Journal of the Korea Nuclear Society
Volume 8, Number 4, December, 1976

Stress Relaxation Behavior of Cold-worked and Annealed Zircaloy-4 Tubing

K. S. Rheem, C. B. Choi and W. K. Park

Korea Atomic Energy Research Institute

(Received September, 4, 1976)

Abstract

Strain rate dependence of the flow stress of cold-worked and annealed Zircaloy-4 was studied by stress relaxation test in temperature range of 200°C to 450°C. The In σ-ln $\dot{\epsilon}$ curves for various temperatures were straight in the $\dot{\epsilon}$ range of 10^{-5} to 10^{-3} sec^{-1}. From the slope of a curve strain rate sensitivity m was obtained. The m in cold-worked Zircaloy-4 had a minimum value at 300°C, while m in annealed Zircaloy-4 had two minimum values, one at 300°C and the other at 450°C. It was found that the temperatures of the minimum m are consistent with the temperatures of strain ageing peaks. The minimum m at 300°C is considered to be due to strain ageing owing to the pinning of glide dislocations by oxygen atoms, while the minimum m at 450°C for annealed specimen is attributed to iron atoms.

요 약

질칼로이-4에 있어서 flow stress의 strain rate 의존성을 냉간가공 및 소둔된 시편에 대해 200°C-450°C 온도 구간에서 응력이완 실험으로 조사하였다. 전체 실험온도에서 In-∂-ln $\dot{\epsilon}$ 관계식은 10^{-5}—10^{-3} sec^{-1} strain rate의 구간에 걸쳐 선형관계를 보였다. 냉간 가공된 질칼로이 -4의 strain rate sensitivity, m값은 300°C에서 최소값을 보인 반면, 소둔된 시편의 경우 300°C와 450°C에서 최소값들을 보였다. 최소값의 m이 나타나는 온도는 가공시효값이 극대값을 보이는 온도와 일치한다는 사실이 확인되었다. m값의 감소는 활동자유전위들이 산소원자에 의해 pinning된 결과로 유발되는 가공 시효와 관련된 것으로 고려된다. 그리고 소둔된 시편이 450°C에서 가지는 최소 m값은 철 원자들로 인한 가공 시효에서 기인된 것으로 생각된다.

1. Introduction

A study on the plastic deformation of a metal with dislocation dynamics requires an explicit knowledge of the stress dependence of the dislocation mobility. This dependence can be determined from direct measurement of dislocation velocities but these experiments are restricted to very few metals. Considerable effort has been devoted to development of techniques for indirect measurement of the stress dependence. The two most common indirect techniques are the strain rate change and stress relaxation experiments. A recent comparison of various indirect tests indicates that the stress relaxation experiment provides the "best" value of strain rate sensitivity¹⁾.

The principal advantage of the stress relaxation experiment is that it covers a wide range of strain rate while straining the specimen by a very small amount. This type of test is generally conducted by loading the specimen in tension until a predetermined load is reached. The machine is then switched off and the applied load is allowed to be relaxed. The same operation is repeated for different loads. Recently, Lee has carried out both the strain rate change²⁾ and stress relaxation experiments³⁾ on Zircaloy-2. He found that there was litte differnce between both of the experimental results. In those experiments he found that strain rate sensitivity of the flow stress was sharply lowered in the temperature range of 300°C in Zircaloy-2. This characteristic was attributed to a strain ageing phenomenon which was explained by the role of oxygen on the plastic flow behavior in terms of dislocation-impurity interaction mechanisms.

The strain ageing behavior of annealed Zircaloy-4 was, however, found to be quite different from that of cold-worked Zircaloy-4.

