FEATURES OF RAPTA-SFD CODE MODELLING OF CHEMICAL INTERACTIONS OF BASIC MATERIALS OF THE VVER ACTIVE ZONE IN ACCIDENT CONDITIONS WITH SEVERE FUEL DAMAGE

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Abstract

A brief description of RAPTA-SFD code intended for computer simulations of VVER-type fuel elements (simulator or absorber element) in conditions of accident with severe damage of fuel. Presented are models of chemical interactions of basic materials of the active zone, emphasized are special features of their application in carrying out of the CORA-W2 experiment within the framework of International Standard Problem ISP-36. Results obtained confirm expediency of phenomenological models application.

1. Brief Description of the Code

Model of damage of a single fuel element of water-cooled power reactor in conditions of accident with severe damage of fuel is important as an element of the general model of damage of the active zone of a reactor describing the first stage of the latter. The object of RAPTA-SFD calculational analysis is a single fuel element (simulator or absorber element) of a rod type.

RAPTA-SFD code is developed basing on RAPTA-5 version intended for calculation of fuel elements behaviour in design accident modes through including of the following models:

- boundary conditions (related to modelling of special experiments, for example CORA-W2);
- model of heat and mass transfer at melting;
- model of boron carbide absorber element with stainless steel cladding;
- models of high-temperature chemical interaction of basic materials of the VVER active zone.

The present version of RAPTA-SFD code is created in 1993 - 94 as a result of works under International Standard Problem ISP-36.

In the code the algorithm of numerical integration of the system of non- stationary equations of balance of heat of elementary volumes of multi-layered cylindric domain is implemented taking into consideration variations of its geometry at each time step. Geometric variations account for thermal elastic deformations of fuel and cladding, creep deformations of the cladding, building up of layers of chemical interactions of materials and displacement of melt. Axial discretization of a cylindric domain can be arbitrary. The problem is solved in axial-symmetric formulation. Within each axial region all parameters describing the state of simulation are assumed independent of the axial coordinate.

Phenomenological model of melt displacement uses conservation laws for mass and heat. Displacement of a melted volume is simulated through variation of lengths and temperatures of corresponding layers of a cylindric domain within the boundaries of adjacent axial regions.

Model of boundary conditions on the outer surface of a simulation takes into consideration convection heat exchange with the stream of single-phase coolant (steam, steam/argon mixture) and radiation heat exchange with surrounding surfaces. Simulation of a 19-rod assembly of CORA-W2 was carried out by means of iterative calculations of single representative simulator from periphery to center and vice versa. With this, calculations for outer layer simulator was carried out simultaneously with calculations of multi-layered heat insulation shroud of the assembly.

Results of RAPTA-SFD calculations are arranged in compliance with requirements of ISP-36 [1] and they include large information concerning energy balance, temperature mode, local and integral characteristics of chemical interactions and displacements of melted volumes.

Usage in this code version of simple phenomenological models for description of interrelated chemical and physical processes enable one to rapidly assess the condition of the fuel. These models imply expert setting of a number of parameters values of which can be determined from the results of experimental modelling. The above applies to a considerable extent to simulation of high-temperature chemical interactions of basic materials of VVER active zone in conditions of accident with severe damage of fuel.

2. Models of High-Temperature Chemical Interactions

Phenomenological models of chemical interactions use parabolic dependence of the measure of reaction W on time t at isothermal conditions:

$$W^2 = K_p\, t, \quad \text{where } K_p = A \exp (-B / T),$$

A, B being empirical coefficients, T - temperature, K.

Kinetics of the reaction in non-isothermal conditions is determined from a recurrent relation taking into account previous reaction history.

In RAPTA-SFD code, models of the following chemical reactions are implemented:

- oxidation of Zr1%Nb by steam (actual dependence);
- interaction of Zr1%Nb with UO_2;
- oxidation of stainless steel X18H10T by steam;
- interaction of stainless steel with B_4C;
- interaction of Zr1%Nb with stainless steel;
- dissolution of UO_2 by melted zirconium.

Empiric coefficients of these models have been determined from the results of laboratory experimental investigation of chemical interaction in temperature range specific for severe accidents [2 - 4] (Table 1).

