MATEC Web of Conferences **110**, 01084 (2017) DOI: 10.1051/matecconf/201711001084
HMTTSC-2017

High temperature x-ray diffraction of zr-2.5nb during thermal cycling in vacuum

Mikhail Tumanov[1,*], *Lyudmila* Lyubimova[1], and *Evgeniy* Puzyrev[2]

[1]National Research Tomsk Polytechnic University, 634050 Tomsk, Russia
[2]Polzunov Altai State Technical University, 656038 Barnaul, Russia

Abstract. The cyclic thermal tests in vacuum of zirconium alloy Zr-2.5Nb in the temperature range 250-350°C is established the presence of anomalies of thermal deformation of the crystal lattice, reducing the efficiency of the fuel rods.

1 Introduction

Zirconium tubes for nuclear reactors undergo irreversible deformation, which on the one hand, creates a threat of destruction of safety barriers due to the non-design stresses, on the other hand, the forming of the cladding restricts the depth of burnup of the fuel and thereby reduces the efficiency of the re-actor [1]. The relationship between the deformation due to radiation swelling that occurs with stress and performance elements of the active zone was the basis for the methodology for thermal cycling tests to determine thermal dilatations of the crystal lattice of the zirconium alloy.

As an object of research used a sample of the channel tube of zirconium-niobium alloy Zr-2.5Nb (known in Russia as E-125 alloy) with the content of niobium in the amount of 2.4-2.7 percent.

Methodology for thermal-cycle tests simulated the processes of thermal fatigue and was playing a sequence of cycles "heating – cooling – heating" with the increase in temperature in each subsequent cycle.

2 Material and methods

Experimental data on changes of the shape and size of the metals showed that the size can both increase and decrease as thermal-cycling [2].

Thermal tests were conducted in vacuum by means of high temperature x-ray diffraction (HT XRD) using x-ray diffractometer and a high temperature diffractometric installation "UVD-2000". The wavelength of the used radiation was λ_{Co} = 0.71069 Å. For analysis of the selected two x-ray diffraction line index (002) and (101), the angles of diffraction which served as the basis for determining the size and crystalline with the hexagonal lattice of the alloy of zirconium, which were determined according to the basic formula of crystallography [3].

* Corresponding author: tumanov_mihail@mail.ru

The relative error in the measurement of the lattice parameters were: $\Delta a/a \leq \pm 0.03\%$ and $\Delta c/c \leq \pm 0.3$ %.

In Fig. 1-3 shows the changes in a, c and the relationship *c/a* unit cell of an alloy of zirconium in the process of thermocycling.

For high-purity zirconium, a = 3.23118 Å, c = 5.14634 Å, the ratio *c/a* is: *c/a* = 1.59271 [4, 5].

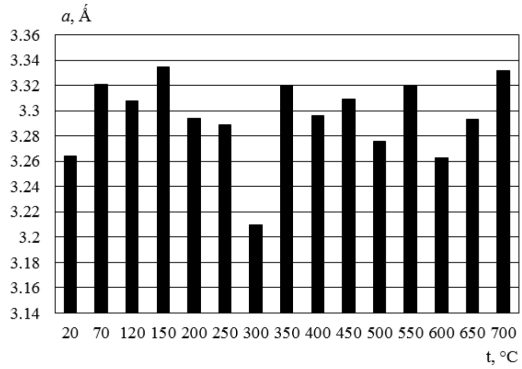

Fig. 1. The dependence of the parameter *a* zirconium alloy Zr-2.5Nb from temperature.

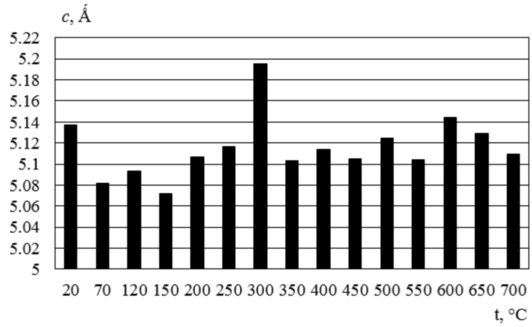

Fig. 2. The dependence of the parameter c of zirconium alloy Zr-2.5Nb from temperature.

Data on the value of *c/a* for various zirconium alloys may differ markedly from each other, because the lattice parameters are significantly influenced by alloying elements, impurities of introduction and substitution, temperature, texture, pre-strain, etc [5]. All this testifies to the need to establish patterns of behavior for samples of similar products the specific composition and shape.

