Characteristics and Properties of Cladding Tubes for VVER-1000 Higher Uranium Content Fuel Rods

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Introduction

To improve the fuel cycle economics and to further increase the VVER fuel usability the work programme is under way to design novel improved fuel, fuel rods and fuel assemblies. Longer FA operation time that is needed to increase the fuel burnup and the related design developments of novel fuel assemblies resulted not only in changing types and sizes of zirconium items and fuel assembly components but also altered the requirements placed on their technical characteristics [1]. To use fuel rods having a larger charge of fuel, to improve their behaviour in LOCA, to reduce fuel rod damageability during assembling the work was carried out to perfect the characteristics of both the cladding (reduced wall thickness and more rigid tolerances for geometry) and its material. To meet the more rigid requirements for the geometry dimensions of cladding tubes an improved process flow sheet has been designed and employed for their fabrication and also the finishing treatment of tube surfaces has been improved. The higher and stable properties of the cladding materials were managed through using the special purity in terms of hafnium zirconium (not higher than 100 ppm Hf) as a base of the E110 alloy and maintaining within the valid specifications for the alloy the optimized contents of oxygen and iron at the levels of (600 – 990) ppm and (250 – 700) ppm, respectively [2]. The work was under way in 2004 – 2008 years; during this period the technology and materials science solutions were mastered that were phased-in introduced into the production of the cladding tubes for the fuels loaded into the 1st unit of the Kalinin NPP.

1 Requirements for Geometry and Material of Higher Uranium Content Fuel Rods

Developments and introduction of higher uranium content fuel rods for VVER required correspondingly to design and optimize the production of novel range cladding tubes. To accomplish this aim the novel requirements have been formulated for the cladding geometry parameters that involved the following:

- an inner diameter increase through a decrease in the minimal wall thickness to 0,54 mm;
- introduction of a requirement for difference in thickness;
- more rigid requirements for the tolerated outer diameter and surface roughness.

On the basis of the formulated requirements novel specifications were worked out for cladding tubes of novel generation fuels having higher uranium content.

Thinner claddings and fuel rod operation in longer fuel cycles with the prospects to reach in future the burnup of up to 80 MW·d/kg U place more rigid requirements on a cladding material also in terms of a margin for a shape change resistance under irradiation. As it is revealed by the available results to provide a property margin for the novel generation fuel rod strength, irradiation induced thermal creep and irradiation growth in comparison to the standard cladding in the Zr sponge base E110 alloy the contents of oxygen and iron have to be respectively maintained at the levels 0f (600 – 990) ppm and (250 -700) ppm [2, 4].

During the design cycle the VVER fuel rod cladding material has to meet the specific requirements that also involve the absolute adherence to the design basis (LOCA, RIA) criteria, specifically, in excluding embrittlement. In view of this primarily zirconium sponge that ensures a more stable behaviour of the alloy under high temperature oxidation conditions is contemplated for use as the base of E110 alloy to be employed for the claddings of the novel generation fuel rods [3].

The practical realization of all the novelties relevant to the claddings of the novel generation fuels proceeded in stages during the period from 2004 to 2008 to produce batches of the cladding tubes for the VVER-1000 fuel rods of unit 1 of the Kalinin NPP.

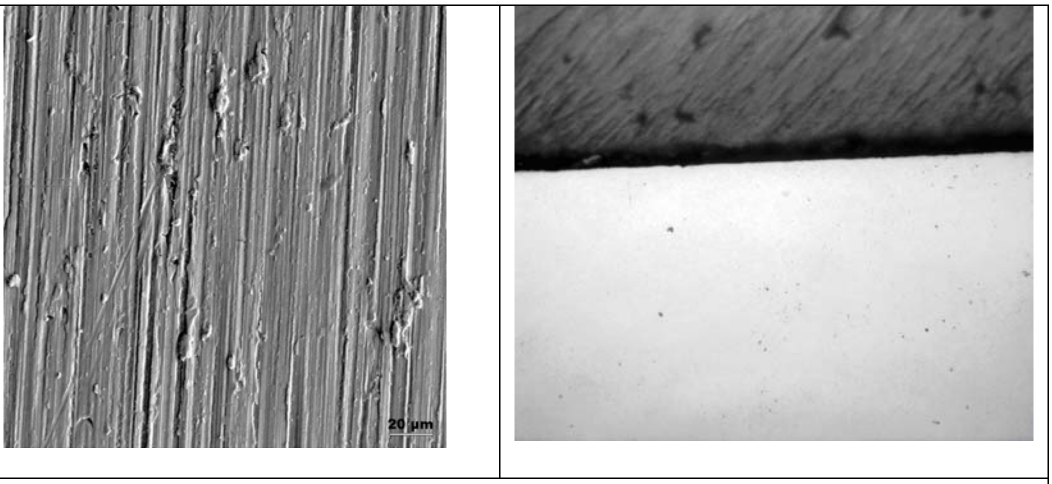
2 Characteristics and Properties of Cladding Tubes from Sponge Base Optimized Composition E110 Alloy Complying with Higher Requirements for Geometry

