

Development of SVECHA/QUENCH Code for Modeling Fuel Cladding Degradation in QUENCH tests

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ABSTRACT

In this paper we review the status of the computer SVECHA/QUENCH (S/Q) code development for modeling of new FZK single-rod quenching tests. The new code allows mechanistic description of the main physico-chemical phenomena of the fuel rod degradation under reflooding from high temperatures characteristic to severe accident conditions (1200–1600°C), and their profound mutual influence. The first results of the S/Q code application to the analysis of the bundle QUENCH tests at the FZK facility are also presented.

INTRODUCTION

The computer SVECHA/QUENCH (S/Q) code was developed in the Nuclear Safety Institute (IBRAE) of Russian Academy of Sciences for the detailed modeling of reflooding phenomena observed in the FZK (Germany) single-rod QUENCH rig tests, in close cooperation with German experimentalists [1,2]. Within the framework of the S/Q code the main physical phenomena occurring during quenching of fuel rods are considered:

- Cladding oxidation,
- Cladding mechanical deformation,
- Hydrogen uptake and release,
- Heat conduction inside the fuel rod,
- Heat and mass exchange in the surrounding two-phase media.

For the adequate description of the profound mutual influence of the above phenomena the self-consistent coupling of the Oxidation, Mechanical Deformation, Hydrogen Absorption, Heat Conduction and Thermal-Hydraulic models in the SVECHA/QUENCH code was performed.

CLADDING OXIDATION MODEL

Zirconium oxidation leads to the heat release and hydrogen generation and affects the mechanical properties of the cladding. The Oxidation model was developed for the description of the Zry-4 cladding oxidation kinetics [3] and further extended to the extreme conditions of high-temperature quenching characterized by rather high cooling rates (> 50 K/s) and steep radial temperature gradients across the cladding [1,2].

The model is based on the system of the partial derivative oxygen diffusion equations with non-equilibrium boundary conditions (corresponding to the high cooling rates) at the phase boundaries. The displacement velocities of boundaries between different phases of the cladding are determined by the oxygen and zirconium flux match conditions at the moving boundaries. Equilibrium oxygen boundary concentrations are determined as functions of temperature in accordance with the zirconium-oxygen binary phase diagram. Under conditions of a fast temperature transient a finite relaxation time is necessary to attain equilibrium values of the oxygen concentration near the interfaces between various phases. This relaxation process at the interfaces is simulated by the first order rate equations for the boundary concentrations, on the basis of experimental investigations [4].

Material properties necessary for the modeling are the oxygen diffusion coefficients and boundary concentrations in various phases. An advanced database for these parameters was developed on the basis of the statistical analysis of the most reliable and mutually compatible data obtained by different methods and/or by different experimental groups. On this base the Arrhenius correlation for the oxygen diffusion coefficients in various phases of oxidized Zry: β -Zr, α -Zr(O), ZrO₂(tetr.), ZrO₂(cub.) with corresponding confidence intervals, were calculated in the temperature range 1000–1825°C [5]. This database was thoroughly tested against various isothermal and transient oxidation experiments [6] and demonstrated a good agreement between experimental and calculated data [3], see Fig. 1.

In order to simplify the numerical solution of the multi-layer diffusion problem with moving boundaries, it is reduced by a coordinate transformation to the convective-diffusion problem in a fixed region. The fully implicit finite-difference time discretisation along with the space discretisation is used. Since the largest concentration gradients are expected near the layer boundaries, non-uniform space grids are used. The obtained tridiagonal system of difference equations is solved by the efficient Thomas' algorithm. As a result, the obtained numerical model is fast and compact and quite appropriate for implementation in various integral codes for the prediction of corrosion layer thickness and oxygen concentration profiles evolution.

The numerical calculations demonstrate that the consideration of the non-equilibrium boundary kinetics is most essential for the adequate simulation of the oxygen stabilized alpha phase growth kinetics (Fig. 2). This result is especially important for the modeling of the cladding mechanical properties: crack propagation through the cladding, ballooning and burst of the cladding, etc.

