SECTION 5. ENERGY AND ENERGY ENGINEERING AND TECHNOLOGIES

5.1 Analysis of safety criterion maked on fuel rod state in design basis accidents, limiting embrittlement of fuel cladding material

The paper reveals briefly experimental studies carried out to validate safety criteria (acceptance criteria). The scope of the data experimentally obtained by research the behaviour and properties of VVER type fuel claddings from Zr1%Nb alloy under loading conditions simulating the stage of core flooding with water in loss of the coolant accident conditions (LOCA) suffices to judge the character and numerical value of criterional parameters of the embrittlement criterion in terms of the cladding stability upon flooding and subsequent implementation of fuel assembly unloading and transportation. Accidents are considered involving loss of coolant by primary circuit which are characterized by conditions of degraded heat transfer from fuels. During accidents loss of tightness by fuel rod cladding is tolerable, however, in this case, the cooling of a distorted fuel rod and the dismantling (unloading) of the core after an accident have to be feasible.

Keywords: loss of the coolant accident conditions, fuel rod, Zr1%Nb alloy, oxidation, deformation, heat resistance.

Introduction

The objective of the work is to validate the fulfillment of safety requirements placed on fuel rod condition in design basis accidents. A design basis accident is the one for which the design defines initial events and final states as well as contemplates safety systems that with the account for a single failure of safety systems or a single staff error, independent of an initial event, provide for the limitation of its consequences via limits established for accidents of this type [253, 254].

Accidents are considered involving loss of coolant by primary circuit which are characterized by conditions of degraded heat transfer from fuels. Under these conditions, the temperature of fuel rods (fuel and/or cladding) rises compared to that under normal operating conditions. During accidents loss of tightness by fuel rod cladding is tolerable, however, in this case, the cooling of a distorted fuel rod and the dismantling (unloading) of the core after an accident have to be feasible.

Experimental validation of embrittlement criterion for Zr1%Nb claddings

In process of LOCA, the inadequate cooling of fuel claddings gives rise to their heating to reach high temperatures. Under these conditions, their intensive steam oxidation is feasible.

The degree of the cladding oxidation is basically governed by the levels of temperatures and pressure, time of oxidation in steam and deformation.

As a results of the cladding-steam interaction, the cladding material becomes brittle.

Because of the cladding material embrittlement, the Zr1%Nb alloy changes its original thermophysical and mechanical properties, and the ductility characteristics become lower. The thickness of the unoxidized cladding material is reduced.

Temperature stresses of cladding arising at the stage of core flooding with water from the emergency cooling system as well as mechanical loads effected by fuel assembly (FA) unloading and transportation may lead to the fragmentation of an embrittled cladding.

The degree of Zr1%Nb cladding-coolant interaction is stipulated by the maximum limit of a fuel rod damage in terms of the tolerable depth of cladding oxidation, namely, the embrittlement criterion [253].

The degree of the Zr1%Nb cladding embrittlement is limited by two criterional parameters $T_{clad}^{\lim}$ and local oxidation depth of cladding (equivalent cladding reacted - ECR) $ECR_{clad}^{\lim}$:

– the maximal cladding temperature T_{clad} does not exceed 1200^0C

$$T_{clad} < T_{clad}^{\lim} = 1200^{o}C.$$

The ultimate value of the fuel cladding temperature $T_{clad}^{\lim}$ characterizes the temperature with the exceeding of which a self-sustaining Zr-steam reaction may begin.

– the maximum ECR is not more than 18% of its original thickness $ECR_{clad}^{\lim}$

$$ECR < ECR_{clad}^{lim} = 18\%.$$

The ultimate magnitude of ECR reflects the degree of the embrittlement of a fuel cladding material and, if it is exceeded, a cladding may fracture in brittle mode as a result of loads produced by the emergency core cooling, discharge, handling and transportation of fuel assemblies.

ECR is an overall thickness of an equivalent Zr layer (that would react with steam assuming the whole locally absorbed oxygen would be spent to form stoichiometric Zr, ZrO_2) related to the original cladding thickness.

If a fuel rod is ruptured (depressurized), the oxidation of both outer and inner surfaces of cladding are taken into account.

The degree of oxidation as applied to Zr1%Nb claddings is assessed from either the oxygen specific weight gain, Δm, or ECR.

