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Cyclic softening mechanisms of Zircaloy-4

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Abstract

Cyclic tests with different total strain amplitudes were conducted on Zircaloy-4 at room temperature. The results show that cyclic softening behaviour is the main mechanical feature observed in this material. A detailed analysis of the hysteresis loops was conducted in order to determine the friction and back stresses. The friction stress corresponds to the resistance which the dislocations have to overcome to keep moving in the lattice. The back stress depends on the density of long-range impenetrable obstacles that are created by the dislocations movement such as pile-ups. The cycling softening that Zircaloy-4 exhibits is related with the decrease of the friction stress. Moreover, the independence of the friction stress behaviour on the total strain amplitude as well as the influence of thermal treatment on interrupted cyclic tests permit the conclusion that the unlocking of dislocations from oxygen atoms is responsible for the observed softening.

1. Introduction

Typically, the cyclic response of most engineering alloys exhibits either a cyclic hardening or softening behaviour depending on the microstructure existing in the material. Well-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. Well-annealed face-centred cubic (fcc) and body-centred cubic (bcc) alloys present a significantly different mechanical behaviour that was related [1–3] to the particular nature of the screw dislocation in bcc metals. Screw dislocations in bcc metals have an extended core structure with a three-fold symmetry and are sessile at low temperature [4]. Compared to the volume of research done on the fatigue of fcc and bcc metals, much less information is available on the cyclic response of hexagonal closed-packed (hcp) metals. In hexagonal transition metals such as Ti and Zr the core of a screw dislocation has also, as in bcc metals, a three-dimensional structure that preferentially moves in the prismatic plane with a high lattice friction. As in bcc metals, the deformation characteristics of these particular hcp metals are strongly influenced by impurity and interstitial contents.

The zirconium alloys Zircaloy-2 and Zircaloy-4 are hcp metals used as the cladding material of nuclear fuel rods in light water reactors. Despite the vast amount of results reported in the literature as a consequence of their importance, it is surprising that some contradictions still remain. In effect, recrystallized Zircaloy-4 was reported to display cyclic hardening [5,6] and also cyclic softening [7] when cycled with different total strain amplitude. For certain strain amplitudes, commercial purity zirconium and titanium, material with similar characteristics to zirconium in several aspects, show a very pronounced cyclic softening stage [8,9]. Up to date, the mechanisms producing this cyclic softening behaviour at room temperature still remain unclear. Some authors [8] suggest the thermally activated overcoming of interstitial solute atoms as the rate controlling mechanism. Other researchers [9] explain this behaviour as associated with the progressive destruction of a planar arrangement of long screw dislocations producing

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high long-range stresses. Consequently, the cyclic deformation progressively increases the density of mobile dislocations. In recent years, the so-called 'texture-rotation induced cyclic softening' model was proposed from studies performed on Zircaloy-4 [7]. After this model, the cyclic softening of the material would be produced by the rotation of textured crystals to an easier direction for prismatic slip.

The purpose of this paper is to give a further insight into the cyclic behaviour of Zircaloy-4 at room temperature, providing some experimental results from tests performed with different total strain ranges and intermediate thermal treatments.

2. Experimental procedure

From 16 mm diameter Zircaloy-4 bars, prepared in accordance with ASTM B550 Grade 704, cylindrical fatigue specimens were machined with a uniform gauge section of 5 mm diameter and 10.5 mm length. The chemical composition of the alloy is (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 fraction of basal poles in the tensile direction was almost unnoticeable. The mean grain diameter was 20 μm.

Total strain controlled cyclic tests with strain ranges 1.0, 1.4 and 2.0% and constant total strain rate 2×10^{-3} s^{-1} were carried out with an electromechanical Instron model 1362 machine using a fully reversed triangular wave form. The tests were performed in air and they were always started in tension. The hysteresis loops were digitally recorded at least with 200 points per loop.

In order to correlate the mechanical behaviour with the dislocation structure of fatigued specimens, thin-foil discs were prepared from sections cut parallel to the tensile axes. The foils were examined in a transmission electron microscope (TEM) operating at 100 kV.

Texture results were obtained by measuring $\{0002\}$, $\{10\bar{1}0\}$, $\{10\bar{1}1\}$, $\{10\bar{1}2\}$, $\{10\bar{1}3\}$ and $\{11\bar{2}0\}$ pole figures by reflection X-ray diffraction in a Philips MPD diffractometer equipped with an Eulerian cradle and CuKα radiation. Such profuse number of pole figures (PF) allowed the calculation of Orientation Distribution Functions and recalculated PFs with a high degree of accuracy and confidence using different starting sets. The analysis was carried out using popLA software [10].