The most reliable measure of steam/zirconium reaction determined in experiments on oxidation of fuel cladding samples is weight increase of oxygen per unit surface area. The absorbed oxygen forms a layer of stoichiometry zirconium dioxide and a layer of a-Zr stabilized by oxygen depths of which can also be used as a measure of reaction. In the result of the reaction hydrogen is liberated, the amount of this hydrogen is determined on the basis of reaction equation. Rate of hydrogen liberation by a structure being oxidized is determined via integration over the structure active surface (assembly of fuel elements and shroud). Heat production of steam/zirconium reaction is estimated as ca 6.5 MJ/kgZr which causes strong influence of steam/zirconium reaction to temperature mode, especially at temperatures above 1500 ^{o}C when acceleration of reaction occurs due to alteration of structure of oxide film.

Eutectic interaction of uranium dioxide with Zr1%Nb alloy (as well as with zircaloy) results in creation of three specific layers (Table 1) in which intermetallic inclusions (U, Zr) can exist in liquid state at relatively not high temperatures. However, within the period from the onset of this reaction to achieving of cladding melting point (characteristic for CORA-W2 experiment), layers of considerable thickness do not manage to build up.

Rate of reaction of stainless steel oxidation by steam is comparable to the rate of steam/zirconium reaction. Reaction can also lead to hydrogen release, the measure of reaction used being the specific weight increase of oxygen. Analysis of values of specific heats of oxidation enables one to conclude that the heat effect of this reaction is an order of magnitude less than that for steam/zirconium one.

Interaction of stainless steel with boron carbide leads to propagation of interaction layer mainly into the depth of the cladding. This leads to emerging of fusible eutectics. Experiments on samples have shown that melting occurs at the temperature of 1200 ^{o}C (melting point of chromium/nickel steels of austenite class constitutes 1400 to 1425 ^{o}C) . The melt of control rod is occured significantly earler than fuel element. The remained boron carbide, evidently, partially flakes off and partially is dissolved in melt. Melt of the control rod coming to adjacent fuel elements initiates the reaction of

TABLE 1. SUMMARY OF MODELS OF CHEMICAL INTERACTIONS OF MATERIALS OF VVER ACTIVE ZONE IMPLEMENTED IN RAPTA-SFD CODE

Interaction type	Measure of reaction, dimension	Temperature range, $\hat{E}^*$	Coefficients		Ref.
			À	Â	
Zr1%Nb + H_2O	Weight gain, mg/ñm^2	1273 - 1773 1773 - 1873	1.59 10^5 9.825 10^5	23040 20800	/1/
	ZrO_2 thickness, ñm	1273 - 1773 1773 - 1873	0.00519 0.01772	15355 14680	/1/
	Thickness of ZrO_2 + a-Zr(O), ñm	1273 - 1773 1773 - 1873	0.0144 0.516	14088 19520	/1/
Zr1%Nb + UO_2	Layer-1 tickness, ñm [a-$Zr(O)_a$+(U,Zr)]	1273 - 1873	5.42	30191	/1/
	Layer-2 tickness, ñm [layer-1 + (U,Zr)]	1273 - 1873	62.68	32247	
	Layer-3 tickness, ñm [layer-2 +a-$Zr(O)_b$]	1273 - 1873	1.8548	22568	
X18H10T + H_2O	Weight gain, mg/dm^2	1073 - 1373	3.607 10^{13}	35210	/1/
X18H10T + B_4C	Layer tickness in cladding, ñm	1073 - 1473	3180	34600	/1/
Zr1%Nb + X18H10T	Layer tickness in cladding, ñì	1173 - 1583	3.717 10^{13}	66140	/2/
Dissolution of UO_2 in Zr melt	wt% UO_2 - 35.8%	2223 - 2523	1.0196 10^{15}	81450	/3/

* Temperature ranges for which experimental data are available

zirconium dissolution which is simulated in RAPTA-SFD code using kinetics of interaction of Zr1%Nb with stainless steel in their solid state.

Zr1%Nb alloy interaction with stainless steel (spacer grids) results in creation of interaction layers. Eutectics (Zr-Fe, Zr-Ni) have low melting point (less than 1000 ^oC) and can be sites of origin for structure melting. This reaction is counteracted by existing oxide films on original materials.

Reaction of uranium dioxide dissolution in zirconium melt is the most essential for determining of the amount and the composition of energy-releasing melt (corium). UO_2 solubility in Zr melt is very high, and the weight concentration of the former essentially instantly reaches 35.8%. For integral features of interaction a major role is played by time and scale factors.

3. Some Peculiarities and Results of Modelling

In Figs. 1 - 7 some results of RAPTA-SFD code simulation of CORA-W2 experiment are shown. In each figure 3 curves are presented: 1 - the plot obtained in the result of direct measurements in the course of the experiment with possible subsequent processing or in the result of post-test investigation of the assembly [5]; 2 - calculational curve obtained as the result of "blind" calculation within the framework of ISP-36; 3 - curve obtained as the result of final corrected calculations.