ECR and weight gain, Δm, are related by the ratio [253-256]

$$ECR = N \times (\delta_e / \delta_o) \times 100, \ \%$$

where N - is the coefficient that takes into account the oxidation of cladding at both the surfaces (N=2 for a ruptured);

δ_o - is original thickness of specimen, cm;

δ_e - is thickness of equivalent layer (calculated thickness of Zr layer that would be spent to form ZrO_2), cm;

$$\delta_e = \mu_{Zr} / \mu_{O2} \times 1/\rho_{Zr} \times \Delta m;$$

μ_{Zr}, μ_{O2} - are molecular masses of zirconium and oxygen, respectively;

ρ_{Zr} - is zirconium density, mg/cm^3;

Δm - is specific weight gain, mg/cm^2,

$\Delta m = \Delta M/S_o$;

ΔM - is oxygen weight gain, mg;

S_o - is area of original cladding surface, cm^2.

It is to be pointed out that the specific weight gain of a cladding is found by dividing the weight gain by the area of the original cladding surface. This gives a conservative assessment of the specific weight gain. The real oxidized area may be larger at the expense of possible cladding deformation.

The ration to determine ECR has the form of

$$ECR = N \times 4{,}355 \times 10^{-2} \times \Delta m / \delta_o, \quad \%. \tag{1}$$

It is recommended to determine the oxygen weight gain of Zr1%Nb claddings via the parabolic ratio [257]

$$\Delta m = 920 \times \exp(-10410/T) \times \sqrt{\tau}, \tag{2}$$

where Δm - is specific weight gain, mg/cm^2;

T - is temperature, K;

τ - is time, s.

The relation (2) recommended for the assessment the degree of Zr1%Nb cladding oxidation is conservative at the temperatures of (900-1200)^{0}C and the time of the alloy-steam interaction up to 900s for the case of the availability of a hydrogen additive in steam, deformation of cladding and irradiation.

Fig.1 presents experimentally acquired values of the oxidation reaction rate constant, relationship (2) for Zr1%Nb alloy, as well as of Zry-4 oxidation rate and Baker-Just ratio obtained for Zr.

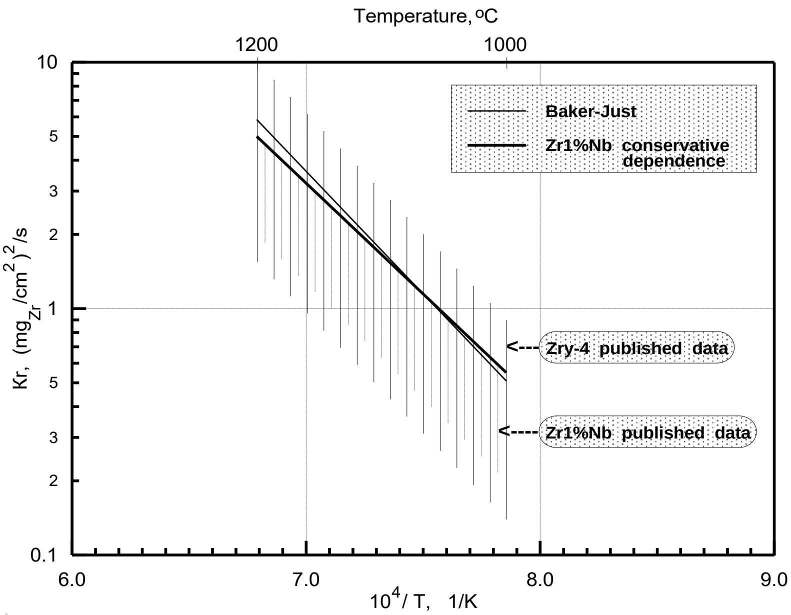

Fig. 1. Zr1%Nb and Zry-4 Reaction Rate Constant vs Temperature.

Results on experimentally validated embrittlement criterion

The embrittlement criterion pertaining to Zr1%Nb fuel claddings is validated by the results of the following experiments [255-260]:

– research of fuel cladding ability to withstand quenching (thermal shock tests);
– estimation of the physico-mechanical state of oxidized specimens after flooding with cool water or after quick removing into cool water (estimation of impact elasticity and residual ductility), metallographic examinations of oxidized specimens subjected to a thermal shock.

Thermal shock tests. Requirements for the procedure used to test for thermal shock (table 1) comprise the following:

- indirect heating of specimens in well thermostatically-controlled facility;
- isothermal exposure; time and temperature being recorded;
- quick removing of a specimen into cool water (or the flooding of a specimen from bottom);
- analysis of a tested simulator state;
- formation of simulator failure map.

