiated	316 ± 27	2
Irradiated	200	1

TABLE 2--Impact test results for CRDF A-2.

Specimen	Temperature of Test °C	Impact Energy, Joules	Number of Specimens
Standard Unirradiated	-40	4.2 ± 0.1	4
Miniature* Unirradiated	23	2.21 ± 0.26	4
Miniature* Unirradiated	99	2.07 ± 0.07	4
Miniature* Irradiated (Outside Area of Interest)	23	0.34 ± 0	2
Miniature* Irradiated (Area of Interest)	24	0.34 ± 0.03	3
Miniature* Irradiated (Area of Interest)	99	0.31 ± 0.03	3

*See Fig. 2. for specimen dimensions

It is seen that irradiation raised the tensile strength of the unnotched tube material to twice that of the unirradiated material, and reduced the elongation by about 33%. In contrast, the tensile strength of a notched bar of the irradiated tube was only 63% of a notched, unirradiated tube. Notch impact energy was reduced from a value of 2.1 Joules for the unirradiated material at 23°C to 0.34 Joules for the irradiated tube.

In a thermal neutron environment aluminum atoms are transmuted to silicon, the fraction of which should be proportional to the thermal neutron fluence. Chemical analyses for silicon made on five pieces of the specimens used in the mechanical tests yielded values of 6.75 to 9.35 wt %, with an average of 8.0 wt %. Allowing for 0.6 wt % Si in the unirradiated condition, the average increase in silicon level corresponds to a thermal fluence of $4.0 \pm 0.6 \times 10^{23}$ n/cm², using the measurements of Sturcken, et al. [2]. This fluence agrees with an estimate of 4.2×10^{23} n/cm² made by Tichler [3] using the calculated spectrum at the CRDF tube. The estimated fast fluence (>0.1 MeV) at the area of interest is 2×10^{22} n/cm².

Figure 3 compares the tensile strength and percent elongation results with those obtained from CRDF's removed earlier in the operating life of the reactor. The error bars represent the actual range of the data obtained from a given CRDF. In accordance with the conclusions of our earlier paper [1], the results are plotted as a function of thermal fluence; however, since these data were all obtained at a constant thermal to fast neutron ratio of approximately 20/1, a plot against fast ($E > 0.1$ MeV) fluence would show a similar trend.

FIG. 3--Tensile strength and total percent elongation data from irradiated CRDF tubes.

Results of Fractographic (SEM) and Microstructural (TEM) Investigations

Typical fractographs of the failure surfaces of tensile and impact specimens from the area of interest of CRDF A-2 are shown in Figures 4 and 5, where they are compared with fractographs of unirradiated specimens of 6061-T6 aluminum broken by the same techniques. Irradiation decreased the percentage of the fracture surface consisting of ductile rupture and increased the percentage of the fracture surface showing evidence of brittle transgranular separation and intergranular fracture. As both the tensile and impact mechanical data and SEM fractographs show, the material retains some residual ductility and ductile rupture behavior in the highly irradiated state.

a. Unirradiated.

b. Irradiated (CRDF A-2)

FIG. 4--SEM fractograph of tensile specimens.

a. Unirradiated

b. Irradiated (CRDF A-2)

FIG. 5--Fractographs of impact specimens.

TEM photographs of thin sections of typical areas of unirradiated and irradiated material are shown in Fig. 6. The original, acicular Mg_2Si precipitate (Fig. 6a), which is responsible for age-hardening of the unirradiated alloy, was destroyed by the irradiation and was replaced by a high concentration of spherical particles about 8 nm diameter (Fig. 6b) consisting primarily of silicon. These spherical particles displayed diffuse diffraction rings rather than discrete spots, implying that the particles are amorphous. There were no irradiation produced voids. Examination of grain boundary regions revealed that the spherical precipitates approached to about 10 nm from the boundaries. On the boundaries there were large non-contiguous, flakes, or islands, of silicon-rich phase that protruded from the foils (Fig. 7). These flakes displayed no diffraction spots. They occupied less than one-fifth of the grain boundary area.

FIG. 6--Microstructures of (a) unirradiated 6061-T6 alloy, and (b) CRDF A-2 tube after irradiation.

FIG. 7--Irradiated CRDF A-2 tube showing large flakes of silicon-rich phase at the grain boundaries.

