



In-situ digital image correlation for fracture analysis of oxides formed on zirconium alloys



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ABSTRACT

Repeated breakdown of the protective oxide layer can be a key factor in the oxidation of zirconium alloys. Hence, accurate measurement of the oxide fracture strength is crucial for simulating the oxidation of these alloys. High resolution digital image correlation was applied to SEM images taken during in-situ tensile loading of oxidised ZIRLO™. Bi-axial strain measurements obtained during crack formation in the oxide films were converted into stress, and fitted to Weibull distributions to predict the oxide failure strength. Analysis highlights the impact of surface roughness. Samples polished prior to oxidation gave a fracture strength of ~1.6 GPa.

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1. Introduction

Since their introduction as nuclear fuel cladding for pressurised water reactors in the 1960s, zirconium alloys have been researched significantly. As a degradation mode, understanding oxidation and corrosion has been of high priority due to its capacity to introduce hydrogen [1,2], induce deformation in the metal substrate [3,4], and thin the cladding by material consumption [5]. More recently, research in this area has been driven by the need for a mechanistic understanding of the corrosion process [6]. Central to this is the development of simulations that can capture mechanical aspects of corrosion as a process [7]. However, for these simulations to be valid an accurate definition of the oxide fracture strength is required.

During oxidation in aqueous autoclave environments, zirconium alloys have been shown to demonstrate cyclical corrosion kinetics every 1.5–4.3 μm of oxide growth [8–14]. The end of each cycle is associated with a breakdown of the protective barrier layer, leading to acceleration in the corrosion kinetics. These accelerations in the corrosion kinetics have repeatedly been linked with the formation of a network of cracks running parallel to the metal-oxide interface [15–19]. The lateral cracks form very close to the metal-oxide interface, and travel through the protective barrier

layer [16–19]. It has also been proposed that the region of tri-axial tensile stress, ahead of these advancing cracks, could destabilise the tetragonal phase present at the metal-oxide interface, leading to further nano-scale cracking [20].

Fracture of the oxide layer has also been shown to be of importance during high temperature oxidation of zirconium alloys. In order to understand oxidation during loss of coolant accidents (LOCA) many authors have researched oxidation of zirconium alloys in the 600–1200 °C temperature range [21–25]. Whether oxidation occurs in air or steam zirconium alloys typically demonstrate a transition to accelerated breakaway corrosion kinetics [21–23]. Research has indicated this acceleration is associated with the formation of large cracks perpendicular to the metal-oxide interface, and the formation of a defective, heavily-fractured oxide layer [21,24,25]. Although in this study oxides are formed at a much lower temperature, the results could still be of value to research carried out at high temperatures.

Digital image correlation (DIC) is a technique that provides full field displacement information on the surface of a sample [26]. In doing so it is possible to calculate the localised strain field of a deforming material under mechanical loading [27]. The sensitivity to localised displacements, and therefore strain fields, makes it an ideal tool for detection of crack nucleation and propagation [28]. As such, it allows for early identification of cracks, and definition of the localised strains at which the cracks form.

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Table 1
Composition of recrystallised ZIRLO™ Sheet (wt%).

Material	Sn	Fe	Cr	Nb
ZIRLO™	0.92	0.09	<0.01	0.91

This work defines a fracture stress and strain for oxides formed on zirconium alloys. It is shown that gold remodelling and in-situ digital image correlation offer a route to analysing the fracture strength of thin oxides and coatings.

2. Experimental method & material characterisation

2.1. Sample preparation and oxidation

For tensile testing, samples of ZIRLO™ material, with composition shown in Table 1 [29], were machined into 7 dog-bone tensile samples, with a gauge volume of $26 \times 3 \times 0.45 \text{ mm}^3$. Two methods of sample preparation were used prior to oxidation. The first three samples were pickled in a dilute hydrofluoric acid solution made up of 10 mL HF (28.9 M), 45 mL HNO₃ (15.8 M), 45 mL de-ionised (DI) H₂O [8,30]. The other four samples were lightly ground to 800 grit, polished with 6 & 1 μm diamond paste, and finished with a colloidal silica polishing solution. These preparations were carried out to remove any damage in the near surface region of the as-received material, and to generate planar surfaces. All samples were subsequently oxidised in a chamber furnace (in air) at 360 °C for 20 days giving an average weight gain of $18.2 \pm 1.7 \text{ mg/dm}^2$. For oxide stress characterisation, five coupons ($30 \times 20 \times 0.45 \text{ mm}^3$) of the same ZIRLO™ material were pickled in a dilute hydrofluoric solution and then oxidised in autoclave at 360 °C for 20 days, in simulated primary water (pure H₂O with 2 wt.ppm lithium hydroxide and 1000 wt.ppm of boric acid), at a pressure of 18 MPa. This gave an average weight gain of $17.3 \pm 1.7 \text{ mg/dm}^2$. These weight gains can be related to oxide thickness using the relationship that 15 mg/dm² weight gain equals 1 μm of oxide thickness [16]. Based on the weight gain, the tensile dog-bone samples demonstrated an average oxide thickness of $1.21 \pm 0.11 \text{ μm}$, and the coupons an average oxide thickness of $1.16 \pm 0.11 \text{ μm}$. The oxidation time of 20 days was chosen to give a substantially thick pre-transition oxide layer that is relatively dense and free of defects such as lateral cracks [16,29].