In the experiments carried out with short-length simulators (table 2), account was taken of many loading factors involved in LOCA. The use of a short-length simulator ensured a uniform in height and time controlled temperature field. The availability of UO_2 pellets (or sintered Al_2O_3) gave essentially real values of temperature effected stresses arising in a cladding upon flooding (quick immersion) with cool water. The tests were implemented using both integer fuel rod simulators internally pressurized with an inert gas (deformation of cladding was simulated) and those having unsealed ends (not pressurized).

The experiments with unirradiated claddings were carried out in two facilities:

– with specimen heating in a shaft-type furnace; the weight gain of a specimen being continuously recorded during of the experiment – UNOPRO;
– with heating simulator cladding placed in the central hole of UO_2 pellets – TEFSAI.

The experiments with irradiated claddings were carried out in all-purpose rigs UVS [255]. The experiments to study the thermal resistance were carried out in a wide temperature range (900-1200)^{0}C; the cladding oxidation degree was different (from 18% to 60% ECR).

Table 1

The parameters of the thermal shock tests were as follows:

Parameters		Facility		
		UNOPRO	TEFSAI	UVS
Simulator cladding temperature,	ºC	(900 - 1200)	(900 - 1300)	(1000 - 1200)
Simulator heating rate,	deg/s	var	10 - 20	1÷3 (from 800ºC)
Steam pressure,	MPa	0,1	0,1	0,1
Steam specific transfer,	mg/cm^2/s	7	50	2 g/min
Flow rate of carrier gas (argon),	cm^3/min	-	-	140 ± 6
Temperature of flooded water (immersion),	ºC	20	20	25÷35
Rate of flooding from bottom (immersion),	m/s s	0,2	0,2	0,5 - 0,8
Cooling rate,	deg/s	~ 100	~ 100	~ 100

Table 2

Type 1-4 simulators consisted of the following components:

Component		Facility		
		UNOPRO	TEFSAI	UVS
Cladding	Material	Zr1%Nb	Zr1%Nb	Zr1%Nb
	Length	60mm	120mm	60mm
Pellet	Material	Al_2O_3	UO_2	UO_2
	Shape	Standard – VVER type		
Plug	Material	Zr1%Nb	-	Zr1%Nb

The data were acquired on the heat resistance of VVER type fuel claddings operated to reach the burn-up of ~ 50 MW•day/kgU.

Simulators having unirradiated (types 1, 2, 3) and irradiated (type 4) claddings

The results of the thermal shock tests are presented as maps illustrating the ability of Zr1%Nb claddings to resist thermal shock – failure maps. The results of the thermal shock tests are plotted on the failure map as temperature of oxidation vs oxidation time.

The boundaries of the allowable ranges of cladding state are:

Maximally allowable temperature T_{clad}^{lim} = 1200°C	Vertical straight line
Maximally allowable degree of oxidation ECR_{clad}^{lim} = 18%	Inclined straight line

(Δm is calculated from ratio (2))

The embrittlement criterion "1200^{0}C – 18% ECR" for Zr1%Nb claddings of VVER type fuel rods was corroborated experimentally.

Estimation of mechanical properties of oxidized claddings subjected to thermal shock

The mechanical properties of oxidized claddings of cold water quenched fuel rod simulators were estimated from the results of impact testing (estimation of impact elasticity) and compression testing in the direction normal to the specimen symmetry axis (estimation of residual ductility).

Compression tests. To assess the mechanical properties of oxidized Zr1%Nb claddings subjected to a thermal shock, use was made of the results of compression testing 30 or 50 mm long pieces cut off from pre-oxidized VVER tubes (∅ 9.15 x 7.72 mm). The compression tests were carried out at room and elevated temperatures in the Instron-TT-DM machine and in a high temperature vacuum facility 1246P – 2/2300, respectively. The grip displacement rate is 2 mm/min. The strain temperature is (20-900)^{0}C.

The results of the compression tests revealed a clear-cut distinction between the specimens having a ductility margin (complete ductility, low ductility) and those that fractured in a brittle mode (fig. 2).

