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Letter to the Editors

High-dose irradiation growth kinetics at 448 K in a zirconium alloy

H.C. González, A.M. Fortis *, G.D.H. Coccoz

Laboratorio Daño por Radiación, Departamento Materiales, Comisión Nacional de Energía Atómica, Av. del Libertador 8250, 1429 Buenos Aires, Argentina

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Abstract

The in situ irradiation growth kinetics at 448 K of a Zr–1.5 at.% Al–0.12 at.% ^{235}U recrystallized specimen are presented. An equivalent fast neutron dose of $\sim 1.4\times10^{26}$ n m^{-2} obtained by a self-fission product irradiation technique was achieved. The in situ growth kinetics are compared with the growth strain measurements performed after each reactor shut-down and remarkable differences both at the beginning of the deformation and during its evolution are observed.

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1. Introduction

After many years of nuclear plants operation it has been possible to obtain a lot of data on growth and creep strains from irradiation programs of reactor component materials. Indeed, data from irradiation experiments in materials testing reactors and specialized accelerators could be compared with data from the nuclear industry to give certain parameters which were taken into account to predict the behavior up to 40 yr, i.e. the potential lifetime of nuclear components. Nevertheless, at present there is not a complete theoretical description of the phenomena but only a general belief that the underlying mechanisms in such processes are more complex than previously inferred and dependent on innumerable variables.

Simulation techniques to study radiation effects with energetic particles, i.e. ions, electrons and protons proved to be successful in revealing mechanisms like void formation in steels. These techniques have been applied to zirconium and zirconium alloys to particular problems such as microstructural evolution with the aim of understanding the mechanisms of processes like irradiation creep and growth [1]. The simulation techniques can be used to accelerate damage processes to reach high fluences in short times, as is the case for heavy ion irradiation, although the type and distribution of the damage is not necessarily the same as that produced during neutron irradiation.

One of the feasible methods to accelerate damage processes in an homogeneous way is to produce displacement damage by means of fission fragments generated in a specimen which has been doped with ^{235}U and submitted to thermal neutrons. Although high equivalent fluxes are obtained, this technique does not exactly represent the processes that take place in the case of fast neutron irradiation, since the mechanisms are not the same (e.g. a higher production rate of point defects that enhance the recombination processes). However, some macroscopic phenomena can be clearly observed and measured with high experimental precision [2–4].

The mechanisms producing deformation of irradiated materials, such as growth, creep and void swelling, are temperature dependent and its variations activate processes which can result in a behaviour different than that obtained at a constant temperature. This is shown in works where the effects on measured growth strain of

* Corresponding author. Tel.: +54-11-47547352; fax: +54-11-47547362.

E-mail address: fortis@cnea.gov.ar (A.M. Fortis).

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changing irradiation temperature between 350 and 550 K are studied [5].

Also during cooling from the irradiation to the post-irradiation temperatures, both the intergranular stresses and the modification of the defect structures should produce changes in the deformation measurement. Therefore, a better assessment of phenomena like irradiation growth should be obtained by following the processes during irradiation and not because of post-irradiation measurements.

Therefore, the theoretical analysis of the mechanisms responsible for irradiation growth should take account of the possible effects of temperature changes and time elapsed without neutron flux. In this work, the in situ irradiation growth kinetics of a Zr–1.5 at.% Al–0.12 at.% ^{235}U specimen obtained by a self-fission product irradiation technique are presented. The growth strain kinetics was measured during 23 CNEA-RA3 reactor periods up to an equivalent fast neutron dose of $\sim 1.4 \times 10^{26}$ n m^{-2} and is compared with growth strain measurements taken after each reactor shut-down.

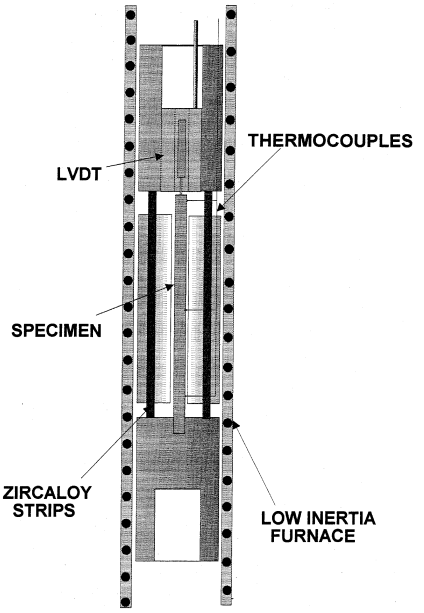

Fig. 1. Schematic view of the measurement device.