2. Theory

The fundamental relationships concerning the loading of a plastic specimen in a tensile testing machine have been described by several authors.⁴˒⁵˒⁶˒⁷⁾ We follow here the treatment given by Hart⁷⁾. The instantaneous "plastic length" of a specimen, i. e., the length of the real gage section, is denoted by L. The load exerted is P. We denote by L_1 the distance between the crosshead and a fixed fiducial point, chosen in such a way that, when $P=O$, $L_1=L$. Then, L is obtained from the equation of

$$P=K(L_1-L)$$

where K is the elastic constant of the machine and specimen. If we differentiate above equation with respect to time t, and designate the time derivative by dots over the respective symbols, we obtain

$$\dot{L}=\dot{L}_1-\dot{P}/K$$

From any experimental record of P as a funcion of t and the value of crosshead speed, we can derive appropriate formulas. If the initial and current specmen cross sections are respectively denoted by A_o and A,

$$A_o/A=L/L_o$$

then

$$\sigma=P/A=PL/A_oL_o$$

and

$$\dot{\epsilon}=\dot{L}/L$$

where L_o is the initial specimen gage length, σ is the flow stress, ϵ is the accumulated natural plastic strain and $\dot{\epsilon}$ the natural strain rate. Specialization to the case of the load relaxation test is accomplished simply by setting $\dot{L}_1$ equal to zero.

3. Experimental

The Zircaloy-4 was supplied by SANDVIK AB Sweden, in tube of usual PWR size 10.7 mm outer diameter, 0.6mm wall thickness. The average composition was:

Sn; 1.37% Fe; 0.22% Cr; 0.09%
0;0.1143% Zr; balance

The as-received material was used as a cold-worked specimen and one group of specimens were annealed in vacuum of 4×

10^{-4} torr for one hour at 750°C and furnace-cooled to room temperature.

The gage length was taken as the distance between both ends of fitting plugs inserted into the tube for tight gripping.

Tests were carried out with a universal tester, Autograph IS-5000 Shimadgu. The crosshead speed during the interval of loading was 10mm/min. For elevaleted temperature tests, the specimen was heated in a heating chamber, through which helium gas was flowed during the entire heating and testing period and the temperature was controlled to within ±0.2°C through each test. The load relaxation test consists in loading a specimen to some predetermined load level, then stopping the crosshead motion, and subsequently recording the load as a function of time at fixed crosshead position.

A typical load-time curve for a specimen is shown in Fig. 1. The loading portion of the curve yields a measure of the elastic constant K, while the remainder of the curve is the relaxation history at fixed crosshead position.

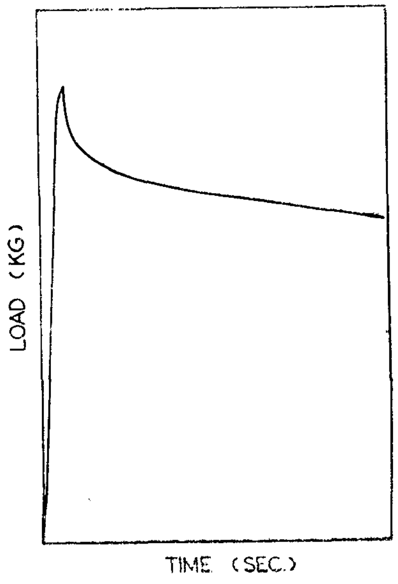

Fig. 1. Schematic Drawing of Load vs Time Relationship.

4. Results and Discussion

The results of the stress vs strain rate relationship obtained from the stress relaxation tests are shown in Fig. 2 and Fig. 3

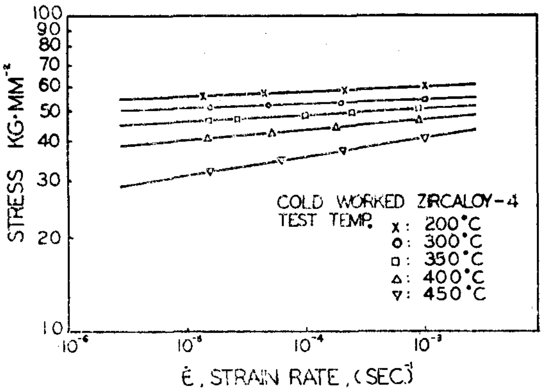

Fig. 2. Stress vs Strain Rate Relationship for Cold-worked Zircaloy-4.