As it was mentioned above, RAPTA-SFD code uses several parameters set forth from expert reasoning based on experimental information. Thus, debugging of the code for calculations under ISP-36 was carried out using the known results of CORA-W1 experiment. In choice of parameter values the following factors were taken into consideration:

- uranium melting point was not reached;
- axial discretization in calculational model should not lead to considerable steps of functions describing processes of hydrogen generation and melt flow down.

Let us focus on choice of calculational parameters of RAPTA-SFD code and on their influence on simulation results.

Mass transfer model uses a parameter essentially influencing the temperature mode and the kinetics of chemical interactions. This is the rate of leaving by a melted ring volume from the boundaries of the axial region which is not to be identified with melt flow rate. Change in volumes of material in melting region and lower adjacent regions is accounted for by changing of layer thicknesses of uniform material. Temperature of the layer similar to incoming melt is adjusted through averaging over mass. With this, the melt "solidifies" if this temperature happens to be less than the preset temperature of motion begining onset. This procedure is applied sequentially for each integration step and for each axial region from top to bottom.

In conduction of "blind" simulation under ISP-36 the rate of melt flow away was assumed 1 mm/s (which proved to be an underestimate), and the relocated melt was joined to a single adjacent region. This led to melt "delay", to considerable overestimation of temperature level after fuel element claddings melting and to an error in determination of lower level of solidified melt. In the corrected calculation the rate was set 3 mm/s and relocated melt being distributed among 2 lower adjacent regions. This permitted us to obtain more satisfactory calculated results in simulation of temperature mode (Fig. 1, 2) and of melt relocation which could be assessed, for example, by height distribution of assembly cros- section blockage (fig. 3) or by share of dissolved uranium dioxide (fig. 4). In both calculation versions higher values of blocking compared to those measured in post-test investigations have been obtained. The following reasons for computer overestimation of blockage: interaction between boron carbide and steam, drop of material parts to outside of bundle active zone, increasing of shroud restricted area due to deformation in the experiment were not take into account. However, form of axial distribution as a result of corrected calculation has fair agreement with experiment. It should be also emphasized that this rate parameter essentially influences kinetics of chemical interactions through time and temperature factors.

Dependencies used for calculation of kinetics of chemical interactions permit to determine their local features. In obtaining of integral features of some materials interactions in conditions of fuel elements assembly one should bear in mind uncertainity in actual values of reagents contact area. This applies first of all to the process of uranium dioxide dissolution by zirconium melt. In the "blind" calculation reagents contact area was assumed to constitute 10 per cent of area of the fuel rod outer surface. This value in corrected calculation being set to 15 per cent. In both cases a realistic estimate of maximum local share of dissolved uranium dioxide was obtained, in the second case the height distribution of this value being closer to measured one (fig. 4).

One of the most important aspects of fuel behaviour in condition of severe accident influencing temperature mode and degradation processes is steam/zirconium reaction. Extrapolation of oxidation law beyond the limits of temperature range of experimental data can lead to the need to account for