2. Experimental

The specimen was taken from a sheet of Zr–1.5 at.% Al–0.12 at.% ^{235}U alloy which had been subjected to a sequence of cold rolling passes up to a 50% reduction in order to obtain a preferential orientation of the grains, the Kearns texture parameter on the rolling direction being $f_L = 0.1$ [6]. A heat treatment of 2 h at 973 K was performed on the specimen after its manufacture in a strip shape, $100 \times 1 \times 3$ mm^3, resulting in recrystallized isotropic grain with mean sizes of $\sim$20 μm. The same alloy was previously utilized to study irradiation growth and it was considered convenient to perform a new study in very different irradiation conditions [7].

The specimen was fastened at the lower end, in the upright position, to the bottom of the measurement device (Fig. 1). The upper part of the specimen is linked to the core of a linearly variable differential transducer (LVDT), whose body is rigidly fixed to the top of the device.

Then, the device with the specimen were placed in the high temperature irradiation facility [8], designed to perform in situ experiments in the CNEA-RA3 experimental reactor. The facility has a low inertia electric furnace in order to compensate for possible temperature variations due to γ-heating during the experiment. The measurement device has temperature homogenizers and the heat transmission by radial conduction towards the reactor refrigerant was assured to avoid significant thermal gradients along the specimen. High purity helium was used as heat transfer gas and all the temperature oscillations caused by power reactor variations were compensated by the electrical furnace. In this way an almost a constant temperature of 448 K was reached; the temperature due to γ-heating alone being 400 K.

2.1. Calculation of damage acceleration

In a previous work the method to obtain the equivalent neutron dose [4,9] was shown. That simple calculation is applied in the present case, although the neutron spectrum and the irradiation position of both experiments are very different. The CNEA-RA3 is operated at a nominal thermal power of 5 MW and the thermal neutron flux, in the assigned irradiation site, is 5.5×10^{17} n m^{-2} s^{-1}, the thermal to fast flux ratio being estimated as 5.5.

The total number of displaced atoms per unit time and volume for incident particles on a target can be approximated by

$$\frac{dN_d}{dt} = \phi N_0 \sigma \upsilon,$$

where ϕ is the flux of the incident particles, N_0 the atomic density of the target material, σ the cross-section for the interaction event and υ is the number of displacements per event.

In these conditions, taking $N_0 = 4.29 \times 10^{22}$ at cm^{-3} for zirconium, $\upsilon = 500$ [10,11], $\phi_{fast} = 1 \times 10^{17}$ n m^{-2} s^{-1} and considering σ equal on the average to the elastic effective cross-section for neutrons of 1 MeV in zirconium, $\sigma_e = 4.5$ barns [12], the production rate per unit volume of atomic displacements due to fast neutrons is

$$\frac{dN_{fast}}{dt} = 9.7 \times 10^{20}\ m^{-3}\,s^{-1}.$$

Finally, considering the thermal flux in the irradiation site, the atomic concentration of ^{235}U, the fission cross-section of ^{235}U, $\sigma_f \sim 583$ barns and 10^5 displacement per event, the production rate of displaced atoms due to the fission fragments in the specimen is

$$\frac{dN_{ff}}{dt} = 1.7 \times 10^{23}\ m^{-3}\,s^{-1}.$$

In these conditions, the ratio between the displacement production rate due to fission fragments and that due to fast neutrons in the irradiated sample, that is, the amplification achieved with a dopant concentration of 0.12 at.% ^{235}U is ≈ 170.

3. Results and discussion

An irradiation lasting ~2300 h, involving 23 periods of ~100 h each, of an annealed specimen of Zr–1.5 at.% Al–0.12 at.% ^{235}U in the experimental reactor CNEA-RA3 was made. Throughout the irradiation, growth strain values were taken every 5 min and from them the measurements performed at temperatures in the range 447.5–449.5 K were only considered. In each period the temperature control was better than ±3 K.

