POSTIRRADIATION EXAMINATION AND TESTING OF THE ORR N_F TRAY

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K. Farrell, E. L. Long, Jr., R. T. King, and A. Jostsons*

*On attachment from Australian Atomic Energy Commission Research Establishment, Lucas Heights, New South Wales.

JUNE 1973

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ABSTRACT

An Oak Ridge Research Reactor (ORR) core lattice component, the N_F tray, became permanently bowed during reactor service. This tray, which was fabricated from recrystallized 1100 aluminum, reached maximum neutron fluences of about 3.0×10^{22} neutrons/cm² (thermal) and 3.2×10^{22} neutrons/cm² ($E > 0.1$ MeV) at an operating temperature of 40 to 50°C. Microstructural examination of the tray revealed voids, many dislocations and a fine precipitate of transmutation-produced silicon. These defects markedly increased the hardness and strength of the 1100 aluminum and also caused a significant loss of ductility. At test temperatures above 100°C ductility was further degenerated by helium embrittlement that resulted in premature intergranular fracture.

The voids made the tray swell by about 1.5% at its center. This swelling was unequal on opposite sides of the tray and thereby resulted in bowing. The degree of bowing was calculated accurately from the swelling data. It is recommended that bowing can be minimized by intermittent annealing at 300 to 350°C whenever the first signs of bowing appear, or, better still, by fabricating the N_F tray from 6061 aluminum alloy in the precipitation-hardened T6 condition.

INTRODUCTION

The Oak Ridge Research Reactor (ORR) is used primarily for experimental studies and for the production of radioisotopes. The reactor is water cooled and is built largely from aluminum components. In 1967, S. S. Hurt III of the Reactor Operations Division of ORNL reported¹ bowing of aluminum components in the ORR lattice, or core. The bowing was apparently caused by irradiation damage and the extent of bowing

*On attachment from Australian Atomic Energy Commission Research Establishment, Lucas Heights, New South Wales.

increased with increasing neutron fluence. The component displaying the greatest distortion was an aluminum box identified as the N_F tray. This tray has now been examined to determine the cause(s) of the bowing.

Most of the observations and measurements described in this report have been made available to interested parties through quarterly progress reports, open literature publications, and informal meetings. This report is the formal compilation and assessment of the data. For easier reading it is divided into sections describing, respectively, the nature of the tray and its irradiation history, swelling and microstructural data, calculation of bowing, effects of postirradiation annealing, and mechanical properties. A further short section summarizes the general observations and conclusions, and contains some recommendations for prolonging the useful lifetime of the N_F tray.

1. THE NATURE OF THE N_F TRAY AND ITS IRRADIATION HISTORY

1.1 Description of the N_F Tray

The N_F tray is a long, narrow, aluminum box made by brazing together two flat plates and two curved plates to produce the same general configuration as an ORR fuel element which it replaces in a core lattice position. Brazing is done at almost 600°C using an Al-Si eutectic brazing alloy. A major function of the tray is to hold targets for the production of radioisotopes. The tray is located in the fast neutron facility in the ORR lattice, as illustrated schematically in Fig. 1. The compass directions N, E, S, and W are used for orientation and for identification of the four long sides of the tray. The maximum neutron flux impinges on the tray from a SE direction.

The dimensions of the tray are available in detail in ORNL Engineering Drawings Nos. D-29701 and D-29702. Figure 2 shows schematically that portion of the tray that we received for examination. A tapered section, 25 cm long, of the tray that lay below the bottom of the fuel plates in the core lattice and was used to seat the tray properly in its site was sawed off and discarded prior to examination of the tray. In this report, distances along the length of the tray are referred to the new "bottom" of

ORNL-DWG 67-7174R

* BERYLLIUM-CADMIUM AND ALUMINUM-CADMIUM SHIM RODS

F = FUEL

E = EXPERIMENT

Be = BERYLLIUM

SR = SHIM ROD

N_F = FAST NEUTRON FACILITY

Fig. 1. Typical ORR Core Lattice Viewed from the Top, Showing Location of the N_F Tray with Respect to Fuel and Other Components.

Fig. 2. Shape and Dimensions of the Portion of N_F Tray Examined in This Study. Note the orientation and type of tensile specimen.

the tray which corresponds to a point about 1.2 cm below the bottom of the fuel plates and 25 cm above the original bottom of the tray. The top of the tray corresponds to a point about 1.2 cm above the top of the fuel plates. The tray is not restrained at its top end.

1.2 Chemical Composition and Original Condition of the Tray

The tray was fabricated from type 1100 aluminum plate of unknown origin for which no certified chemical analysis is available. Type 1100 aluminum is simply commercial quality, 99% aluminum with about 1% of Si + Fe as major impurities. The high temperature achieved during brazing of the plates would undoubtedly ensure that the finished tray was in a recrystallized condition. This was confirmed by transmission electron microscopy examination of specimens from low fluence sections which showed equiaxed grains and no evidence of retained plastic deformation.

1.3 Bowing Measurements

The degree of bowing or distortion in the N_F tray after irradiation was measured by Hurt¹ using a dial gauge micrometer calibrated against a reference plate which was flat to within ± 0.0013 cm. The results are shown in Fig. 3. In general the tray was bent like a four-sided banana with the center of curvature beyond the NW edge. The sign of the deflection refers simply to whether a convex surface (positive) or a concave surface (negative) was presented to the dial gauge as the tray was rotated to measure each side. The important features of Fig. 3 are the absolute magnitudes of the deflections, the general shape of the curves and the axial positions of the deflection maxima. These features will be referred to subsequently to draw attention to the relations between bowing and fast neutron fluence and swelling.

1.4 Neutron Flux and Fluence Distribution in the N_F Tray

The neutron environment in the ORR changes with time during a normal operating cycle and is also influenced by the arrangement of fuel and of experimental assemblies in the lattice. Consequently, the N_F tray is expected to have been exposed to considerable radial and axial flux gradients which make it difficult to obtain precise flux and fluence data. A number of studies of neutron flux distribution have been made

Fig. 3. Bowing Measurements on the N_F Tray, After Hurt.¹

on the N_F tray but only two^{2,3} of these allow a partitioning of the flux into fast ($E > 0.1$ MeV) and thermal ($E < 0.0253$ eV) in which terms it is easier to understand and discuss neutron damage.

O'Brien² measured the fast and thermal neutron flux variations at the maximum axial flux position, across the S and E sides of the tray. These measurements were made over one reactor cycle of about 22 days. Subsequently Kam³ repeated these measurements in greater detail to give the flux variations across all sides of the N_F tray at all axial locations. Kam's measurements were based on only a 16-hr irradiation at the beginning of a reactor cycle and hence his axial flux profiles are not representative of that expected after a full reactor cycle since movement of the control rods, particularly at the beginning of a cycle, cause an axial shift in the maximum flux. The peak axial flux position in Kam's study is depressed towards the bottom of the N_F tray.

Both O'Brien and Kam studied the maximum fast and thermal fluxes at the SE corner of the N_F tray. In order to compare the magnitudes of these fluxes, we have recalculated O'Brien and Kam's data using High Flux Isotope Reactor (HFIR) neutron spectrum averaged reaction cross sections for their flux monitors in Table 1. The HFIR spectrum is believed⁴ to describe adequately the ORR spectrum in a lattice position for neutron energies in excess of 0.1 MeV. The positions of the flux monitors across the N_F tray at the maximum axial flux position are illustrated in Fig. 4, and the recalculated flux values are listed in Table 2.

Table 1. Reaction Cross Sections Used in ORR N_F Tray Neutron Flux Calculations

Reaction	$\bar{\sigma}$, $E > 0.1$ MeV (barns)
$^{58}\text{Ni}(n,p)^{58}\text{Co}$	0.074
$^{54}\text{Fe}(n,p)^{54}\text{Mn}$	0.050
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	0.00045
$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	37.0 (thermal neutrons)

ORNL-DWG 71-14070

Fig. 4. Location of Flux Monitors at the Maximum Axial Flux Position.

Table 2. Summary of Fast ($E > 0.1$ MeV) and Thermal ($E < 0.0253$ eV)
Neutron Flux Measurements at the Maximum Axial Flux
Position for 30 MW Operation

Position	Fast Flux ($\times 10^{14}$ n/cm ² /sec)					Thermal Flux ($\times 10^{14}$ n/cm ² /sec)		
	O'Brien Fe	Fe	Kam Ni	Al	Selected Value	O'Brien	Kam	Selected Value
1)	1.26	1.56	1.27	1.24	1.25	1.62	1.20	1.4
2)	1.21	1.84	1.46	1.46	1.45	1.77	1.25	1.5
3) E	1.62	2.04	1.62	1.75	1.65	1.92	1.38	1.6
4)	1.77	2.19	1.79	1.94	1.85	1.89	1.45	1.7
5)	1.89	2.40	1.92	1.94	1.90	2.09	1.45	1.8
6) S	1.48	1.56	1.39	1.34	1.40	1.85	1.25	1.6
7)	1.14	1.21	0.95	0.90	1.05	1.60	1.21	1.4
8)		1.12	0.82	0.85	0.95		1.26	1.4
9) W		0.95	0.77	0.73	0.80		1.21	1.4
10)		0.80	0.58	0.63	0.65		1.16	1.3
11) N		0.73	0.92	0.56	0.75		1.14	1.3
12)		1.12	0.87	0.75	0.90		1.14	1.3

In the absence of reliable axial flux data for a full reactor cycle for the A7 position in the ORR core, we use the values obtained by O'Brien for the C3 partial fuel element position. The normalized axial thermal and fast neutron flux profiles obtained this way for the N_F tray are plotted in Fig. 5. While there is obviously some uncertainty in the derivation of these profiles they are considered to be the best approximation of the average fluxes experienced by the tray during its lifetime in the reactor. The specific flux value for any position on the tray is obtained from the product of the relevant flux value in Table 2 and the normalized axial flux distribution in Fig. 5, assuming a constant radial flux distribution at all axial positions. The tray was irradiated to an integrated power of $67,550 \text{ Mwd}^1$ in the ORR core, mostly at 30 MW power, for a total time of 1.95×10^8 sec. The neutron fluences, or integrated neutron fluxes over this period, averaged across each of the four sides of the tray at the axial maximum flux region, are given in Table 3.

Fig. 5. Fast and Thermal Flux Profiles Along the Length of the N_F Tray, Normalized to the Maximum Axial Fluxes.

Table 1. Average Fast and Thermal Neutron Fluences
in the ORR N_F Tray at Maximum Axial Flux Position

Side	Fast Fluence ($n/cm^2 > 0.1$ MeV)	Thermal Fluence ($n/cm^2 < 0.0253$ eV)
East	3.2×10^{22}	3.1×10^{22}
South	2.8×10^{22}	3.1×10^{22}
West	1.6×10^{22}	2.6×10^{22}
North	1.9×10^{22}	2.5×10^{22}

1.5 Irradiation Temperature

During reactor operation, the inner and outer surfaces of the tray were exposed to flowing coolant water at $38 \pm 5^\circ\text{C}$ at the inlet (top) end to $46 \pm 5^\circ\text{C}$ at the peak flux region and $54 \pm 5^\circ\text{C}$ at the outlet end. Allowing for the effect of oxide films on thermal conductivity and the nuclear heating rate in aluminum it is concluded that the N_F tray operated within 5°C of the coolant temperature, i.e., roughly 40 to 50°C .

