The Growth of Grain Boundary
Cavities Under Applied Stress
and Internal Pressure

by

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Abstract:

Experiments were performed to investigate the growth of grain boundary cavities in nickel under internal pressure and applied stress. Under the experimental conditions the growth kinetics was found to be controlled by stress induced mass transfer. The growth rate was found not to be a linear function of the applied stress. The growth rate was found to depend on grain boundary orientation and grain boundary sliding.

Introduction

This paper reports an experimental investigation of cavity growth at grain boundaries in nickel under internal gas pressure and applied tensile stress. Cavity growth under internal gas pressure is an important damage mechanism during hydrogen attack at elevated temperatures [1], while cavity growth under applied tensile stress is an important damage mechanism during creep at lower stresses and can lead to intergranular failure [2].

This work addresses cavity growth due to stress induced diffusion. The kinetics of this phenomenon have been considered by several investigators, who have shown that the driving force for growth is the normal stress at the grain boundary. They concluded that the radial growth rate should be linearly proportional to the applied stress [3,4,5]. However, this dependence has not been observed experimentally even at low stress where cavity growth due to grain boundary shearing is not important [6]. A major objective of this experiment is to investigate the reported non-linear stress dependence under conditions where diffusion controlled cavity growth is important. It is expected that the results of this work will be applicable to the development of hydrogen attack damage models as well as to creep-to-rupture analyses.

In order to investigate cavity growth under applied stress without the influence of cavity nucleation, we have prepared nickel specimens with pre-existing cavities for the growth studies. The initial cavities were produced by exposing the specimens to high pressure (3.5 - 14.5 MPa) hydrogen gas at elevated temperatures (300-500°C). Growth rate was determined for a number of combinations of applied stress and temperature. Special attention was given to determining how the growth rate varied with the angle between the grain boundary surface and the applied stress axis.

Experiment

The detailed description of the experimental procedure is given in Reference [7]. Nickel 270 specimens were fabricated from 2.4 cm diameter bar stock obtained from International Nickel Inc. The bar was swaged to a diameter of 0.36 cm and recrystallized to achieve the desired grain size. The final specimen length was approximately 6 cm for growth studies without applied stress and 3.1 cm for growth studies under an applied stress.

Cavities were produced by methane reaction resulting from exposing the nickel specimens containing dissolved carbon to high pressure hydrogen gas at 500°C [1]. Two types of exposure conditions were used. One produced grain boundary cavities that contained excess gas pressure which provided a driving force for continued cavity growth upon subsequent annealing in the absence of a hydrogen environment. The other set of exposure conditions yielded cavities with an equilibrium gas pressure. These cavities grew only under an applied stress in the absence of a hydrogen environment.

Growth studies without applied stress were performed by vacuum anneals at 500, 550 and 600°C with the internal methane pressure of the cavity as the driving force for growth. For each experimental sequence at a given temperature a single 6 cm rod was used. A section of the rod was removed at different time intervals to determine cavity sizes as a function of time. The cavity cross-sectional areas were determined from intergranular fracture surfaces by scanning electron microscopy (SEM). The detailed method of preparing the fracture surfaces is described in Reference [7]. A typical SEM photomicrograph of cavities on a grain boundary is shown in Figure 1. An example of the cavity size distributions obtained is shown in the histograms in Figure 2. More than 100 cavities were counted for the determination of an average cavity size.

The study of cavity growth under applied tensile stress employed specimens that had a 3.1 cm gauge length and contained cavities with an equilibrium methane gas pressure. The experiments were performed under constant load at 500 and 550°C. Strain of the specimen was monitored during the experiments. The amount of cavity growth due to the applied stress was determined by comparison of cavity cross-sectional areas in the stressed and unstressed portions of the specimens. Stereographic SEM of the fracture surface was used to determine cavity size as a function of the grain boundary orientation as determined by a stereographic technique [7,8].

