## ISTC Project No # 3592

## Investigation of Corium Melt Interaction with NPP Reactor Vessel Steel (METCOR-P)

## Annual Project Technical Report

## on the work performed from 01.04.2008 to 30.03.2009

(Second year)

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# **1.** Brief description of the work plan: objective, expected results, technical approach

Main project objective is to enhance the safety of NPP reactors at severe accidents resulting in the core meltdown. Concrete subject is an experimental and theoretical study of physicochemical phenomena taking place at the interaction between a molten pool of prototypic corium and reactor vessel steel. The study is aimed at determining steel corrosion at the in-vessel melt retention (IVR) and external cooling of reactor vessel with boiling water.

The studies of molten corium interaction with vessel steel are carried out in the following conditions:

- Vertical position of interacting surface of the vessel steel specimen to determine the influence of orientation on the corrosion process by comparing with the data provided for the horizontally-oriented interaction surface, other conditions being the same.
- Transient oxygen potential in the system during the interaction achieved by replacing neutral atmosphere (argon) by oxidizing (steam, air), which in reactor conditions can lead to the melt temperature growth, inversion of the metallic and oxidic layers in the melt, increase of heat flux into the vessel, also due to the focusing effect, increase in the aerosol and fission product release.
- Interaction of molten corium with vessel steel of European reactors and comparison with data on the Russian vessel steel performance in similar conditions. Expected results of the project are:
- Experimental data on the vessel steel corrosion kinetics and depth at its interaction with molten coria having different compositions.
- Mechanisms of physicochemical phenomena influencing the corrosion.

- Experimental data to determine the qualitative influence of vertical orientation of interacting steel surface on the parameters of interaction between molten corium and vessel steel and on the kinetics of this interaction.
- Experimental data on the steel corrosion and history of the molten pool kinetics in the non-stationary conditions of the corium melt oxidation.
- Qualitative and quantitative characteristics of European vessel steel corrosion in comparison with Russian vessel steel.
- Correlations for calculating corrosion kinetics in the studied parameter range.
   The provided data can be used for:
- modeling of corrosion physicochemistry by numerical codes for severe accidents;
- numerical code verifications;
- insight into the temperature and stress-and-strain conditions of reactor vessel at severe accidents;
- development and justification of different concepts for the molten pool retention at a severe accident with core meltdown and, primarily, for the in-vessel melt retention (IVR).

Experimental studies within the projects are carried out on the RASPLAV-2 and RASPLAV-3 rest facilities, which have shown efficient operation during 16 and 6 years respectively. The melt is produced by the method of induction melting in a cold crucible (IMCC). It can produce melts with temperatures up to 3300 K. The method of IMCC provides a contact-free power deposition in the melt and melt layer heating to a certain depth.

RASPLAV-2 facility can be used for experiments with oxidized and suboxidized systems, and RASPLAV-3 – for metal-oxidic systems. The RASPLAV-2 and 3 facilities can produce up to 8 or up to 2 kg of high-temperature melt respectively in the inert, air and steam atmospheres.

Experimental studies for investigating interaction mechanisms use the following methods:

- Visual polythermal analysis in a cold crucible (VPA IMCC) and in the Galakhov microfurnace (VPA MG).
- Differential thermal analysis (DTA).
- High-temperature microscopy (HTM).

The following methods are used for the physicochemical analysis of corium samples:

- Analysis of elemental composition (X-ray fluorescence [XRF], chemical analysis [ChA], mass-spectrometry with inductively-coupled plasma [ICPMS], spark massspectrometry [SSMS]).
- Analysis of the phase composition (X-ray diffractometry [XRD], energy dispersion X-ray [EDX]).
- Metallo- and ceramography (optical microscopy [Opt M], scanning electron microscopy [SEM]).

### 2. Technical progress during the first year

In accordance with the Work plan the METCOR-P experimental matrix foresees the implementation of 5 tasks. Their main goal is the availability of experimental and analytical data on the vessel steel corrosion kinetics at its interaction with molten corium depending on:

- orientation of interacting surface,

- non-stationary processes of melt oxidation at the replacement of inert atmosphere by oxidizing (air or steam),

- brand of steel used for European and Russian reactor vessels.