1.6 Transmutation Products

Transmutation products affect the physical and mechanical properties of an irradiated metal. They are also a useful measure of the neutron fluences, provided reliable, spectrum-averaged neutron absorption cross-sections are available for the transmutation reactions.

The major transmutation products in aluminum are the gases hydrogen and helium resulting from fast neutron (n,p) and (n, α) reactions with aluminum, and silicon produced from the $^{27}\text{Al}(n,\gamma)$ reaction with thermal neutrons followed by a β -decay to ^{28}Si . Past experience has shown that chemical analyses for transmutation-produced hydrogen are not satisfactory because of interference from hydrated oxide films that persist on aluminum, so no attempt was made to analyze for hydrogen. However, based on the spectrum-averaged cross-section⁵ for the $\text{Al}(n,p)$ reaction for $E > 0.1$ MeV of 2.75 mb, we would expect 82 at. ppm of hydrogen to be created by a fast fluence of 3×10^{22} neutrons/cm².

Helium analyses were made in a high-sensitivity mass spectrometer calibrated against a known volume of ^3He . The results are presented in Table 4. The spectrum-averaged cross-section for the $\text{Al}(n,\alpha)$ reaction for $E > 0.1$ MeV is 0.469 mb (ref. 5) which would give 14 at. ppm of helium for a fast fluence of 3×10^{22} neutrons/cm 2 . Since a small quantity of the measured helium may have derived from impurities in the 1100 aluminum the close agreement between the calculated and measured helium concentrations gives confidence in the estimated fast neutron fluence values. Analyses of specimens after postirradiation annealing for periods up to 20 hr at 300 and 560°C did not reveal any loss of helium during the anneals.

Table 4. Transmutation Products in the N_F Tray

Specimen	Neutron Fluence ($\times 10^{22}/\text{cm}^2$)	Measured Amount (at. ppm)
C10	2.8 (fast)	14.0 He
B62	3.2 (fast)	18.3 He
B65 (postirradiation annealed 20 hr at 300°C)	3.0 (fast)	18.3 He
B72 (postirradiation annealed 20 hr at 560°C)	2.6 (fast)	15.9 He
B49	Control (< 0.1 thermal)	3560 Si
B36	1.01 (thermal)	5480 Si
B79	1.97 (thermal)	7200 Si
B97	2.01 (thermal)	7200 Si
B73	2.63 (thermal)	8940 Si

The results of chemical analyses for silicon are also summarized in Table 4. The silicon content of specimen B49, which was irradiated to a thermal fluence of less than 1×10^{21} neutrons/cm², is taken as the reference value for the unirradiated material. It can then be seen that on the average each increment of thermal fluence of 1×10^{22} neutrons/cm² introduces about 1900 at. ppm silicon (~ 0.2 wt %). This indicates an effective cross section for the silicon reaction of 190 mb. But the calculated, spectrum-averaged cross-section for $E < 0.0253$ eV is 235 mb.⁶ Assuming the latter cross-section to be correct, the estimated thermal fluences are too high. The error could arise from the silicon analysis of specimen B49 or from the fact that we derived the thermal fluences by averaging the flux measurements of O'Brien and Kam which show significant differences. Multiplication of the selected thermal flux values in Table 2 by 0.87 would give good agreement with the thermal flux data derived from Kam's measurements and would lead to an almost exact agreement between the measured and calculated silicon reaction cross sections. Thus, the thermal fluences may be overestimated by about 15%. Because of the uncertainty, however, we will adhere to our estimated thermal fluences throughout this report.

1.7 Discussion of the Nature of the N_F Tray and Its Irradiation History

This discussion permits us to draw attention to several points that will be important in digesting and assessing the irradiation-induced changes in the physical and mechanical properties described in subsequent sections of this report.

The first of these points is that the general shape of the axial fast flux distribution (Fig. 5) is very similar to that of the measured bowing along the length of the tray (Fig. 3), suggesting a strong connection between the two. This observation supports Hurt's original contention that bowing increases with increasing irradiation, but is more definitive in that it indicates that there is a better correlation of bowing with the fast flux than with the thermal flux. This implies an association of bowing with displacement damage rather than with, say, silicon production from the thermal neutrons.

The second point centers on the transmutation-produced silicon. This silicon is readily distinguishable from any silicon present in the aluminum prior to irradiation. It has been demonstrated⁷ that preexisting silicon in 1100 aluminum is combined with iron and aluminum in the form of large, easily recognizable inclusions that are not responsive to thermal treatment. The silicon introduced by transmutation takes the form of a fine precipitate that coarsens during annealing and which has been identified by electron diffraction, microprobe analyses and chemical analyses as elemental silicon.⁷ Silicon is almost insoluble in aluminum at the irradiation temperature,⁸ and the density of the silicon precipitate is only 2.3293 g/cm^3 (ref. 9) which is less than the density of aluminum, 2.71 g/cm^3 (ref. 10), so it causes some swelling of the aluminum. A simple rule of mixtures indicates that each 1 wt % silicon in the form of precipitate has a volume fraction of 0.0116 and causes 0.16% swelling. In the next section we will see that this swelling is small in comparison with that caused by radiation-induced voids. But the silicon has considerable effects on the mechanical properties of the aluminum.

The third point is that the irradiated tray contains significant quantities of transmutation-produced gases in addition to any gases that may be present before irradiation. At the irradiation temperature these gases do not form visible gas bubbles so they are not the cause of the bowing. At elevated temperatures, however, the gases do form bubbles that cause swelling, and helium in particular can cause premature intergranular fracture and embrittlement.

1.8 Summary of the Nature of the N_F Tray and Its Irradiation History

1. The N_F tray was fabricated from 1100 grade aluminum originally in a recrystallized condition.
2. The tray was irradiated in contact with the coolant water at 40 to 50°C to estimated maximum fluences at the mid-point of its SE edge of about $3.0 \times 10^{22}/\text{cm}^2$ ($E > 0.1 \text{ MeV}$) and somewhat less than 3.1×10^{22} neutrons/ cm^2 ($E < 0.0253 \text{ eV}$).
3. The axial fast neutron flux distribution along the tray has the same shape as that of the bowing measured on the tray, implying an association of bowing with displacement damage.
4. Significant quantities of the transmutation products hydrogen, helium and silicon were generated in the tray.

2. MICROSTRUCTURE AND SWELLING MEASUREMENTS

2.1 Microstructure

The effects of neutron irradiation on 1100 grade aluminum from this N_F tray have been described qualitatively by Stiegler and co-workers.¹¹ In addition to radiation-induced voids, they observed a high density of randomly distributed dislocations and a fine precipitate of transmutation-produced silicon. We have now extended these observations and have made quantitative measures of the various radiation-induced defects.

We did not find voids in the N_F tray for fast fluences less than 7×10^{21} neutrons/cm². At a fluence of 7×10^{21} neutrons/cm² a few voids were present near to grain boundaries. Immediately adjacent to the grain boundaries were zones about 0.1 μm wide on each side of the grain boundaries that were free of damage structure (Fig. 6). At fluences greater than 7×10^{21} neutrons/cm² regions abutting these denuded zones contained a significantly higher concentration of voids and the average void diameter was larger than that in regions well removed from grain boundaries (Fig. 6). At low fluences the heterogeneous distribution of the voids prevented meaningful measurements of void concentrations but at a fast fluence of about 1×10^{22} neutrons/cm² the grain interiors began to fill with voids and it was possible to measure a mean concentration of $1 \times 10^{13}/\text{cm}^3$ of average diameter ~ 400 Å. Eventually, at higher fluences the void distribution within the grains became more uniform, Fig. 7, and the difference between the matrix and near grain boundary regions, except for the void-denuded zones, was less obvious. The void size distributions in regions removed from grain boundaries are shown in Fig. 8. The data in Fig. 8 represent about 1000 voids for each condition. Measurements were made with the aid of a Zeiss particle size analyzer on photomicrographs at a magnification of 10^5 . Stereo electron microscopy was used to determine the foil thickness which usually ranged from 2000 to 5000 Å. The voids were predominantly {100} cubes frequently truncated on {111} planes.

Fig. 6. Grain Boundary Denuded Zones and Enhanced Void Formation Near Grain Boundary in Specimen Irradiated to 2.1×10^{22} neutrons/cm² ($E > 0.1$ MeV). Arrow indicates the grain boundary.

Fig. 7. Voids and Transmutation-Produced Silicon Precipitate in Specimen Irradiated to 3.2×10^{22} neutrons/cm² ($E > 0.1$ MeV). Arrow indicates a grain boundary.

Fig. 8. Variation of Void Size and Concentration with Fast Neutron Fluence. The size interval is 50 $\text{\AA}$.

It has been shown previously¹² that void nucleation during neutron irradiation is enhanced in aluminum injected with protons prior to irradiation, at least in material neutron irradiated at 150°C. Since the N_F tray operated in contact with the reactor coolant water, some pick-up of hydrogen may be expected from oxidation of aluminum in water and from proton recoil resulting from elastic scattering of neutrons in water. Consequently, if there was significant hydrogen contamination at the specimen surfaces it may have resulted in a hydrogen concentration gradient through the thickness of the tray which may cause changes in the concentration and mean size of voids through the thickness of the wall. However, the concentrations and sizes of voids measured at different depths at one location on the wall of the tray were essentially identical (Fig. 9) indicating a uniform distribution of voids through the wall thickness and implying an absence of hydrogen gradients.

The tray contained a high concentration of interconnected dislocations, Fig. 10, presumably formed from the precipitation of irradiation-produced point defects. Only a few small dislocation loops were observed in the higher fluence specimens but loops were more readily apparent at the lower fluences. From this observation and from the observation of Stiegler *et al.*,¹¹ who found a high concentration of small loops in 1100 aluminum irradiated to a fast fluence of only 1.6×10^{21} neutrons/cm² in the HFIR at $\sim 50^\circ\text{C}$, we deduce that the dislocation segments in the N_F tray specimens are portions of loops that have grown to a size larger than the thickness of the electron microscopy foil. The concentrations of dislocations were measured from photographic prints using the linear intercept counting technique. The photographs were taken under strong [200] or [111] diffraction conditions. Assuming all Burgers vectors to be the $\langle 110 \rangle$ type, the dislocation counts were multiplied by 1.5 to allow for out-of-contrast dislocations. Foil thickness was taken into consideration. Typically, the concentration of dislocation lines was found to be 0.5 to $1.0 \times 10^{10}/\text{cm}^2$ and was independent of fast neutron fluence in the range investigated, 0.7 to 3.2×10^{22} neutrons/cm².