Results and Discussion

1. Cavity Growth Under Internal Pressure

Experimental data obtained from the vacuum annealing experiments are shown in Figure 3 which contains a plot of the mean cross sectional area of the cavities against a reduced (annealing) time, Dt , where D is the grain boundary diffusion constant [9] and t is the time at the annealing temperature. The time dependence of cavity size suggest diffusion controlled growth. In the same plot, the calculated growth curves based on the diffusion controlled growth model are shown, whereby the rate of growth is expressed by [4,5]:

$$\frac{dr}{dt} = \frac{wD}{kT} \left(\frac{\Omega}{r^2} \right) \frac{(\sigma_n - [\frac{2\gamma}{r} - P][1 - (\frac{2r}{a})^2]) [(\frac{2r}{a})^2 - 1]}{2 [\frac{3}{4} + \frac{1}{4}(\frac{2r}{a})^4 + \ln(\frac{2r}{a}) (\frac{2r}{a})^2]}$$

where

D is the grain boundary diffusion coefficient

w is the width of the grain boundary

r is the radius of the cavity

σ_n is the normal stress on the grain boundary

P is the internal gas pressure in the cavity

γ is the surface tension

- Ω is the atomic volume
- a is the center-to-center cavity spacing
- k is Boltzman's constant
- T is the absolute temperature

The internal pressure has been estimated by using the ideal gas law, the asymptotic value of the cavity size for long annealing times and the equilibrium gas pressure relationship, $P = \frac{2\gamma}{r}$. The values of D and γ were obtained from the literature [9,10]. The solid lines represent numerical integrations of the growth rate over time. The comparison between the calculated curves and the data suggests that stress induced diffusion is the cavity growth mechanism. More important to the purpose of the present work this comparison supports the validity of using the values of D and γ found in the literature for the calculation of cavity growth under applied stress.

2. Cavity Growth Under Applied Stress

2a. Average Cavity Growth Rate

Cavity growth rates averaged over all grain boundary orientations are plotted versus the applied tensile stress in Figures 4 and 5 for the two test temperatures. These data exhibit a non-linear stress dependence. The conditions of the experiments were such that grain boundary shear was not expected to be an important mechanism for cavity growth [7]. The solid lines in the same figures are growth predictions based on Equation 1, the values of D and γ established in the previous section and the assumption that $\sigma_n = T/2$ where T is the applied tensile stress and $T/2$ is the average normal stress on the grain boundaries. The data in Figs. 4 and 5 suggest that the agreement between the model predictions and the experimental results improves as the applied stress increases. These data suggest also the possibility that the linear stress dependence expected from the growth model (Equation 1) will be exhibited for stresses higher than those employed in the present work, provided other growth mechanisms do not become important.

In the next section we will attempt to show how the non-linear stress dependence observed in this work is related to grain boundary sliding as suggested by the analysis of the effect of grain boundary orientation on the cavity growth rate.

2b. Effects of Grain Boundary Orientation

It is well established that grain boundary sliding is an important deformation mechanism at low strain rates for creep at temperatures near one-half the melting point. Under the same conditions one usually observes intergranular failure with reduced ductility in a creep-to-rupture test. The existence of grain boundary sliding has been demonstrated in specimens used to obtain data for Figures 4 and 5. A detailed discussion of the evidence for grain boundary sliding is given in Reference 7. The most direct indication of the existence of grain boundary sliding in these specimens was the emergence of grain boundary steps on the surface of the specimen after creep.

Grain boundary sliding is believed to produce stress concentrations at grain corners. A schematic representation of the expected stress concentration (taken from Reference 11) is shown in Figure 6. Qualitatively the stress concentrations that are produced by sliding will lead to redistribution of the stresses at the grain boundaries such that the grain boundary normal stresses will be reduced. At higher stresses where grain boundary sliding is not important the normal stress at the grain boundary will essentially be equal to $T \cos^2 \theta$ where θ is the angle between the grain boundary normal and the tensile stress axis and T is the applied uniaxial stress. The above relation suggests the maximum cavity growth to occur at grain boundaries lying perpendicular to the applied stress axis ($\theta = 0$), while no growth should occur at boundaries parallel to the stress axis ($\theta = 90^\circ$).

Using the $\cos^2\theta$ dependence for σ_n and Equation 1 with the values of D and γ discussed previously we can calculate the cavity growth rate as a function of the grain boundary orientation (θ) at a given temperature and applied stress. The results of this type of calculation are shown as solid lines in Figures 7 and 8, in plots of cavity growth rate versus grain boundary angle. Cavity growth data are also shown in the same figures.