DISCUSSION

From Fig. 3 it appears that the tensile strength of the CRDF tubes is continuing to increase at decreasing rate with time and fluence and is approximately proportional to the square root of the fluence,

$$T.S. = 50 + 24.5 (\phi_{th})^{1/2} \times 10^{-23} \text{ in ksi, or} \quad (1)$$

$$= 345 + 170 (\phi_{th})^{1/2} \times 10^{-23} \text{ MPa} \quad (1a)$$

At high fluences the ductility continues to drop slightly, although the minimum of the scatter band has remained constant (at approximately 6.5%) since the second CRDF was examined in 1976 (at $\phi_{th} = 1.7 \times 10^{23} \text{ n/cm}^2$). The exceptionally high ductility results obtained in June 1969 (at $\phi_{th} = 4 \times 10^{22} \text{ n/cm}^2$) remain an anomaly, since the tensile strengths measured on the same specimens do not show a corresponding weakening. The original records from 1969 were reexamined and a small (approximately 30%) correction (for gauge length) made, but no satisfactory explanation can yet be made for this result. It is not regarded as an example of radiation softening of the type reported by Ismail [4] in precipitation-hardened aluminum because strength was increased, not decreased.

Attempts to fit ductility loss to the remaining four points (including the results for unirradiated material) led to the equation,

$$\%E = 10.7 - (0.69) (\phi_{th} \times 10^{-23}), \quad (2)$$

which, along with the more conservative approximation of a line drawn through the averages of only the 1984 and 1989 data,

$$\%E = 13.5 - (1.5) (\phi_{th} \times 10^{-23}), \quad (3)$$

are being used to predict future behavior and to provide guidance for selecting a conservative basis for removing the next CRDF for surveillance measurements.

Although it is clear from the unnotched tensile data that the irradiated alloy retains a respectable fraction of its original ductility, it is also evident from the notch tensile data and the notch impact data that the irradiation has raised the notch sensitivity and strain rate sensitivity of the material. The notch impact energy is seen to have been reduced to approximately 1/6 of its pre-irradiation value; these results were surprisingly reproducible. Values for K_{IC} were estimated by Kassir [5] from these and the notched tensile data; his results are shown in Table 3. The mean estimated values he obtained were $8.7 \text{ MPa(m)}^{1/2}$ from the impact data and 7.7 to $8.8 \text{ MPa(m)}^{1/2}$ from the notch tensile data. These are quite similar from the two sets of experimental measurements, and are probably within the experimental accuracy.

TABLE 3--Estimated Values of K_{IC} for Irradiated 6061-T6 Aluminum from CRDF A-2.

Sample	K_{IC} , MPa(m) ^{1/2}
Unirradiated 6061-T6	21.75
Irradiated (CRDF A-2) notch impact data	8.72 ± 0.8
From notch tensile data	7.7 - 8.8

These notch toughness data are the first such data for heavily irradiated 6061-T6 alloy. There are many smooth bar tensile data but toughness measurements are a rarity in irradiated aluminum alloys. Only Albertin and DeMastry [6] have explored the fracture toughness behavior of irradiated 6061-T651 alloy, but they were primarily interested in cryogenic conditions. Their irradiations were made at 77K to fluences up to only 1×10^{19} fast n/cm², and they found a loss of fracture toughness at 77K but not at higher temperatures. DeVries and Cundy [7] have examined 5154 aluminum (Al-3.5 wt % Mg) from the old core box of the HFR at Petten exposed to fluences of 6.9×10^{22} n/cm², fast and 7.5×10^{22} n/cm², thermal. Hardness and tensile strength were increased, and fracture toughness was decreased, with increasing level of radiogenic silicon. The lowest value of pseudo K_{IC} at room temperature was 17.7 MPa(m)^{1/2}.

The changes in mechanical properties observed in the CRDF-A2 tube are no doubt due to the radiation-induced changes in microstructural details in the alloy. The original Mg₂Si precipitates were almost erased and were superseded by a fine dispersion of silicon-rich particles. A comparison of these particles with those observed earlier [8] in 6061-T6 alloy irradiated to similar thermal fluences in the light water, High Flux Isotope Reactor (HFIR) at Oak Ridge revealed them to be much smaller and more numerous. The mean diameter of the particles in the HFIR material was 23 nm, vs 8nm for the CRDF tube. It was also noted that there were no voids formed in the CRDF tube whereas there were many voids in the HFIR material. These differences in microstructural features are attributed to differences in neutron exposure conditions. In a previous paper [1] Weeks, et. al., observed that there were significant differences in the effects of irradiation in HFIR, and in two different areas of HFBR. They suggested that the primary effects were due to the thermal neutrons, but that higher fast fluxes caused less pronounced changes in mechanical properties at the same thermal fluence. Although the irradiation temperatures and thermal neutron fluences were similar in both cases, the neutron fluxes were higher in the HFIR and the fast fluence for the HFIR material was about ten times higher than for the CRDF tube. To account for the dissimilar mechanical and microstructural responses from the two different reactors, Farrell [9] has proposed that the ratio of the thermal to the fast fluxes (or fluences) is a potent factor. A high ratio, as in the CRDF tubes, will induce a high concentration of insoluble silicon per unit atomic displacement and, hence, a high degree of supersaturation and profuse nucleation of silicon-rich precipitates. On the other hand, a low thermal-to-fast ratio, i.e., a relatively high fast flux or fluence as in the HFIR, will provide more point defects per silicon atom thereby encouraging atomic transport and raising the effective solubility of

