CYCLIC DEFORMATION OF ZIRCALOY-4 AT ROOM TEMPERATURE

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Fully annealed metals usually harden during the first cycles of low cycle fatigue. However, fatigued specimens of Zircaloy-4 at room temperature show an evident cyclic softening. From the analysis of the cycles it can be inferred that the unlocking of dislocations from interstitials atoms is responsible for the observed softening.

Keywords: Cyclic softening; Zircaloy-4; friction stress; dislocations.

1. INTRODUCTION

It is well known that fully annealed metals usually show cyclic hardening as a consequence of dislocation multiplication, while hardened metals manifest cyclic softening resulting from the elimination or weakening of the obstacles to dislocation movements. Numerous studies have been devoted to analyze the mechanical behaviour of fully annealed face centered cubic (FCC) and base centered cubic (BCC) materials. Nevertheless, scarce information is available about the cyclic softening / hardening behaviour of hexagonal closed packed (HCP) metals.

In this respect, a pronounced cyclic softening was previously observed in Titanium [1] and then in commercial purity Zirconium and its alloy Zircaloy-4 [2,3]. Some researchers [2] explain this behaviour as associated with the progressive destruction of a planar arrangement of long screw dislocations producing high long range stresses. Studies in annealed Titanium [1], material with similar characteristics to Zirconium in several aspects, report that the cyclic softening of the material has to be caused by the unlocking of dislocations from their interstitials atoms. Therefore, cyclic deformation progressively increases the density of mobile dislocations. Recently, another proposal called “texture-rotation induced cyclic softening” model considers that cyclic softening of Zircaloy-4 [3] would be produced by the rotation of textured crystals to an easier direction for prismatic slip.

The aim of this paper is to clarify the causes of the cyclic softening of Zircaloy- 4 at room temperature.

2. EXPERIMENTAL PROCEDURE

Low cycle fatigue specimens were machined from Zircaloy-4 forged bar. The principal alloyin elements were (in wt %): Sn-1.37, Fe-0.14, Cr-0.10, C-0.01, O-0.14, N-0.004, H-20 ppm, Zr-balance. Samples were tested in a recrystallized condition. The texture of the rod was such that the resolved fraction of basal poles in the tensile direction was less than 0.1. The mean grain diameter was 20 microns.

Total strain controlled cyclic tests with strain ranges 1.0, 1.4 and 2.0 % and constant total strain rate 2 x 10^{-3} s^{-1} were carried out using a fully reversed triangular wave. The tests were performed in air and they were always started in tension.

Thin foils were prepared from sections cut parallel and perpendicular to the tensile axis. The foils were examined in a transmission electron microscope operating at 100 kV.

3. RESULTS AND DISCUSSION

Fig.1 shows the cyclic stress response of Zircaloy-4 at room temperature for different total strain ranges. This figure exhibits the variation of the Peak Tensile Stress with the number of cycles. It should be noticed that, independent of the strain range, all the curves show, after the first cycles of the test, a pronounced softening. For comparison purposes, the tests performed with total strain ranges 1.0 and 1.4 % are represented only up to cycle 1000. A remarkable initial cyclic hardening is present only at the highest total stress amplitudes. It is inferred from this figure that the characteristic cyclic mechanical behaviour of annealed Zircaloy-4 is a pronounced cyclic softening occurring almost from the beginning of the life of the specimen.

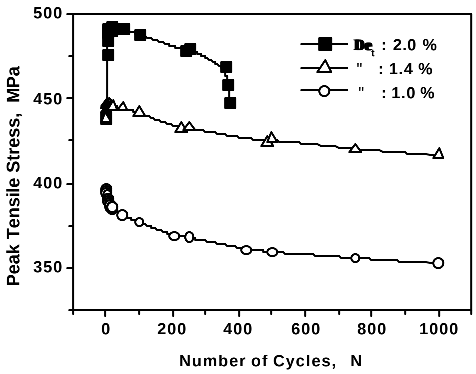

Fig. 1 Cyclic softening observed in Zircaloy-4 at three different total strain ranges.