The data from Figure 7 were obtained from the same specimen used to measure the average cavity growth rate at the highest stress in Figure 5. We have pointed out in a previous discussion that the agreement between the measured growth rate and the model predictions improves at this stress level. This observation is consistent with the data in Figure 7 where the measured orientation dependence of the cavity growth rate approximates that predicted by a $\cos^2\theta$ dependence of the normal stress. This agreement supports diffusion controlled cavity growth. This type of orientation dependence suggests that, at the higher stress levels, grain boundary sliding is not important and that significant redistribution of the stress at the grain boundaries will not occur.

In contrast the data in Figures 8a and 8b were obtained at lower stresses shown in Figures 4 and 5 where the average cavity growth rate differs significantly from the model predictions. The observed deviations in the direction of lower growth rates compared to the calculated values in Figure 4 and 5, are believed to be a consequence of the measured orientation dependence in Figures 8a and 8b. The measured cavity growth rate at lower stresses in these figures shows essentially none of the orientation dependence predicted by using the geometrical factor $\cos^2\theta$. These data suggest that grain boundary sliding is more important at lower stresses such that stress redistribution will occur leading to the observed results. We may conclude that the non-linear stress dependence of the cavity growth rate observed here and in reported work [12] is a consequence of

stress redistribution at grain corners due to grain boundary sliding.

3a. Discussion

The results of this work confirm the previous observations that the average cavity growth rate is not a linear function of the applied stress in the stress range where cavity growth is controlled by stress induced mass transport. These results suggest the need to account for the effect of grain boundary sliding on the stress redistribution at the grain boundary so that we can have better cavity growth calculations in order to estimate properly the rate of creep damage accumulation. At present the required model for this effect does not exist. The type of data obtained in this work on the orientation dependence of grain boundary cavity growth can be used to experimentally determine the extent of stress redistribution which can be used as a useful basis for future model development.

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Figure Captions

- Figure 1: SEM photomicrograph of typical grain boundary cavities produced by hydrogen attack of nickel specimens.
- Figure 2: Examples of cavity size (cross-sectional area) distributions observed. The distributions shown above were obtained in a cavity annealing experiment at 500°C. They show that cavities created with greater than equilibrium internal gas pressure will grow upon subsequent anneals in the absence of a hydrogen atmosphere. (T = annealing time and A = mean cavity cross-section).
- Figure 3: Plot of cavity cross-sections versus the reduced annealing time, Dt . Data that were obtained after 500, 550 and 600°C annealing are shown as symbols. The solid lines represent growth predictions based on a diffusional growth model (Equation 1) and the estimated initial cavity gas pressures.
- Figure 4: Plot of average cavity growth rates (cross-sectional area/time) vs. the applied tensile stress for creep tests performed at 500°C. Data are shown as symbols while the solid line represents the results of calculations based on Equation 1 and the average normal stress on the grain boundaries for a given applied stress.
- Figure 5: Plot of average cavity growth rates (cross-sectional area/time) vs the applied tensile stress for creep tests performed at 550°C. Data are shown as symbols while the solid lines represent the results of calculations based on Equation 1 and the average normal stress on the grain boundaries for a given applied stress.
- Figure 6: Two dimensional idealization of the stress concentrations produced by grain boundary sliding in a polycrystalline material [11].

Figure 7: Plot of the stress induced cavity growth rate versus θ (the angle between the grain boundary normal and the applied stress axis) for creep experiments performed at 550°C. These data (circles), obtained at the highest stress shown in Fig. 5, show good agreement with the calculated growth rates based on Equation 1 and the relationship $\sigma_n = T \cos^2 \theta$.

Figure 8: (a,b): Plots of the stress induced cavity growth rate versus θ for low strain rates. Predictions based on Equation 1 and the relationship, $\sigma_n = T \cos^2 \theta$, are shown as solid lines in the figures. These data show poor agreement with the model predictions.

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig.5

Fig. 6

Fig. 7

Fig. 8a

Fig. 8b