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Determination of residual stress fields in a thermally grown oxide under thermal cycling loadings, using XRD and Raman spectroscopy. Correlations with microstructural states

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Abstract. The presence of residual stresses in thermal oxide layers has been recognized for a long time. In the present work, the mechanical fields for chromia oxide are determined either by XRD or Raman spectroscopy. In addition, the microstructure of the chromia films is investigated ant its influence on the evolution of the stress release processes is analyzed.

Introduction

NiCr alloys are currently used at high temperatures because it develops a dense chromia surface oxide film which slows down the oxidation process. In turn, the material durability depends on the ceramic film integrity. The isothermal oxide layer growth or the cooling steps usually induce the development of high residual stress in the ceramic film. The determination of such growth or residual stress has already been undertaken [1-7]. And subsequent stress release may induce buckling or spalling phenomena which will renew oxidation of the metallic alloy [8-10]. In addition, stress release may also proceed by creep which should be less detrimental for the system. This behaviour has been suggested by [2,11] from growth stress evolution measurements in Ni-30Cr alloys. In order to increase the durability of chromia forming alloys, it is mandatory to better understand such stress build up and relaxation mechanisms. The aim of the present work is a first attempt to evaluate quantitatively the stress release processes in Ni-30Cr alloys. To this end, in addition to stress evaluation, the delamination rates (buckling and spalling) will be determined. The influence of different metallurgical parameters (cooling rate, oxidation temperature and duration) which in turn modify the microstructure, will also be investigated.

Experimental procedure

Specimens 1.5mm thick providing from a Ni-30Cr rod have been prepared. Its composition is given in [10]. A 1 hour initial thermal treatment at 1000°C has been achieved to homogenise the metal. Specimens are mechanically polished until 4000 grade, washed and dried. The oxidations have been done at 800, 900 and 1000°C in a muffle furnace, for 3 or 18 hours. Then, specimens are cooled by using different supports; the associated cooling rates being in the range [80°C/min – 500°C/min]. The later different steps constitute the thermal cycling loading. Stress determination has been undertaken either by XRD or Raman spectroscopy. The corresponding methodology has been described in [12]. Due to the good agreement between these two methods, an average value will be used. For both methods, the stress value always provides from the adherent part of the chromia films. Delamination situations (buckling, spalling) are observed by optical microscopy. The normalised delamination rates are obtained from Eq. 1 [8] :

$$T_d = \frac{\sum_0^N S_i^s + \sum_0^{N'} S_j^b}{S} \tag{1}$$

with S_i^s surface of spall number i, S_j^b surface of buckle number j, S total surface, N and N' respectively total number of buckles and spalls. The later are observed thanks to optical microscopy and the quantitative measurements are performed through image analysis.

Residual stress

Oxidation condition influence. Stress magnitudes in chromia films are reported in Fig. 1. It is clear that residual stress increase with the oxidation temperature whatever are the experimental conditions. This behaviour is directly related to the evolution of the thermal stress. The later has been calculated from Eq. 2 and also reported in Fig. 1. However, systematically the measured residual stresses are inferior to the thermal stress. It implies the activation of stress release mechanisms in the adherent parts of the ceramic films.

$$\sigma_{th} = -\frac{E_{ox}(\alpha_{ox} - \alpha_{met})(T_f - T_i)}{1 - \nu_{ox}} \tag{2}$$

α_{ox} and α_{met} are the thermal expansion coefficients for oxide and metal, T_f and T_i are the final and initial temperatures, E_{ox} and ν_{ox} are the oxide elastic coefficients. Eq. 2 holds when the oxide is thinner than the metal.

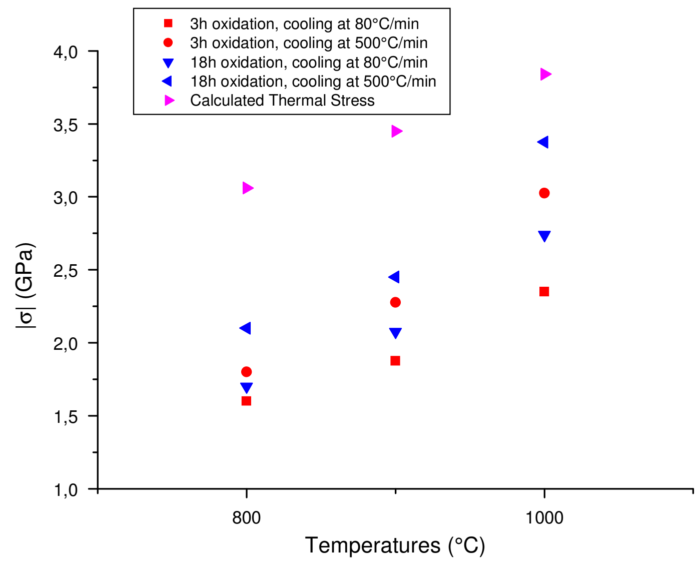

Fig. 1 : Residual stress evolution vs the oxidation temperature, for different oxidation times and cooling rates

In addition, for each oxidation temperature the stress magnitude variation is the same for the two other parameters: it increases with the cooling rate, and it also increases with the oxidation time. The latter observation is in good agreement with the results in [3].