The transmutation-produced silicon precipitate, seen as numerous small dark spots in Fig. 10, was distributed homogeneously through the matrix except at the denuded regions at grain boundaries. Some large

Fig. 9. Distribution of Voids Through the Wall Thickness.

Fig. 10. Microstructure of Specimen Irradiated to a Fast Fluence of 3.2×10^{22} neutrons/cm², Viewed Under Diffraction Contrast to Reveal the Dislocations and Silicon Precipitate.

particles were present on the grain boundaries but were usually widely spaced. Because of the high concentration and small size of the particles it was difficult to make accurate measurements of them. As far as we could tell, however, the concentration of particles was 5 to $6 \times 10^{15}/\text{cm}^3$ and was independent of thermal fluence over the range investigated, i.e., 1 to 3×10^{22} neutrons/ cm^2 (thermal). The mean size of the particles appeared to increase slightly with fluence, from about 100 Å diam. at 1×10^{22} neutrons/ cm^2 (thermal) to about 150 Å diam. at 3×10^{22} neutrons/ cm^2 (thermal). The range of particle sizes was about 40 to 250 Å. These measurements of the silicon particles are in good agreement with what is expected from the chemical analyses for transmutation-produced silicon. These analyses (Table 4) indicate that each 1×10^{22} neutrons/ cm^2 (thermal) produces about 0.2 wt % silicon, or a volume fraction of 0.0023 if all the transmutation-produced silicon is in the form of precipitate. Assuming this precipitate occupies 5×10^{15} identical spheres/ cm^3 then the diameters of the spheres should be 100 Å and 140 Å for thermal fluences of 1×10^{22} neutrons/ cm^2 and 3×10^{22} neutrons/ cm^2 , respectively, which agrees with the values observed for the particles.

2.2 Swelling Measurements

The presence of voids in high fluence regions of the tray will cause the aluminum to expand or swell, which is reflected in a decrease in the physical density of the aluminum. Such swelling is measured simply and accurately by comparing the density, ρ_1 , of a specimen from a distorted region of the tray to that of a reference specimen, ρ_0 , that showed little or no volume change. The relative volume change, or degree of swelling, is

$$\frac{\Delta V}{V} \% = 100 \frac{\rho_0 - \rho_1}{\rho_0} . \quad (1)$$

Swelling calculated from transmission electron microscopy measurements of voids agreed within $\pm 20\%$ with the results from the density measurements.

Specimens for density measurements were cut from the E side of the tray. Material in the vicinity of brazed seams was excluded and two types of specimens were prepared. Initially, 5.84 cm long and 0.635 cm wide

strips were cut lengthwise across the plate but the neutron fluence varied along their length by $\pm 20\%$ of the average value for the specimen. Later, the fluence variation along a specimen was reduced to $\pm 8\%$ by reducing the size of specimens to 2.54 cm long by 1.27 cm wide.

Densities were determined by Archimedes method from weighings in air and in bromobenzene ($\rho \cong 1.49$ gm/cc) at 25°C . The density of the solution was determined before and after each sample weighing by calibration against a standard quartz block. The precision of each density determination was about $\pm 0.1\%$ of the density of aluminum in the initial measurements, but improvements in technique resulted in a precision of about $\pm 0.015\%$ in later measurements. Oxide films, formed from the coolant water in the ORR, were removed chemically with a solution of 900 ml H_3PO_4 and 100 ml HNO_3 at 60 to 80°C . None of the original material from which the tray was manufactured was available for reference purposes, so a reference specimen of density (ρ_0) was cut from the top of the E side of the tray where the fast neutron fluence is estimated to have been only about 3×10^{20} neutrons/cm². This fluence does not cause voids or measurable swelling in annealed 1100 aluminum irradiated at $\sim 50^\circ\text{C}$.¹¹ The density of this specimen before oxide stripping was 2.7010 g/cm³, and after stripping, 2.7108 g/cm³.

A list of the swelling measurements made on the E side of the tray is given in Table 5. The locations of the specimens and the variation of swelling with axial distance along the tray are shown in Fig. 11. The data points on the left-hand side of the curve represent the average swellings for two, three, or four specimens from each of four radial positions, rows 1 through 4. Those on the right-hand side of the curve are from single specimens, each of which spans almost the full side of the tray so they are also really average values across the face of the tray. Note that the degree of swelling in Fig. 11 follows the same pattern as the bowing (Fig. 3) and the fast neutron flux (Fig. 5). The axial position of maximum swelling corresponds with the positions of maximum bowing and maximum fast flux.

The variation of swelling with fast neutron fluence is shown in Fig. 12. The data for the smaller specimens in rows 1 through 4 are presented in Fig. 13. We can see no obvious reason why the swelling

Table 5. List of Swelling Measurements Made on the East Side of the N_F Tray.
The locations of the specimens are indicated in Fig. 11. Swelling from the silicon precipitate is calculated from the thermal fluence and the measured silicon production rate. The difference between columns 4 and 7 gives the swelling from voids only.

Specimen Number	Row Number	% Swelling		Neutron Fluence ($\times 10^{22}$ n/cm ²)		% Swelling from Silicon Precipitate
		With Oxide	Oxide Stripped	(>0.1 MeV)	(<0.0235 eV)	
B1	--	1.37	1.29	3.1	3.0	0.10
B2	--	1.42	--	3.1	3.0	0.10
B5	--	1.52	1.50	3.2	3.1	0.10
B10	--	1.41	1.49	3.0	2.9	0.09
B11	--	1.41	--	3.0	2.8	0.09
B15	--	1.33	1.17	2.6	2.6	0.08
B16	--	--	1.12	2.6	2.5	0.08
B17	--	--	1.06	2.5	2.4	0.08
B18	--	--	1.03	2.4	2.4	0.08
B20	--	0.90	--	2.3	2.2	0.07
B25	--	0.61	--	1.9	1.8	0.06
B29	--	--	0.31	1.5	1.5	0.05
B30	--	0.35	--	1.4	1.3	0.04
B33	--	--	0.15	1.1	1.2	0.04
B36	--	--	0.024	0.8	1.0	0.03
B40	--	0.0	0.0	0.3	--	--
B60	1	--	0.73	2.5	2.7	0.09
B61	2	--	1.30	2.9	2.9	0.09
B62	3	--	1.52	3.2	3.0	0.10
B63	1	--	0.69	2.4	2.6	0.08
B64	2	--	1.21	2.7	2.8	0.09
B65	3	--	1.53	3.0	3.0	0.09
B66	1	--	0.67	2.2	2.4	0.08
B67	2	--	1.15	2.5	2.5	0.08
B68	3	--	1.40	2.8	2.7	0.09
B69	4	--	1.44	3.1	2.8	0.09

Table 5. (Continued)

Specimen Number	Row Number	% Swelling		Neutron Fluence ($\times 10^{22}$ n/cm ²)		% Swelling from Silicon Precipitate
		With Oxide	Oxide Stripped	>0.1 MeV)	(<0.0235 eV)	
B70	1	--	0.52	2.0	2.2	0.07
B71	2	--	1.01	2.3	2.3	0.07
B72	3	--	1.26	2.6	2.5	0.08
B73	4	--	1.33	2.8	2.6	0.08
B75	2	--	0.84	2.1	2.1	0.07
B76	3	--	1.06	2.3	2.3	0.07
B79	2	--	0.62	1.9	2.0	0.06
B80	3	--	0.84	2.1	2.1	0.07
B82	1	--	0.17	1.4	1.7	0.06
B83	2	--	0.46	1.6	1.8	0.06
B84	3	--	0.58	1.8	1.9	0.06
B85	4	--	0.64	1.9	2.1	0.07
B87	2	--	0.23	1.3	1.8	0.06
B88	3	--	0.30	1.4	1.9	0.06
B91	2	--	0.11	1.0	1.7	0.06
B92	3	--	0.12	1.1	1.8	0.06
B94	1	--	0.06	0.6	1.7	0.05
B95	2	--	0.04	0.7	1.8	0.06
B96	3	--	0.05	0.8	1.9	0.06
B97	4	--	0.10	0.8	2.0	0.06

Fig. 11. Swelling Data on Density Specimens Cut from the East Side of the Tray. The boxes under the curve indicate the fast neutron fluences and the axial and radial locations of the specimens. The data points on the left hand side of the figure are the average values for rows 1 through 4.

Fig. 12. Dependence of Swelling on Fast Neutron Fluence in East Side of Tray. Large specimens only.

Fig. 13. Dependence of Swelling on Fast Neutron Fluence for the Small Specimens in the East Side of the Tray, Showing Effect of Specimen Position. Row 1 specimens are located farthest away from the sources of neutrons.

data for row 1 specimens deviate from those for rows 2 through 4. It is noted that the row 1 specimens are farther from the neutron source than rows 2 through 4, and their thermal to fast fluence ratios are higher.

2.3 Effect of the Oxide Scale

With the oxide scale intact, the swelling in the metal is given by

$$\frac{\Delta V}{V} \% = 100 \frac{\rho_{0+ox} - \rho_{1+ox}}{\rho_{0+ox}}, \quad (2)$$

where the subscript ox denotes the presence of oxide films. This is almost the same as Eq. (1), as shown by the triangular data points in Figs. 11 and 12. But if ρ_{1+ox} is compared with the oxide-stripped reference specimen, ρ_0 , there is an apparent additional swelling of 0.3 to 0.4% for specimens from the E side of the tray. These increments occur because the oxide films increase the wall thickness of the tray. This is a purely external effect and appeared to be of constant magnitude along a given side of the tray. Optical metallography showed the oxide scale to be of uniform thickness of 0.005 cm on each exposed face of the E side. From the geometry of the specimens and the density decrease due to oxide layers, it can be shown that the ratio $\rho_{oxide}/\rho_{metal} \approx 0.82$. Hence, the effective oxide density, for $\rho_{metal} \sim 2.71$ g/cc, is 2.22 g/cc. The nature of the oxide was not established, but x-ray diffraction studies¹³ of oxides from similar reactor components suggest that the oxide is bayerite whose density is 2.53 g/cc. Optical metallography showed that the oxide layer was porous, so the low value of 2.22 g/cc deduced from our measurements is not surprising.

2.4 Discussion of Microstructural and Swelling Observations

Voids are caused by the agglomeration of excess vacancies created by displacement events. Each event creates an equal number of vacancies and interstitial atoms. Most of these point defects annihilate one another but a small fraction survives and forms dislocations and voids. It is believed that voids can develop only if the interstitial atoms are preferentially removed from the lattice, leaving a supersaturation of vacancies. The dislocation structure developed during irradiation is thought to provide preferred sinks for interstitial atoms.

