Influence of Neutron Irradiation on Radiation Swelling of Aluminum Alloys

Sh. Alikulov, F. Akhmedjanov, S. Baytelesov, F. Kungurov, U. Salikhbaev
INP AS RUz, pos. Ulugbek, 100214, Tashkent, Uzbekistan

William O’Connor, Bruce Reid, Eric Gauerke Pacific Northwest National Laboratory – USA

ABSTRACT

Influence of neutron irradiation on radiation swelling of aluminum alloys of two types: 97.8Al+0.9Mg+0.8Cu and 96.8Al+2.4Mg+0.3Cu, samples which had right forms: parallelepiped, cylinder and disk were studied. The samples were irradiated with fast neutrons in vertical channels of WWR-SM research reactor of INP AS RU (fluences of $1.3\cdot10^{18}$, $1.5\cdot10^{19}$ and $1.2\cdot10^{20}$ n/cm²). It was discovered, that irradiation leads to the change of linear dimensions of samples and corresponding radiation swelling. Relative volume change of samples was calculated and concentration of excess vacancies, generated by fast neutrons, was assessed. Mechanism, limiting maximum size of pores in irradiated samples, conditioned by surface tension is offered. It was presented, that in wide range of fluences of fast neutrons super-linear dependence of swelling of samples from dose was observed. Observed deviation from linear dependence could be caused by deposit of interstitial atoms.
Influence of neutron irradiation on radiation swelling of aluminum alloys

Sh. Alikulov, F. Akhmedjanov, S. Baytelesov, F. Kungurov, U. Salikhbaev
INP AS RUz, pos. Ulugbek, 100214, Tashkent, Uzbekistan

William O’Connor, Bruce Reid, Eric Gauerke
Pacific Northwest National Laboratory, USA
e-mail: salikhbaev@inp.uz

SUMMARY

Influence of neutron irradiation on radiation swelling of aluminum alloys of two types: 97.8Al+0.9Mg+0.8Cu and 96.8Al+2.4Mg+0.3Cu, samples which had right forms: parallelepiped, cylinder and disk were studied. The samples were irradiated with fast neutrons in vertical channels of WWR-SM research reactor of INP AS RU (fluences of 1.3 \times 10^{18}, 1.5 \times 10^{19} and 1.2 \times 10^{20} \text{n/cm}^2). It was discovered, that irradiation leads to the change of linear dimensions of samples and corresponding radiation swelling. Relative volume change of samples was calculated and concentration of excess vacancies, generated by fast neutrons, was assessed. Mechanism, limiting maximum size of pores in irradiated samples, conditioned by surface tension is offered. It was presented, that in wide range of fluences of fast neutrons super-linear dependence of swelling of samples from dose was observed. Observed deviation from linear dependence could be caused by deposit of interstitial atoms.

INTRODUCTION

Aluminum alloys have good resistance to radiation and are widely used as a construction material for devices operating under the influence of high doses of ionizing radiation and high temperatures. This leads to changes in their structure and the corresponding change in many parameters defining operational performance materials, such as elasticity, thermal conductivity, electrical conductivity, etc. Simultaneously, the high neutron fluxes irradiated in structural materials nucleate and grow vacancy pores, which leads to a noticeable increase in the volume of material - swelling radiation [1,2]. Vacancy porosity is formed in almost all metallic materials under the action of radiation in the temperature range (0.3-0.5) Tm. Thus, in [3] it was shown that the amount of austenitic steels irradiated at an operating temperature of 450 °C linearly increases with a neutron flux and may increase by 20% or more. Swelling is particularly enhanced by the cluster in the micropores of gases formed during irradiation. Practical interest in the phenomenon of radiation swelling and radiation creep of structural materials, working in the fields of ionizing radiation, due to the danger of the consequences associated with changes in the mechanical properties of these materials. The emergence and development of radiation in materials porosity, accompanied by swelling of the material may cause instability of structures, and reduce the reliability of their work. From a fundamental point of view of radiation swelling is pronounced manifestation of competitive interaction forces in the defect structure of the crystal. Its study provides information on the interaction of point defects with dislocations, pores, coherent and incoherent boundaries and redistribution of point defects between uniform and non-uniform flow distribution in the different efficiency of structural materials.
METHODS AND RESULTS OF EXPERIMENT

In this paper we studied the impact of neutron irradiation on radiation swelling of aluminum alloys of two compositions: Al + 0.9% Mg + 0.8% Cu and Al + 2.4% Mg + 0.3% Cu. Studies were conducted on samples prepared from these alloys having different correct form: parallelepiped (height 40 mm, width 5 mm), a cylinder (height 40 mm, diameter 5 mm) and a disk (diameter 15 mm, thickness 5 mm). X-ray analysis and the determination of impurities in the initial samples of the alloy produced by X-ray microanalyzer "Jeol" JSM 5910 IV (Japan). Density of samples was determined by hydrostatic weighing on an analytical balance with an accuracy of 0.2%. Linear dimensions were measured with absolute precision micrometer ±2 microns.