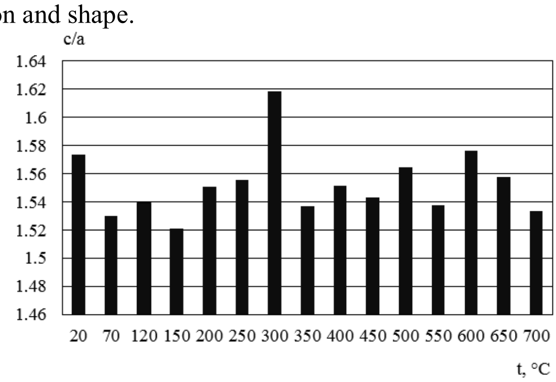

Fig. 3. The ratio *c/a* of zirconium alloy Zr-2.5Nb.

DOI: 10.1051/matecconf/201711001084

From Fig. 1-3 it is seen that the lattice parameters of the alloy reveal a very complex temperature dependence.

Thus, the lattice parameter *a* oscillates, showing some temperature negative creep (Fig. 1): about of 100°C, in the range of temperatures in the 350-400°C, 450-500°C, 550-600°C. the temperature anomaly for lattice parameter as stated in the service temperature for the alloy of zirconium 250-350°C. The temperature anomaly for lattice parameter as stated in the service temperature for the alloy of zirconium 250-350°C.

For parameter c in addition to negative effects creep there is an anomalous jump in the thermal strains in the temperature 300°C (Fig.2).

The dependence of the ratio c/a of crystal lattice is also from differs the structural instability is most pronounced for the temperature range 250-350 °C (Fig.3).

Anomalous effects of thermal deformation of the crystal lattices relates to the field of polymorphic transformations [4].

The feature of polymorphic transformations in structural materials as a kind of phase transitions is that in the process of transformations, for example, under the action of tempera-tours, a substance changes the density, while in solid-house condition. When this occurs, the change of microhardness, coefficient of hydrogen diffusion, heat capacity, there is a temperature hysteresis.

Observed anomalous changes in the properties of coefficient of Li-Nanoha the extension of the young's modulus, elongation, ductility and heat capacity in strictly certain intervals of temperature [6]. Thus the symmetry of the crystal lattice in the region of the anomalies is not changed, therefore, to attribute the anomalies to the polymorphic phase transitions cannot, accordingly, not found out the cause. It is important to note that anomalies are inherited with chemical compounds and alloys like structural polymorphism. For example, if the Zirconia has phase transitions of the first kind and the anomalous effects of thermal deformation in the studied temperature region, but the composition of the alloy is, for example, Nickel having this effect, the alloy of zirconium with the addition of Nickel inherits this feature [7]. In particular, the Nickel included in the zirconium alloy Zr-2.5Nb has an anomaly of thermal deformation of the crystal lattices in the region of 300-400 °C, i.e. approximately the same as an alloy of zirconium [4].

It is fair to assume that the presence of Nickel in alloy composition can have a negative impact on its service properties, as in the anomalous temperature points have been the following material properties and processes:

– superplasticity – a sharp decline in the tensile strength and tough-ness in zirconium alloys (iron, titanium);

– 10 to 20 – fold stepwise elongation of the metal without the necking and rupture;

– punching leads to a smaller grain;

the quenching retains the properties of the high temperature phase, which started cooling;

– annealing of the deformed metal below the abnormal point leads to small grain, and at temperatures above the anomalous pointing to large (different speed of grain growth);

– thermal cycling through the anomaly causes irreversible deformation of the products;

– increase the sorption properties of the material and azoproite the cost;

– with increasing degree of deformation increases the rate of oxidation and dissolution of metals in chemical reactions [2].

Negative creep is associated with bond breaking and formation of microcracks and porosity [8]. The occurrence of porosity and microcracks in the structure of the alloy of zirconium is of great importance, because the zirconium is practically the only metal (even titanium), dis-solving large quantities of oxygen, which forms with the zirconium as a chemical compound ZrO_2, and the phase of introduction of oxygen into the crystal lattice. Non-metallic impurities (O, H, C, N) significantly alter the properties of zirconium. The dissolution of oxygen in zirconium is an important process associated with the formation of protective oxide films. Corrosion behavior of zirconium and its alloys in a steam environment is characterized by the presence of two periods. In the first period to fracture on the surface of the zirconium oxide film is formed, providing a high protective function, and the oxidation rate of zirconium stabilizirue-most. After reaching a certain critical thickness of the oxide layer suddenly collapses – this is the second period of oxidation, which oxide layer will no longer protect the metal and it is the rapid destruction [1, 5, 9].