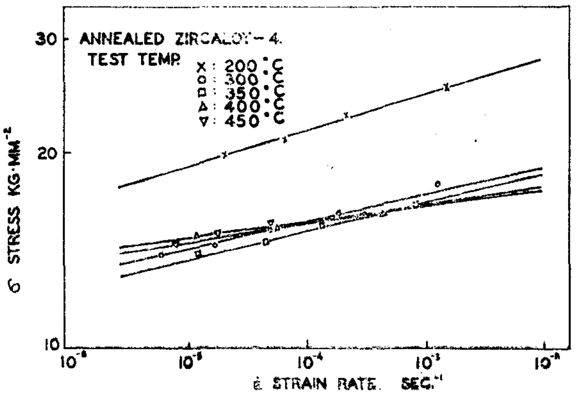

Fig. 3. Stress vs Strain rate Relationship for Annealed Zircaloy-4.

The $\ln\sigma$-$\ln\dot{\epsilon}$ curves for both cold-worked and annealed specimens were straight. The strain rate sensitivity, m, which was calculated from the slope of the straight line, is plotted as a function of temperature as shown in Fig. 4.

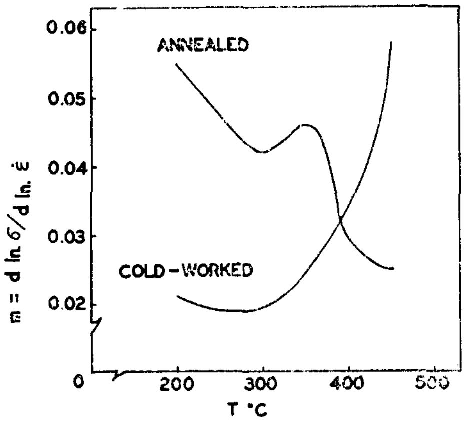

Fig. 4 The Temperature Dependence of Stran Rate Sensitivity in Cold-worked and Annealed Zircaloy-4.

Keeler[8] reported that annealed zirconium shows little change in rate sensitivity with increased temperature, whereas severely cold-reduced material shows considerable increase in rate sensitivity with increased temperature. The cold-worked Zircaloy-4 had similar temperature dependence. However, annealed Zircaloy-4 showed large decrease of rate sensitivity with the increase of temperature.

Furthermore, some minimum values of rate sensitivity were detected in Zircaloy-4. The value of m for cold-worked specimen is lowest at 300°C, while annealed specimen shows two minimum values of m at 300°C and 450°C. These results show the strain rate dependence of flow stress over the temperature range of 300°C to 450°C; the flow stress increases with the increase of the strain rate. The strain rate sensitivity shows an anomalous temperature dependence, i.e., there are two transition points at 300°C and 450°C.

It is of interest that the minimum value of m is found at 300°C in cold-worked Zircaloy-4 and at 300°C and 450°C in a annealed specimen. These temperatures are consistent with strain ageing peak temperatures which were confirmed by Rheem and Park[9,10]. Luban and Pugh[11,12,13,14] suggested that a reduced strain rate sensitivity is associated with strain ageing behavior. During stress relaxation, the total strain is constant and the elastic strain ϵ_e is continually replaced by the plastic strain ϵ_p. It is considered that at the vicinity of minumum m the replacement of elastic strain to plastic strain is inhibited due to the dislocation-impurity interactions and thus stress relaxation is suppressed.

It is suggested that the minimum value of m at 300°C is due to the segregation of interstitial oxygen atoms into cell walls which were produced during straining. These oxygen atoms will make glide dislocations pinned, thus immobilizing the glide dislocation and reducing the stress relaxation.

Veevers and Snowden[15] suggested that strain ageing peak at 450°C in annealed Zircaloy-2 is attributed to iron atoms. Recently Veevers[16] prepared binary alloys of Zr-Fe, Zr-Cr and Zr-Ni and investigated the strain ageing peaks for each alloy. He found that Zr-Fe alloy showed a peak at 450°C, whereas other alloys had peaks between 275°C and 325°C. It is suggested that the minimum value of m in annealed Zircaloy-4 at 450°C is attributed to iron atoms.

