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Full Length Article

On the determination of growth stress during oxidation of pure zirconium at elevated temperature

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ABSTRACT

An experimental approach have been proposed to evaluate growth of stress during high temperature oxidation of pure zirconium. The development of stress in the oxide scale has been investigated experimentally in in-situ conditions by combining the Deflection Test in Monofacial Oxidation (DTMO) with Acoustic Emission analysis (AE). Microstructure of the sample were studied by using Scanning Electron Microscopy technique. Oxidation experiments were performed continuously during 24 h at 400 °C and 500 °C in air under normal atmospheric pressure. Taking into account purely elastic behaviour of the material, primary evolution of growth stress developed in the oxide scale during oxidation process have been estimated. Presented study of the Zr/ZrO_2 system revealed two opposite phenomena of stress relief when cooling from 400 °C and 500 °C to room temperature. This study is presented as a tool to understand the phenomena of stress evolution in the zirconia layer during isothermal treatment at high temperature and after cooling.

1. Introduction

Zirconium alloys due to their excellent mechanical properties, transparency to neutrons and sufficient corrosion resistance are considered as very promising materials to be used in current and future generations of nuclear power plants [1]. It is known, that oxide scale developing during corrosion process on zirconium consists of two phases (tetragonal and monoclinic) [2,3]. These phases can be stabilized be several factors among which compressive stress seems to be the most important [4]. Reported finding potentially may lead to slowing down oxidation kinetics by better understanding mechanics of the zirconium/zirconia system, by controlling of oxide phase formation. Consequently, lifetime of the material can be extended (zirconium is used as a cladding material in current NPP's, it is also considered as a component for Gen IV nuclear reactors). However, working environment of the reactor is highly corrosive. It is known that during oxidation, various chemical, mechanical and thermal processes may appear. Among numerous phenomena occurring in the material, one must consider growth stress developed during oxidation and fluctuation of the thermal strains which are related to the differences in thermal expansion coefficients between metal and oxide. In order to better understand these mechanisms and their consequences, an experimental approach reported by Przybilla et al. [5] is often used.

It is known that stress developed during isothermal oxidation and cooling to room temperature can accumulate and lead to the delamination of the oxide scale [6,7]. Moreover, it has been reported that during even very slow, yet continuous oxidation, growth stresses can accumulate and reach critical level at which micro-cracking of the material begins. Despite large amount of studies aiming to understand different types of strains appearing during zirconium oxidation [8–12], phenomenon of stress accumulation and relaxation is still under discussion. In addition to that, most of the conducted research tackle the problem of the stress estimation in ex-situ conditions (hence, specimen is already partially relaxed). Therefore, studies performed in in-situ conditions are highly desirable.

Main aim of our study was to experimentally determine values of stress developed in zirconia due to the oxidation process at two different temperatures. As the first step, determination of intrinsic growth stress by the method successfully applied to the Ni/NiO system by Przybilla et al. [5], the Deflection Test in Monofacial Oxidation (DTMO) has been performed. In the second stage of the research, measurement of the Acoustic Emission (AE) activity in

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in-situ conditions was performed. Finally, the third part of work was devoted to the registration of the sample bending by using high speed camera mounted outside the furnace. Implementation of these complementary techniques allowed us to investigate behaviour of oxide scale developing on zirconia and better understand mechanisms of the studied metal/oxide system.

In the present work the level of intrinsic growth stress has been presented, as well as high temperature stress relaxation and accumulation of thermal strains during cooling process were discussed. Reported phenomena are very often neglected in available literature concerning behaviour of zirconia scale submitted to the influence of isothermal oxidation and cooling processes. Therefore, our aim is to present more complete explanation of interactions between observed phenomena and determine the values of the stress.

Mechanical and structural tests were performed on the metal/oxide system of zirconium/zirconia. The measurements were performed in air at 400 and 500 °C under normal atmospheric pressure for 24 h. Conducted experiments allowed us to investigate stress in the function of oxidation time and combined it with acoustic emission data. Thus a correlation between stress level, displacement of the sample and acoustic activity is presented. Afterwards, microstructural investigation of the specimens were performed. According to our knowledge, reported in this paper experimental methodology is one of the few attempts to combine and apply reported previously three independent techniques: SEM, AE and deflection. The main goal of the conducted research is to clarify the phenomena occurring during oxidation.