At JSC “ChMP” for the production of cladding tubes Ø9,10x7,93 mm from the sponge base E110 alloy having more rigid tolerances for geometry and the optimized contents of oxygen (600 – 990) ppm and iron (250 – 700) ppm an advanced deformation scheme was designed that involved hot pressing a larger billet, radial forging pressed sleeves followed by cold rolling in four stages. Aside from this, the finishing treatment of tube surfaces has been improved via grinding outer surfaces and jet etching inner surfaces of tubes.

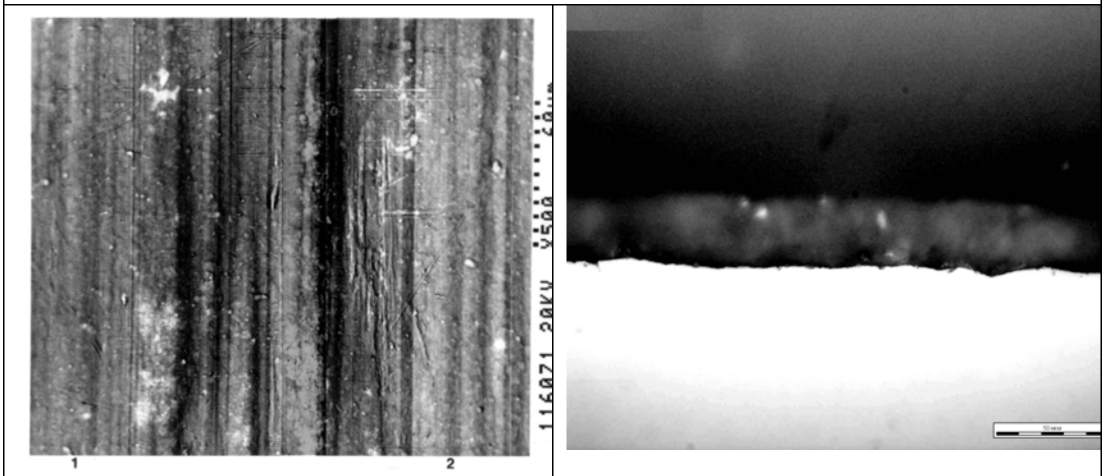
The analysis of the geometric characteristics of tubes commercially produced from the E110 alloy optimized in oxygen and iron using the designed deformation schema and the improved finishing treatment of the surface has demonstrated that the scatter in the dimensions along the tube lengths is in the following ranges: the outer diameter – 0,001 ÷ 0,008 mm; the inner diameter - 0,001 ÷ 0,009 mm; the wall thickness - 0,001 ÷ 0,008 mm; the thickness difference - 0,001 ÷ 0,016 mm; the ovality - 0,001 ÷ 0,008 mm. The difference in the thickness does not exceed 0,050 mm, the ovality – 0,040 mm. The more rigid tolerances for the outer diameters and the reduced roughness of the tubes lead to a ~ 15 % lower mean force applied during the fuel rod assemblage and narrows the range of the applied forces.

The characteristics and properties of the novel cladding tubes were assessed by comparing to the standard tubes.

The scanning electron microscope examinations of the outer ground and etched surfaces of the E110 alloy tubes revealed some differences between them (figure 1).



Ground surface



Etched surface

Figure 1 – Surface condition of E110 alloy cladding tubes (x 500)

The surface of the ground tube has a more even and smooth microrelief in comparison to that of the etched tubes. Besides, the outer surface in the ground tubes features the availability of a work hardened layer 12 – 15 µm deep (figure 2). In this case with an increase in the oxygen content the microhardness of the outer surface and the strain hardening degree increase.