Samples were irradiated with fast neutrons in vertical channels at the WWR-SM reactor of INP, Uzbekistan. During irradiation the temperature of the samples did not exceed 60°C. Changes in linear dimensions of the samples were determined under the influence of the integral neutron flux (fluences $1.3 \cdot 10^{18}$, $1.5 \cdot 10^{19}$ and $1.2 \cdot 10^{20}$ n/cm²).

The results of research on the effects of neutron radiation on the linear dimensions of the sample alloy Al + 2.4% Mg + 0.3% Cu are shown in Fig. 1.

Relative changes %

![Graph showing relative changes in linear dimensions of samples](image)

Fig. 1. The relative change in linear dimensions of the samples of aluminum alloys having the shape of a parallelepiped (1 and 4), a cylinder (2, 5) and drive (3, 6) depending on the fast neutron fluence. The solid line shows the variation along the long side of the samples (4-6), the dashed - thickness of samples (1-3).

It is seen that under the influence of neutron radiation changes in linear dimensions in all studied cases is the same, regardless of the shape of the sample. At the same time along the long side of the sample (for disk - its diameter) was first observed shrinkage at a fluence of $1.3 \cdot 10^{18}$ n/cm², and then with increasing doses from $1.5 \cdot 10^{19}$ to $1.2 \cdot 10^{20}$ n/cm² swelling occurs. Along the short side of the samples made in the form of the bar and the cylinder first swelling is observed at a fluence $1.3 \cdot 10^{18}$ n/cm², and then with increasing doses up to $1.5 \cdot 10^{19}$ n/cm², there is a reduction in linear dimension, and further increase of dose to $1.2 \cdot 10^{20}$ n/cm² linear size increases again. Only the sample having a disk shape, there is a difference - at all doses, a
monotonic increase in the linear dimension of the thickness. Almost the same pattern is observed for aluminum alloy samples
Al + 0.9% Mg + 0.8% Cu, except that the sample disk shrinkage fluence $1.3 \cdot 10^{18}$ n / cm$^2$ is not observed at all dose levels and there is a monotonic increase in the linear dimension of the diameter.

Thus, the effect of neutron irradiation on the linear dimensions of the tested aluminum alloys depends not only on the composition and concentration of the impurities but also the size and shape of the samples. Regardless of the shape of the sample, the linear size of its long side under the influence of neutron radiation, first decreases and then increases with increasing neutron fluence. Along the line of small sample size (5 mm) resizing occurs only in the region of swelling irrespective of the shape and composition of the impurities.

Results were used to calculate the relative change in the volume of the samples $\Delta V / V_0$ exposed to neutron radiation. Calculation was performed using the relation, which is easily derived from mathematical formulas for the volume of the investigated bodies, and it is common for them

$$\Delta V / V_0 = [(1+A)(1+B)^2 - 1],$$

where $A = \Delta L / L_0$ - the relative change in length $L$ (the disc - is its thickness), $B = \Delta d / d_0$ - the relative change in the thickness of the sample (for disk - its diameter). Calculation results are shown in Table 1 and Fig. 1.

**Table 1. Relative change in volume of the samples of aluminum alloys, $\Delta V / V_0, \%$, depending on the fluence neutron radiation.**

<table>
<thead>
<tr>
<th>Neutron fluence, n/sm$^2$</th>
<th>97,8Al+0,9%Mg+0,8%Cu</th>
<th>96,8Al+2,4%Mg+0,3%Cu</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Bar</td>
<td>Cylinder</td>
</tr>
<tr>
<td></td>
<td>Bar</td>
<td>Cylinder</td>
</tr>
<tr>
<td>$1,3 \cdot 10^{18}$</td>
<td>1,6</td>
<td>1,1</td>
</tr>
<tr>
<td>$1,5 \cdot 10^{19}$</td>
<td>1,2</td>
<td>0,9</td>
</tr>
<tr>
<td>$1,2 \cdot 10^{20}$</td>
<td>1,5</td>
<td>1,1</td>
</tr>
</tbody>
</table>

Swelling $\Delta V / V_0, \%$
Fig. 2. Relative change in volume of the samples of aluminum alloy 96.8Al + 2.4% Mg + 0.3% Cu, having a parallelepiped shape (1), a cylinder (2) and the disc (3) according to the fast neutron fluence.