3. Results

The cyclic stress response of Zircaloy-4 at room temperature for different total strain ranges is shown in Fig. 1. This figure shows, in linear scale, the variation of the peak tensile stress with the number of cycles. It is relevant to remark that all the curves show, after the first cycles of the test, a pronounced softening independent of the strain range. For comparison purposes, the tests performed with total strain ranges 1.0% and 1.4% are represented only up to cycle 1000. An initial cyclic hardening, depending on the amplitude of the test, is observed for total strain ranges 1.4% and 2.0%. It is evident from Fig. 1 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.

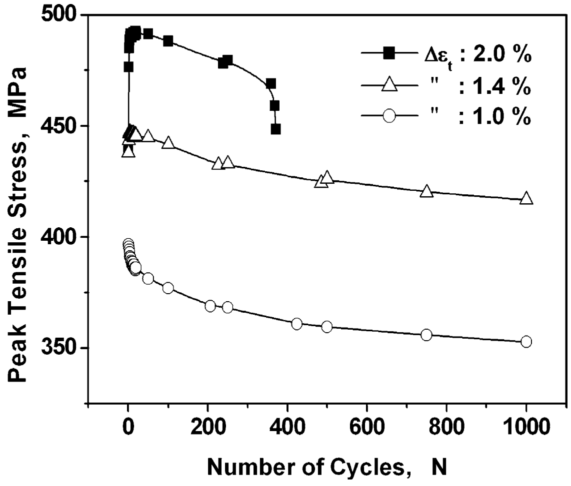

Fig. 1. Cyclic softening observed in Zircaloy-4 at three different total strain ranges.

From the analysis of the pole figures obtained from texture measurements of non-fatigued (threaded part) and fatigued (gauge length) parts of a sample cycled with total strain range 1.0% up to 2000 cycles, no difference was found that could confirm the rotation of the grains as proposed by the 'texture rotation induced model' [11].

A detailed analysis of the flow stress, as originally suggested by Cottrell [12] and employed by Kuhlmann-Wilsdorf and Laird [13] as well as Handfield and Dickson [14], was used to determine the mechanisms responsible for the softening observed in this material. Upon this method, the flow stress obtained from the hysteresis loops is the result of two kinds of resistance to plastic deformation: the 'friction stress', σ_F, and the 'back stress', σ_B. The friction stress corresponds to the resistance which 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 Fig. 2. At the peak stress, the applied stress σ_P is the sum of the friction stress and the back stress. On lowering the ap-

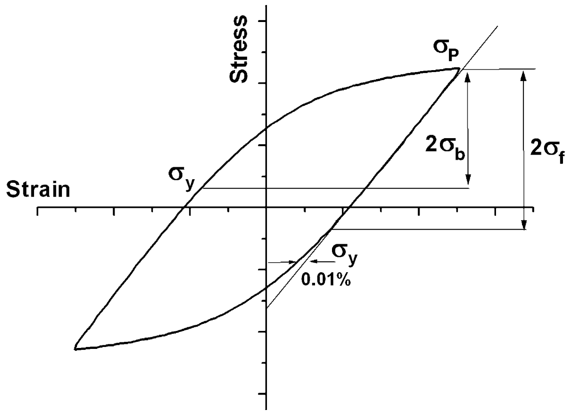

Fig. 2. Method to obtain the friction and back stresses.

plied stress, the friction stress will oppose the backward motion of dislocations. Reversed plasticity will be obtained when the applied stress, yield stress σ_y, aided by the back stress, can overcome the friction stress. The friction stress and the back stress are simply determined as follows:

$$\sigma_P = \sigma_f + \sigma_b, \tag{1}$$

$$\sigma_y = \sigma_f - \sigma_b. \tag{2}$$

The asymmetry found in the hysteresis loops of this material was in all cases lower than 8%, with the flow stress in compression higher than that in tension.

Information on the kind of obstacles to dislocation movement can be obtained from the measurement of these stresses as given in Fig. 2. The figure shows a hysteresis loop on which is drawn a straight line (determined from a least-squares fit) coincident with the most linear part of one side of the loop. The value of the yield stress, just as in monotonic tensile tests, is somewhat arbitrarily determined. In the present analysis, σ_y for each hysteresis loop was defined as the stress corresponding to an offset of the loop of 0.01% strain (indicated in Fig. 2) with the drawn line representing the elastic part of the loop. This analysis was performed on each digitally recorded loop (more than 200 points per loop) by a computer program.