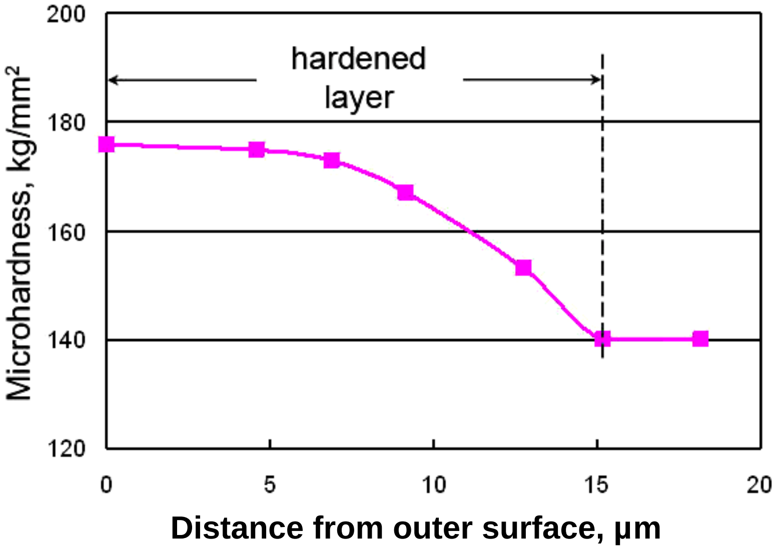


Figure 2 – Microhardness changes in depths of outer surfaces of ground tubes

The investigations reveal that the mechanical processing and, hence, the resultant work hardened layer did not result in a lower corrosion resistance of tubes in water or steam (table 1).

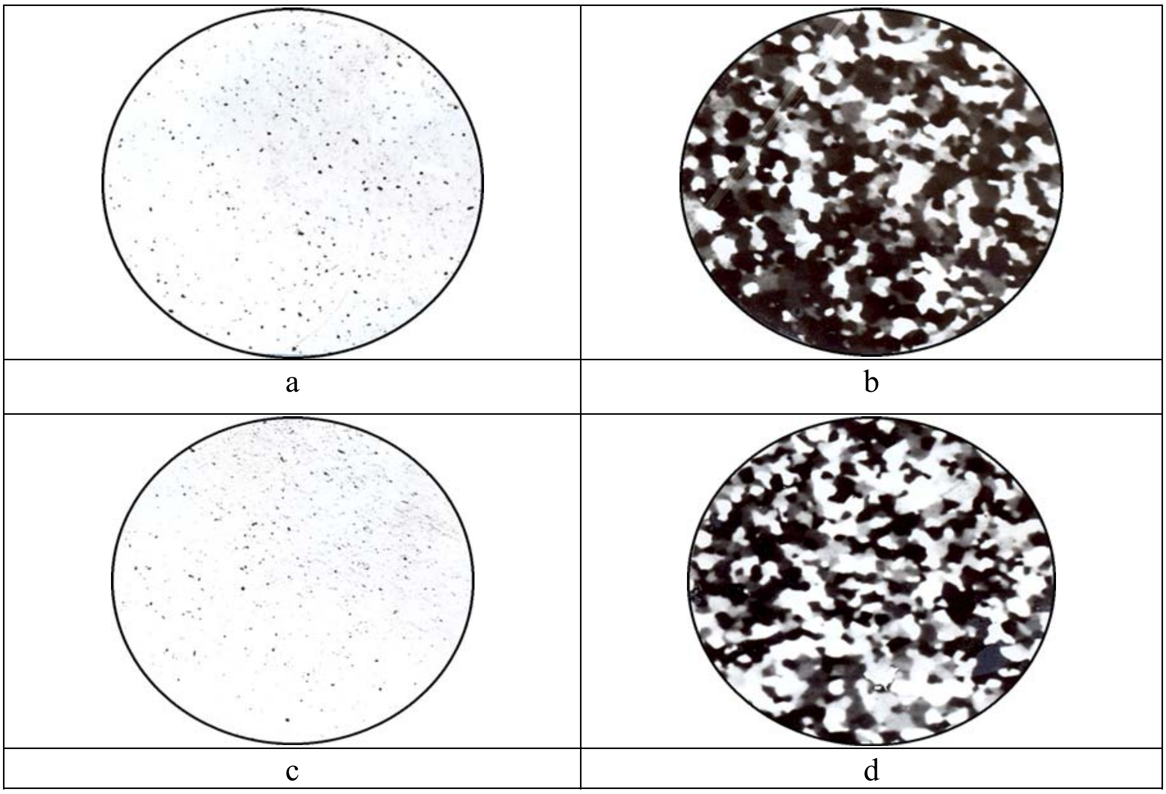
Table 2 - Results of Roughness Measurement and Corrosion Tests