Voids are responsible for most of the observed swelling. The bowing, the fast neutron flux and the swelling profile along the axis of the tray are related. The degree of swelling increases with fast neutron fluence. Consequently, the swelling is non-uniform along the length of the tray and also across the tray, being greatest at the SE edge and decreasing towards the NE edge, following the neutron flux profiles. Figure 14 shows the swelling profiles derived for each side of the tray using the fast neutron fluence data (Fig. 5 and Table 3) and the fluence dependence of swelling measured on the E side (Fig. 12). The differences in swelling between opposite sides of the tray cause the tray to bend. No other mechanism of bowing, e.g., sagging by radiation-enhanced creep or by unidirectional growth, would involve the observed swelling. The silicon precipitate contribution to swelling is incorporated in Figs. 11 through 14, but is small (Table 5). At the maximum swelling of 1.5% the contribution from the silicon precipitate is about 0.10%, the remaining 1.4% being due to voids.

The oxide scale on the surfaces of the tray is not considered to subscribe to bowing but may make the tray bind more readily.

The dislocation structure in the tray is not evidence of plastic deformation (creep). The dislocations observed are common in unstressed and undistorted, irradiated aluminum, and are developed from the precipitation of radiation-produced point defects. In fact, the dislocations and the silicon precipitate actually strengthen the aluminum and make plastic deformation more difficult, as we shall see in Section 5 of this report. The dislocations affect bowing or swelling only in the sense that they are believed to encourage void formation by absorbing interstitial atoms.

2.5 Summary of Microstructural and Swelling Measurements

1. Neutron irradiation of the 1100 aluminum N_F tray causes the formation of voids, a dislocation structure and a fine precipitate of silicon.

2. Voids are found only in those portions of the tray whose fast neutron fluence exceeds 7×10^{21} neutrons/cm². They increase in size and concentration with increasing fluence, being about 500 Å diam. and about 1×10^{14} /cm³ at the maximum fast fluence of 3.2×10^{22} neutrons/cm².

Fig. 14. Swelling Profiles Derived for Each Side of the Tray.

3. The concentration of dislocations is about 6×10^9 lines/cm² and is apparently independent of fast fluence in the range 1 to 3×10^{22} neutrons/cm².

4. The concentration of silicon particles is about 5×10^{15} /cm³ and is independent of thermal fluence over the range 1 to 3×10^{22} neutrons/cm². The average size of the particles increases slightly from about 100 Å diameter to about 140 Å over this range of fluence. These particles cause only a small amount of swelling.

5. The voids cause the major part of the swelling. Swelling increases with fast neutron fluence and differs on each side of the tray according to the radial fluence distribution.

6. It is proposed that unequal swelling on the four sides of the tray is responsible for the observed bowing.

7. An oxide scale on the surfaces of the tray causes a small amount of swelling which is considered not to affect bowing.

3. CALCULATION OF BOWING

3.1 The Hypothesis

The creation of voids non-uniformly along the length of the tray and radially across the tray cause non-uniform swelling. Because of the unequal swelling in the radial direction the four sides of the tray grow by different amounts; the differences in length between opposite sides of the tray induce bending.

3.2 The Method

The extent of bowing is estimated by deriving the increases in length on opposite sides of the tray and translating these lengths into curvature. Since this curvature is non-uniform along the length of the tray, the tray is partitioned into equal, adjoining sections, in each of which the bowing is assumed to be uniform. The deflection of each section is computed, and the deflections are summed over all sections. Bowing is allowed to occur independently in two planes, NS and EW only.

The unirradiated tray is considered to consist of ten equal sections of original length, ℓ_0 , and width w , as shown in Fig. 15. After irradiation each section is bent through a radius of curvature, R , and subtends an angle θ at the center of curvature. Changes in w are ignored; ℓ_0 increases to $\ell_0 + \Delta\ell$. The bowing deflection from a reference plane is x . The subscript letters, N, S, etc., identify the appropriate side of the tray, and the subscript numerals denote the particular section.

The sections are arranged symmetrically on each side of the radial midplane of the tray as shown in Fig. 15. The two sections adjacent to the radial midplane receive the highest fluences and have the greatest curvature. Their ends are the fulcrums for bowing in the adjacent sections, and the plane of reference for the bowing deflection in each adjacent section is the tangent to the surface at the junction of the sections. At each junction the total deflection from the original horizontal reference plane is x plus multiples of x from the previous sections, e.g., at the junction of sections 6 and 7 in Fig. 15 the total deflection on the S side is x_{S6} ; at the junction of sections 7 and 8 the total deflection is $[x_{S7} + f(x_{S6})]$.

Fig. 15. Geometrical Parameters and Construction Used in Calculation of Bowing.

3.3 The Variables

The known variable, $\Delta\ell$, in these calculations is derived from the swelling, $\frac{\Delta V}{V}$. Since $V = \ell^3$, $dV = 3\ell^2 d\ell$ and $\frac{dV}{V} = \frac{3\ell^2 d\ell}{\ell^3}$. Therefore,

$$\Delta\ell = \frac{\ell_0}{3} \cdot \frac{\Delta V}{V} \quad (3)$$

The required, unknown variable, x , is given by

$$x = y \cos \theta. \quad (4)$$

y is determined by the relationship $\cos \theta = \frac{R_S}{R_S + y}$, which gives

$$y = R_S \left[\frac{1}{\cos \theta} - 1 \right]. \quad (5)$$

The radius of curvature is obtained through the relationship

$$\ell_0 + \Delta\ell = R\theta, \quad (6)$$

with θ in radians. To eliminate θ we take the ratios of the lengths of opposite sides of the tray:

$$\frac{R_S \theta}{R_N \theta} = \frac{\ell_0 + \Delta\ell_S}{\ell_0 + \Delta\ell_N}.$$

Since

$$R_S = R_N + \omega, \quad (7)$$

$$\frac{R_N + \omega}{R_N} = \frac{1 + \frac{\Delta\ell_S}{\ell_0}}{1 + \frac{\Delta\ell_N}{\ell_0}},$$

and from Eq. (3),

$$1 + \frac{\omega}{R_N} = \frac{1 + \frac{1}{3} \left(\frac{\Delta V_S}{V} \right)}{1 + \frac{1}{3} \left(\frac{\Delta V_N}{V} \right)},$$

which is rearranged to

$$\frac{\omega}{R_N} = \frac{\frac{1}{3}(\frac{\Delta V_S}{V}) - \frac{1}{3}(\frac{\Delta V_N}{V})}{1 + \frac{1}{3}(\frac{\Delta V_N}{V})},$$

and thence

$$R_N = \frac{\omega[3 + (\frac{\Delta V_N}{V})]}{[(\frac{\Delta V_S}{V}) - (\frac{\Delta V_N}{V})]}. \quad (8)$$

To obtain θ in terms of $\frac{\Delta V}{V}$ or $\Delta \ell$, we again use Eq. (6) and note that the difference in length of opposite sides is $(\ell_0 + \Delta \ell_S) - (\ell_0 + \Delta \ell_N) = R_S \theta - R_N \theta$. Therefore,

$$\theta = \frac{\Delta \ell_S - \Delta \ell_N}{R_S - R_N} = \frac{\Delta \ell_S - \Delta \ell_N}{\omega}.$$

In terms of degrees, rather than radians,

$$\theta = \frac{180(\Delta \ell_S - \Delta \ell_N)}{2\omega\pi}. \quad (9)$$

3.4 Swelling Values

The values of $\Delta V/V$ used in the computations for each section were read from the swelling profiles in Fig. 14 at distances corresponding to the mid point of each section and are listed in Table 6. Values of $\Delta \ell$, θ , R , and x are also listed. For small angles the values of x and y are essentially identical.

3.5 Summation of Bowing Deflections

To a close approximation the total deflections at the section junctions increase in the order s_6 ; $[x_7 + f(x_6)]$; $[x_8 + f(x_7) + f(x_6)]$, etc., as the axial distance from the radial mid plane increases. The function, f , is given by the approximate relationship $\frac{x}{a} = \frac{f(x)}{a + \ell_0}$, where

Table 6. Parameters Used in Calculation of Bowing Deflections.

Section	Swelling ($\Delta V/V$)		Δl_S (cm)	Δl_N (cm)	θ (deg.)	R_S (cm)	R_N (cm)	x_S (cm)	x_N (cm)
	S Side	N Side							
1	0.0009	0.0004	0.00186	0.00083	0.0078	45,670.01	45,662.40	0.00041	0.00041
2	0.0038	0.0013	0.00785	0.00269	0.0389	9,141.17	9,133.56	0.00210	0.00210
3	0.0076	0.0032	0.01571	0.00661	0.0685	5,198.43	5,190.82	0.00371	0.00370
4	0.0107	0.0053	0.02211	0.01095	0.0842	4,238.33	4,230.72	0.00457	0.00456
5	0.0126	0.0065	0.02604	0.01343	0.0949	3,753.43	3,745.82	0.00515	0.00514
6	0.0120	0.0060	0.02480	0.01240	0.0933	3,815.61	3,808.00	0.00506	0.00506
7	0.0095	0.0044	0.01964	0.00910	0.0793	4,486.67	4,479.06	0.00430	0.00430
8	0.0065	0.0024	0.01345	0.00496	0.0639	5,577.66	5,570.05	0.00347	0.00346
9	0.0035	0.0009	0.00724	0.00186	0.0405	8,789.42	8,781.81	0.00220	0.00219
10	0.0010	0.00035	0.00207	0.00072	0.0101	35,132.30	35,124.69	0.00056	0.00056
	E Side	W Side	Δl_E (cm)	Δl_W (cm)	θ (deg.)	R_E (cm)	R_W (cm)	x_E (cm)	x_W (cm)
1	0.0013	0.0002	0.00268	0.00041	0.0167	21,253.79	21,246.00	0.00091	0.00091
2	0.0054	0.0008	0.01116	0.00165	0.0699	5,088.75	5,080.96	0.00379	0.00378
3	0.0096	0.0018	0.01984	0.00372	0.1186	3,004.63	2,996.84	0.00643	0.00642
4	0.0131	0.0034	0.02707	0.00703	0.1474	2,418.12	2,410.33	0.00801	0.00798
5	0.0150	0.0045	0.03100	0.00930	0.1596	2,234.79	2,227.00	0.00867	0.00864
6	0.0144	0.0040	0.02976	0.00827	0.1574	2,256.06	2,248.27	0.00852	0.00849
7	0.0117	0.0027	0.02418	0.00558	0.1368	2,605.13	2,597.34	0.00742	0.00740
8	0.0084	0.0014	0.01736	0.00289	0.1064	3,346.96	3,339.17	0.00577	0.00576
9	0.0050	0.0008	0.01033	0.00165	0.0638	5,572.65	5,564.86	0.00346	0.00345
10	0.0013	0.0002	0.00269	0.00041	0.0165	21,253.79	21,246.00	0.00089	0.00089

a is the distance shown in Fig. 15, and f at any junction n sections from the midplane is given by $f_n = (a + n\ell_0)/a$. The distance a is approximately equal to $\ell_0/2$, thus $f_n \cong 1 + 2n$. The total deflections appear as the simple algebraic progression in Table 7.