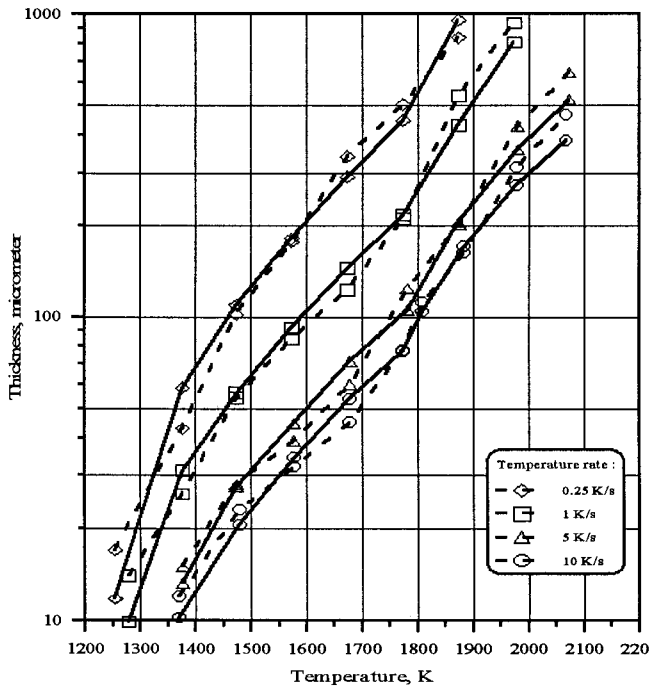


Fig. 1. Validation of the oxidation model against transient tests [6] with various heat-up rates: dependence of oxide scale thickness on maximum temperature.

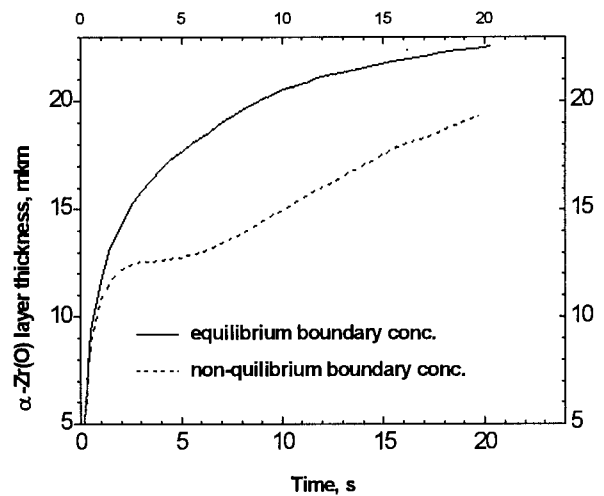


Fig. 2. α -Zr(O) layer growth kinetics during cooling from 1773 to 1273 K with 100 K/s.

HYDROGEN ABSORPTION MODEL

In order to simulate the hydrogen absorption by the Zr cladding observed in the FZK [7] and KFKI [8] separate-effect tests on the high temperature cladding interactions with pure hydrogen and pure steam atmospheres, respectively, two separate kinetic models were developed [1,2].

In order to simulate the hydrogen absorption by the Zry cladding observed in the FZK quench tests, a general model considering the cladding interactions with hydrogen/steam mixtures and comprising the two particular models as the limiting cases, was developed. The model was tightly coupled with the Oxidation model, and the two models were implemented in the SVECHA/QUENCH code as a combined module.

Hydrogen Uptake and Release by Zircaloy at High Temperatures

For quantitative description of the hydrogen uptake and release by Zircaloy, mass transfer in the gas and metal phases as well as at the oxide/metal interface, is considered. The hydrogen diffusion equation in the Zry sample is solved with non-linear boundary conditions determined by the Sieverts' law. In the gas phase a standard approach for modeling multi-

component transport through the diffusion boundary layer in the gas mixture is completed by consideration of additional net (Stefan) fluxes occurred owing to the hydrogen absorption.

The obtained system of equations is analyzed qualitatively and quantitatively. It is analytically shown that the hydrogen uptake and release can occur in different ways. Under conditions of the FZK separate-effect tests [7] with abrupt variation of the hydrogen gas pressure between zero and a finite value (0.5 bar) at various temperatures, the hydrogen release is described by a slow (hyperbolic) function of time, whereas the hydrogen uptake obeys a rapid (exponential) time law, in accordance with experimental observations. Under conditions of the hydrogen pressure variation between two finite values, the model predicts similar (exponential) kinetics in both cases of the hydrogen release and uptake. Later this conclusion was confirmed by the FZK observations.

The simplified qualitative consideration is completely confirmed by numerical calculations. Numerical solution of the problem furnishes a satisfactory fitting between measured kinetic curves [7] and calculations (Fig. 3).