The contents of tasks are defined below:

Task 1	-	Adjustment and testing of the test facility and measurement system. Development and testing of experimental methodologies and postexperimental physicochemical and metallographic analyses.	
		Specification of experiments.	
Task 2	-	Interaction of suboxidized molten corium with vertically positioned vessel steel specimen. Interaction of oxidized melt C-100 and $UO_{2+x}$ -ZrO <sub>2</sub> -FeO <sub>y</sub> with a horizontal surface of vessel steel in the oxidizing atmosphere (air, steam).	
Task 3	-	Non-stationary processes (change of the pool structure, heat effects, gas and aerosol release) taking place during oxidation of suboxidized oxidic and metal-oxidic melt – achieved by replacing inert atmosphere (argon) with oxidizing (steam, air).	
Task 4	-	Interaction of suboxidized and oxidized melts with a European vessel steel specimen in the neutral (argon) and oxidizing (air) above-melt atmosphere.	
Task 5	-	Integrated physicochemical, thermodynamic and thermophysical analyses of completed experiments, experimental series and the whole complex of experimental studies. Preparation of deliverables. Development of numerical models and correlations. Preparation of papers and presentations at conferences.	

Each task has been divided into stages including experiments, primary analysis of experimental data, pre- and posttest calculations, physicochemical analysis and integrated analysis of produced results.

#### The scope of work completed during the 1<sup>st</sup> year.

#### Task 1

• 3 new designs of RASPLAV-3 induction furnaces were developed for experimental investigations of interaction between vertically positioned steel surface and suboxidized corium (Fig. 1), each of the designs has advantages and disadvantages.

Advantages of 1.a schematics are a convenient location of the specimen and a possibility to use the ultrasonic sensor (USS). But in this case the volume of pool is too large, a crust can be present in the pyrometer sighting spot and furnace air-tightness is difficult to achieve in experiments with controlled compositions.



- a)
- 1 crucible sections;
- 2 water-cooled pyrometer shaft;
- 3 inductor;
- 4 vessel steel specimen;
- 5 specimen calorimeter;
- 6 ultrasonic sensor
- 7 -K-type thermocouples;
- $8 \pm melt;$

b)



1 – water-cooled lid; 2 – – water-cooled pyrometer shaft; 3 – specimen calorimeter; 4 – water-cooled shaft of ultrasonic sensor; 5 – ultrasonic sensor of distance; 6 – quartz tube; 7 – crucible sections; 8 - melt; 9 – inductor; 10 – calorimeter; 11 – vessel steel specimen; 12 –K-type thermocouples; 13 – ultrasonic sensor; 14 – protective casing.

#### Fig. 1. Design schematics of modified RASPLAV-3 test facility.

Design option 1.6 has an extensive interaction surface and a possibility to use the USS. Drawbacks of this scheme are low reliability and efficiency of inductor, too shallow molten pool and complicated provisions for inductor air-tightness; also problematic is its electric insulation in the place where the inductor is in contact with the specimen. Option 1.c uses the proven crucible design, but it has limitations for installing a USS into the specimen. Along with that the modeled temperature profile in the specimen

turned out to be non-symmetrical.

- The decision-making about the experimental setup was based on the numerical analysis of modifications proposed for the required generator power, dimensions of cold crucible and inductor, technological properties of the overall design, specimen cooling system, molten pool volume and the minimum inventory of measurements (without USS). The chosen test facility modification had the traditional configuration of the vertical crucible and vertical, axially positioned cylindrical vessel steel specimen having an internal cooling channel (Fig. 2).
- For the experiments with a vertically-positioned specimen new components and induction furnace of RASPLAV-3M setup were manufactured; the test facility was assembled, test runs were performed, the HF generator was adjusted for the operation with a new furnace, power deposition and specimen temperature conditions were

determined experimentally in blank tests without melt. The RASPLAV-3M setup was fully prepared for the first MCP-1 test with a vertically-positioned steel specimen.



1 – specimen cooling weir, 2 – water-cooled shaft-screen, 3 – water-cooled lid, 4 – quartz tube, 5 – crucible section, 6 – inductor, 7 – vessel steel specimen, 8 – bottom calorimeter, 9 – calorimeter support, 10 – uncooled screen.

## Fig. 2. Schematics with a traditional design of the vertical crucible and with a vertical steel specimen

#### Task 2

#### Subtask 2.1

An experimental study on the interaction of suboxidized melt with a vertically positioned cooled vessel steel specimen has been conducted (experiment MCP-1).

The MCP-1 experimental objective is to determine the sensitivity of steel surface (its corrosion) to the orientation of its interacting surface. For this reason other conditions of MCP-1 were to be completely identical to those of MC-6 (Project #833.2 METCOR): the interaction of suboxidized corium melt C-30 with a horizontally positioned vessel steel specimen.