In constructing the bowing curve as we do in Fig. 15, the midplane is the reference point of zero bowing. This is the reverse of the real case as measured by Hurt where the ends of the tray are the zero bowing reference points. The calculations are brought into correspondence with Hurt's measurements by subtracting the total deflections in Table 7 from the maximum deflections at the appropriate end of the tray, i.e., sections 1 through 5 are subtracted from section 1, and sections 6 through 10 from section 10. The plotting distances in Table 7 are the distances of the section junctions from the bottom of the tray. The calculated bowing deflections are shown as points superimposed on Hurt's bowing curves in Fig. 16.

3.6 Conclusions

The excellent agreement between these calculations and the measured bowing deflections strongly support the contention that bowing is a result of non-uniform swelling.

3.7 Summary of Bowing Calculations

1. A simple model of bowing is derived based on differential length changes between opposite sides of the N_F tray caused by non-uniform swelling.
2. Utilizing the swelling data the model accurately predicts the measured bowing curves.

Table 7. Summation of Calculated Bowing Deflections.

	Section									
	1	2	3	4	5	6	7	8	9	10
Plotting Distance (cm)	0	6.2	12.4	18.6	24.8	37.2	43.4	49.6	55.8	62.0
Summation of f(n)	9x ₅ 7x ₄ 5x ₃ 3x ₂ x ₁	7x ₅ 5x ₄ 3x ₃ x ₂	5x ₅ 3x ₄ x ₃	3x ₅ x ₄	x ₅	x ₆	3x ₆ x ₇	5x ₆ 3x ₇ x ₈	7x ₆ 5x ₇ 3x ₈ x ₉	9x ₆ 7x ₇ 5x ₈ 3x ₉ x ₁₀
S Side										
Total Deflection (cm)	0.103	0.072	0.043	0.020	0.005	0.005	0.019	0.042	0.069	0.100
Plotting Deflection	0	0.031	0.060	0.083	0.098	0.095	0.081	0.058	0.031	0
N Side										
Total Deflection (cm)	0.103	0.072	0.038	0.020	0.005	0.005	0.019	0.042	0.069	0.100
Plotting Deflection	0	0.031	0.065	0.083	0.098	0.095	0.081	0.058	0.031	0
E Side										
Total Deflection (cm)	0.178	0.124	0.074	0.034	0.009	0.008	0.033	0.071	0.117	0.169
Plotting Deflection	0	0.054	0.104	0.144	0.169	0.161	0.136	0.098	0.052	0
W Side										
Total Deflection (cm)	0.178	0.123	0.073	0.034	0.009	0.008	0.032	0.070	0.117	0.168
Plotting Deflection	0	0.055	0.105	0.144	0.169	0.160	0.136	0.098	0.051	0

Fig. 16. Calculated Bowing Deflections Superimposed on Measured Bowing Curves.

4. EFFECTS OF POSTIRRADIATION ANNEALING

4.1 Annealing Behavior of Voids

Voids are thermodynamically unstable and shrink during annealing at temperatures low enough to prevent the formation of gas bubbles.^{11,14,15} To determine the annealing times and temperatures suitable for removing swelling in the N_F tray, and also to glean some information on the kinetics of void annihilation in 1100 aluminum, a series of postirradiation anneals was performed on some of the density specimens.

The effects of isochronal heat treatment in air are shown in Fig. 17. Recovery from swelling was not detected until after 20 hr at 170°C. Almost complete recovery occurred at about 300 to 350°C (~ 0.6 of the absolute melting temperature). Annealing above 350°C caused gas bubbles to form with accompanying swelling which at high temperatures exceeded the original irradiation-induced swelling.

Transmission electron microscopy showed that recovery from swelling at 300°C and below was associated with the elimination of irradiation-induced voids, Figs. 18 and 19. Most of the small amount of swelling remaining after isochronal anneals in the temperature range 300 to 350°C is due to the transmutation-produced silicon which remains precipitated but coarsens during annealing (Fig. 19). A comparison of Figs. 18 and 19 with Fig. 7 gives a general view of the microstructural changes occurring during annealing. The change in void size and concentration during annealing at 237°C is seen in Fig. 20. The reduction in void concentration occurs by the preferential elimination of the smallest voids; both the mean and maximum void sizes show a small but experimentally significant increase during postirradiation annealing. The overall microstructural changes are summarized¹⁶ as follows:

- (a) The smallest voids anneal out preferentially. The larger voids grow slightly before eventually shrinking.
- (b) Voids near grain boundaries are also eliminated preferentially causing a widening of the grain boundary denuded regions.
- (c) The transmutation-produced silicon precipitate coarsens simultaneously with the disappearance of voids.

Fig. 17. Effect of Isochronal Annealing on Swelling in Specimens Irradiated to about 3.0×10^{22} neutrons/cm² ($E > 0.1$ MeV).

Fig. 18. After Annealing for 20 hr at 237°C. The silicon particles have coarsened, the void denuded region near the grain boundary has expanded and the smallest voids have been eliminated. Compare with Fig. 7.

Fig. 19. Coarsening of Silicon Precipitate and Elimination of Voids in Specimen Annealed for 20 hr at 300°C after Irradiation to 3.2×10^{22} neutrons/cm² ($E > 0.1$ MeV). Compare with Fig. 7.

Fig. 20. Effect of 20 hr Postirradiation Anneal at 237°C on the Void Size and Concentration Within the Grains in Specimen Irradiated to 3.2×10^{22} neutrons/cm² ($E > 0.1$ MeV).

(d) The dislocation density is reduced, but many dislocations remain and form irregular networks that frequently link the remaining voids and silicon particles.

(e) On annealing at 350°C and above gas bubbles are formed causing a resurgence of swelling. These bubbles appear as crystallographically faceted cavities of a wide range of diameters on grain boundaries, inclusion matrix interfaces and dislocations (Fig. 21). The association of these cavities with grain boundaries as well as their growth with increasing temperature confirms that they are gas bubbles. The bubbles have the shape of regular polyhedra with faces formed from {111}, {110}, and {100} planes. Optical metallography revealed the existence of much larger bubbles in addition to those detected by transmission electron microscopy.

4.2 Kinetics of Void Sintering

The kinetics of void annihilation were investigated in greater detail¹⁷ using a combination of isochronal and isothermal anneals on bulk specimens. The results are shown graphically in Figs. 22 and 23 in terms of percentage recovery of irradiation-induced density decrease due to voids after correction for swelling from the transmutation-produced silicon precipitate. The initial swelling of specimens used in these kinetic measurements was 1.2 to 1.4% and the swelling from the silicon precipitate was about 0.10%.

The isothermal and isochronal data were combined by means of a method developed by Howe¹⁸ to yield an activation energy for void annealing from the equation

$$\ln t = c - \frac{E}{kT} \quad (10)$$

where t is the time taken in an isothermal anneal for an irradiated sample to recover an amount equal to that recovered by a sample isochronally annealed at temperature T , c is a constant, k is Boltzmann's constant, and E is the activation energy. Plots of $\ln t$ versus T^{-1} , shown in Fig. 24 for the 230°C isothermal data combined with the 1 hr and 20 hr isochronal annealing measurements, yield activation energies for void annealing of 2.0 and 1.8 eV, respectively.

Fig. 21. Gas Bubbles in Specimen Annealed 20 hr at 560°C After Irradiation to 3×10^{22} neutrons/cm² ($E > 0.1$ MeV). Bubbles on grain boundaries are larger than those within the grains.

Fig. 22. Isochronal Recovery of Swelling from Radiation-Induced Voids.

Fig. 23. Isothermal Recovery of Swelling from Radiation-Induced Voids.

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Fig. 24. Plot of Logarithm of Annealing Time Versus the Reciprocal of the Annealing Temperature for the Determination of the Activation Energy for Void Annihilation.

4.3 Discussion of Postirradiation Annealing Effects

Recovery from irradiation-induced swelling occurred in the density specimens in 1 hr at 350°C or 20 hr at 300°C. Therefore, suitable annealing should retard or reduce swelling in the N_F tray. However, we have no evidence that removal of swelling will return a highly bowed tray to its original shape. If annealing should be adopted as a preventative policy, we suggest that the anneals be done at frequent intervals, more specifically, whenever the first signs of bowing appear. This should prolong the working lifetime of the tray.

Void annihilation occurs because the voids emit vacancies to suitable sinks. The measured activation energy for void shrinkage of 1.8 to 2.0 eV is higher than that derived for the shrinkage of an isolated void in the vicinity of perfect vacancy sinks.¹⁹ The latter derivation is based on a simplified model that predicts the activation energy for void annealing to be slightly smaller than the self-diffusion activation energy, which is about 1.3 eV for aluminum.²⁰ This implies that void annealing in bulk specimens of irradiated 1100 aluminum cannot be described in terms of the simple kinetic equations derived for the isolated void. The simplified model does not consider interactions of vacancies with other defects and impurities during annealing. Such interaction is common in the irradiated 1100 Al.

Growth of some of the voids during annealing is proof that voids themselves entrap vacancies during annealing. Similar growth of voids has been reported elsewhere for pure aluminum,¹⁵ and is predicted by the Gibbs-Thomson equation which dictates that the total energy of a material containing a range of void sizes is lowered by the transfer of vacancies from a small void to a larger one. This effect will be observed only if the spacing of the major vacancy sinks is much larger than the mean-free path between voids. The major vacancy sinks in the 1100 aluminum are the grain boundaries, as evidenced by the preferential disappearance of all voids near grain boundaries irrespective of void size (Fig. 19). Another source of vacancy trapping is the transmutation-produced silicon precipitate which coarsens concurrently with void annihilation. The silicon-vacancy binding energy in aluminum is about 0.2 eV^{21,22} and evidence for

strong silicon-vacancy complexes has been observed in Al-Si alloys in precipitation studies²³ and during resistivity recovery of irradiated alloys.²⁴ Vacancy trapping is also expected by the transmutation-produced gas atoms.

The complexity of the various vacancy trapping processes during void annealing in bulk specimens of irradiated 1100 aluminum precludes the setting up of simple kinetic equations to describe the overall process. Consequently, the measured activation energy cannot be given a physical meaning and is to be regarded as an "effective" activation energy.

Annealing at 300 or 350°C does not cause the formation of visible gas bubbles even though significant quantities of gases are present in the aluminum. The development of bubbles and the consequent renewal of swelling occurs at higher annealing temperatures. This thermally activated swelling could, in theory, continue until the gas was released. The helium analyses show that no helium is released, at least for annealing treatments up to 20 hr at 560°C. Nothing is known about the role of hydrogen.

4.4 Summary of Postirradiation Annealing Effects

1. During postirradiation annealing in air, elimination of voids and recovery from swelling occur in 1 hr at 350°C or 20 hr at 300°C.

Concurrently the silicon precipitate coarsens.

2. Annealing at 300 to 350°C will reduce swelling in the N_F tray but may not return a badly bowed tray to its original shape. To be effective in minimizing bowing annealing should be performed periodically or, at least, whenever a tray shows the first signs of bowing.