The reactor cycles consisted of 5 days at full power followed by shut-downs of 2 days. During reactor shutdown the temperature of the specimen was set at 323 K and after 48 h the strain was measured. Fig. 2 shows the in situ growth kinetics at 448 K and the post-irradiation growth strains measured at 323 K; on the x-axis the equivalent fast neutron dose obtained by multiplying the fast neutron dose times the amplification factor is represented.

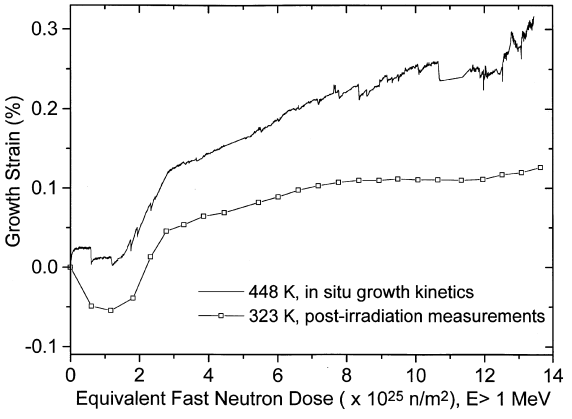

Fig. 2. In situ growth kinetics at 448 K and post-irradiation growth strain measurements at 323 K of the Zr–1.5 at.% Al–0.12 at.% ^{235}U specimen irradiated in the CNEA-RA3.

An increasing in situ growth can be observed after a rather complex initial transient where the growth strain decreases after the first period. The growth rate is $8 \times 10^{-29}\ m^2\ n^{-1}$ up to a fast neutron equivalent dose of $\sim 3 \times 10^{25}\ n\ m^{-2}$ when a nearly linear growth behaviour with a growth rate of $1.8 \times 10^{-29}\ m^2\ n^{-1}$ begins.

The irradiation growth measured at 323 K (125 K below irradiation temperature) referred to the initial value of growth strain at this temperature, shows an initial contraction with dose. It reaches a minimum value of approximately -5.4×10^{-4} at a dose of $\sim 1.2 \times 10^{25}\ n\ m^{-2}$. Then a reversal in the strain direction occurs, the measured strain being again zero at a dose of $\sim 2.3 \times 10^{25}\ n\ m^{-2}$, when a slow continuous change of the growth rate is observed that continues up to a dose of $\sim 1.2 \times 10^{26}\ n\ m^{-2}$, after which an incipient tendency to an accelerated growth is observed.

For the in situ growth kinetics and after a fast neutron equivalent dose of $\sim 1.1 \times 10^{26}\ n\ m^{-2}$ a local increase of the neutron flux resulted in an increase of the γ-heating in the specimen. Therefore, it was necessary to reduce the electrical heating to maintain the specimen temperature constant, i.e. the temperature distribution was changed. This caused a difference in the relative expansion of the specimen with respect to the measurement device, resulting in strain values lower than those recorded up to the above-mentioned dose, which cannot be attributed to a variation of the growth rate. For the post-irradiation measurements at 323 K the thermal conditions remained invariable.

From a fast neutron equivalent dose of $\sim 1.2 \times 10^{26}\ n\ m^{-2}$, an increase on the growth rate of both the in situ growth kinetics at 448 K and the post-irradiation growth measurements at 323 K can be observed, suggesting an onset of growth breakaway. This effect was not previously reported at these temperatures.

The most important observation is the clear-cut distinction in the growth behaviour of the specimens according to the way of performing the strain measurements: during irradiation or post-irradiation. The growth rate obtained for the in situ measurements is different to the classical behavior of the lower growth rate reported for recrystallized materials [13]. Furthermore, as it can be seen in Fig. 2, in situ growth strains are more than 100% that of those obtained in the post-irradiation measurements, under the present experimental conditions.

The temperature jump ($\Delta T \sim -125$ K) undergone by the specimen between the irradiation and post-irradiation temperatures, corresponding to different recovery stages, seems to result in a modification of the intergranular stresses generated during the deformation as well as of the defect structures, responsible for the dimensional variations.

Growth kinetics measurements in specimens with other mechanical treatments and at temperatures near to

those of the power reactor operation are in progress with the aim of validating different theoretical models proposed to predict the in-service dimensional behavior of the reactor components.

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