Cooling conditions influence. The cooling step is also very significant for the stress release processes. In particular, it is on cooling that delamination may appear [12]. The influence of the cooling rate has been studied in details by imposing the other parameters. Results are presented in Fig. 2.

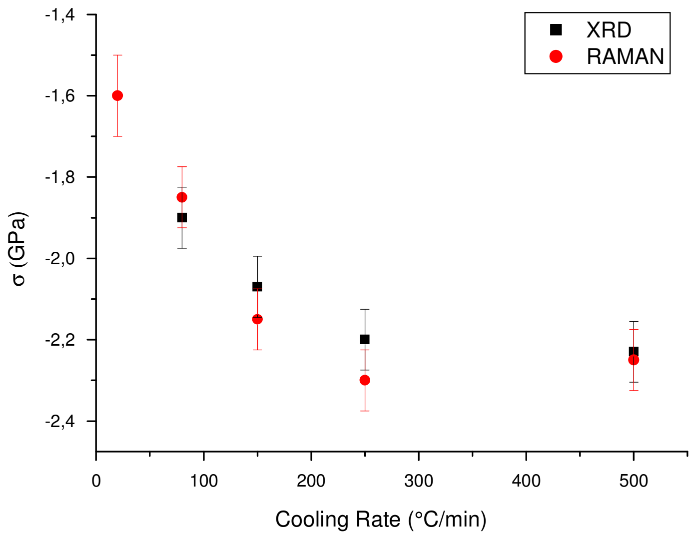

Fig. 2 : Residual stress evolution vs the cooling rates. 3 h upon oxidation at 900°C

It can be observed that stress release mechanisms in the adherent oxide film are more efficient for low cooling rates. From [2], after 3 hours upon oxidation at 900°C, the ceramic film has developed its specific microstructure with an associated compressive growth stress magnitude of about 2.3 GPa. In these conditions, none stress relaxation e.g. by creep of the oxide has been activated yet. On the other hand, results in Fig. 2 suggest that such release mechanism can appear during the subsequent cooling step, and that the available stress release amplitude is correlated to the cooling rate. The present behaviour is in agreement with previous qualitative work by Kemdehoundja [1]. It can also be compared to the work of Tolpygo and Clarke [13] in alumina ceramic films. The stress magnitude also increases with the cooling rate, before a plateau is attained.

Stress relaxation by delamination

Delamination damage is a destructive phenomenon which locally leads to stress release. In the present work, we do not focus on the reasons allowing the presence of a necessary initial debounding patch at the metal / oxide interface which in turn may induce either buckling or spalling. The aim is to quantify the situations of delamination and to relate it to the stress magnitude. Circular buckles are characterised by a typical colour darker or whiter compared to the adherent part of the film (Fig. 3), while spalls leave the metal appear.

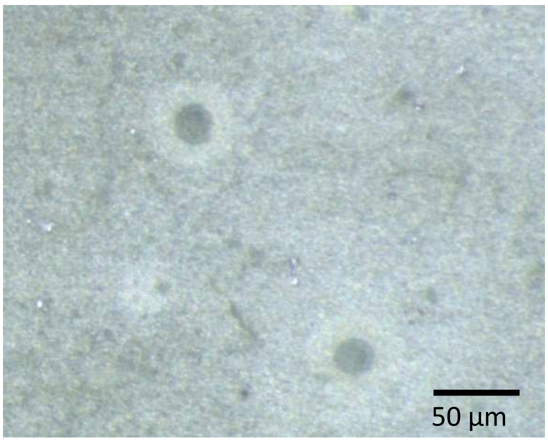

Fig. 3 : Circular buckles observed by Optical Microscopy. 3 h at 1000°C

Oxidation and cooling conditions influence. The delamination rates are presented in Fig. 4.

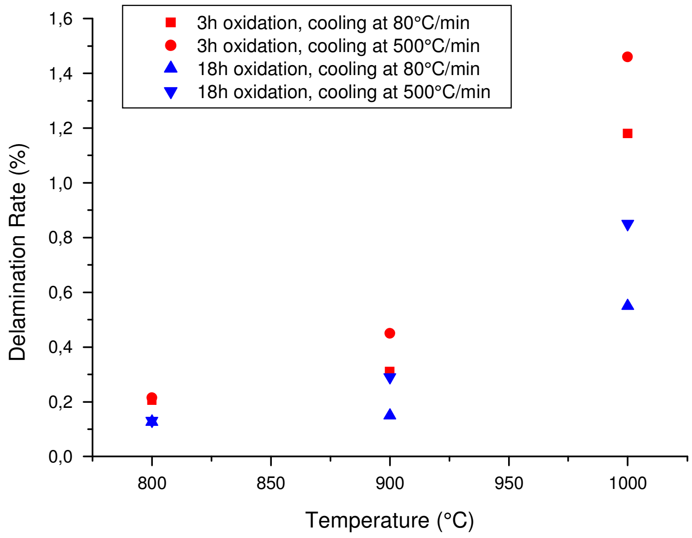

Fig. 4 : Delamination rate vs the oxidation temperature. 3 and 18 hours upon oxidation for different cooling rates