Type of testing	Parameter	Improved tube	Standard tube
Determination of roughness	Outer surface R$_a$, µm	0,22 - 0,45	0,21 - 0,59
	Inner surface R$_a$, µm	0,25 - 0,67	0,22 - 0,69
Corrosion tests in steam, 400 °C - 72 h	Weight gain, mg/dm^2	14 - 17	11 - 16
	Surface condition	Adequate	Adequate
Corrosion tests in water, 360 °C – 300 days	Weight gain, mg/dm^2	50 - 54	51 - 54
	Oxide coat	dark	dark
Corrosion tests in steam, 400 °C – 300 days	Weight gain, mg/dm^2	230 - 235	231 - 236
	Oxide coat	Dark grey	Dark grey

The characteristics of the microstructure, texture and hydride orientation in tubes ∅9,10×7,93 mm, from the E110 alloy having higher contents of oxygen and iron fabricated by the novel process flow sheet also remained at the level of the standard products (table 2). The microstructures of the improved and standard tubes were fully recrystallized ones having equiaxial grains of the mean sizes ~ 7 μm (figure 3). The hydride phase precipitates in the improved and standard tubes primarily have the tangentional orientation (figure 4) that provides for the hydride orientation coefficient within 0,11-0,33 which is below the specified requirements (≤ 0.4).

Table 2 – Microstructure and Texture Parameters of E110 Alloy Tubes

Testing conditions	Parameter	Improved tube	Standard tube
Microstructure parameters	Mean grain size, μm	6,8-7,0	7,4-7,7
	Microhardness	140-155	100-125
Texture parameters	fr	0,55-0,59	0,54-0,59
	ft	0,35-0,37	0,35-0,37
	fl	0,05-0,10	0,06-0,10
Hydriding	Coefficient of hydride orientation	0,11-0,26	0,14-0,33



a

b

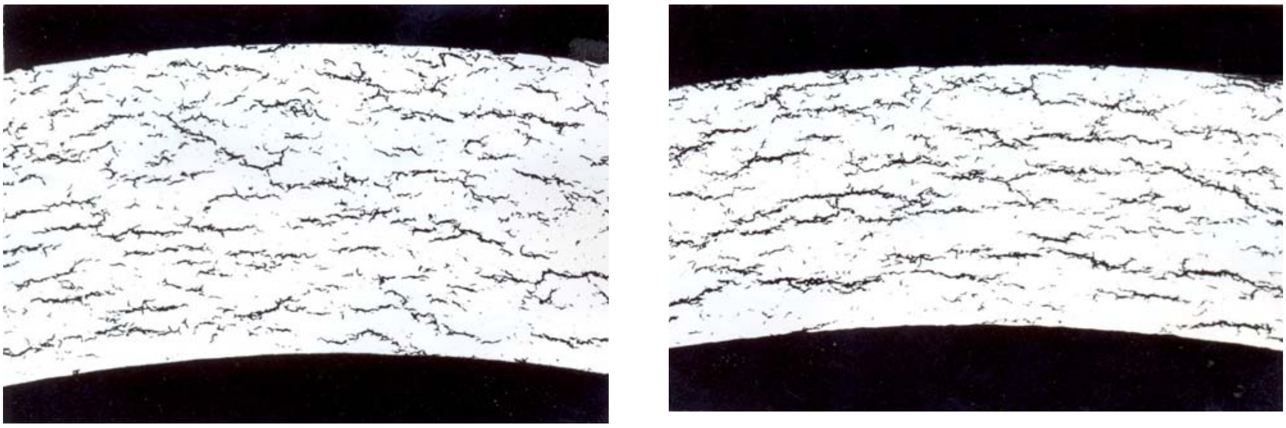
c

d

a, b – tubes ∅9,10×7,73 mm

c, d – tubes ∅9,10×7,93 mm

Figure 3 – Microstructure of E110 alloy tubes in reflected and polarized light, × 500



a
Fn = 0,18

b
Fn = 0,12

a – tube Ø9,10×7,73 mm, b – tube Ø9,10×7,93 mm

Figure 4 – Typical orientation of hydrides in tubes hydrided in a gas saturation facility, × 63

