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High temperature creep properties of zirconium and Zircaloy-4 in vacuum and oxygen environments

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Abstract

A special set-up has been used to follow the evolution of mechanical parameters under various applied stresses in in situ conditions (controlled temperature and atmosphere). This experimental set-up is used to study the creep behavior of zirconium and Zircaloy-4 in the temperature range 723–823 K. The influence of applied stresses, atmosphere and alloy grade on the deformation and oxidation processes are specifically analyzed. The results underline the presence of two distinct deformation domains for both alloy grades, depending on the applied stress value and the temperature. The results show that the presence of an oxide scale only leads to slight modifications of the creep behavior.

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1. Introduction

High temperature oxidation of pure zirconium and zirconium alloys is very sensitive to many metallurgical, physico-chemical and mechanical factors. In industrial applications, high temperature is usually combined with applied stresses, which leads to creep deformation.

Generally, the creep of materials comprises three successive domains:

- The primary creep where the specimen begins to deform relatively quickly whereupon the creep rate decreases with time.
- The secondary creep which is characterised by a constant strain rate.
- The tertiary creep which corresponds to an increase of the strain rate and finally leads to the failure of the material.

At high temperature, there are two kinds of creep mechanism: creep controlled by dislocation movements, and creep controlled by diffusion. For the first kind, when the plastic flow is mainly ensured by the thermally activated movement of dislocations, creep follows a power law of the Norton type [1]:

$$\dot{\varepsilon} = A\sigma_0^n \exp\left(-\frac{\Delta E}{RT}\right), \tag{1}$$

where $\dot{\varepsilon}$ is the strain rate, σ_0 is the applied stress, A and n are constants, ΔE is the molar activation

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enthalpy, R the gas constant and T is the absolute temperature.

When creep is controlled by core diffusion, the power-law does not apply anymore and the strain rate is a function of the applied stress, σ_0, the vacancies diffusion coefficient, D, and the grain size, d. In the case of core diffusion (Nabarro–Herring creep) [2], the strain rate, $\dot{\varepsilon}$, is given by [3]:

$$\dot{\varepsilon} = K\frac{\sigma_0 D}{d^2}\exp\left(-\frac{\Delta E}{RT}\right), \quad (2)$$

where K is a constant.

The levels of applied stress and temperature used in the present study should normally lead to dislocation-controlled creep of zirconium and Zircaloy-4 [4,5]. So the analysis of the creep results will focus on power-law creep (Eq. (1)).

The purpose of this paper is to quantify the effect of an oxide scale on the creep behavior of zirconium and Zircaloy-4. In order to accelerate the experiments and to obtain sufficiently thick oxide scales, the creep temperatures are chosen in the range 723–823 K. These temperatures are higher than the operating temperatures used in the nuclear industry. But the applied stress values are chosen to yield similar creep mechanisms as in the normal operating conditions.

2. Materials and experiments

The materials used in this work are cold rolled zirconium from Goodfellow and cold rolled Zircaloy-4 from Cezus. The chemical compositions of these materials are given in Table 1. Both metals exhibit the classical rolled texture of zirconium alloys which corresponds to a misorientation of the basal plane (0 0 1) of 30° as referred to the sample surface. The as-received plates are machined to form test-plates with a rectangular useful length of 12 mm in the rolled direction and a thickness of 1 mm. The width of the test-plates is 2 mm. After cutting, the test-tubes are first polished with abrasive paper (SiC) until grade 2500 followed by a 'polished mirror' with OPS solution (suspension of colloidal silica). The sample is then cleaned and dried. After polishing, the samples are annealed during 5 h under primary vacuum at 973 K to stabilize the microstructure and to remove the residual stresses. Finally the surface is polished again with a felt cloth and OPS solution.

The creep tests were performed in a special set-up (Fig. 1) which allows to apply micro-deformations (a few μm) or to impose loads on the samples. In the present study, the set-up has been configured to perform uniaxial sollicitations. Note that the configuration can be switched to a four point bending geometry. The sample is fixed between two tongs (one is mobile, the other one is fixed), at the end of an alumina cane. This cane is then introduced into a quartz tube which is hermetically closed and connected to a primary vacuum pump. The atmosphere within the quartz tube can be modified using a source of dry $^{16}O_2$ or an $^{18}O_2$ tank. This tank is a container of zeolite which traps oxygen when it is at low temperature. To release oxygen 18 and inject it into the quartz tube, it is thus necessary to heat the tank. After a test, oxygen 18 is recovered by cooling the tank with liquid nitrogen. Another reserve of this type is used to trap the remaining impurities after pumping to improve the vacuum. The pressure in the enclosure is measured using a primary vacuum gauge. The temperature around the sample is controlled with a furnace which slides around the quartz tube. The maximum temperature of the sample could be 1273 K.

The displacement of the moving tong is ensured by a small engine controlled by a computer and located outside the assembly. Still outside the assembly, there are force and displacement sensors, and acoustic emission sensors which allow the detection of the acoustic signal emitted by the sample and transmitted through the alumina cane which is used as a wave guide. It is thus possible to follow, in situ, the evolution of the force, the displacement and the mechanical damages in temperature under mechanical sollicitation and controlled atmosphere. To obtain the creep behavior of zirconium and Zircaloy-4, the test-plates are heat treated for 4 h under controlled temperature ($723 < T < 823$ K), and under controlled atmosphere (primary vacuum

Table 1
Chemical composition of zirconium and Zircaloy-4 in weight %

	Sn	Fe	Cr	Ni	Hf	O	C	N	H	Zr
Zr	–	0.02	0.020	–	0.250	0.10	0.025	0.01	0.001	Bal
Zry-4	1.34	0.22	0.110	0.004	0.0044	0.120	0.0119	0.0034	<0.0003	Bal