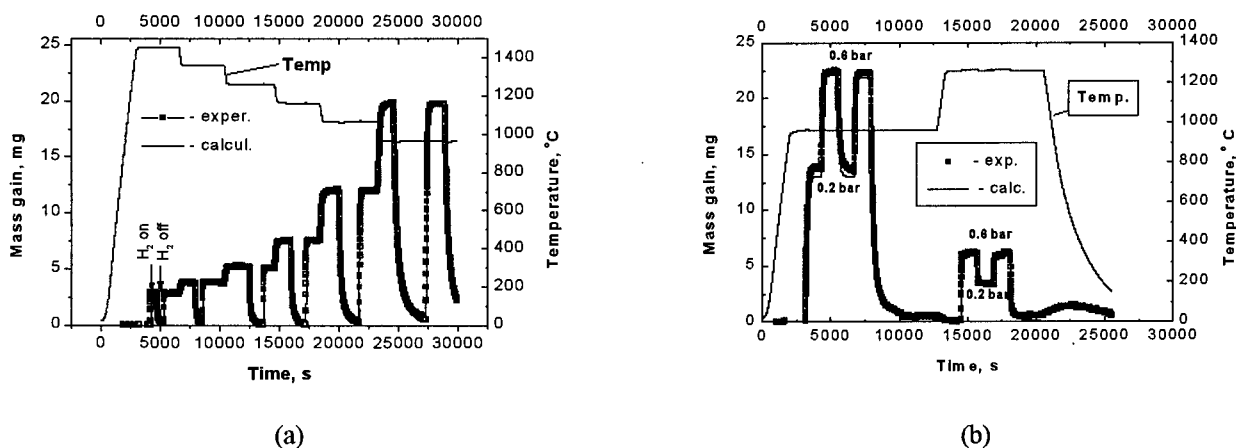


Fig. 3. Simulations of FZK separate-effect tests on H₂ uptake by Zry-4 tube segment in H₂/Ar gas mixtures. (a) post-test: P_{H2} = 0/0.5 bar; (b) pre-test: P_{H2} = 0.2/0.6 bar.

Hydrogen Absorption by Zirconium Alloys during High Temperature Oxidation in Steam

The model was developed for the description of hydrogen absorption by Zr alloys during oxidation in steam. It is essentially based on the experimental results of the KFKI (Hungary) tests [8] on the kinetics of hydrogen absorption by Zr1%Nb cladding during steam oxidation in the temperature range from 900 to 1200 °C.

The model takes into account that hydrogen may intrude into oxide as the result of H₂O dissolution process in the form of positively charged protons. Discharge of protons by electrons occurs at the oxide/metal interface (rather than at the gas/oxide interface as considered in the standard approach) after diffusion transporting of highly mobile protons through the oxide scale.

For quantitative description of hydrogen behavior by this mechanism, mass transfer in the three layers: gas, oxide and metal, and at corresponding interfaces: gas/oxide and oxide/metal, is considered.

The obtained system of equations is qualitatively analyzed explaining the main features of the observed complicated kinetics of hydrogen uptake: (i) continuous increase of hydrogen concentration in the metal phase being roughly proportional to the oxidation mass gain, in the initial stage of the process; (ii) desorption of hydrogen to the gas phase accompanied by slow decrease of measured hydrogen concentration in the metal phase, in the late stage of the process; (iii) decrease of maximum hydrogen concentration in Zr attained at the absorption/desorption regime transformation, with temperature increase.

Numerical solution of the problem generally confirms the main conclusions of the simplified analytical treatment and furnishes a satisfactory fitting between measured kinetic curves [8] and calculations. Comparison of the experimental data and numerical calculations are presented in Fig. 4