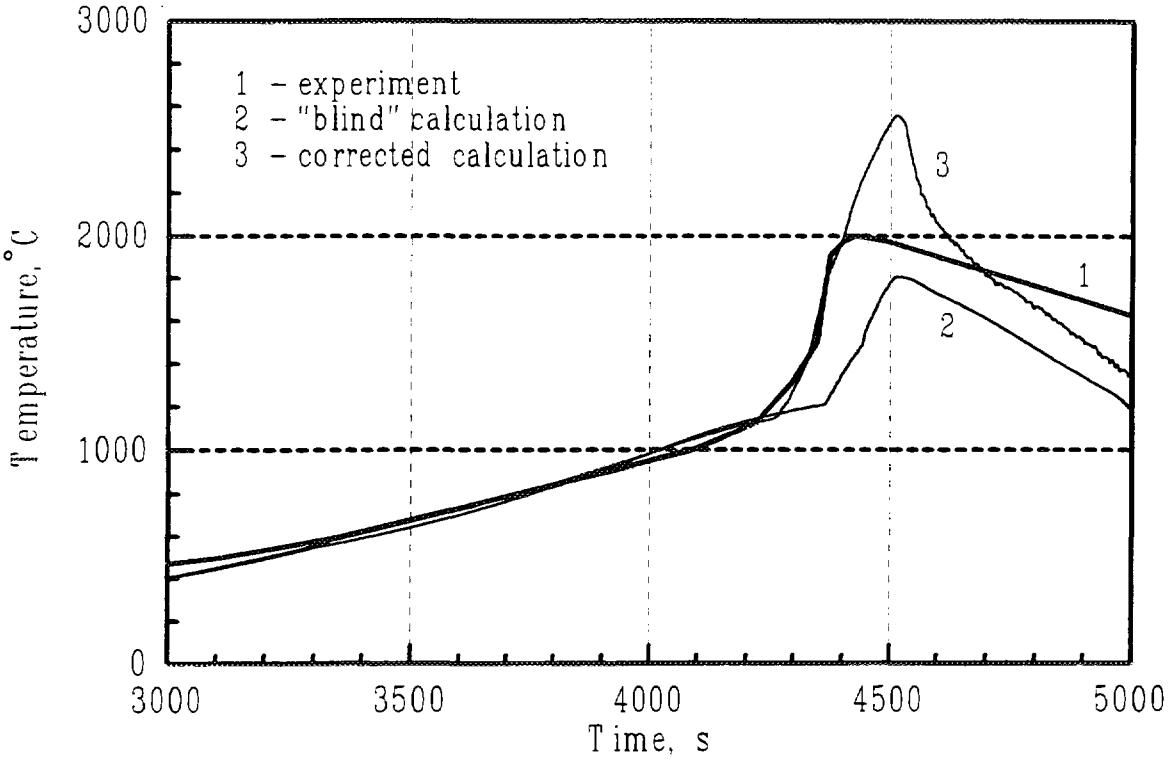

Fig. 1. CORA-W2 Experiment. Temperature mode of fuel elements simulation. Height coordinate 550 mm.

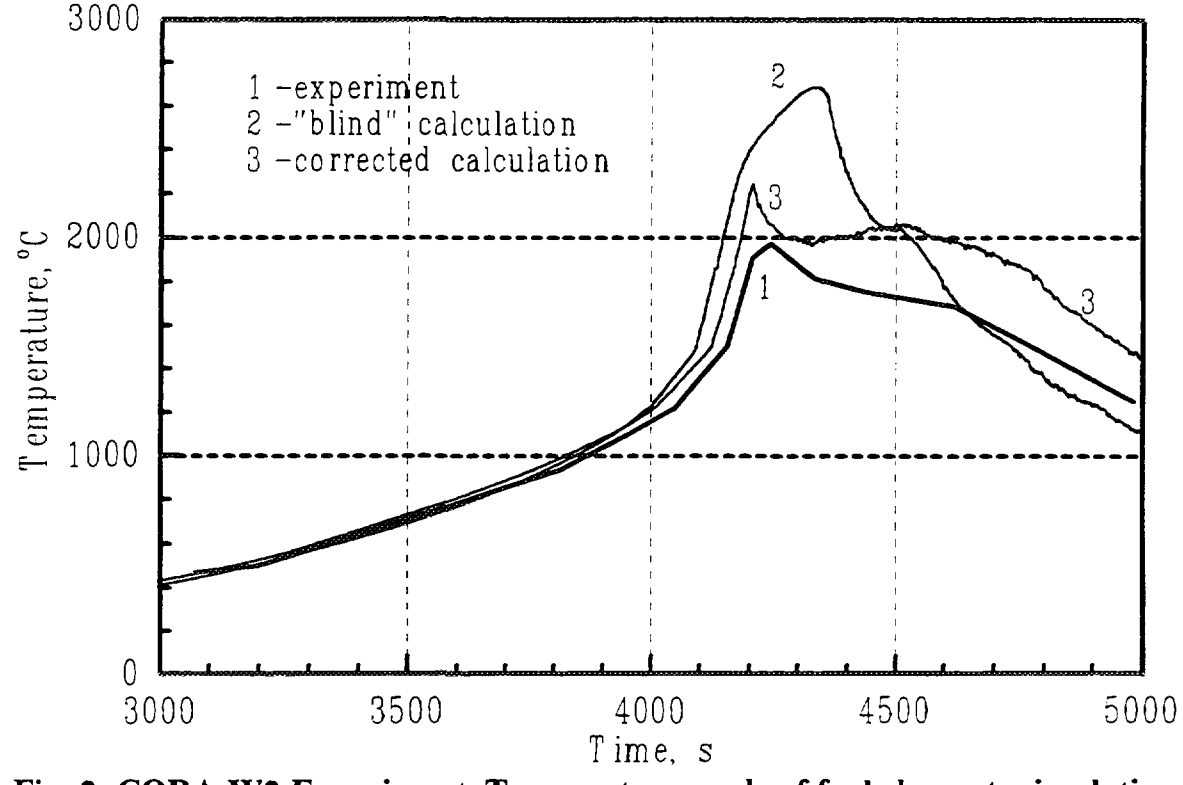

Fig. 2. CORA-W2 Experiment. Temperature mode of fuel elements simulation. Height coordinate 950 mm.