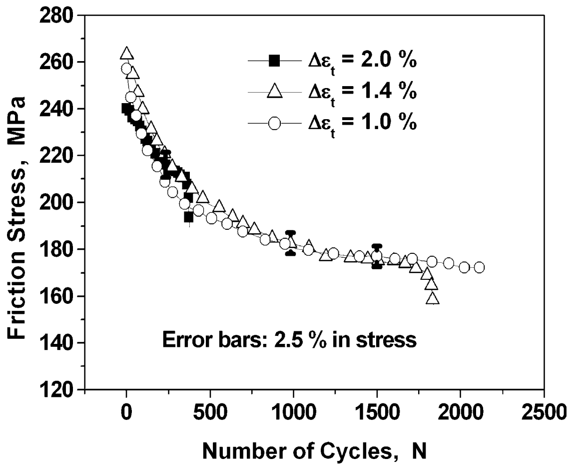

Fig. 3. Variation of the friction stress with cycles in Zircaloy-4.

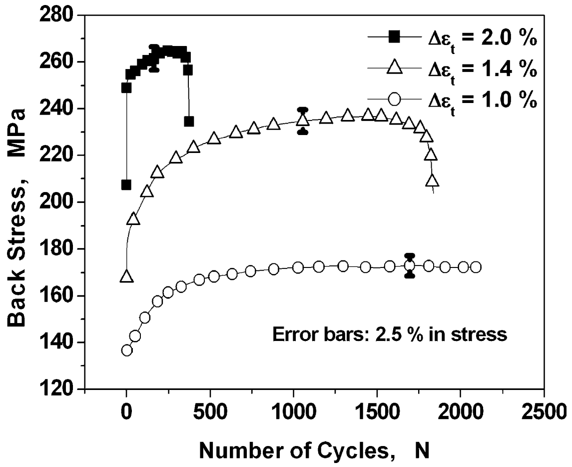

Fig. 4. Variation of the back stress with cycles in Zircaloy-4.

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. Due to the inaccuracy of the method, these values of stresses exhibit a scatter band which was represented by error bars in the diagram. Despite this inaccuracy, the trends of the curves are clearly defined. The cyclic behaviour of the friction stress for three different total strain amplitudes is observed in Fig. 3. A pronounced cyclic softening and the overlapping of the curves are evident from this figure. The cyclic softening rate is independent of the total strain amplitude of the tests. Meanwhile, as shown in Fig. 4, the back stress is strongly dependent on the strain amplitude at the beginning of the test. As cycling proceeds no remarkable difference is observed for the three total strain ranges.

In order to analyze the influence that thermal treatment has on the cyclic behaviour of the material, interrupted tests with intermediate annealing were performed. Fig. 5 shows the peak and the corresponding friction and back stresses behaviour during an interrupted test performed with a total strain range 1% and total strain rate 2×10^{-3} s^{-1}. The specimen was cycled up to near saturation, point A in Fig. 5, annealed in vacuum at 573 K during one hour and cycled again up to point B in the figure. At that point the sample was annealed in vacuum at 473 K during one hour and cycled again. From this figure it is also evident that the cyclic

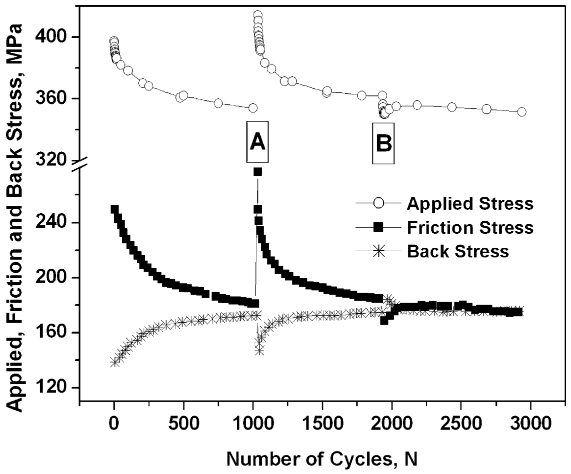

Fig. 5. Applied, friction and back stresses during a test with interruptions. Point A, test interruption with thermal annealing one hour at 573 K. Point B, test interruption with thermal annealing one hour at 473 K (see text).

softening of this material corresponds to a decrease in the friction stress. It is worthwhile to remark 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. After the 473 K thermal treatment, only a small disturbance was present in the cyclic behaviour.