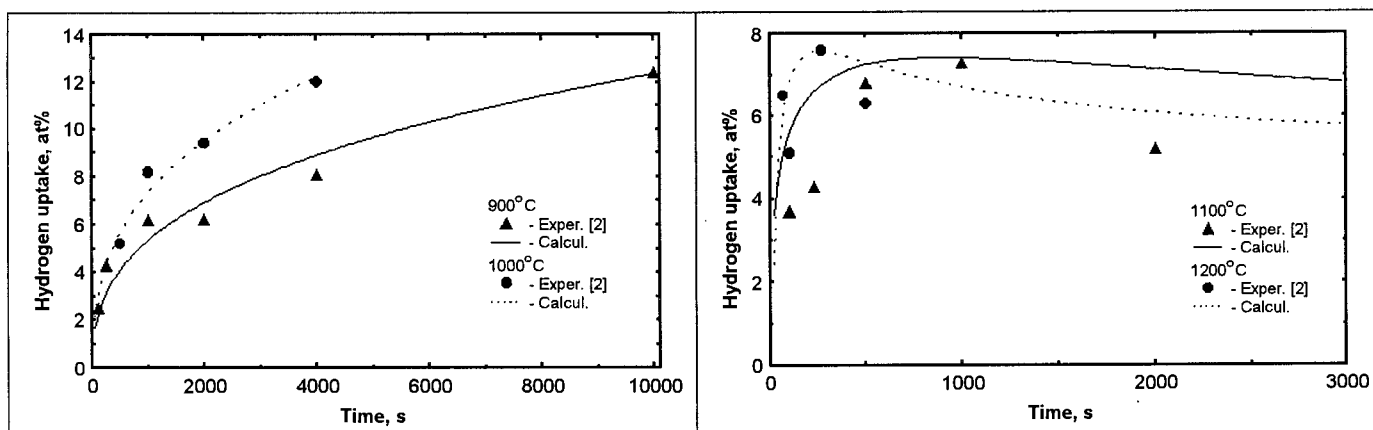


Fig. 4. Simulations of KFKI experiments on hydrogen uptake during $Zr/steam$ interactions at different temperatures.

Hydrogen Absorption by Zirconium Alloys during High Temperature Oxidation in Steam-Hydrogen mixtures

In the above presented models the two different cases: direct Zirconium reaction with molecular hydrogen (gaseous hydriding) and absorption of hydrogen liberated by the decomposition of steam during the oxidation process, were considered separately. However, in many real cases, especially under conditions of fuel element quenching by water, both these mechanisms should be considered simultaneously and self-consistently, since in this case Zircaloy cladding interacts with the steam/hydrogen mixture and the two mechanisms act competitively. For these reasons, a more detailed model considering the general situation and including both mechanisms as the limiting cases is developed.

Similarly to the previous case, the model considers the mass transfer in the three layers: gas, oxide and metal, and at corresponding interfaces: gas/oxide and oxide/metal. The model is qualitatively and numerically analyzed. It is shown that, at first, the dilution of steam by hydrogen leads to the increase of hydrogen absorption by the cladding. At some H_2/H_2O ratio (1:1) hydrogen absorption attains its maximum value and then decreases. For the steam content in the gas mixture lower than 1 mol.%, hydrogen absorption by the cladding becomes comparable with that in the pure steam atmosphere, and continues to decrease with the further decrease of the steam content in the mixture (Fig. 5a). However, at low steam contents in the gas mixture (≤ 0.1 mol.%) steam starvation is sustained during a short initial time interval of oxidation. During this period while the oxide scale is rather small ($\leq 1 \mu m$), the direct gaseous hydriding from the H_2 reach atmosphere occurs leading to a significant increase of the integral value of the absorbed hydrogen (Fig. 5b).