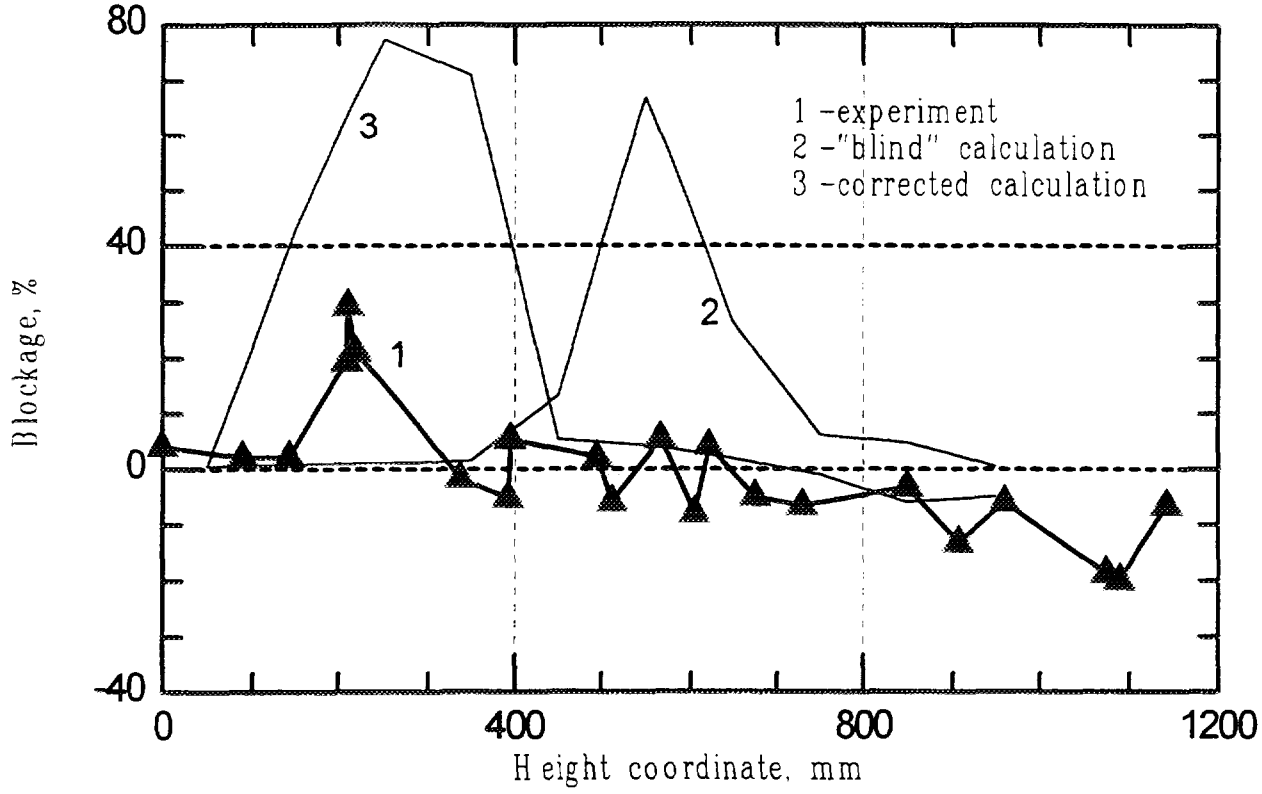

Fig. 3. CORA-W2 Experiment. Blockage distribution of cross section over height of the bundle.