The direct pole figures of the improved tubes do not differ from the pole figures of the standard tubes (figure 5) which is also corroborated by the values of the texture parameters (the values of Kearns parameters of all the tubes in the radial and tangential directions make up 0,54 – 0,59 and 0,35 – 0,37, respectively).

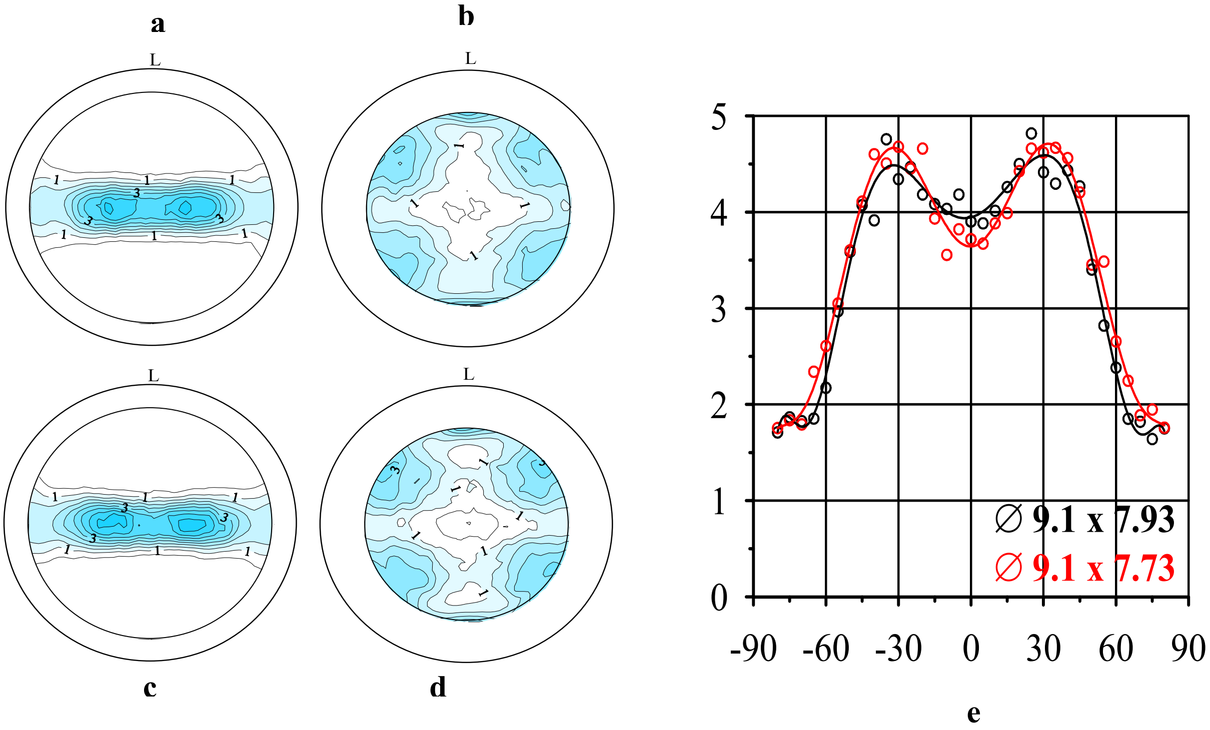


Figure 5 – Direct pole figures (0001) (a, c) and {11.0} (b, d) and distribution of basal normals in RT - section (e) of tube specimens ∅9,10x7,93 mm (a, b) and ∅9,10x7,73 mm (c, d)

As far as the tensile properties, in the range of the valid specifications for E110 alloy the contents of oxygen and iron maintained at the levels of (600 – 990) ppm and (250 – 700) ppm, respectively, lead to the strengthening of the cladding material at room and operation temperatures if compared to the standard tubes (table 4). For instance, at the room temperature the ultimate strength upon the iniaxial tension in the cross direction increased by 4 – 8 kg/mm^2 while upon the biaxial stress in testing under the internal pressure the increase was 4 – 5,5 kg/mm^2. In the uniaxial tension tests at 380 ºC the strength characteristics in the longitudinal and cross directions increased insignificantly (by 1 – 2 kg/mm^2) while the total hoop creep strain at 400 ºC and the stress of 100 MPa after 3000 h testing under the internal pressure decreased from 4,2 to 3,2 %.