4. Discussion

The cyclic behaviour of the peak stress observed in Fig. 1 is the result of the behaviour of the friction and back stresses. Particularly, the initial cyclic hardening observed at the larger total strain ranges is a consequence of the back stress. However, the softening behaviour exhibited in all the amplitudes is a consequence of the friction stress.

The overlapping of the friction stress curves observed in Fig. 3 could be rationalized thinking that the friction stress is equivalent in each cycle to the yield stress in a monotonic 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. As the initial structure of the samples is considered to be almost the same, the yield stress for the first cycle (friction stress) should be the same independent of the total strain amplitude. Therefore, if the cyclic softening rate is independent of the strain amplitudes of the tests, the curves would overlap.

Fig. 3 shows that the friction stress decreases with the number of cycles and that such a decrease is independent of the imposed total strain range. This is a striking result that is in conflict 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, $4 \cdot \gamma_P \cdot N$, 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 that would cause a larger rotation of the crystals; hence, a larger total strain range should produce a larger decrease rate in the friction stress. The results presented in Fig. 3 indicate that the microstructure, on which the friction stress behaviour depends, becomes softer with cycles but independent of the plastic strain amplitude.

The back stress depends on the density of long-range impenetrable obstacles that are created by the dislocations movement such as pile-ups. Therefore, the larger the total strain range, the larger the density of these obstacles. As a result of this effect, the back stress will be also larger. The observed increase following this initial hardening (Fig. 4) could be attributed to the increased amount of dislocations produced during cycling.

Some researchers [15] propose 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.

The recovery of the sample after thermal treatment at 573 K, as observed in Fig. 5, would suggest that annealing at that temperature distributes the obstacles to the dislocation movement as in the original state. Annealing treatments at 573 K could produce atomic movements and dislocation recovery but not a significant change in the texture [16]. In this respect, the proposal based on texture changes or rearrangement of the dislocation structure during cycling do not seem to be appropriate to explain the behaviour observed in the friction stress after thermal treatments. At 573 K, corresponding to $0.27T_m$ (melting temperature in K 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. Annihilation of dislocations by recovery processes does decrease the flow stress but does not increase, as shown in Fig. 5.

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 has to be accommodated by only a few dislocations. During cycling not only new dislocations are created, but also existing ones are unlocked from their pinning points. 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 trying to pin the dislocations again. The longest distance which 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 wt% that corresponds to 0.8 at.%, that is, one interstitial atom per 125 atoms of matrix. If the crystal structure were simple cubic, the average distance between two atoms of oxygen would be 5 atoms of the matrix. The hexagonal closed packed structure, with a highest atomic packing factor, should have a smaller average distance between two atoms of oxygen. So, the longest distance which the pinning atoms must travel to catch the dislocations again, should be between two and three atomic distances (an atomic distance is considered to be equal to the atomic diameter of the zirconium atom, which is 0.32 nm [17]). The relation between the travelled distance $\bar{x}$ in the time t and the diffusion coefficient D is given by [18]:

$$\bar{x} = 2 \cdot \sqrt{D \cdot t}. \tag{3}$$

Upon the results obtained by Ritchie and Atrens [19] for temperatures up to 923 K the diffusion coefficient can be expressed as

$$D = 6.61 \times 10^{-6} \exp(-Q/RT)\ \mathrm{m}^2/\mathrm{s} \tag{4}$$

with $Q = 184$ KJ/mol, R the gas constant and T the absolute temperature. In this expression D is attributed to the jumps of oxygen interstitial atoms in the basal plane.

With expressions (3) and (4) it can be obtained that the necessary temperature for the atoms to travel a distance of three atomic distances in one hour should be 560 K. Our results show that, after 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 present results allow to conclude 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. After 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 of 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.

5. Conclusions

The present work analyses the low cycle fatigue behaviour of Zircaloy-4 cycled at room temperature. According to the results reported in this paper it can be concluded that the material shows a pronounced cyclic softening which agrees with previous results found in the literature. From the analysis of the hysteresis loops and after interrupted tests it can be assumed that the cyclic softening is caused by the decrease of the friction stress. This work proposes that the unlocking of dislocations from their oxygen interstitial solute atoms during cycling is responsible for such behaviour.

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