Preparation by polishing or pickling resulted in different surface profiles. This largely resulted from the strong preferential etching effect of the pickling process. After oxidation raised areas could be observed that correlate with the size and shape of the underlying metal grain structure. The surface roughness was defined using a Keyence VK-X200K 3D laser scanning microscope. This was done by taking 10 line profiles, 96 μm in length, for a pickled sample and a polished sample after oxidation. The parameter used to define the roughness was the root mean square of the profile slope (R_{dq}) as discussed in [16], defined using Eq. (1).

$$R_{dq} = \sqrt{\frac{1}{N} \sum_{i=1}^n \Delta_i^2} \quad (1)$$

where Δ is the slope between two given points, taken at N number of intervals across the interface. The polished samples gave a median R_{dq} of 0.101 ± 0.007 , whilst the pickled sample gave a median R_{dq} of 0.569 ± 0.051 . This indicates that the oxide surface is much rougher for samples prepared via the pickling processes than the polished samples. R_{dq} is a hybrid parameter, as it is defined by the slope of the intervals between points, which is based on both the amplitude and width of a peak. The parameter has been applied extensively to the analysis of the interface roughness between the

Table 2
Residual strains and tetragonal phase fraction for oxides formed on ZIRLO™ after 20 days at 360 °C, in simulated primary water.

	σ_m (MPa)	σ_t (MPa)	f_{tet}	σ_{avg} (MPa)
Sample1	-1200 ± 57	-3085 ± 163	0.110	-1407 ± 69
Sample2	-1161 ± 45	-3687 ± 473	0.120	-1465 ± 96
Sample3	-1133 ± 48	-3264 ± 419	0.123	-1396 ± 94
Sample4	-1357 ± 38	-3267 ± 336	0.110	-1568 ± 71
Sample5	-1267 ± 60	-3020 ± 292	0.099	-1441 ± 83

oxide and metal substrate in [16]. Rather than the absolute values, it is the difference in roughness between the two sample preparations that is of note here.

2.2. Residual stress/strain characterisation

Characterisation of the residual stress in oxides formed on the 5 ZIRLO™ coupons was performed using synchrotron X-ray diffraction (SXRD) at the EDDI beam-line at BESSY II (Berlin, Germany) [31]. The experimental setup allowed for rapid acquisition of the diffraction peaks in the energy range of 20–150 keV utilizing a polychromatic beam in energy dispersive mode, more details can be found in [30]. In-plane residual stresses in the oxide layer ($\sigma_{11} = \sigma_{22}$) were determined by tilting the samples through a range of ψ angles, measuring the diffraction pattern at each angle and applying the $\sin^2 \psi$ technique. The oxide reflections of interest for residual strain characterisation are $(\bar{1}11)_m$ and $(101)_t$. Additionally, the $(111)_m$ reflection is necessary for calculation of tetragonal phase fraction using the Garvie-Nicholson formula (Eq. (2)) [32].

$$f_{tet} = \frac{I_{t(101)}}{I_{t(101)} + I_{m(111)} + I_{m(\bar{1}11)}} \quad (2)$$

where I is the integrated intensity for each reflection, t denotes the tetragonal phase and m denotes the monoclinic phase. Such residual stress and phase fraction analysis methodology has been extensively applied previously to these materials in [3,30]. Table 2 shows the residual stresses calculated for the monoclinic and tetragonal phases, the tetragonal phase fractions and the average in-plane stress in the oxide layer. This average in-plane stress is calculated by weighting the in-plane stresses in each phase against the phase fraction (Eq. (3)), as described in Ref. [3].

$$\sigma_{avg} = (f_t \sigma_t) + (f_m \sigma_m) \quad (3)$$

The average in-plane stress was converted to strain by assuming that a biaxial stress condition exists (i.e. stress perpendicular to the specimen surface, σ_{33} , is assumed to be zero and $\sigma_{11} = \sigma_{22}$). Thus the in-plane strains, ε_{11} and ε_{22} , can be obtained by:

$$\varepsilon_{11} = \varepsilon_{22} = \frac{\sigma_{11}}{E} (1 - \nu) \quad (4)$$

Using the elastic properties of the bulk material adopted from previous work [3,30], E is the elastic modulus for the oxide (253 GPa), ν is the Poisson ratio for the oxide (0.282), and σ_{11} is the in-plane residual stress in the oxide layer. Taking the mean of the 5 samples in Table 2 gives a characteristic compressive residual stress of $-1455 \pm 83 \text{ MPa}$.

Using Eq. (4) gives an in-plane strain of -0.00413 ± 0.00024 , which should be the tensile strain required to reduce either of the σ_{11} or σ_{22} residual stress components to zero. Although Eq. (4) has been used previously to relate residual stress to strain in biaxial compressed thin film oxides [33], definition of this problem can be calculated by a number of routes [3], which could impact the final value for residual strain.

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