The Influence of Thermal Shocks on the Thermophisics Properties of the Zircaloy-4

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This paper presents the researches on the influence of thermal shocks on the heat transfer properties of the zircaloy-4, now used in fuel element cladding of third generation nuclear reactors. Thermal shock testing was performed using solar energy at high temperatures, up to 1350°C, with 1, 3 and 6 thermal cycles of 60s. The determination of the thermal diffusivity of the tested samples was made by the flash method at 350 °C, the operating temperature of the third-generation nuclear reactors.

Keywords: zircaloy, solar energy, thermal shock, microstructure, thermal diffusivity.

The transition from third generation reactors, in operation today, to a new generation of reactors involves significant researches in the field of materials [1-7]. Romania will build the prototype of the lead cooled fourth generation reactors at the Institute of Nuclear Research, Pitesti – RATEN. The fuel element cladding must provide a long lasting running in contact with the moderator, an efficient heat transfer under normal operating conditions, at 350^0C for CANDU reactors, and 550^0C for the lead cooled reactors respectively, and to withstand to temperature variations under running and accident conditions.

The research presented by this paper shows the impact of the heat shock and heat cycles at high temperatures on the microstructure and the thermal diffusivity of the Zy-4 alloy which is used in the construction of the fuel element cladding in the CANDU reactor.

Experimental part

The tests were performed on Zy-4 alloy samples of a 10 mm diameter. The chemical composition of the used alloy is Sn-1.31%, Fe-0.20%, Cr-0.11%, the main part being Zr . The mechanical properties of the delivered alloy are as follows: the flow limit is 533.5 MPa, tensile strength is 744 MPa and elongation is 21.25%. In cross-section, hardness has an average value of 228 HV, whereas in longitudinal section it has an average value of 258 HV. The microstructure of the delivered alloy is polyhedral, the average grain size being of 6.9 µm in cross section and of 7.7 µm in longitudinal section (fig.1).

Thermal fatigue tests were performed in the PROMES Solar Furnace at Odeillo - Font Romeu, France [8], on cylindrical samples with a 10 mm diameter and a 8 mm thickness, through thermal cycles, in the air, with a shock, respectively, three and four successive thermal shocks of 30 and 60s.

The specimens were characterized using scanning electron microscopy and EDS, on unprepared samples at the surface and in the cut section, after mechanical preparation and attack with a 45% HNO_3, 45 % H_2O °i 10 % HF solution, and determining the thermal diffusivity at 350°C.

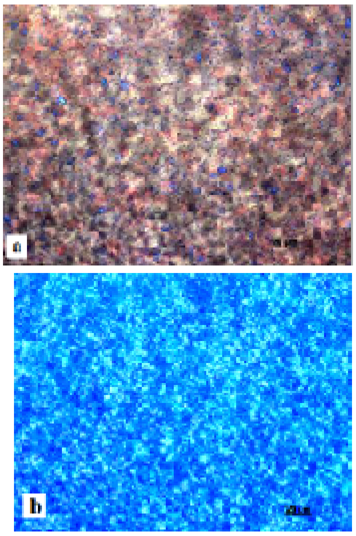

Fig.1 Microstructure of Zy-4 alloy in longitudinal section - a and cross section - b

Results and discussions

The metallographic analysis showed the development of the oxide layers and of the microstructure of the base metal, according to the temperature and the number of thermal shocks.

The surface microscopic analysis showed the microstructure of the oxide layers and the intensification of their degradation process as the temperature of the thermal shocks rises (fig.2. a,b). The rapid growth of the layers at high temperatures leads to high internal tensions resulting in oxide exfoliation which takes place simultaneously with the formation of a new oxide layer. (fig 2c) [9-15].

For the same thermal shock temperature, by increasing the number of the applied thermal shocks, a significant intensification of the surface degradation process was emphasized. (fig. 2a, d).

The section microstructure analysis showed the structure of the oxide layers, the development of their degradation and of the oxide metal interface quality (fig.3).