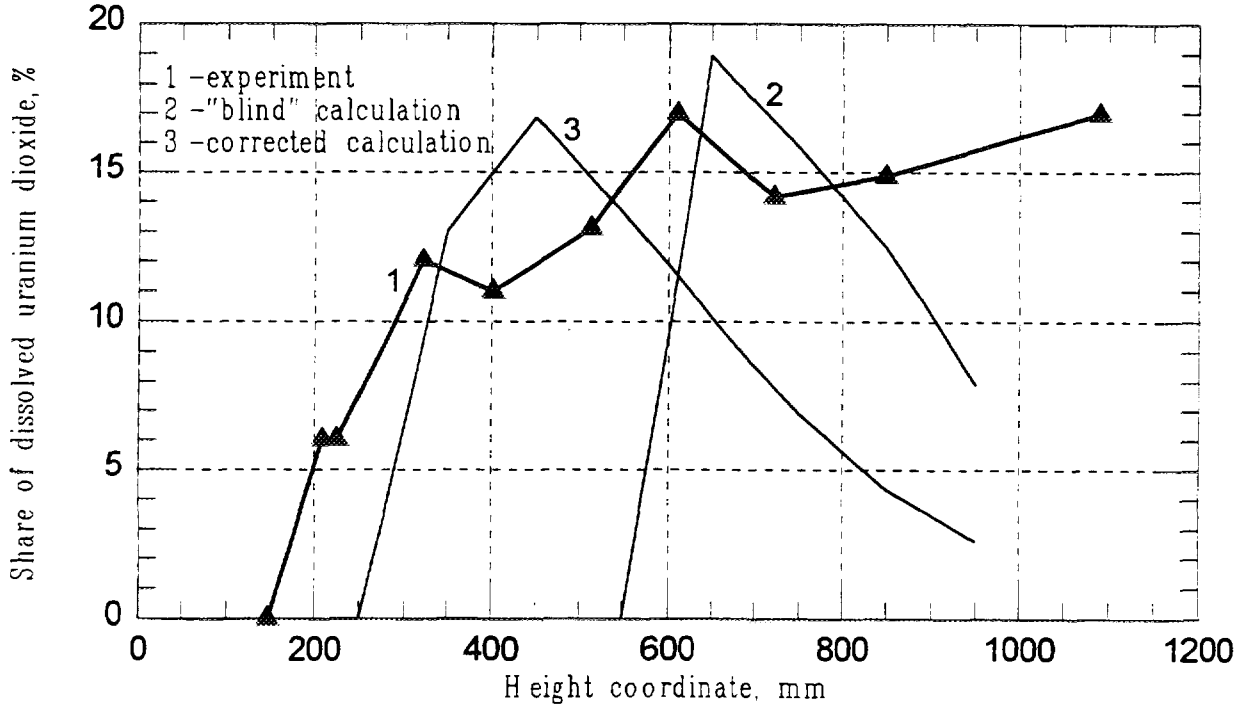

Fig. 4. CORA-W2 Experiment. Distribution of the share of uranium dioxide dissolved by zirconium melt over height of the bundle.