Table 4 – Results of Tensile Tests

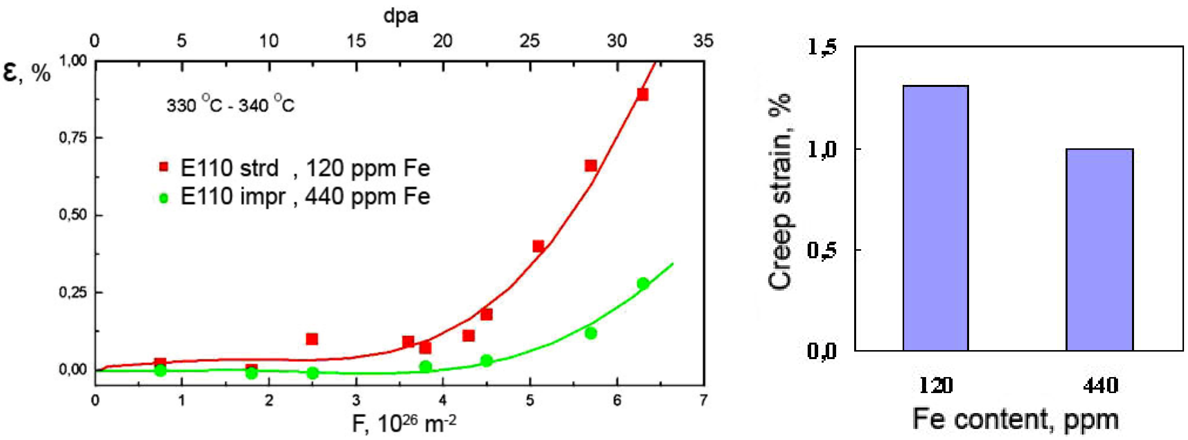
Testing conditions	Parameter	Improved tube	Standard tube
At 20 °C uniaxial tension in cross direction	σ_b, kg/mm^2	40 - 46	35 - 41
	$\sigma_{0.2}$, kg/mm^2	35 - 39	31 - 34
	δ, %	26 - 35	31 - 37
At 380 °C uniaxial tension in cross direction	σ_b, kg/mm^2	19 - 21	18 - 19
	$\sigma_{0.2}$, kg/mm^2	17 - 18	16 - 17
	δ, %	39 - 44	35 - 41
in longitudinal direction	$\sigma_{0.2}$, kg/mm^2	10 - 12	10 - 11
At 20 °C for rupture under internal pressure	σ_b, kg/mm^2	57 - 59	52 - 54
	δ, %	74 - 83	83 - 99
At 400 °C and 130 MPa for creep under internal pressure (after 240 h)	Strain, %	1,8 - 2	2,2 - 2,5
At 400 °C and 100 MPa for creep under internal pressure (after 3000 h)	Strain, %	3,1 - 3,3	4,1 - 4,4

Along with the alloy hardening a 2 – 4 % increase in the relative elongation in the cross direction at 380 °C and some decrease in ductility at the room temperature are observable.

The effect of the E110 alloy composition optimization is also revealed by BOR-60 irradiated samples; in this instance an increase in the iron content of the alloy also influences the properties of the claddings [2, 4]. The cladding tubes fabricated from the optimized alloy have higher strength properties (table 5), resistances to irradiation induced creep and growth (figure 6).