Under the oxide layers there were evident the structure of the base metal and its changes due to air heating at high temperatures, the formation and the increase of the layers of alpha solid solution stabilized by oxygen dissolution in

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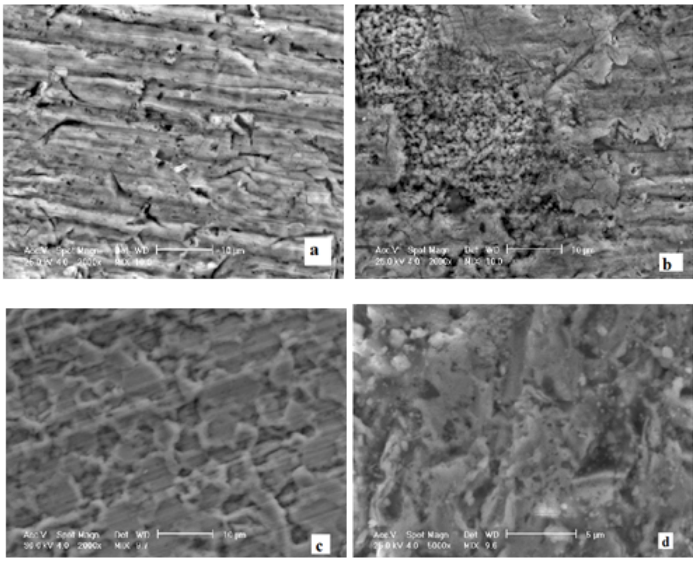

Fig.2 Surface microscopy of oxide layers formed at different temperatures: for a single thermal shock at 1000°C-a, 1300°C- b and 1450°C- c and for 6 cycles at 1000°C - d.

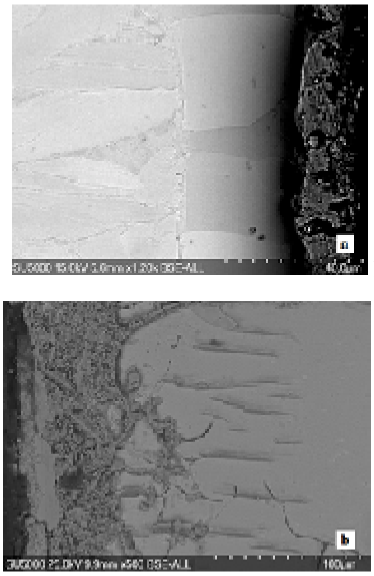

Fig.3 Oxide layers formed at : a- 900°C, 6 cycles, 30s; b- 1150°C, 3 cycles, 30s.

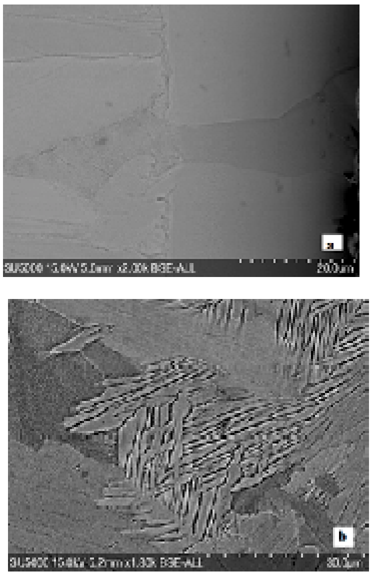

Fig. 4 Microstructure of the sample tested at 900°C with cycles of 60s: a-alpha solution layer and b-core with needles and plaques

metal under the oxide layer (fig. 3.a, fig.4.a) and the appearance of some needles and plaques structures in the core (fig.4b) which continuously enlarge as the temperature increases.

The degradation of the oxide metal interface through cracks which progress mainly perpendicularly on the interface and the degradation of the oxide layers open new ways of oxygen diffusion and allow for the thickening of the oxide layers and of the alpha solid solution (fig. 5) [16-20].

The influence of the thermal shocks and cycles on the thermal diffusivity was determined on samples as thick as 1.9-2 mm. Diffusivity measurements were performed using a FlashLineTM 3000 system which allows the measuring temperature to be programmed at a very close resolution ($+0.5\ ^0C$), a $\pm 2\%$ results reproducibility and $\pm$ 4% accuracy.