the silicon, resulting in a coarser distribution of the silicon precipitates. Coarsening should be further aided by cascade dissolution of the precipitates, which will increase with decreasing thermal-to-fast flux ratio. Since improvement in mechanical strength will be less for coarser microstructures, the strength should be dependent on thermal-to-fast flux ratio, as well as on the thermal fluence. This is demonstrated in Fig. 8, where data that were seemingly in conflict in the past are now rationalized via their thermal-to-fast flux ratios. Another consequence of a low thermal-to-fast flux ratio is that the fast flux will generate more helium and hydrogen per unit displacement, promoting void formation at high fluences, hence explaining the presence of voids in the HFIR material and their absence in the CRDF tube.

FIG. 8--Effect of thermal to fast flux ratio on the mechanical properties of Type 6061-T6 aluminum.

The retention of about 6% tensile elongation in this CRDF tube confirms the trend of diminishing rate of loss of ductility in 6061-T6 alloy at high fluences recognized earlier [1,8]. This reduced loss of ductility is presumably due to the establishment of the relatively stable radiation-induced microstructure dominated by the spherical, silicon-rich precipitates. It is not known whether the amorphous nature of the precipitate phase plays a part. It seems unlikely that continued irradiation will nucleate additional precipitate particles. Rather, further irradiation will probably simply increase the size of the existing particles. In which case, the slow rate of ductility loss will be maintained. However, the possibility of interference by the flakes of silicon-rich precipitates on the grain boundaries can not be ignored. A scenario in which the fracture path shifts to a predominantly intergranular route must be considered but much will depend on whether the flakes grow by thickening or by increasing their area. More microstructural information is needed to investigate such issues.

CONCLUSION

1. Following irradiation to a fast fluence of 2×10^{22} n/cm² (E>0.1 MeV) and a thermal fluence of 4.2×10^{23} n/cm² at 333K the 6061-T6 alloy CRDF A-2 tube contained 8 wt % radiogenic silicon.
2. In room temperature mechanical tests the tube displayed a twofold increase in mechanical strength, a reduction in tensile elongation of about 33% to a value of 7%, and a 37% reduction in notched tensile strength.
3. The notch impact energy of non-standard specimens was reduced to 1/6 of the unirradiated value at room temperature.
4. Fractographic examination showed a mixture of ductile and brittle transgranular separation.
5. Transmission electron microscopy revealed that the original Mg₂Si precipitates were replaced with a high concentration of 8 nm diameter, amorphous particles rich in silicon.
6. The changes in mechanical properties are attributed to the development of the silicon-rich precipitates; an influence of thermal-to-fast flux (or fluence) is recognized in which higher ratios cause finer distribution of silicon precipitates and higher strengths.
7. Consideration of earlier data in terms of thermal-to-fast flux ratios allows a rational representation of previously disparate data.

ACKNOWLEDGEMENTS:

This work was performed under the auspices of the U.S. Department of Energy, Office of Energy Research. The flux calculations and personal encouragement of P. R. Tichler of the Brookhaven National Laboratory (BNL) Reactor Division, the hot cell work of M. H. Schuster, T. Roberts, and L. W. Milian, the stress analysis of M. K. Kassir of the Department of Nuclear Energy, BNL, and the silicon analyses of J. L. Botts, Oak Ridge National Laboratory (ORNL) are all gratefully acknowledged.

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LIST OF NEGATIVES/FIGURES FOR J. WEEKS' PAPER ENTITLED: "Effects of High Thermal Neutron Fluences on Type 6061 Aluminum"

Figure 1 - Tensile test specimens cut from CRDF tube.

Figure 2 - Miniature impact specimens cut from CRDF tube.

Figure 3 - Tensile strength and total percent elongation data from irradiated CRDF tubes.

Figure 4 (Negative #'s: a) 5-346-92 and b) 5-344-92) - SEM fractograph of tensile specimens.

Figure 5 (Negative #'s: a) 5-347-92 and b) 5-345-92) - Fractographs of impact specimens.

Figure 6 (Negative #'s: a) 6-328-92 and b) 10-70-91) - Microstructures of (a) unirradiated 6061-T6 alloy, and (b) CRDF A-2 tube after irradiation.

Figure 7 (Negative # 10-72-91 [right half]) - Irradiated CRDF A-2 tube showing large flakes of silicon-rich phase at the grain boundaries.

Figure 8 - Effect of thermal to fast flux ratio on the mechanical properties of Type 6061-T6 aluminum.

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