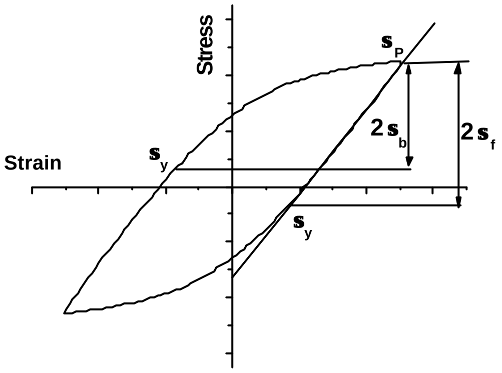

Fig. 2 Method to obtain the Friction and Back Stresses

Moreover, some researchers [4] assume that fatigue softening could be associated with a reduction of the precipitates size during cycling, until they become thermodynamically unstable and revert to the solid solution. If this were the case, the distribution of precipitates in the non-fatigued (threaded part) and fatigued (gauge length) parts of the specimen would be different for a cycled specimen, being less numerous in the gauge length of the specimen due to cycling. This difference was not observed after transmission electron microscopy observations carried out in all the fatigued specimens.

Upon the analysis of the flow stress, as originally suggested by Cottrell [5], the “Friction Stress”, σ_F, and the “Back Stress”, σ_B, calculated from the hysteresis loops, could be obtained. The Friction Stress corresponds to the resistance the dislocations have to overcome to keep moving in the lattice. The Back Stress is associated with piled-up dislocations that were created after overcoming the Friction Stress.

This well-known method is illustrated in Figure 2. At the peak stress, the applied stress σ_P is the sum of the Friction Stress and the Back Stress. On lowering the applied stress, the Friction Stress will oppose the backward motion of dislocations. Reversed plasticity will be obtained when the applied stress, σ_y, aided by the Back Stress, can overcome the Friction Stress. Information on the types of obstacles to dislocation movement can be obtained from the measure of these stresses as shown in Fig. 2.

The variation of the Friction and Back Stresses with the number of cycles for the different total strain amplitudes is shown in Figs. 3 and 4. The yield stress, σ_y, was determined from a least-squares fit for each hysteresis loop. Due to the inaccuracy of the method this value of stress presents a scatter band which was represented by error bars in the diagram.