The thermal diffusivity values are correlated with the thermal diffusion of oxygen in the metal and the structural transformations induced by high temperatures [2-3, 21-28].

The influence of structural transformations induced by thermal shocks of 60s, whose duration was determined by diffusivity measurements made at CANDU reactor operating temperatures (350°C), is shown in figure 6.

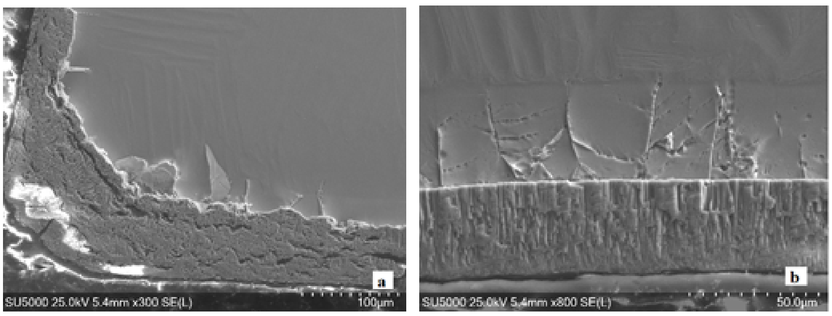

Fig.5 Degradation of the oxide-metal interface at 1000°C, with the increase in the number of thermal shocks: a-3 cycles, b-6 cycles

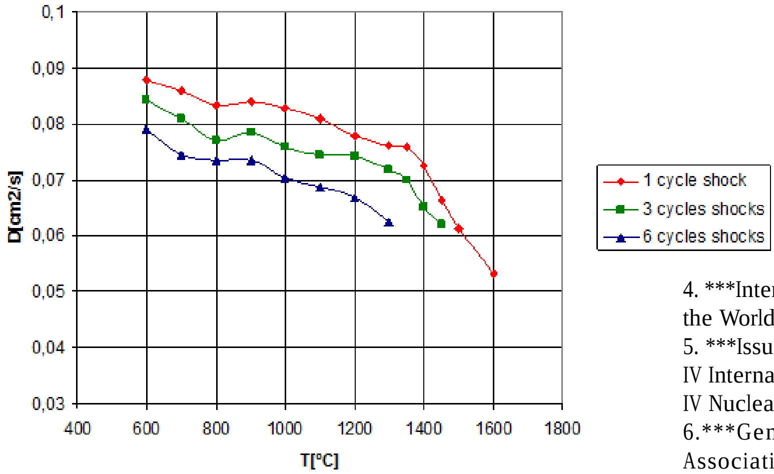

Fig.6 Variation of thermal diffusivity determined at 350°C depending on the temperature and number of shocks of 60s duration, relative to the untreated sample diffusion

Thermal shock temperature increase leads to diffusivity decrease, a phenomenon associated with grain size increase and with oxide layer degradation processes [16, 29].

As the temperature of allotropic transformation is reached, the values of thermal diffusivity present a narrow range of sudden growth, a phenomenon typical of metals in the field of structural transition, which was also emphasized in the case of Zr-Nb alloys [26].

Conclusions

The zircaloy – 4 specimens were tested to thermal shocks of 60s, at temperatures up to 1350°C and also to cyclic thermal stress of 3 or 6 treatment cycles.

The development of the oxide layers, the structural transformations of the metallic mass and the degradation process of the oxide metal interface were shown.

The diffusivity determinations for the specimens treated with a single thermal shock showed that thermal diffusivity decreases continuously as the shock temperature increases, a phenomenon which can be correlated with the increase of the metal mass grain size [16,29]. According to the existing literature, this phenomenon can be correlated with conductivity decrease through the increase of the oxide layer porosity [30].

For cyclic stresses, at the same treatment temperature and duration of the thermal shock, the diffusivity values decrease according to the number of the thermal cycles applied.

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