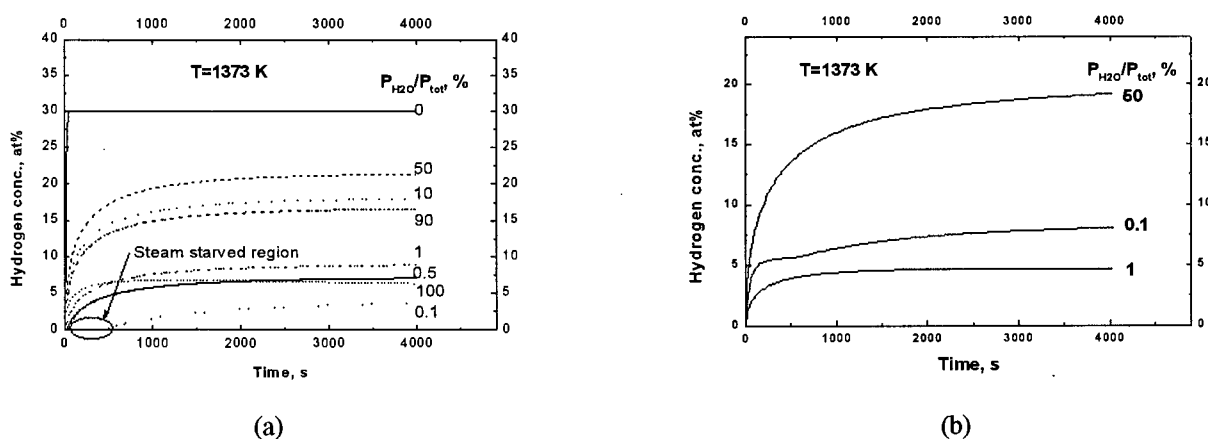


Fig. 5. Hydrogen absorption by $Zr1\%Nb$ cladding for various gas mixtures: (a) high $H_2:H_2O$ ratios, lacking steam starvation; (b) effect of steam starvation at low $H_2:H_2O$ ratios.

MECHANICAL DEFORMATION MODEL

The Mechanical Deformation model was initially developed to predict the deformation and rupture behavior of Zircaloy fuel cladding under severe accident conditions [3]. High temperature heating leads to an increase of the internal pressure of the fuel rods and, hence, to the ballooning of the fuel cladding. The clad ballooning influences heat generation during oxidation due to:

- reduction of the coolant channel cross-section;
- increase of the oxidized external cladding surface;
- internal oxidation of the cladding caused by steam penetration into the fuel rod gap;
- increase of the fresh metallic surfaces accessible for steam due to oxide cracking.

The deformation behavior of the oxide scale influences further accident progression. After the melting of the metal Zircaloy, the oxide shell prevents the molten Zircaloy from relocation, rapid oxidation and blockage formation until the oxide scale is broken due to the "flowering" mechanism [3].

After burst or local melting of the clad the external and internal pressures equalize. In this situation the oxide scale cracks due to:

- volumetric expansion of Zircaloy due to oxidation;
- different thermal expansion of α -phase, β -phase and oxide layers;
- non-uniform temperature distribution;
- fuel pellet thermal expansion;
- hydrostatic pressure induced by the molten materials.

In order to describe a complicated behavior of the cladding under quenching conditions observed in the FZK quenching tests, the model was further developed to account for:

- beta-to-alpha phase transformation of Zircaloy during quenching;
- tetragonal-to-monoclinic phase transformation of oxide during quenching;
- additional stresses generated by steep temperature gradients,

and their influence on the stress state of the oxidized cladding, crack formation and spalling [1,2].

Model description

The fuel rod is divided into meshes along the axial direction. Each mesh comprises the fuel pellet and Zircaloy cladding (Fig. 6). For each mesh the cladding is considered as a cylindrical shell consisting of three layers: external oxide layer, α - and β -Zircaloy layers. Each layer is considered as a thin-walled cylinder.

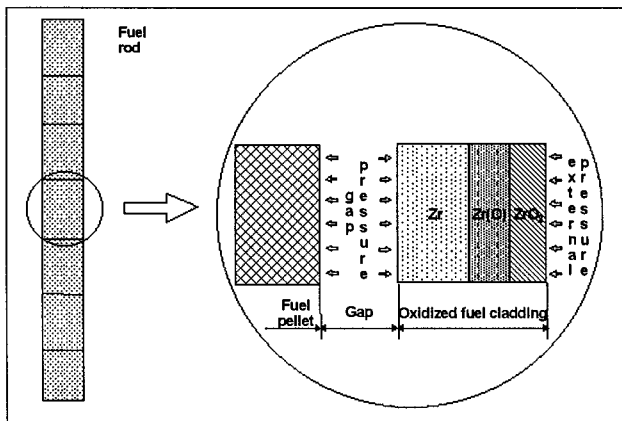


Fig. 6. Mesh structure of the Zircaloy fuel cladding.