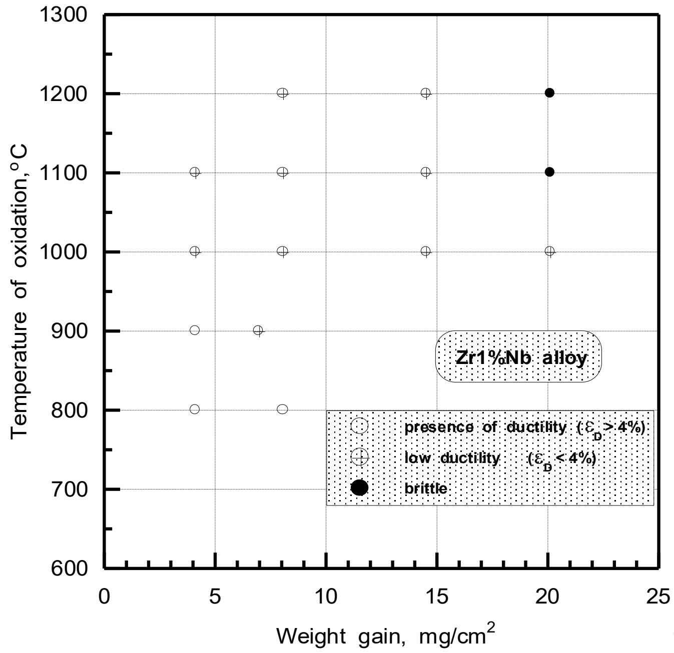

Fig. 2. Compression tests (T_{test} = 20^0C).

A strong dependence of the "ductility boundary" on a testing temperature and a specimen weight gain can be seen [259].

The results of the compression testing showed that the ductility characteristics of Zr1%Nb and Zry-4 alloys differed at a room temperature [260]. This difference reduces with the rise of the compression testing temperature.

Impact tests. In our view, the estimation of the oxidized Zr1%Nb cladding elasticity from the results of impact tests is most unbiassed.

In the experiments assessing the impact elasticity of cold water quenched oxidized claddings, original specimens were used; they were prepared from claddings of fuel rod type 2 simulators that remained integer after testing for thermal shock.

A notch 0.5 mm wide and (1.0 – 1.5) mm deep was cut in a simulator cladding 100 mm long using a diamond disc. The impact elasticity tests were carried out using a pendulum hammer of the PSV-1.5 type:

- Testing temperature 20 ^{o}C;
- Maximum impact energy ~ 15 J;

- Impact speed ~ 3-4 m/s.

The impact strength was calculated via the formula

$$a_k = W / F,$$

where W - is fracture strength, J;

F - is a specimen cross area at a point of shock load application, cm^2.

The impact elasticity of Zr1%Nb specimens in the original state equals 64.2-89.3 J/cm^2. This value is a factor of ~ 2 higher than that of Zry-4F specimens.

The tough fracture mode shown by Zr1%Nb claddings oxidized at (900-1300)^{0}C takes place upon oxidation to 5% ECR in impact tests (fig. 3).

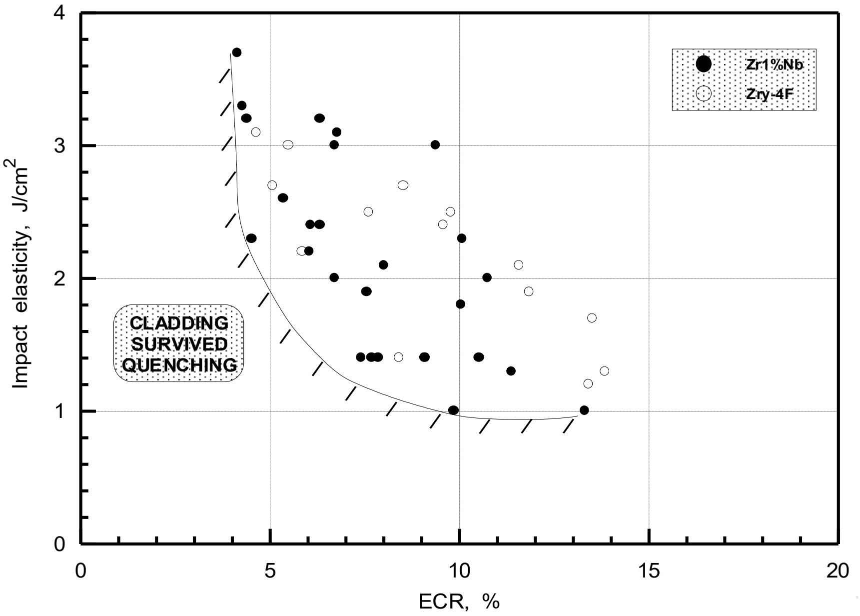

Fig. 3. Impact elasticity of oxidized claddings vs oxidation degree.

At a higher oxidation degree, claddings fracture in a mixed mode while at the oxidation degree more than 7% ECR, the fracture of claddings is brittle.