Table 5 – Results of Tensile Testing Annular Tube Samples BOR - 60 irradiated at ~330 °C to Neutron Fluence of $2 \cdot 10^{26}$ m^{-2}

Testing conditions	Parameter	Improved tube	Standard tube
At 20 °C initial tension in cross direction	σ_b, kg/mm^2	57 - 60	47 - 49
	$\sigma_{0.2}$, kg/mm^2	52 - 55	41 - 44
	δ, %	13 - 16	14 - 19
At 350 °C initial tension in cross direction	σ_b, kg/mm^2	35 - 36	32 - 33
	$\sigma_{0.2}$, kg/mm^2	32 - 33	30 - 31
	δ, %	25 - 27	18 - 21



a b

Figure 6 – Irradiation growth strain during 4200 h vs neutron fluence [4] (a) and irradiation creep strain at T_{irr} = 330 °C for 2900 h (F =1 × 10^{26} m^{-2}) (b) of E110 alloy cladding tubes (irradiation in BOR-60)

The use of sponge zirconium for fabricating tubes by the novel process using the E110 alloy of the optimized composition made it possible to improve and stabilize the corrosion resistance of the tubes under conditions of the high temperature oxidation. Samples of tubes fabricated on a sponge base when tested at 1200 ºC to 18 % ECR (local oxidation depth) have a dark lustrous coat as distinct from the dark coat having whitish isolated nodules on the standard tubes (figure 7). The residual ductility of the improved tubes and the standard ones meets the design requirements. At ISC “VNIINM” the work is currently under way to increase the design margin of the high temperature oxidation of the standard E110 alloy.

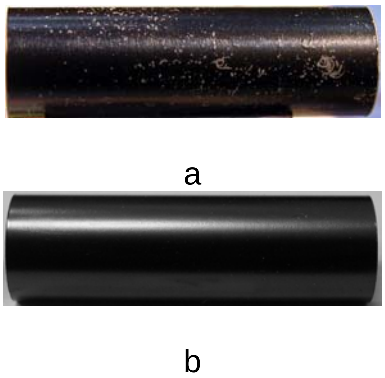


Figure 7 – Appearance of steam oxidated (1200 ^{O}C) cladding tube from E110 alloy on bases of electrolytic and iodide (a) and sponge (b) Zr

Conclusion

1 To increase the fuel charge of VVER-1000 rods and to promote their serviceability in long-term fuel cycles at the burnup up to 80 MW·d/kg U:

- the inner diameter of claddings is increased via decreasing the minimal wall thickness to 0,54 mm;

- more rigid requirements are placed on the geometrical parameters of cladding tubes;

- the oxygen and iron contents of the E110 alloy were optimized to the level of (600 – 990) ppm and (250 – 700) ppm, respectively, to provide for the margin of strength, irradiation-thermal creep and irradiation induced growth;

- zirconium sponge was used as a base of the alloy to increase its margin for promoting criteria of design basis accidents LOCA and RIA.

2 To produce cladding tubes complying with higher requirements for the geometry the process flow sheet of their fabrication was upgraded to include at the finishing stages the tube outer surface grinding and the tube inner surface jet etching.

3 In 2004 – 2008 some individual options of the developments were phased-in introduced into the VVER-1000 assembly fuel rods in unit 1 of the Kalinin NPP.

List of References

1. Новиков В.В. Твэлы водоохлаждаемых реакторов. Перспективы развития. // Тезисы докладов НТК-2008 «Ядерное топливо нового поколения для АЭС. Результаты разработки, опыт эксплуатации и направления развития», Москва, 19-21 ноября 2008 г., ВНИИНМ, с. 33.

2. Маркелов В.А., Новиков В.В., Коньков В.Ф., Перегуд М.М., Шишов В.Н., Гусев А.Ю. Развитие работ по разработке сплавов циркония для оболочек твэлов и деталей ТВС // Тезисы докладов НТК-2008 «Ядерное топливо нового поколения для АЭС. Результаты разработки, опыт эксплуатации и направления развития», Москва, 19-21 ноября 2008 г., ВНИИНМ, с. 43

3. Novikov V.V., Markelov V.A., Shishov V.N., Tselishchev A.V., Balashov A.A. ”Results of post–irradiations examinations (PIE) of E110 claddings and alloy upgrading for VVER” // 2006 International Meeting on LWR Fuel Performance “Nuclear Fuel: Addressing the Future”, 22-26 October 2006, Salamanca, Spain / Transactions Topfuel 2006 - Poster Session II, p.p. 97-101.

4. Маркелов В.А., Новиков В.В., Перегуд М.М., Коньков В.Ф., Шишов В.Н., Балашов А.А. «Оптимизация сплава Э110 для оболочек твэлов ВВЭР-1000» // Сборник тезисов 5-й международной научно-технической конференции «Обеспечение безопасности АЭС с ВВЭР», 29 мая – 1июня 2007, Подольск, Россия, с. 72.