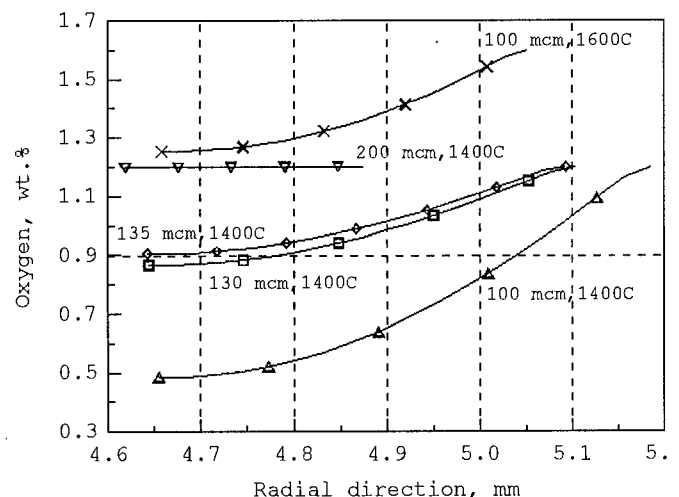


Fig. 7. Calculated oxygen radial distribution in the β -layer for different temperatures and extents of preoxidation.

The α and β phases are assumed to have different visco-elastic properties up to the melting temperature. Oxide is assumed to have elastic properties up to the melting temperature. The tensile and compressive strengths of oxide have different values. Mechanical properties and phase composition of the cladding layers depend on the oxygen content.

The dimensions and oxygen content of the fuel cladding layers change due to oxidation or dissolution. The oxidation kinetics is calculated by the Oxidation model of the S/Q code (Fig. 7). Temperature distribution inside the cladding is calculated by the Heat Conduction model.

The oxide layer of the oxidized fuel cladding can crack. Cracking leads to the reduction of the deformation strength of the oxide layer. Microcracks are assumed to be formed when the equivalent stress (which is the combination the first and the third principle stresses) attains the value of the tensile strength limit.

Simulation Results

The main simulation results of the fuel cladding deformation behavior under reflooding conditions are as follows:

A «large strain» criterion may be used to predict the time-to-rupture under burst. In this case the limit value of the circumferential strain is set to a certain large strain value (50 % in the present model). The S/Q code calculations and the experimental results show that the onset of ballooning occurs after a fraction of a second..

Oxidation can manifold increase the time-to-rupture or even prevent it. This is owing to the substantial decrease of the creep rate of the oxygen-stabilized α phase.

«Flowering» of the oxide scale is the result of the stress induced by the radial temperature gradient and the molten material hydrostatic pressure. Before the «flowering» occurs, the thickness of an uncracked oxide scale depends on the load history: temperature and pressure evolution, oxidation kinetics:

During quenching of the oxidized fuel cladding the tensile stress is generated in oxide due to the radial temperature gradient and the difference in the thermal strains of the α -phase, β -phase and oxide layers. This tensile stress leads to the onset of microcracks in the oxide scale at temperatures above the tetragonal-to-monoclinic phase transition (Fig. 8a).

At temperatures below the tetragonal-to-monoclinic phase transition (attained during quenching) the high level compressive stress occurs in the oxide layer and the high level tensile stress – in the alpha layer. Low oxygen content in the metal layer is the main reason that prevents the through wall crack formation. In this case the Chung-Kassner criterion can be applied: to withstand a thermal shock during quenching the thickness of the cladding with ≤ 0.9 wt. % oxygen should be greater than 0.1 mm (Fig. 7). If this layer is thinner and additionally the oxide scale with the residual deformation strength is thick enough to generate in the α -layer the stress exceeding the tensile limit, then the through wall cracks appear. Otherwise microcracks appear in the oxide scale (Fig. 8b). Oxide microcracking and creep strain in the metal layers lead to the relaxation of high level stresses.

The developed approach to the through wall crack density evaluation furnishes a satisfactory agreement with experimental data. This allows calculation of the hydrogen generation due to the through wall crack surface oxidation.