As far as Zry-4F claddings, the critical oxidation degree at the tough to brittle fracture transition is not less than 7% ECR. The same procedure was used to test the specimens of both the alloys.

According to, the critical impact elasticity of Zry-4 claddings was 1.25 J/cm^2. If this value is normalized to the cross section area of the transformed β-Zry it will be ~ 1 J/cm^2. Fig. 8 indicates the impact elasticity range within which Zr1%Nb cladding does not fracture when cold quenched (subjected to thermal shock). At the degree of Zr1%Nb cladding oxidation to 18% ECR, the impact strength remains not lower than 1 J/cm^2.

Metallographic examinations. Examinations of the cladding microstructure allowed determination of the thickness of Zr1%Nb – steam interaction layers (ZrO_2, α-Zr(O), β-Zr) formed as a result of the oxidation. The estimation of the degree of the cladding oxidation based on the results of the metallographic measurements of interaction layer thicknesses is an indirect one.

The knowledge of the oxygen distribution within the interaction layer thickness gives an idea of the influence produced by absorbed oxygen on the ductile (plastic) properties of claddings. The indirect estimation of the oxygen content and distribution within interaction layers was carried out based on the results of measuring the microhardness of layers.

The microhardness of claddings was measured with PMT-3 microhardness tester at the load of 50 g. The error of the measurements was ± 8 kg/mm^2.

Metallographic examinations revealed that the (metal + α-Zr(O)) layer thickness averaged hardness H_{50} of the claddings of type 1-4 simulators oxidized at temperatures < 1050^{0}C to 43 % ECR amounts up to 600 kg/mm^2 at the cladding thickness up to two times reduced compared to the original thickness (fig. 4).

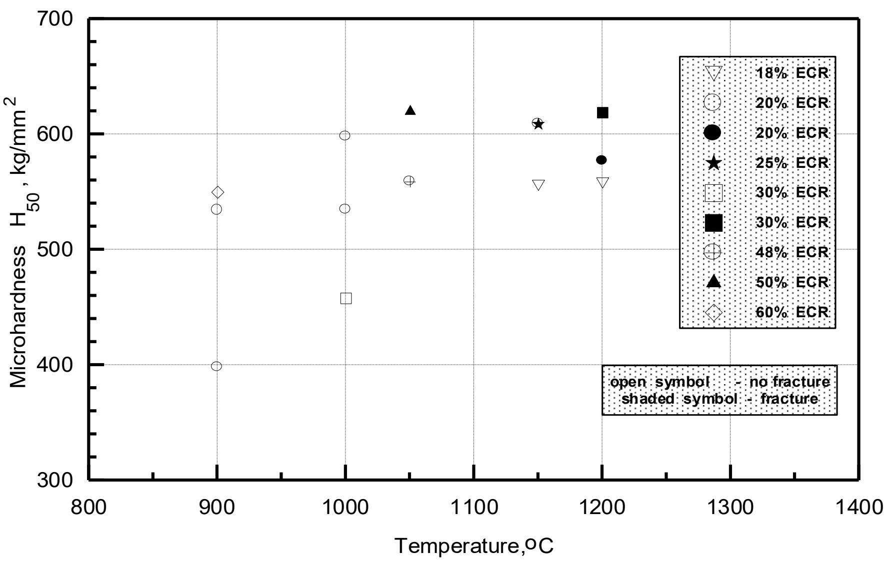

Fig. 4. Variation in (metal + α-Zr(O)) layer thickness averaged microhardness of Zr1%Nb claddings vs oxidation temperature.

The microhardness of the metal part of the Zr1%Nb claddings oxidized at < 1050^0C increases with the oxidation degree The microhardness of claddings oxidized at $(1100\text{-}1200)^0$C (fig. 4) is weakly dependent upon the oxidation degree.

Conclusions

The analysis of the data experimentally obtained by research the behaviour and properties of VVER type fuel claddings from Zr1%Nb alloy under loading conditions simulating the stage of core flooding with water in LOCA suffices to shows the character and numerical value of criterional parameters of the embrittlement criterion in terms of the cladding stability upon flooding and subsequent implementation of fuel assembly unloading and transportation.

The representative maximal design limit of fuel rod damage in terms of oxidation (embrittlement criterion) comprises all together the maximum temperature of the fuel cladding heating and the local depth of cladding oxidation.

The numerical values of the criterional parameters $T_{clad}^{\lim}$ =1200°C and $ECR_{clad}^{\lim}$ =18% are validated by the experimental data obtained from studies into the kinetics of the Zr-steam reaction and from the specially implemented thermal shock experiments.