2. Experimental procedures

2.1. Concept of deflection test in monofacial oxidation

Deflection Test in Monofacial Oxidation (DTMO) is a method which was originally developed by Evans [13]. It has been implemented already for investigation of the mechanical properties of nickel/nickel oxide system [5]. Concept of this method is based on the fundamental approach assuming that investigated thin metallic foil is protected against oxidation on one side and oxidizes on the other side. Due to the oxide growth, an internal foil deformation appears which causes deflection of the specimen (deflection test – DT). Registered specimen deflection allows one to estimate the intrinsic oxide scale stress.

However, it should be noted that this method provides a stress analysis assuming non-linear creep deformation. It considers the situation where the oxide experiences a net compressive stress while the metallic substrate net tensile stress component. The advantage of this method is the fact that it can be implemented for metal/oxide systems where relatively thick oxide scale is developed.

In order to assess the level of stress during isothermal oxidation by using experimental set-up of DTMO technique, an Eqs. (1) and (2) were used:

$$\sigma_s = -\frac{E_m d_m^3}{6R(1-\nu_m)d_s(d_m+d_s)} - \frac{E_s d_s^2}{6R(1-\nu_s)(d_m+d_s)} \tag{1}$$

$$R = \frac{L^2 + D^2}{2D} \tag{2}$$

where σ_s represents the average stress in the oxide scale, E_m and E_s are the Young's modulus of the metal and oxide, respectively, d_m and d_s are metal and oxide thicknesses, ν_m and ν_s are the respective Poissons ratios and R is the bending radius. Detail analysis of these equations can be found in [14].

It should be explained that according to DTMO approach, calculated stress is based on purely elastic deformation behaviour. However, it is known that at least metal should show some creep or plastic deformation at high temperatures. From the qualitative point of view, reported result may overestimate stress level, yet

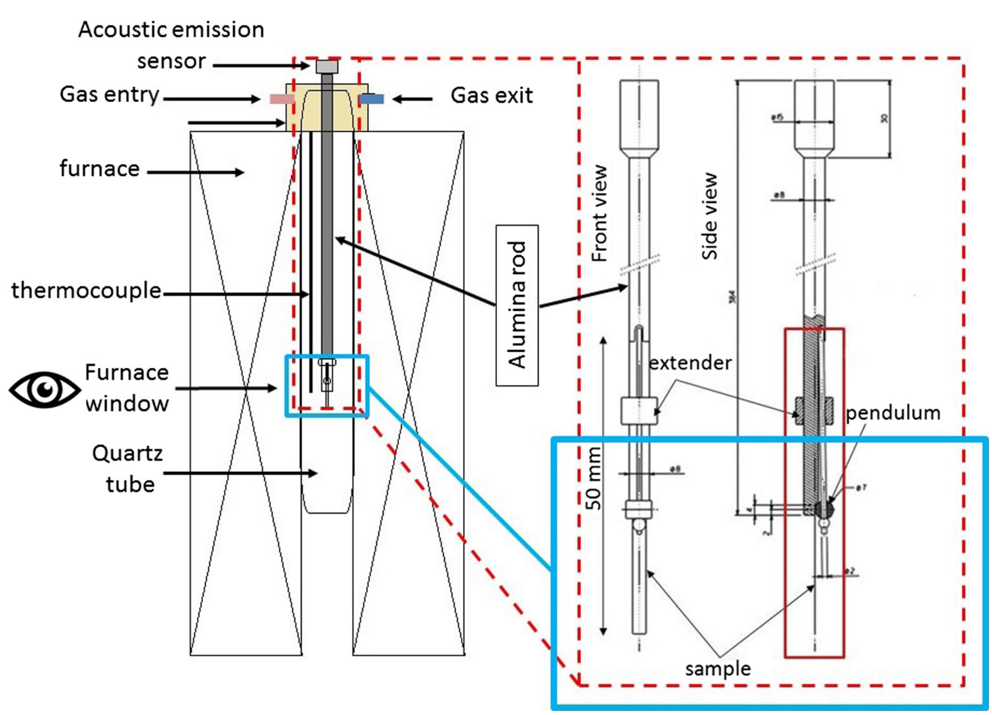

Fig. 1. Schematic representation of the test equipment used in the present investigation. Red dotted line depict the interior of the vertical high temperature furnace with a special emphasis to the alumina rod to which specimen is attached. Details of the DTMO set-up can be found in [15]. The blue square depict the inlet window through which specimen positon is observed by using HR camera – represented by the "eye" figure. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)