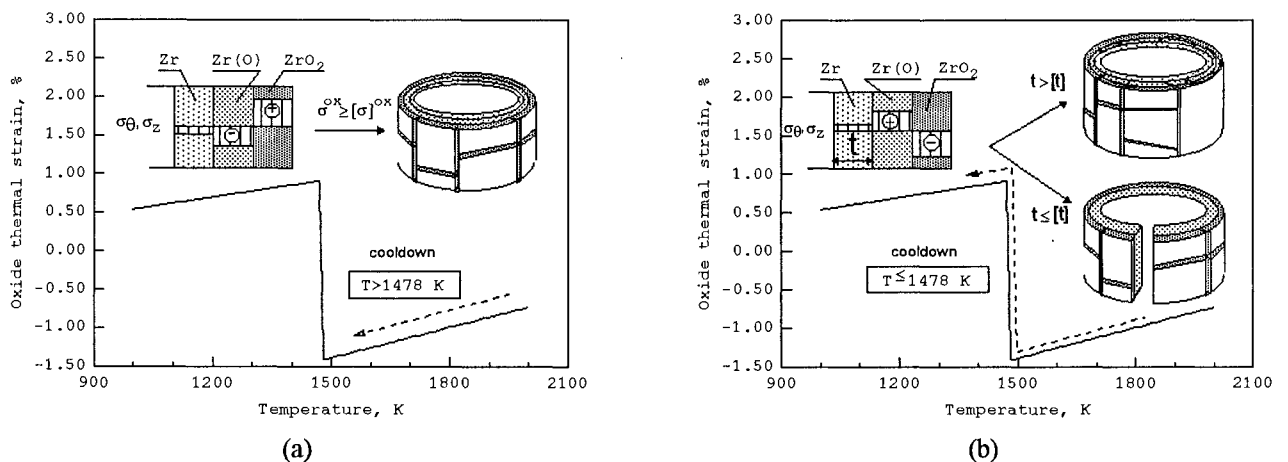


Fig. 8. Scheme of the stress state and failure modes of oxidized cladding on cool down: (a) above the oxide phase transition temperature; (b) below the oxide phase transition temperature.

HEAT CONDUCTION MODEL

The Heat Conduction model describes the heat conduction process in the multi-layered structure of an oxidizing fuel rod (pellets, gap between pellets and cladding, internal oxide layer, metal layer, external oxide layer of cladding) with account for different thermal properties of various layers [1]. The sequence of layers may be arbitrary; the number of layers is not limited.

The temperature distribution is assumed to be axially symmetrical, so the 2-D finite differences numerical scheme is used. The adaptive grid accounts for steep axial temperature gradients in the quench region. The application of the axial meshing refinement in the region of steep temperature gradients allows more accurate calculation of the temperature distribution in comparison with the uniform meshing. The adaptive grid also accounts for the axial variation of the different layer thickness due to thermal expansion and Zr oxidation.

The spatial distribution of the heat sources due to Zr oxidation is included in the model. The density of the heat sources is calculated by the Oxidation model. The boundary conditions for the heat conduction problem are provided by the Thermal-Hydraulic model.

THERMAL-HYDRAULIC MODEL

The Thermal-Hydraulic model of the S/Q code provides the description of the heat- and mass-exchange processes in the coolant channel [1,2]. The model considers the simultaneous existence of several coolant-gas regions: Nucleate Boiling (NB), Inverted Annular Film Boiling (IAFB), Churn Boiling (CB) and Gas Mixture (GM) with different properties at increasing elevations. The model calculates the heat transfer rates for the different regions and determines the non-stationary motion of inter-region boundaries.

The temperature evolution of the cladding surface temperature at the upper TC elevation (122 mm from the bottom) in the different FZK small-scale experiments with the initial temperatures 1200 -1600°C is shown in Fig. 9. The water inlet temperature was close to the saturation value (90°C). One can recognize four different portions of the temperature evolution curves with different characteristic heat exchange rates: I-II (GM region), II-III (CB region), III-IV (IAFB region) and IV-V (transition from film boiling to nucleate boiling).

The presented model considers the non-stationary motion of IAFB/CB and CB/GM boundaries, this allows an adequate analysis of the FZK non-stationary quench experiments and prediction of the rod temperature evolution. The model considers steam/hydrogen/argon mixtures and provides the boundary conditions for the Oxidation and Hydrogen Absorption models. The implementation of the gas mixture model makes possible a correct description of the oxidation kinetics in a wide range of steam concentration values, prediction of steam starvation and thus adequate calculation of the temperature evolution of the rod surface in the situation of a possible temperature escalation due to oxidation (Fig. 10).

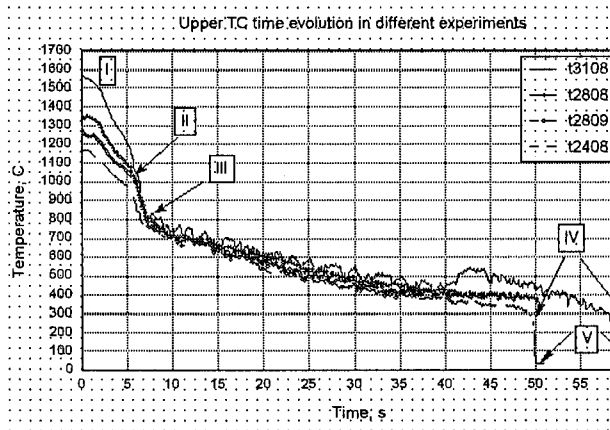


Fig. 9. Temperature evolution of the upper TC in FZK tests with different initial temperatures. I-II curves portion: GM region; II-III: CB region; III-IV: IAFB region; IV-V: transition from film boiling to nucleate boiling.