3. The activation energy for void sintering in the N_F tray is 1.8 to 2.0 eV which is higher than the value of about 1.3 eV calculated for sintering of a single void. The discrepancy is believed to be caused by temporary trapping of vacancies by voids, silicon atoms and gas atoms.

4. Annealing at temperatures above 350 to 400°C causes the development of gas bubbles and a resurgence of swelling.

5. MECHANICAL PROPERTIES

Mechanical property measurements were not considered necessary to explain the problem of bowing in the N_F tray. However, such measurements extend our knowledge of radiation damage in metals, and provide information for our surveillance program of radiation damage studies of aluminum in water-cooled reactors. More importantly in the present case, it was considered prudent to determine the conditions under which mechanical failure, rather than bowing, would limit the operating life of the N_F tray and similar irradiated 1100 aluminum components.

5.1 Tensile Tests

Tensile specimens with gauge lengths of 1.9 cm and of rectangular cross section, 0.635×0.51 cm, were machined from the flat N and S sides of the N_F tray. The orientation of the specimens with respect to the tray is shown in Fig. 2. Most of the specimens were tested in air at a slow strain rate of 16%/hr over a range of temperatures to simulate creep that might occur during an in-reactor temperature excursion. Specimens were held at the test temperature for 20 min prior to testing. Yield or flow stresses were measured at 0.025% plastic offsets. Since none of the original material was retained in the unirradiated state, data from the Alcoa Aluminum Handbook²⁵ are used as a guide to the tensile properties of annealed, unirradiated 1100 aluminum. The Alcoa specimens were probably tested at a faster strain rate than ours and their yield stresses were measured at 0.2% offset. The data for all tensile specimens are compiled in Table 8.

5.1.1 Effects of Neutron Fluence and Test Temperature. The interesting features of the data are emphasized in Figs. 25 and 27. The tensile strengths and elongations at two test temperatures are plotted in Fig. 25 versus thermal neutron fluence since, as will be argued later, the silicon particles are responsible for much of the irradiation-induced strengthening and the silicon is produced largely by the thermal neutrons. At both test temperatures the higher fluence irradiations caused at least a twofold increase in ultimate tensile strength and more than a fourfold increase in yield stress over the Alcoa values for the unirradiated material.

Table 8. Tensile Data on Specimens Cut from the South and North Sides of the N_F Tray

South Side Number	Neutron Fluence ($\times 10^{22}$ n/cm ²)		Strain Rate (%/hr)	Test Temperature (°C)	Yield Stress (kg/mm ²)	Ultimate Stress (kg/mm ²)	Elongation (%)		Comments
	(E > 0.1 MeV)	(E < 0.0253 eV)					Total	Uniform	
C1	0.7	1.8	16	107	11.5	13.2	16.0	3.8	
C2	1.0	1.8	untested						
C3	1.3	1.8	16	107	11.5	12.4	21.0	5.5	{ Postirradiation annealed 1 hr 537°C before testing
C4	1.5	1.9	16	205	2.25	4.43	38.8	11.9	
C5	1.8	2.1	16	370	2.90	3.12	6.1	0.7	
C6	2.1	2.3	16	93	13.4	15.3	17.9	3.6	
C7	2.3	2.5	16	147	11.7	13.8	8.0	5.0	
C8	2.5	2.8	16	25	15.7	18.1	23.1	5.4	
C9	2.6	2.9	16	25	16.1	18.4	23.6	5.1	
C10	2.8	3.1	16	205	10.4	11.2	3.2	0.9	
C11	2.8	3.1	16	258	7.32	7.88	3.3	0.7	
C12	2.7	3.0	16	317	4.64	4.85	7.6	0.5	
C13	2.6	2.9	untested						
C14	2.4	2.7	16	205	10.0	10.4	3.0	0.9	
C15	2.3	2.5	16	25	14.6	18.7	24.1	5.5	
C16	2.1	2.3	16	73	12.9	15.0	26.2	5.0	
C17	1.9	2.1	16	25	14.5	17.9	25.0	5.7	
C18	1.7	1.9	16	147	10.4	11.9	25.0	6.1	
C19	1.5	1.6	16	107	11.6	14.9	24.5	4.6	
C20	1.3	1.4	16	205	7.74	8.86	16.4	2.6	
C21	1.0	1.2	16	25	13.1	16.9	27.5	7.0	
C22	0.7	1.0	untested						
C23	0.4	0.9	16	78	12.7	16.1	29.0	6.1	{ Fluences uncertain
C24	0.1	0.9	16	205	4.92	6.75	48.2	11.3	
C25	?	~0.9	16	25	8.23	14.3	20.0	14.4	
C26	?	~0.9	16	78	6.69	11.3	33.0	9.5	
11-55	0	0	16	25	1.53	8.04	38.0	25.1	{ Unirradiated. Round section specimens. Recrystallized 30-min. at 400°C. Yield stresses difficult to measure.
11-54	0	0	16	25	1.70	8.18	38.0	24.4	
11-44	0	0	16	205	1.53	2.74	48.2	13.1	

Table 8. (Continued)

North Side Number	Neutron Fluence ($\times 10^{22}$ n/cm ²)		Strain Rate (%/hr)	Test Temperature (°C)	Yield Stress (kg/mm ²)	Ultimate Stress (kg/mm ²)	Elongation (%)		Comments
	(E > 0.1 MeV)	(E < 0.0253 eV)					Total	Uniform	
A37	0.5	1.4	untested						
A36	0.7	1.4	untested						
A35	0.8	1.4	untested						
A34	1.0	1.5	untested						
A33	1.2	1.6	untested						
A16	1.8	2.4	200	25	16.0	20.5	27.9	7.3	After testing, the head of specimen A17 was held in a vice and struck repeatedly with a hammer. It bent but did not break.
A17	1.9	2.5	2000	25	14.9	21.1	31.3	8.9	
A18	1.8	2.4	20,000	25	17.7	21.4	29.5	8.5	
A19	1.8	2.3	untested						
A20	1.7	2.2	untested						
A21	1.6	2.1	untested						
A22	1.5	2.0	untested						
A23	1.4	1.8	untested						
A24	1.3	1.7	untested						
A25	1.1	1.5	untested						
A26	1.0	1.3	untested						
A27	0.8	1.1	untested						
A28	0.7	1.0	untested						
A29	0.6	0.9	500	25	13.5	20.5	36.0	14.0	
A30	0.5	0.8	untested						
A31	?	?	untested						
A32	?	?	untested

Fig. 25. Tensile Properties at Two Test Temperatures as Functions of Thermal Neutron Fluence.

Fig. 26. Tensile Curves for Unirradiated 1100 Aluminum and Irradiated N_F Tray Specimens at Two Test Temperatures. The irradiated specimens were exposed to 2.7×10^{22} neutrons/cm² (fast) and 3.0×10^{22} neutrons/cm² (thermal).

Fig. 27. Effect of Test Temperature on Tensile Properties for a Constant Neutron Fluence.

Strengthening was not greatly dependent on fluence over the small fluence range investigated.

Irradiation-induced loss of ductility occurred at all fluence levels and was much greater at a test temperature of 205°C than at 25°C. As with the strength, the ductility was not very sensitive to change in neutron fluence over the range investigated, but was more sensitive at 205°C than at 25°C. Annealing for 1 hr at 537°C prior to testing at 205°C restored the tensile strength to the unirradiated level but did not fully recover the ductility.

The degree of strengthening from the irradiation is actually somewhat higher than that depicted in Fig. 25 and the loss in ductility is a little less severe when the irradiated specimens are compared with unirradiated specimens tested under the same conditions rather than with the Alcoa Handbook data. We tested two unirradiated specimens to obtain strain hardening exponents. Their test curves are shown in Fig. 26 together with those for the irradiated specimens. Here radiation-induced strengthening and embrittlement are seemingly most pronounced at 205°C. The work hardening exponent, n , was obtained from the tensile curve by assuming constant volume of gauge section at stresses up to the ultimate tensile strength, and a power law of the form $\sigma = K \epsilon^n$; where σ and ϵ are true stress and true strain. These exponents are listed in Table 9. These are only rough measurements but the value of n is about equal to the uniform strain, which it should be if the above power law holds for these tests. The work hardening exponent is severely reduced by irradiation.

The broader dependence of the tensile properties of the most heavily irradiated specimens on test temperature is shown in Fig. 27. Tensile strengths were markedly reduced with increasing test temperature but at all temperatures were several times greater than those for the Alcoa unirradiated reference material. Testing above 100°C caused a further decline in ductility, quite contrary to the enhanced ductility in the Alcoa reference material. At test temperatures below 100°C the specimens displayed significant necking and their fracture surfaces exhibited shear dimples typical of transgranular failure. At test temperatures above 150°C the amount of nonuniform strain in the irradiated specimens decreased markedly and intergranular fracture became more predominant with increasing

Table 9. Work Hardening Exponents for Specimens Tested
at a Strain Rate of 16%/hr

Specimen Number	Thermal Fluence ($\times 10^{22}$ n/cm ²)	Test Temperature (°C)	Uniform Strain	n
11-54	Unirradiated	25	0.22	0.28
C21	1.2	25	0.068	0.073
C17	2.1	25	0.055	0.053
C9	2.9	25	0.050	0.052
C6	2.3	93	0.035	0.029
C7	2.5	147	0.049	0.035
11-44	Unirradiated	205	0.12	0.13
C10	3.1	205	0.009	0.015
C4	1.9; annealed 1 hr 537°C	205	0.11	0.15
C12	3.0	317	0.005	0.012
C5	2.1	370	0.007	0.007

test temperature. The change in fracture morphology is illustrated in Fig. 28. At 317°C there was a suggestion (Fig. 27) of an uptrend in the elongation of the irradiated specimens but it remained low in comparison with that of the unirradiated material.

5.1.2 After-Test Microstructures. Some of the specimens that provided the data in Fig. 27 were studied by transmission electron microscopy after fracture. The microstructural changes incurred from the test temperature alone were obtained by examining the unstressed heads of the specimens. The gauge sections of the specimens provided information on the combined effects of temperature and tensile strain. The combined preheating and testing periods amounted to about 1 hr.

