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Viscoplastic characteristics of thermally grown chromia films obtained from in situ 2D synchrotron X-ray diffraction

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ABSTRACT

This work takes place in the general context of a better understanding of materials degradation mechanisms in severe environments. More particularly, its purpose is to correlate microstructural elements to growth stress magnitude evolution and stress release mechanisms for chromia thin films thermally grown on model NiCr alloys. X-ray elastic strains have then been measured in situ, as the chromium oxides develop and evolve, by using synchrotron X-ray diffraction, coupled with an induction furnace. 2D diffraction patterns were continuously recorded by applying different temperature variation procedures between 1000°C and 700°C. Thermal stresses imposed on Cr_2O_3/NiCr system by abruptly reducing the sample temperature, i.e. by exploiting the thermal expansion difference between oxide and substrate, showed noticeable subsequent stress relaxation by creep. The main results obtained from these experiments were the kinetic of the growth stress from the isothermal measurements, and the study of the stress release mechanism after the temperature jumps to lower values. In complement, the oxide microstructure development during the course of oxidation was also investigated from both the peaks intensity and peaks width evolution. The creep coefficients could be estimated for the first time for chromia under the thin film form. It was proposed that the stress release by diffusion creep in the chromia films is very likely governed by a mechanism involving diffusion of oxygen ions at the chromia grain boundaries.

1. Introduction

Degradation of metallic materials under high temperature conditions represents 3% of the whole worldwide corrosion phenomena. In such fields like aeronautical turbines or power plants, chromium and aluminum alloys are used in order to reduce high temperature material oxidation. These alloys have been developed to form at high temperature an oxide layer, e.g. chromia or alumina, at the metal surface [1,2], playing the role of a protective diffusion barrier for the underlying metal. However, it is well known that high stresses are generated in the thermally grown oxide (TGO), related to the scale growth during isothermal oxidation. It has also been observed that a highly compressive growth stress arises from oxide formation within the scale as the diffusing cations and anions meet and may react, particularly along transversal grain boundaries. In addition, residual stresses also result from the thermal expansion mismatch between the substrate and the TGO during cooling or heating [1–3]. As a consequence of this high stress level in the scale, relaxation phenomena may also occur [3,4]. Two main types of stress release mechanisms can then be distinguished: non-destructive stress relaxation, like viscoplastic deformation where the scale remains adherent and protective, and destructive ones, like spalling or buckling, which lead to scale damage [4,5].

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Therefore, metal durability depends on the oxide film integrity through the balance between destructive and non-destructive stress release processes. The microstructure in the TGO also affects the growth stress magnitude and the relaxation mechanisms. Indeed, for stressed scales at sufficiently high temperatures, oxide creep is expected to occur for fine grained oxides, rather than delamination phenomena. However, none of these mechanisms is fully understood. Thus, it appears essential to better characterize the strain state and associated creep response in the TGO. Moreover, while creep has been extensively studied in bulk ceramics [6–12], only a few measurements have been reported for the TGO [13–17]. Thus, the present work focuses on the study of nondestructive relaxation phenomena, with the aim of a better understanding of the creep behavior. Indeed, it does not lead to scale degradation and favoring this kind of stress release phenomenon, for example through the control of initial oxide grain size, may ameliorate the oxide scale durability. In particular, measuring the stress magnitude in thermally grown chromia film is important because it is the primary oxide encountered in widely used stainless steels. Among chromia-forming alloys, NiCr is an oxidation resistant alloy whose oxidation behavior has been particularly studied [18–21]. It is indeed of particular interest for fundamental investigations since chromia is the only oxide developed during high temperature oxidation. For the specific case of chromia thermally grown oxide, the evolution of the stress has already been carried out for different isothermal oxidations [22]. These first results have also been modeled and associated to an activation of diffusion-creep in the ceramic film, either at grain boundaries or in the bulk [23]. In addition, recent results have reported the activation of grain boundary sliding as a companion mechanism of diffusion-creep in this system [24]. However, these analyses were unable to assess the respective influences of either the grain size effect or the thermal activation in the diffusion-creep mechanism.

In complement to previous works, the specific purpose of the present study is to separate the thermal activation from the microstructure effect in the creep behavior of the thermally grown oxide. The selected approach consisted, in a first step, to build different initial microstructural states by oxidizing NiCr alloy at several high temperatures; this in particular allows to fix the grain size of chromia. Then, in a second step, temperature jumps towards lower temperature values were applied in order to impose a mechanical loading. The oxide creep response could then be investigated along each subsequent isothermal temperature plateau. Finally, the stress build-up and release mechanisms will be affected, which should be evidenced from the values of the thermo-mechanical parameters. Thus, for the different initial microstructures, several temperature jumps were successively imposed down to the inferior effective limit for creep thermal activation, estimated about 700 °C for Cr_2O_3 [25].