In this regard, one of the problems is the loss of protective proper-ties of oxide films of ZrO2. This important issue is debated until the pre-sent time. To explain the second period in the oxidation kinetics of zirconium involved various theories and mechanisms, in particular the theory of mechanical cracking of the oxide film [1, 5, 9].

On the basis of the obtained experimental results (Fig. 1-3) it is believed that the properties of the oxide films zirconium and their destruction affects the anomalous behavior of the alloy at the operating temperature and accumulation of damage in the presence of even minor temperature gradients. Accumulation of damage leads to a limit state surface, its destruction and the destruction of the protective oxide film. The trigger to the destruction of the oxide film are the processes of destruction of the zirconium alloy, which can not affect the structure of the formed protective oxide ZrO_2.

Thus, the determining process in the destruction of the oxide-film Noah, or important, may be internal structural state of the alloy, the mechanisms and rate of degradation.

Impurities, texture and nature of the structure of intra - and inter-grain boundaries will have a significant impact on the other properties, such as hydrogenation. It is known that zirconium alloys actively absorb hydrogen, and the deformation of specimens of alloy of zirconium, as shown in Fig. 1-3, will only lead to increase of hydrogen absorption.

3 Conclusions

The result of the research established the following.

1. The crystal lattice of the zirconium alloy Zr-2.5Nb he-have a complex dependence on temperature, showing some values, negative creep, which is associated with micro cracking.

2. The crystal lattice of the zirconium alloy Zr-2.5Nb exhibit the effects of abnormal thermal expansion of the crystal re-bars in the absence of phenomena of polymorphism in the region of temperatures of the experiment. The effect is most evident in the field of operating temperatures of 250-350°C and can be the cause of the destruction of protective oxide films.

Thus, speaking on the anomalous characteristics in relation to thermal stress to structural elements of nuclear power reactors can be expected that the study about anomalous phenomena provides the most complete picture of the behaviour of multicomponent metallic systems at high temperature

The reported study was supported by RFBR grant "Fundamentals of engineering Sciences" No. 15-08-99544a.

References

1. L. A. Belyanin, V. I. Lebedev, Yury V. Garusov, *Bezopasnost' AEHS s kanal'nymi reaktorami. Rekonstrukciya aktivnoj zony* [Safety of nuclear power plants with channel reactors. Reconstruction of the active zone] (MEI, Moskov, 1997) [in Russian]

2. V.F. Kovalenko, *Teplofizicheskie processy i ehlektrovakuumnye pribory* [Thermophysical processes and electrovacuum devices] (MEI, Moskov, 1975) [in Russian]
3. S. S. Gorelik, Y. A. Skakov, L. N. Rastorguev, *Rentgenograficheskij i ehlektronno-opticheskij analiz* [Radiographic and electron-optical analysis] (MEI, Moskov, 2002) [in Russian]
4. V.A. Finkel. *Vysokotemperaturnaya rentgenografiya metallov* [High-temperature radiography of metals] (MEI, Moskov, 1968) [in Russia]
5. D. Duglas, *Metallovedenie cirkoniya* [Metallography of zirconium] (MEI, Moskov, 1975) [in Russian]
6. L.L. Lyubimova, A.A. Makeev, A.A. Tashlykov, Steel in Translation, **2** (2014)
7. Y.D. Vishnyakov, D*efekty upakovki v kristallicheskoj structure* [Defects of packing in the crystal structure] (MEI, Moskov, 1970) [in Russian]
8. A.S. Zavorin, L.L. Lyubimova, A.A. Tashlykov, R.N. Fisenko, R.B. Tabakaev, Sposob rentgenometricheskoj ocenki temperaturnyh uslovij ehkspluatacii trubnyh ehlementov kotlov [Method radiometric evaluation of temperature operating conditions of tube elements of boilers]. Patent RF, no. 2012147388/28, 2006
9. B.G. Parfenov, V.V. Gerasimov, G.I. *Venediktova, Korroziya cirkoniya i ego splavov* [Corrosion of zirconium and its alloys] (MEI, Moskov, 1967) [in Russian]