In agreement with the earlier postirradiation annealing studies, no significant microstructural changes were noted in the heads of the specimens for all test temperatures up to and including 205°C. In the

Fig. 28. Change in Fracture Mode with Test Temperature in Specimens Irradiated to 2.7×10^{22} neutrons/cm² (fast) and 3.0×10^{22} neutrons/cm² (thermal); Upper, Transgranular Tearing at 25°C; Lower, Intergranular Fracture at 205°C.

strained regions of the specimens changes were observed. At room temperature and 147°C the concentration of dislocations was increased by a factor of about three. These dislocations were uniformly distributed just as in the as-irradiated condition. They intersected one another and many of them were bent around silicon particles. Small loops and dislocation cells, the normal configuration of dislocations in deformed, unirradiated 1100 aluminum were absent in the tested irradiated material. Neither was there any evidence of dislocation channeling, i.e., concentrated bands of deformation running through the matrix. Testing at 205°C caused about a twofold increase in dislocation density but no noticeable change in the silicon particles or the voids. The new feature in this specimen was the presence of large cavities on the grain boundaries (Fig. 29) that ranged in size from about $500 \text{ \AA}$ to $>> 3000 \text{ \AA}$. Stereomicroscopy and the perturbations in the grain boundary diffraction contours (Fig. 29a) confirmed that the cavities were located on the grain boundaries. Most cavities were intersected by the surfaces of the thinned foils but some of the smaller ones were retained completely within the foil and were seen to be bounded by crystallographic facets parallel to those on the voids in one of the adjacent grains, usually $\{111\}$ and $\{100\}$. The incidence of cavitation was greatest on those grain boundaries that were clearly flanked by bands of voids, but not all such boundaries were cavitated. There was no evidence of any association of cavities with the voids. Some grain boundaries had cavities at fairly uniform intervals over the visible area of the boundary; others had clusters of cavities at just one or two places on the boundary; more than half of the boundaries showed no cavities. There was no preferred incidence of cavitation at triple grain junctions. Most of the cavities had their largest dimension in the plane of the boundary and within the clusters of cavities many were obviously coalescing (Fig. 29b).

At a test temperature of 317°C the silicon particles coarsened markedly, the voids were almost eliminated and many of the dislocations were removed. The concentration of silicon particles was reduced by a factor of about 400 to $1.5 \times 10^{13} \text{ cm}^{-3}$. The microstructure was very similar to that shown in Fig. 18. Some of the silicon particles had plate- or ribbon-like shapes, the more equiaxed particles having a typical

Fig. 29. Cavities on Grain Boundaries in Specimen Tested to Failure at 205°C. Coalescence of cavities is obvious in the lower photograph.

size of about 600 Å. In the strained portion of the specimen there was a roughly twofold increase in the dislocation density over that in the head of the specimen. Grain boundary cavities were larger and less numerous than those in the 205°C specimen. None of these cavities was small enough to lie entirely within a thinned foil of 3000 Å thickness; they intersected the foil surfaces and caused large etched-out holes in the foils.

5.1.3 Effects of Strain Rate. Many metals whose tensile properties are normally strain rate dependent become more sensitive to shock loading and notch-embrittlement after neutron irradiation. 1100 grade aluminum displays little or no dependence on strain rate but we were not sure how it would behave after irradiation so we tested a number of specimens over a range of strain rates at room temperature. The specimens were irradiated to a common thermal neutron fluence of about 2.4×10^{22} neutrons/cm². Some unirradiated 1100 aluminum specimens were tested for comparison purposes.

The results are presented in Table 10 and show no significant effect of strain rate. Ductility is certainly not reduced by increasing strain rate. If anything, it seems to be increased. But some of this effect, and the apparent increase in ultimate strength with increasing strain rate, may be due to a difference in the thermal to fast neutron fluence ratios: Specimens C17 and C9 have ratios of 1:1 while those of specimens A16 through A18 are > 1:3. This change in ratio will probably affect the radiation-induced damage structures.

As a further check on notch sensitivity the head of specimen A17 after tensile testing was gripped in a vice with the portion containing the 1/4-in. diam pinhole protruding. It was then struck six times in rapid succession by a hammer wielded with all the force of the hot cell operator through his manipulator. The specimen bent slightly but showed no sign of fracture.

5.2 Hardness Data

Hardness measurements were made on the density specimens as part of the postirradiation annealing study described in Section 4. A diamond pyramid microhardness tester was used, with a load of 500 g. All

Table 10. Effect of Strain Rate on Tensile Properties at Room Temperature

Specimen	Neutron Fluence ($\times 10^{22}$ n/cm ²)		Strain Rate (%/hr)	Yield Stress (kg/mm ²)	Ultimate Stress (kg/mm ²)	Elongation (%)		n
	(E > 0.1 MeV)	(E < 0.0253 eV)				Total	Uniform	
1100-1	0	0	16	0.895	7.56	63.3	36.5	0.42
1100-3	0	0	160	2.23	7.60	54.5	33.0	0.28
1100-4	0	0	16,000	1.78	7.42	57.7	35.2	0.31
C9	2.6	2.9	16	16.1	18.4	23.6	5.1	0.052
C17	1.9	2.1	16	14.5	17.9	25.0	5.7	0.053
A16	1.8	2.4	200	16.0	20.5	27.9	7.3	0.080
A17	1.9	2.5	2,000	14.9	21.1	31.3	8.9	0.095
A18	1.8	2.4	20,000	17.7	21.4	29.5	8.5	0.053

measurements were made at about 28°C. Specimens irradiated to 3.2×10^{22} neutrons/cm² (0.1 MeV) and 3.1×10^{22} neutrons/cm² (thermal) had a hardness of about 59 kg/mm², which is more than double the value of 23 kg/mm² expected for unirradiated, recrystallized 1100 aluminum.¹⁰ During annealing softening began when the temperature exceeded 200°C (Fig. 30). The minimum hardness occurred after 20 hr at 460°C, with a small tendency for higher values at higher temperatures. At higher temperatures there was significant swelling from the development of gas bubbles but these clearly had little effect on the hardness.

5.3 Discussion of Mechanical Test Data

Irradiation induces a marked increase in strength and loss in ductility in the previously annealed 1100 grade aluminum of the N_F tray. But at the normal reactor operating temperature of ~45°C the changes in short term properties are not regarded as cause for alarm. More than 20% elongation is retained at test temperatures below 100°C, which should provide a cushion against sudden failure under normal conditions. In the unlikely event that the tray should experience temperatures greater than 100°C, brittle-type failure is possible but would require the application of stresses much beyond the design stress levels. Slower strain rates than those used in this work are likely to exaggerate the loss in ductility at elevated temperatures. The effects of shock loading, notch sensitivity, etc., were not specifically investigated, but the lack of strain rate sensitivity in the tensile tests at room temperature suggests an absence of notch sensitivity. However, as always with an irradiated reactor component, impact conditions should be avoided at all times.

A detailed discussion of the temperature and neutron fluence dependence of these mechanical test data has been published elsewhere.²⁶ It is shown that the increase in hardness or tensile strength is caused by the irradiation-defect structure which impedes the movement of dislocations during plastic straining. The major defects are the silicon precipitate, the dislocation structure and the voids. Each defect will make a contribution to the strength of the aluminum. The strengthening contribution from each type of defect in terms of the concentrations and sizes of the defects were calculated from existing theories of defect

Fig. 30. Variation of Hardness with Annealing Temperature for 20 hr Isochronal Anneals.

hardening²⁷⁻³⁰ and are summarized in Table 11. The calculated and observed values are in satisfactory agreement and are taken as evidence that the strength of the N_F tray specimens can be adequately described in terms of the observed defect structure. The change in yield stress with increasing neutron fluence over the small range of fluence examined is caused by an increase in the volume fraction of transmutation-produced silicon and by an increase in the contribution from voids. The dislocation density appeared to remain constant over this fluence range.

Much of the decrease in strength of the irradiated aluminum with increasing test temperature (Fig. 27) is probably due to matrix softening, as displayed also by the unirradiated Alcoa aluminum. Above 200°C coarsening of the silicon precipitate in the irradiated aluminum and the annealing out of some of the irradiation-induced dislocation structure causes further softening. This latter point is demonstrated most clearly by the hardness measurements in Fig. 30 which we made at room temperature and show a large degree of softening due solely to coarsening of the microstructure.

Two factors determine the observed ductility loss or embrittlement in the tensile tests. One is a consequence of the strengthening effect of the irradiation-induced defects which retard dislocation motion. This source of ductility loss is a very significant one in the present work. It is believed to occur at all temperatures at which radiation-induced hardening persists but is dominant at the lower test temperatures below about 100°C where the fracture mode is transgranular tearing. Defect hardening embrittlement is indicated by a marked reduction in the strain hardening exponent (Table 9). At higher test temperatures the second form of embrittlement, premature intergranular fracture by cavity coalescence, enters the picture in addition to defect hardening embrittlement.

Intergranular fracture by cavity coalescence is common in metals tested under creep conditions and is considerably accelerated by the presence of certain gases, particularly helium.^{31, 32} As little as 0.1 at. ppm helium can cause pronounced ductility loss in irradiated metals.³³⁻³⁸ The specimens used to obtain Fig. 27 contained 10 to 16 at. ppm helium. Full consideration of the detailed observations on

Table 11. Calculated Contributions to Room Temperature Yield Stress from the Observed Structural Defects

Defect	Thermal Fluence	
	1.2×10^{22}	2.95×10^{22}
	(n/cm ²)	(n/cm ²)
	Hardening Contribution, kg/mm ²	
Silicon precipitate	4.2	5.6
Dislocations	6.0	6.0
Voids	1.0	3.5
<hr/>		
Unirradiated yield stress	1.5	1.5
<hr/>		
Total	12.7	16.6
Measured yield stress after irradiation	13.1	16.1

the cavities and the test data²⁶ lead to the conclusion that the cavities are initiated by helium, possibly through grain boundary sliding, and they grow and coalesce by a stress-assisted influx of vacancies.^{39,40} The irradiation-induced defect structure permits high stresses to be sustained by the specimens, thus accelerating cavity growth. It follows that the extent of cavitation, and hence the degree of embrittlement, will be increased by increasing helium content and by increasing tensile strength.

5.4 Summary of Mechanical Property Tests

1. Irradiation more than doubles the hardness and ultimate tensile strength and more than triples the flow stress of the N_F tray at all test temperatures between 25 and 370°C.

2. The increase in strength is caused largely by the transmutation-produced silicon precipitate and by the radiation-induced dislocation structure.

3. The work hardening exponent is significantly reduced.

4. The tensile properties are not strongly sensitive to strain rate at room temperature.

5. There is considerable ductility loss which is most severe in tests above about 150°C, where the fracture mode changes from transgranular to intergranular.

6. Intergranular fracture is caused by the development and coalescence of cavities on the grain boundaries.

7. The grain boundary cavities are nucleated by helium and grow by vacancy condensation under the influence of the applied stress. A strengthened matrix therefore encourages intergranular failure.

8. These mechanical property changes are not regarded as being seriously detrimental to the short-term properties of the N_F tray provided the operating temperature of the tray does not exceed $\sim 100^\circ\text{C}$. Brittle type failure can be induced at higher temperatures.

6. GENERAL SUMMARY AND CONCLUSIONS

6.1 General Summary of Observations

An estimation of the fast and thermal neutron flux distributions in the 1100 aluminum ORR N_F tray indicates a peak flux at the mid length of the SE edge. Maximum bowing occurred in this region. The fast neutron flux profile along the length of the tray has the same general shape as the axial bowing profile measured by Hurt.

Radiation-induced microstructural damage is manifest as dislocations, voids and a fine precipitate of transmutation-produced silicon. The presence of the voids causes the aluminum to swell. It is shown that the degree of swelling increases with fast neutron fluence, and that the swelling profile along the length of the E side of the tray is compatible with the axial bowing profile. Bowing is caused by non-uniform swelling on opposite sides of the tray, and it is demonstrated that the bowing deflection can be accurately calculated from swelling curves derived for each side of the tray.