However, such a study requires an experimental setup able to realize in situ measurements. An extensive study of the considered phenomena is possible providing a coupling between in situ X-ray diffraction and high-temperature oxidation in a furnace. In addition, the use of synchrotron radiation, and its high brilliance, appears mandatory, first because of the slow oxide growth rate and weak X-ray scattering of the chromia scale, secondly because of the small diffracting volume (very thin oxide scale) and the rapid strain change during oxidation. Furthermore, since the samples under investigations are composed of a stacking of the ceramic film and the metallic substrate (Cr_2O_3/NiCr), tuneable wavelength is required to avoid peaks overlapping on the diffractograms.

2. Material and experimental methods

Material of investigation is a Nickel containing Chromium alloy (Ni-30Cr) provided by Aperam, which is a non-textured material. Longitudinal rods of 6 mm diameter and 4 mm thickness were cut by electro-polishing from an initial 12 mm diameter bar. The alloy nominal composition is reported on Table 1. The samples were previously annealed for 1 h at 1000 °C under Argon and SiC polished until the 4000 grade.

Table 1
Alloy composition (weight %).

NiCr	Ni	Cr	Si	Mn	C (ppm)	P (ppm)	S (ppm)
Ni-30Cr	balance	30.22	<0.01	<0.01	230 ppm	30 ppm	40 ppm

The experiments were carried out at the European Synchrotron Radiation Facility (ESRF, Grenoble, France) on the BM02 beamline. Oxidations of the metallic samples were realized in ambient air in the range [700–1000 °C] with a high temperature induction furnace provided by the Sample Environment Laboratory from ESRF. The samples were placed on the specimen-holder of the goniometer and were heated and cooled at the same linear rate of 150 °C/min. A thermo-couple with PID controller was used in order to regulate the temperature in the furnace and a pyrometer was positioned above the sample to control the surface temperature. Hence, the temperature variations were continuously monitored. A 2D FreLON detector was used to record the Debye-Scherrer rings from both the chromia oxide films and the NiCr alloy. The reflection mode was used with a fixed incident angle ω of 5°. The X-ray energy was 20 keV with corresponding dimensions of the X-ray beam (300 μm * 1500 μm) allowing to maximise the diffracted intensity from the earlier oxidation steps. In addition, with such an energy no overlapping of the oxide film and metallic substrate peaks is observed. Diffraction in reflection mode also maximised the signals coming from the chromia film. The latter developed all along the course of oxidation under ambient air. XRD diagrams were recorded using a detector, large enough to get sufficient Psi values to determine the stress evolution in the chromia films [26]. The use of Synchrotron radiation permitted to collect a diffraction diagram every 11 s, duration including 5 s for acquisition, the image readout time a sleeping time. This frequency was considered as sufficient to obtain real time evolution, compared to the phenomena

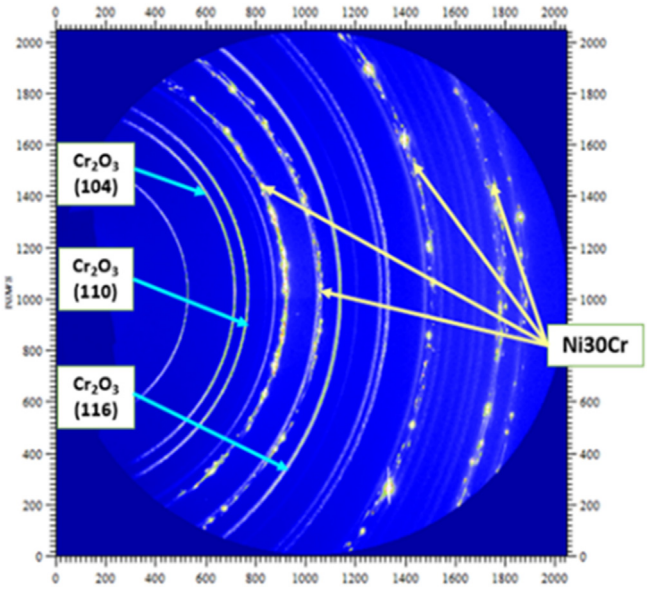

Fig. 1. Typical 2D pattern showing the Debye-Scherrer rings of Cr2O3-TGO film and Ni-30Cr.