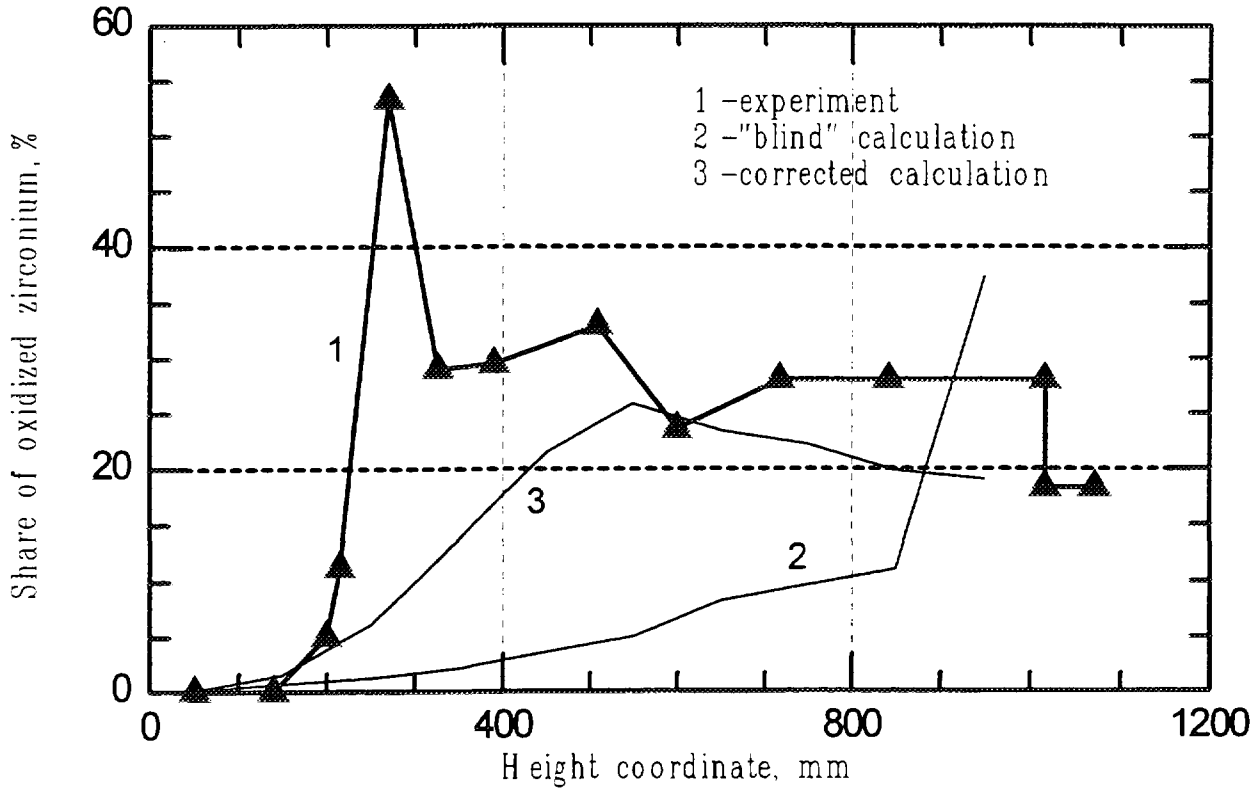

Fig. 5. CORA-W2 Experiment. Distribution of the share of oxidized zirconium in original position over height of the bundle

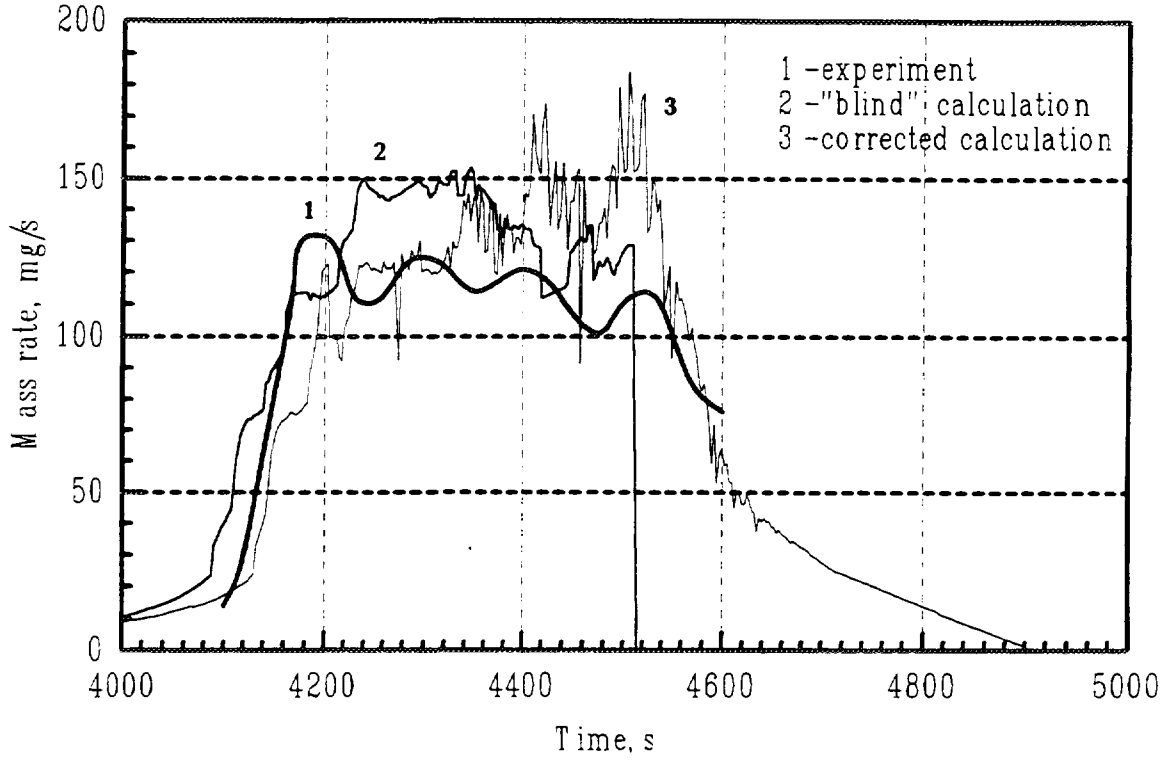

Fig. 6. CORA-W2 Experiment. Rate of hydrogen production (Bundle + Shroud).