Postirradiation annealing studies show that voids and swelling can be eliminated in 1 hr at 350°C or 20 hr at 300°C and the silicon precipitate coarsens. Annealing at higher temperatures causes the formation of bubbles of transmutation-produced gas and a resurgence of swelling.

Irradiation more than doubles the hardness and tensile strength of the 1100 aluminum, even at elevated test temperatures. The increase in strength is accounted for quantitatively in terms of defect hardening by the radiation-induced dislocation structure and the silicon precipitate. The defect structure also reduces the work hardening exponent, resulting in significant loss of ductility. These mechanical property changes are not very sensitive to strain rate. At test temperatures $> 100^\circ\text{C}$ helium embrittlement via grain boundary cavitation causes a further loss in ductility.

6.2 Recommendations for Improving the Working Lifetime of the N_F Tray

Since it is concluded that bowing of the 1100 aluminum N_F tray is caused by non-uniform swelling from the development of voids during fast neutron irradiation, it follows that any means that will prevent or

minimize void formation will accordingly minimize bowing. There are at least two ways to achieve this.

The first way is by periodically annealing at 300 to 350°C. As discussed in Section 4, this annealing will give recovery from swelling but may not recover the original shape of the tray in a highly bowed tray. To successfully minimize bowing it is probably necessary to anneal the tray whenever the first signs of bowing appear, and to repeat as required. Such heat treatment is recommended only for an existing 1100 aluminum tray where temporary or emergency relief from swelling is required.

The second and better way to minimize bowing is to build the N_F tray from a material that is more resistant to void formation than 1100 aluminum. In the past two years we have measured the degree of swelling in a number of irradiated aluminum alloys,⁴¹ with the results shown in Fig. 31. The N_F tray data are shown as 1100 Al (annealed). It is clear that the 6061 alloy in the precipitation-hardened T5 or T6 conditions offers the greatest resistance to swelling. At a given neutron fluence the degree of swelling in 6061-T6 alloy is only about one tenth of that in recrystallized 1100 aluminum. The serviceable lifetime of the tray ought to be prolonged accordingly. The 6061 alloy has given good service in the high flux regions of the HFIR.⁴¹⁻⁴⁴ The 6061 alloy is not amenable to post-irradiation annealing.

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Fig. 31. Swelling in Pure Aluminum and Some Commercial Aluminum Alloys as a Function of Neutron Fluence. Specimens irradiated at about 50°C in HFIR except for the N_F tray specimens (1100 Al, annealed). After Farrell et al. ^{4.1}

REFERENCES

1. S. S. Hurt III, "Experience with Neutron Damage to Beryllium and Aluminum Lattice Components at the ORR," Internal Report, Oak Ridge National Laboratory; Trans. Am. Nucl. Soc. 10, 11-12 (1967).
2. H. A. O'Brien, Jr., Fast and Thermal Neutron Flux Measurements in the ORR and LITR, USAEC Report ORNL-TM-1163, Oak Ridge National Laboratory (1966).
3. F.B.K. Kam, Neutron Flux Spectra Measurement in the ORR, USAEC Report CF-67-8-75, Oak Ridge National Laboratory (1967).
4. J. D. Jenkins, Oak Ridge National Laboratory, private communication.
5. G. W. Cunningham and J. D. Jenkins, CASDAR, A Cross-Section and Neutron Spectrum Direct Access Retrieval System, Oak Ridge National Laboratory Technical Memorandum to be published.
6. M. D. Goldberg, S. F. Mughabghab, B. A. Maguano, and V. M. May, Neutron Cross Sections, BNL-325 (second edition, Suppl. 2) (1966).
7. K. Farrell, J. O. Stiegler, and R. E. Gehlbach, "Transmutation-Produced Silicon Precipitates in Irradiated Aluminum," Metallography 3, 275 (1970).
8. M. Hansen, Constitution of Binary Alloys, McGraw-Hill, New York, 1958, p. 132.
9. Handbook of Chemistry and Physics, 52nd ed., R. C. Weast, editor, Chemical Rubber Publishing Company, Cleveland, 1971.
10. Aluminum - Vol. 1 - Properties, Physical Metallurgy and Phase Diagrams, ed. by K. R. Van Horn, American Society for Metals, Metals Park, Ohio, 1967.
11. J. O. Stiegler, K. Farrell, C.K.H. DuBose, and R. T. King, "High Fluence Neutron-Irradiation Damage in Aluminum," Radiation Damage in Reactor Materials, Symposium Proceedings, Vienna, Austria, June 2-6, 1969, IAEA, Vienna, 1969, vol. II, p. 215.
12. K. Farrell, A. Wolfenden, and R. T. King, "The Effects of Irradiation Temperature and Preinjected Gases on Voids in Aluminum," Radiation Effects 8, 107 (1971).
13. A. E. Richt, Oak Ridge National Laboratory, private communication.
14. R. T. King, E. L. Long, Jr., J. O. Stiegler, and K. Farrell, "High Neutron Fluence Damage in an Aluminum Alloy," J. Nucl. Mater. 35, 231-243 (1970).

15. J. T. Houston and K. Farrell, "Void Coarsening in High Purity Aluminum During Postirradiation Annealing," J. Nucl. Mater. 40, 225 (1971).
16. A. Jostsons, E. L. Long, Jr., J. O. Stiegler, K. Farrell, and D. N. Braski, "Annealing of Voids in Aluminum," Radiation-Induced Voids in Metals, Proc. Intern. Conf., Albany, New York, June 9-11, 1971, AEC Symposium Series 26 (1972), p. 363; also ORNL-TM-3494 (October 1971).
17. A. Jostsons and E. L. Long, Jr., "Radiation Damage and the Effects of Postirradiation Annealing in 1100 Grade Aluminum," Radiation Effects 16, 83 (1972).
18. L. M. Howe, The Annealing of Irradiation Damage in Zircaloy-2 and the Effect of High Temperature Irradiation on the Tensile Properties of Zircaloy-2, Atomic Energy of Canada Report, AECL-1024, Chalk River National Laboratory (1960).
19. T. E. Volin and R. W. Balluffi, "Annealing Kinetics of Voids and the Self-Diffusion Coefficient in Aluminum," phys. stat. sol. 25, 163 (1968).
20. A. Seeger and H. Mehrer, "Analysis of Self-Diffusion and Equilibrium Measurements," Vacancies and Interstitials in Metals, Proc. Intern. Conf., Jülich, Germany, September 23-28, 1968, North-Holland Publishing Company, Amsterdam, 1970, p. 1.
21. J. Takamura, "Specimen Size and Quenched-in Lattice Defects," Lattice Defects in Quenched Metals, Intern. Conf. Proc., Argonne National Laboratory, Argonne, Ill., June 15-17, 1964, Academic Press, New York, 1965, p. 521.
22. J. Burke and A. D. King, "A Determination of the Binding Free Energy Between Vacancies and Silicon Solute Atoms in Aluminum Using an Equilibrium Method," Phil. Mag. 21, 7 (1970).
23. E. Ozawa and H. Kimura, "Excess Vacancies and the Nucleation of Precipitates in Aluminum-Silicon Alloys," Acta Met. 18, 995 (1970).
24. S. Ceresara, T. Federighi, and F. Pieragostini, "Recovery of Al-Si Alloys Neutron Irradiated at 78 K," phys. stat. sol. 11, 779 (1965).
25. Alcoa Aluminum Handbook, Aluminum Company of America, Pittsburgh, Pa., 1967, p. 33.
26. K. Farrell and R. T. King, "Radiation-Induced Strengthening and Embrittlement in Aluminum," Metallurgical Transactions, (May 1973).
27. M. F. Ashby, "On the Orowan Stress," Physics and Strength and Plasticity, ed. A. S. Argon, MIT Press, Cambridge, Mass., 1969, p. 113.

28. J. E. Bailey and P. B. Hirsch, Phil. Mag. 5, 485 (1960).
29. D. Kuhlmann-Wilsdorf, Trans. AIME 224, 1047 (1962).
30. P. Coulomb, "Sur le Blocage des Dislocations par des Cavities on de Petits Precipites," Acta Met. 7, 556 (1959).
31. E. D. Hyam and G. Sumner, Radiation Damage in Solids, I., IAEA, Vienna, 1962, p. 323.
32. W. H. Chatwin and E. D. Hyam, Grain Boundary Holes in Irradiated Uranium, UKAEA TRG Report 325 (w), 1962.
33. R. S. Barnes, Nature 206, 1307 (1965).
34. D. R. Harries, J. Brit. Nucl. Energy Soc. p. 74 (January 1966).
35. A. F. Rowcliffe, G.J.C. Carpenter, H. F. Merrick, and R. B. Nicholson, "An Electron Microscope Investigation of the High-Temperature Embrittlement of Irradiated Stainless Steels," ASTM Spec. Tech. Pub. 426, The Effects of Radiation on Structural Metals, p. 161.
36. J. O. Stiegler and J. R. Weir, Jr., "Effects of Irradiation on Ductility," Ductility, Proc. ASM Seminar, October 1967, p. 311.
37. D. Kramer, H. R. Brager, C. G. Rhodes, and A. G. Pard, J. Nucl. Mater. 25, 121 (1968).
38. R. T. King, "Cyclotron Simulation of Neutron-Transmutation Produced Gases in Reactor Cladding and Structural Materials," Intern. Conf. The Use of Cyclotrons in Chemistry, Metallurgy and Biology, Oxford, England, September 1969, ed. by C. B. Amphlett, Butterworths, London.
39. D. Hull and D. Rimmer, Phil. Mag. 4, 673 (1959).
40. M. V. Speight and J. E. Harris, Metal.Sci. J. 1, 83 (1967).
41. K. Farrell, A. Jostsons, E. L. Long, Jr., and J. E. Ferguson, "Swelling in Aluminum and Its Alloys," presented at the 1971 Fall Meeting of Met. Soc. AIME, Detroit, October 1971, to be published.
42. K. Farrell, R. T. King, and A. Jostsons, Examination of the Irradiated 6061 Aluminum HFIR Target Sleeve, USAEC Report ORNL-4139, in press.
43. R. T. King, A. Jostsons, and K. Farrell, "Neutron Irradiation Damage in a Precipitation-Hardened Aluminum Alloy," Effects of Radiation on Substructure and Mechanical Properties of Metals and Alloys, ASTM Symposium, Los Angeles, June 1972, ASTM STP 529, in press.
44. A. E. Richt and K. Farrell, "Examination and Testing of the Irradiated 6061-T6 Aluminum HFIR Hydraulic Tube," work in progress at ORNL.

45. R. T. King, A. Jostsons, and K. Farrell, "Neutron Irradiation Damage in a Precipitation-Hardened Aluminum Alloy," in Effects of Radiation on Substructure and Mechanical Properties of Metals and Alloys, ASTM Symposium, Los Angeles, June 1972, ASTM STP 529, in press.
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