The delamination rate increases with the oxidation temperature for a given oxidation time. It is also higher for the shortest oxidation times. It can be reminded that the stress magnitude increases both with the oxidation temperature and the oxidation time (Fig. 1). Thus, residual stress in the ceramic film and delamination rates doest not seem to be directly correlated in particular when the oxidation time evolves. This behaviour is in good agreement with the results obtained by Siab [8] for the temperature range [700-900°C]. In this work, the delamination rate was compared with the growth stress evolution. The present results obtained from residual stress measurement after cooling, confirm this tendency also at 1000°C. Finally, whatever the oxidation conditions, the delamination rate increases with the cooling rates. This behaviour is also in good agreement with the first qualitative results given in [1].

Result analysis. Mechanical approach of delamination rate evolution

From buckling mechanical models, one may try to analyse the delamination rate evolution. It has to be taken into account that damage mainly occur by buckling, in the present case. Eq. 3 giving the critical conditions for buckle formation has to be considered. It is assumed a size distribution of the initial debounding patch (radius b) present between the oxide film and the substrate. The transformation of such a debounding region into a buckle needs a circular buckling index $\Pi \geq 1{,}22$ [14, 15]. Thus, from Eq. 3 a high stress magnitude should a priori favour buckle formation. But, the film thickness also appears in this relation. Then, if the influence of the oxidation temperature is considered, it may modify both the stress magnitude and the film thickness, and in turn the buckling index.

$$\pi = \frac{\sigma_{ox}(1-\upsilon_{ox}^2)}{E_{ox}}\left(\frac{b}{e_{ox}}\right)^2 \qquad (3)$$

with σ_{ox}: residual stress, E_{ox} and ν_{ox}: oxide elastic coefficients, b: initial debounding patch radius, e_{ox}: oxide film thickness

From this analyse, buckling should be easier at 900°C compared to 1000°C. Indeed, from Eq. 3, one may associate a buckling index for each debounding patch of radius b, considering both the thickness of the ceramic film and the stress magnitude in this layer. It has been done in Fig. 5 for different radius b after 3 hours upon oxidation at 900°C and 1000°C. A minimum value of 5 µm has been chosen for b because for the majority of the buckles observed the radius is superior to this dimension. It clearly appears in Fig. 5 that it is at 900°C that the buckling index is the most often superior to the critical value. Thus, from this direct application of the buckling mechanical model, delamination by buckling should be promoted at the lowest temperature. But, the results from Fig. 4 indicate the opposite behaviour.

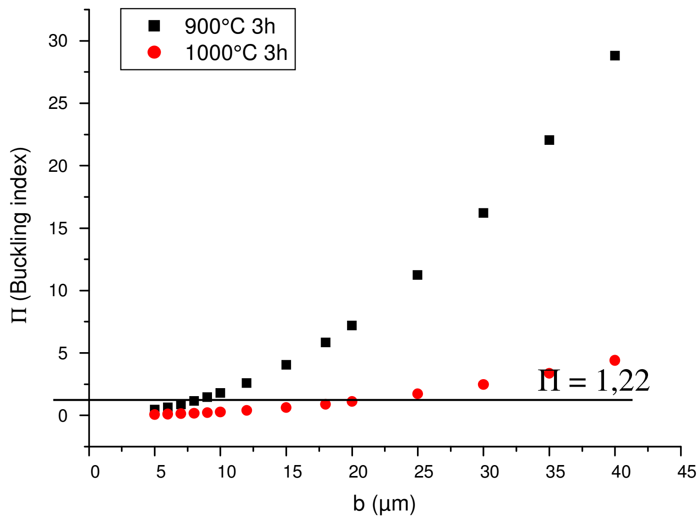

Fig. 5 : Buckling index vs radius b of the initial debounding patch. The critical buckling index value is also indicated

Summary

In conclusion, it has been shown that the residual stress magnitude in a chromia ceramic film after cooling depends on the growth and thermal source terms but also the stress release processes, which also appear either during the isothermal domain but also during cooling. Moreover, it is important to consider all the dynamic and spatial repartitions to get a clear description of the phenomena. From the delamination rates measurement and confrontation with buckling mechanical models it is clear that the non-destructive release mechanisms are sensitive to the oxidation temperature and seem to be more efficient at the lowest temperature.

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