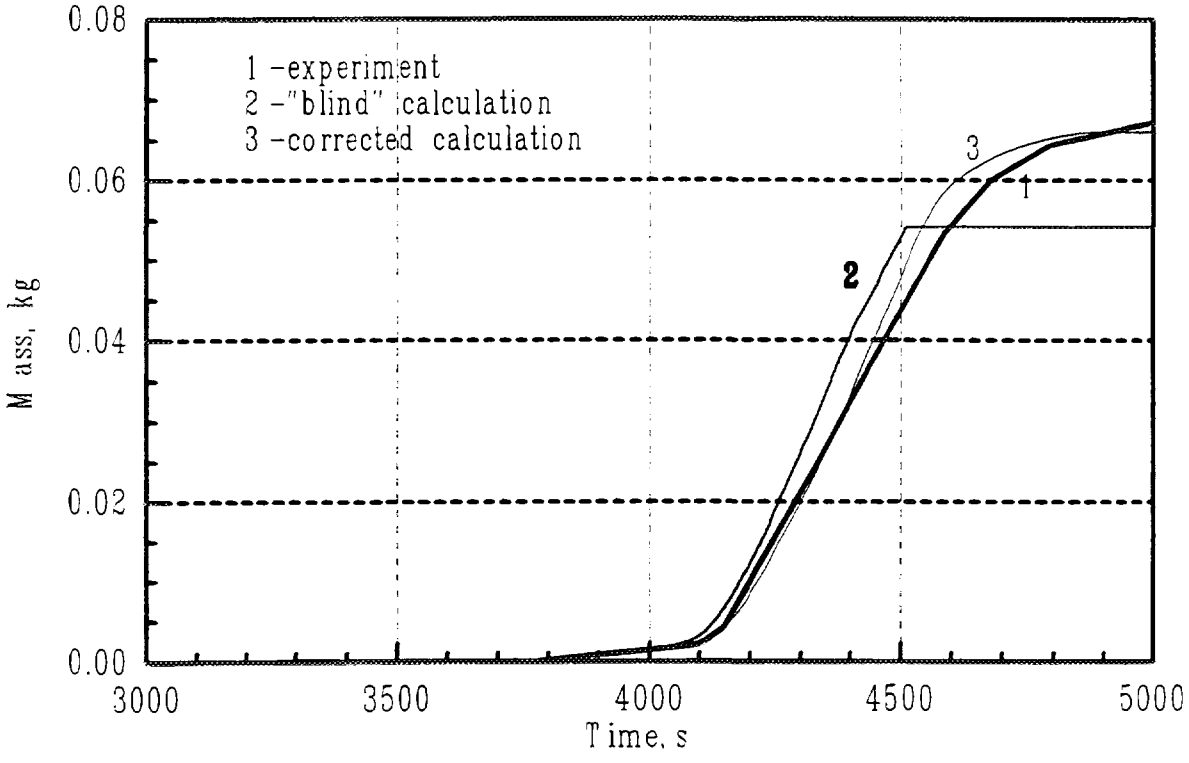

Fig. 7. CORA-W2 Experiment. Total hydrogen yield (Bundle + Shroud).

"steam hunger". In conduction of "blind" and corrected calculation the rate of specific weight gain oxygen was limited by the value of steam flow rate through the assembly divided by total zirconium surface area of the assembly. In corrected version the residual steam flow after valve shutting was taken into account. In the "blind" calculation it was assumed that after melt relocation onto an axial region oxidation of original zirconium in this region stops and oxidation of relocated melt continuing. In the corrected calculation oxygen weight gain in the axial region containing relocated melt was divided to original and relocated zirconium proportionally to their masses. This permitted to obtain better result in assessment of height distribution of assembly oxidized zirconium prior to its melting (fig. 5). In both calculations fair agreement of hydrogen generation rate and of its total yield with experiment values has been obtained (fig. 6,7).

Conclusion

Henceforth, simulation of CORA-W2 experiment by RAPTA-SFD code has shown that usage of relatively simple phenomenological models could give good results. Application of phenomenological models of chemical interactions and mass transfer is related to the necessity of assuming of several expert parameters. Reliable values of these parameters could be determined on the basis of integral experiments, such as CORA-W1, CORA-W2. The similar possibilities are possessed by the experimental unit PARAMETER (Research Scientific Institute of Scientific and Production Amalgamation "Looch", Moscow, RF) on which two similar experiments with 7-rod assemblies have been carried out in 1993 - 94. It seems expedient to continue such experimental investigations of fuel behaviour expanding the range of loading conditions (rate of heating, inner fuel element pressure, heat transport medium parameters). This would strengthen the base of experimental data for assessments of possibilities to control severe accident as well as for development and verification of calculational codes intended for fuel licensing.

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