The effect of heat treatment on the stress relaxation was also investigated. The stress relaxation vs initial stress is plotted in Fig. 5 and Fig. 6. The curves in these figures are straight. They show that the slope of the curve increases with the increase of the test temperature in cold-worked Zircaloy-4 while in a annealed specimen the slope

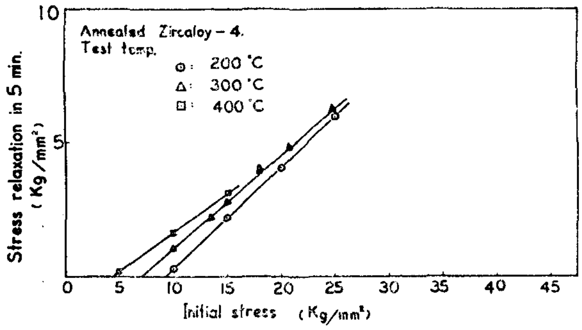

Fig. 5. Stress relaxtion at various temperatures as a function of initial stress in annealed Zircoloy-4. tubing.

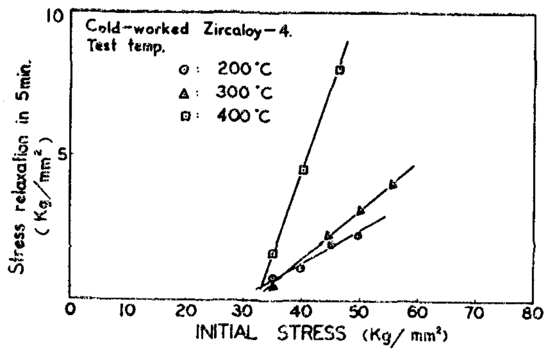

Fig. 6. Stress relaxtion at various temperatures as a function of initial stress in cold-worked Zircaloy-4 tubing.

decreases with the increase of the temperature. It is considered that this interesting phenomenon is related to different pinning processes of glide dislocations by oxygen atoms.

5. Conclusions

(1) The $\ln\sigma$-$\ln\dot{\epsilon}$ curves in both cold-worked and annealed Zircaloy-4 showed linear relationships.

(2) The strain rate sensitivity in cold-worked Zircaloy-4 was lowest at 300°C, while in annealed Zircaloy-4 it had two minimum values, one at 300°C and the othe at 450°C. These temperatures are consistant with the temperatures of strain ageing peaks.

(3) The lowest strain rate sensitivity at 300°C is supposed to be due to the oxygen atoms.

(4) The minimum strain rate sensitivity in annealed Zircaloy-4 at 450°C is attributed to iron atoms.

Acknowledgements

The authours are grateful to K. S. Yeum for helpful discussions.

References

1) C. Y. Cheong and U. F. Kocks, Sec. International Conf. on the Strength of Metals and Alloys. ASM. Metal Park, Ohio (1970)
2) D. Lee, Met. Trans. 1. (1970) 1607.
3) D. Lee and E. W. Hart, Met. Trans. 2(1971) 1245
4) F. W. Noble and D. Hull, Acta Met. 12 (1964) 1089
5) D. Hull and F. W. Noble, Disc. Faraday Soc. 38 (1964) 251
6) G. B. Gibbs, Phil Mag. 13(1966) 317
7) E. W. Hart, Acta. Met. 15(1967) 351
8) J. H. Keeler, Trans. ASM 47 (1955) 157
9) K. S. Rheem and W. K. Park, J. Kor. Nuc. Soc. 8, 3 (1976) 151
10) K. S. Rheem and W. K. Park, J. Kor. Nuc. Soc. 8(1976) 19
11) J. D. Lubahn, Trans. ASM 44 (1952) 643
12) J. W. Pugh, Trans. ASM 47 (1955) 984
13) J. W. Pugh, Trans. ASM 48 (1956) 677
14) J. W. Pugh, Trans. ASME, 205(1957) 1243
15) K. Veevers and K. U. Snowden, J. Nucl. Mater. 47 (1973) 311
16) K. Veevers. J. Nucl. Mater. 55 (1975) 109