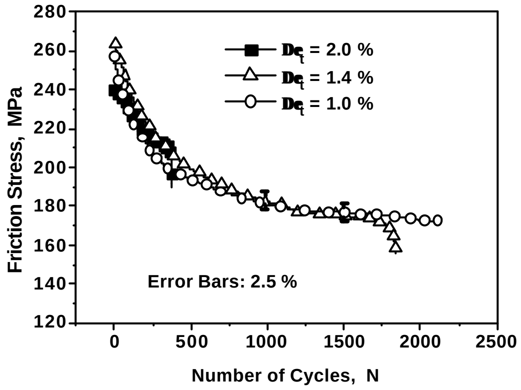

Fig.3 Stress with Cycles in Zircaloy-4

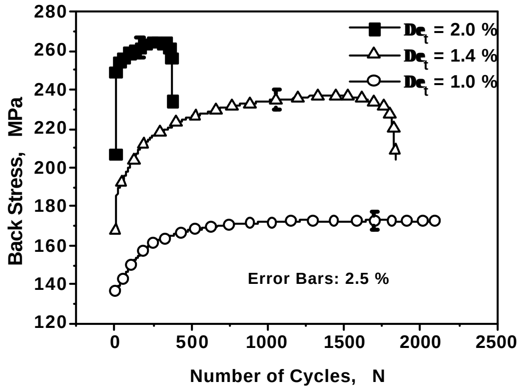

Fig. 4 Variation of the Back Stress with Cycles in Zircaloy-4

It should be noticed, (Fig. 3), that the Friction Stress appears to be almost independent of the total strain range of the test. This effect could be rationalized thinking that the Friction Stress is equivalent to the yield stress in a uniaxial tensile test. It will depend on the obstacles that the initial internal structure of the metal imposes to the dislocation movement. These obstacles can be the lattice friction, precipitated particles, other dislocations and foreign atoms. On the contrary, the Back Stress depends on the obstacles that are created by the dislocations movement such as pile ups. Therefore, the larger the total strain range, the larger the amount of dislocations piled-up. As a result of this effect, the Back Stress will be also larger. Additionally, Fig. 3 shows that the Friction Stress decreases with the number of cycles and that such a decrease is also independent on the imposed total strain range. This is a striking result is incompatible with the "texture-rotation induced" model. The rotation of the crystals as a result of cyclic deformation should be dependent on the applied plastic strain which, in the case of cyclic deformation, is represented by the cumulative plastic strain, $4\ \gamma_P\ N$, where γ_P is the plastic strain amplitude of the cycle and N the number of cycles. For a certain amount of cycles, tests with larger plastic strain amplitude will produce a larger cumulative plastic strain causing a larger rotation of the crystals; hence, a larger total strain range should produce a larger decrease rate in the Friction Stress. It should be emphasized (Fig. 3) that the microstructure, on which the Friction Stress behaviour depends, becomes softer with cycles but independent on the plastic strain amplitude. On the other hand, the Back Stress is strongly dependent on the plastic strain amplitude at the beginning of the test. As cycling proceeds no remarkable difference is observed for the three total strain ranges. The increase of the Back Stress could be attributed to the increased amount of dislocations produced during cycling.

The cyclic behaviour of the Peak stress curve observed in Fig. 1 is the result of the behaviour of the Friction and Back Stresses. Thus, in Zircaloy-4 the initial cyclic hardening observed at the larger total strain ranges seems a consequence of the Back Stress meanwhile the softening behaviour exhibited in all the amplitudes appears as a consequence of the Friction Stress.

Interrupted tests with intermediate annealing were performed so as to analyze the influence that thermal treatment has on the cyclic behaviour of the material. Fig. 5 shows the Peak and the corresponding Friction and Back Stresses behaviour during an interrupted test. The specimen was cycled up to near saturation, point A in Fig. 5, annealed in vacuum at 573K during one hour and cycled again up to point B in the figure. At that point the sample was annealed in vacuum at 473K during one hour and cycled again. It is important to notice that the heat treatment at 573 K increased the Peak Stress to a value similar to that of the flow stress corresponding to the first cyclic loading. Nevertheless, after the 473 K thermal treatment only a small disturbance was present in the cyclic behaviour.

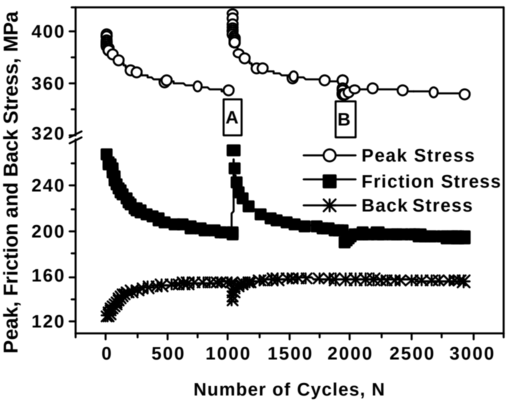

Fig. 5 Peak, Friction and Back Stresses for Zircaloy-4 cycled at Room Temperature with test interruptions.