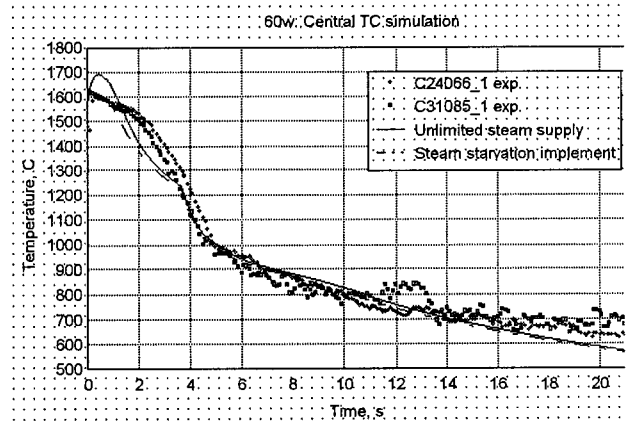


Fig. 10. Effect of steam starvation on temperature evolution of rod surface, central TC (1600°C initial temperature, 0 μm pre-oxidation). Solid line – unlimited steam supply; dashed line – with account for steam starvation.

CODE VALIDATION

The S/Q code has been verified against the FZK small-scale experimental data [1,2] (23 tests with water quenching and 6 tests with cooling by steam). The code demonstrates the satisfactory reproduction of the experimentally observed temperature evolution curves in the full range of the single rod experiments conditions (different pre-oxidation and initial temperatures) without tuning and adjusting of the code parameters.

Code Application to Bundle Tests

The 'effective channel' approach to the QUENCH bundle tests simulation by the S/Q code was developed. This approach assumes the usage of the experimentally measured temperatures of heated rods and shroud for the formulation of the central rod boundary conditions. On the basis of such conditions the S/Q code allows the correct solution of the temperature problem inside the rod and so, allows detailed description of the cladding mechanical deformation, oxidation and hydrogen absorption processes during reflooding (Figs. 11 and 12).

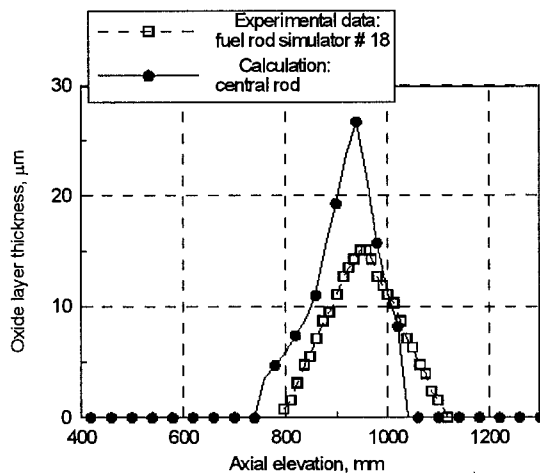


Fig. 11. Calculated and measured inner oxide layer thickness axial profiles after reflooding.

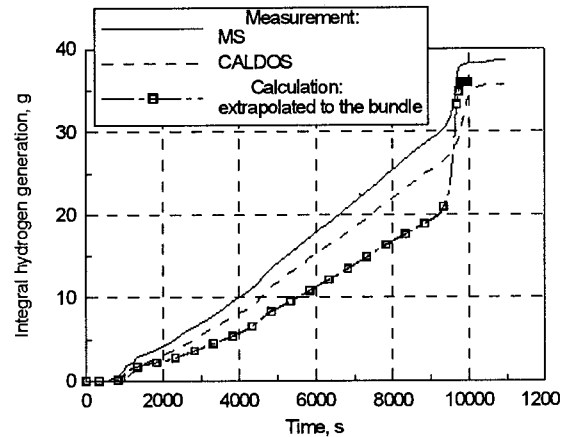


Fig. 12. Calculated and measured hydrogen generation extrapolated to the bundle.

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