Figure 2 shows that for all samples irradiated aluminum alloy 96.8Al + 2.4% Mg + 0.3% Cu swelling is observed, increasing from 0.3% to 2% with an increase in fast neutron fluence for samples of 97.8Al + 0.9% Mg + 0.8% Cu swelling occurs almost as well.

Detected radiation swelling of aluminum alloys under the influence of fast neutrons, obviously due to the formation and growth of pores in the crystal volume. The theoretical description of the integral effect of radiation swelling was done in [4], in which it was shown that the pore size distribution has the character of a Gaussian distribution, and due to the simultaneous birth then due to violation of the stability of the system of excess vacancies irradiated material. The reason for the further growth of the pores after the formation of the nucleus is larger than the critical vacancy flow caused by the gradient of vacancies under the influence of neutron radiation. As a result, the authors of [4] obtained an expression for the vacancy concentration at the boundary pores of radius R, due to which the growth of pores and the corresponding swelling of the material:

\[ C_i = C_i \exp \left[ \frac{2\gamma \cdot \Omega}{k \cdot T} \left( \frac{1}{R} - \frac{1}{R_{crp}} \right) \right] \]  \( (2) \)

where \( C_i \) - concentration of excess vacancies away from the pores, \( \gamma \) - surface tension, \( \Omega \) - atomic volume, \( R_{crp} \) - critical pore radius.

During the growth of the vacancy pores on stream time increases due to, firstly, reduce the concentration of vacancies near the pore, secondly, increase the surface pore area. Increasing vacancy flow should lead to a super linear dependence of pore volume of the time.

In [5], the method of small-angle neutron scattering super atomic aluminum alloy structure CAB-1 was studied (the original sample and irradiated with fast neutron fluence), similar in composition to the investigated aluminum alloys. As a result, the irradiated material is detected in a marked decrease in the volume fraction of pore radius of 30-50 nm, which is largely compensated by increasing the share of the total pore radius less than 20 nm.

Using the data [5], and Table 1, it is easy to determine the excess amount attributable to the small and large pores. Then, assuming that the observed swelling caused by the formation and growth of pores in the volume of the entire sample, it is possible to estimate the concentration of excess vacancies \( C_i \), due to which, according to the model [4] and there is swelling materials. The radius of the vacancies in the studied aluminum alloys and their volume is determined by analogy with the work [6]. The result is a value of the expected concentration of excess vacancies arising under the influence of neutron radiation \( C_i = 7 \cdot 10^{21} \text{ mol}^{-1} \), which is the estimated and true order of magnitude.

According to [5], the main sinks of vacancies in aluminum alloys are nano-sized pores with characteristic dimensions of 5-10 and 30-50 nm. In this case, the vacancy flow on time increases as the area of the pores and at first there is a further increase in the pores [4]. In an effort to minimize its free energy, the boundary layer pores creates a pressure difference from different sides and provides a dynamic equilibrium due to additional pressure \( \Delta P \), due to surface tension and counteracts the pressure generated site and interstitial atoms. This additional pressure depends on the mean curvature of the surface at this point and is given by the formula of Laplace. For a spherical cavity the radius R of the formula is written is known as:

\[ \Delta P = \frac{2 \cdot \gamma}{R} \]  \( (3) \)

Were \( \gamma \) – the surface tension.
Obviously, with increase of pore size $\Delta P$ additional pressure decreases and when it reaches a certain critical radius, large pores are broken into a number of smaller clusters. This conclusion is consistent with the data [5], in which, after irradiation with fast neutrons aluminum alloy SAV-1 decrease in the proportion of large pores was observed with radii of 30 - 50 nm, and increased the proportion of pores with radii of 5 - 20nm. However, the mechanism for this behavior vacancy voids in [5] is not specified.

Theoretical studies [1, 7] point to a linear dependence of the effect radiation swelling of the radiation dose, at the steady stage. Our results (Fig. 2) indicate that a fairly wide range of fast neutron fluence dependence of the swelling of the dose superlinear. This fact indicates that the description of the integral effect of radiation swelling must take into account the contribution of interstitial atoms. The flow of interstitial atoms on pore can significantly change the outcome, because near the surface of the pores generated by neutron radiation interstitial atoms have only preferred stock on pore.

REFERENCES