Annealing treatments at 573K could produce atomic movements and dislocation recovery but not a significant change in the texture [6]. With this in view, the model based on texture changes does not seem to be appropriate to explain the behaviour observed in the Friction Stress after thermal treatments. At 573 K, corresponding to $0.27\ T_M$ (melting temperature of the material), the envisaged mechanisms are significant migration of interstitial solute atoms to sinks, such as edge dislocations, and some recovery through decreasing dislocation density. As annihilation of dislocations by recovery processes do decrease the flow stress, the increase in the flow stress shown in Fig. 5 is not consistent with this process. The mechanism proposed in this paper is related to the movement of interstitial oxygen atoms which initially pin the dislocations in the annealed metal. At the beginning of the test, the plastic deformation is accommodated by only a few dislocations. During cycling not only new dislocations are created, but also existing ones are unlocked from their pinning points. Thus, the increase of the density of mobile dislocations would produce the softening observed in the material. The arrangement of the interstitial solute atoms is unchanged during cycling. Solute atoms will be immobile in the low temperature range but on increasing the temperature, interstitial atoms would become mobile in order to decrease the free energy of the metal pinning the dislocations again. The longest distance the atoms should travel to catch the dislocations again is half the average distance between two interstitial atoms. This distance can be calculated through the concentration of oxygen atoms. The concentration of oxygen for the present alloy is 0.14 weight per cent that corresponds to 8000 atomic parts per million, that is, one interstitial atom per 125 atoms of matrix. Thus, the average distance between two atoms of oxygen is 5 atoms of matrix. So, the longest distance the pinning atoms must travel to catch the dislocations again should be between two and three atomic distances. The relation between the travelled distance $\overline{x}$ in the time t and the diffusion coefficient D is given by [7]:

$$\overline{x} = 2 \cdot \sqrt{D \cdot t} \qquad (1)$$

Upon the results obtained by Ritchie et al. [8] for temperatures up to 923 K the diffusion coefficient can be expressed as:

$$D\,[\mathrm{cm^2/s}] = 0.66 \exp\,(-44000/RT) \qquad (2)$$

and is attributed to the jumps of oxygen interstitial atoms in the basal plane.

Taking into account (1) and (2) the necessary temperature for the atoms to travel a distance of three atomic distances (ϕ (Zr) = 0.32nm) in one hour should be 530 K. The results of this paper indicate that, after a thermal annealing at a temperature between 473 and 573 K, a cycled sample of Zircaloy -4 will recover the value of Friction Stress it had at the beginning of the test, Fig. 5.

The analysis of the results explained above lead to the conclusion that the cyclic softening, which takes place in Zircaloy-4, is a consequence of the decrease in the Friction Stress caused by the unlocking of dislocations from oxygen interstitials solute atoms present in the annealed metal. Considering this mechanism, it can be rationalized the results observed in Fig. 3 where the decrease of the Friction Stress appears as almost independent of the total strain range. Indeed, the Friction Stress in cyclic tests is equivalent to the yield stress in uniaxial tensile test. To start moving dislocations in the lattice, the applied stress must be, at least, equal to the Friction Stress in each cycle. Similar to the yield stress in uniaxial tensile tests, the Friction Stress is also independent on the applied total strain. As was mentioned above, the Friction Stress depends, principally, on short range obstacles which are known to produce a dependence of the stress on the strain rate and temperature but not on the amplitude of the test. However, if dislocations or segments of dislocations are unpinned in each cycle, the Friction Stress will be reduced for the next cycle due to an increase in the density of mobile dislocations. The amount of unlocked dislocations or segments of dislocations in each cycle would not depend on the plastic strain amplitude of the test. As cycling proceeds more dislocations are unlocked from their pinning points reducing the Friction Stress of the next cycle. If, by mean of thermal energy, the interstitial solute atoms have the possibility to move, they will travel up to the nearest dislocation in order to decrease the elastic energy of the lattice and to anchor the dislocations again.

4. CONCLUSIONS

- Zircaloy-4 shows a pronounced cyclic softening which agrees with previous results found in the literature.
- The cyclic softening is caused by the decrease of the Friction Stress. The unlocking of dislocations from their oxygen interstitial solute atoms during cycling is responsible for such behaviour.

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