The crucible charge had a required melt composition. But, due to a thick oxidic crust on the melt surface, which failed to disappear at the maximum plate voltage of the generator, and due to a different melt hydrodynamics, experimental conditions of MCP-1 and MC-6 were not similar, in particular, melt composition and thermal characteristics in the corium-steel interaction zone. E.g. the MCP-1 oxidation index was typical of C-17, and for MC-6 it was C-32; in the first case the maximum heat flux into the specimen was  $\approx 2 \text{ MW/m}^2$ , and in MC-6 – 1,25 MW/m<sup>2</sup>; maximum temperature on the interaction interface was  $\approx 1435^{\circ}$ C; and in MC-6 – 1400°C. These differences in experimental conditions are likely to explain non-similarity of

corrosion interaction characteristics. In particular, differently from MC-6, the bottom part of corium ingot contained a massive ring-shaped metallic inclusion with a composition different from that of the specimen interaction zone. Also differently from MC-6 the interaction zone had two layers with different compositions and microstructure. Temperature of the interaction interface, at which corrosion stops, for MCP-1 was 1000÷1090°C (note that in absence of on-line diagnostics in MCP-1 the final position of corrosion front is determined indirectly), and for MC-6 it is 1120÷1200°C; the composition of metallic interaction zone (IZ) in MCP-1 has a higher content of U and Zr in comparison with MC-6. As mentioned before, no direct measurement of steel specimen corrosion was foreseen in MCP-1. Therefore, its corrosion rate was evaluated by posttest calculations using the data of thermocouples and final position of corrosion front. Taking into account all mentioned above, the MCP-1 data cannot be used directly for making final conclusions about the influence of interaction interface orientation on corrosion characteristics.

Posttest physicochemical and metallographic analyses of the IZ and ingot samples, which were conducted within Task 5, as well as numerical studies, enabled a detailed analysis of MCP-1 outputs.

The completed analysis of MCP-1 data and posttest studies, also using the data of MC-6÷MC-9 (Project # 833.2 METCOR) as well as the OECD MASCA project, interpreted the MCP-1 data disregarding the spatial orientation of the interacting surface.

To explain differences between MCP-1 and MC-6, other than those of initial conditions, a hypothesis was proposed, which explains the final position of the interaction front by a chemical equilibrium in the established oxidic-metallic system in the thermal gradient conditions. This hypothesis enables to determine the final temperature of corrosion interaction, which, depending on the specific interaction conditions (melt mass and composition, specimen temperature conditions), is equal either to the eutectic temperature or to the IZ liquidus temperature.

For practical reasons and following the conservative approach as a first approximation we can use the eutectic temperature of  $\approx 1060^{\circ}$ C as a final temperature of corrosion interaction.

Still, in order to specify the steel corrosion kinetics at the interaction of suboxidized corium and vertically positioned steel, and for stronger experimental justification of abovementioned conclusions, it is advisable to have another experiment with ultrasonic measurements of corrosion kinetics.

A report on the MCP-1 results has been prepared.

#### Subtask 2.2

An experimental study of  $UO_{2+x}$ -ZrO<sub>2</sub>-FeO<sub>y</sub> melt interaction with a vessel steel specimen having a horizontal orientation in air and steam atmospheres at a high temperature on the specimen surface (1000÷1135°C) (experiment MCP-0, alias MC-12).

The experimental objectives were as follows: confirm vessel steel corrosion acceleration at interaction interface temperatures higher than  $\approx 1050^{\circ}$ C; check influence of air and steam atmospheres on the corrosion parameters and get experimental data on the vessel steel corrosion rate at temperatures > 1050°C on the interaction interface.

The analysis of MCP-0 (MC-12) data and posttest physicochemical and metallographic analyses; as well as the results of modeling studies have determined the following:

• The new data on vessel steel corrosion kinetics and mechanism at the interaction with UO<sub>2+x</sub>-ZrO<sub>2</sub>-FeO<sub>y</sub> corium in the air and steam atmospheres at 1000÷1135°C on the interaction interface, as well as the data of #833.2 METCOR testify to the absence of considerable influence of the oxidizing atmosphere (steam, air) on the steel corrosion kinetics in the studied temperature range.

- For the first time the following differences between steel corrosion in the mentioned conditions and corrosion in the oxidizing atmosphere (without corium) have been identified:
  - corrosion rate is time-independent, if temperature on the interaction interface is steady. It is explained by the constant thickness of corrosion and crust layer on the steel surface;
  - corrosion intensifies, if temperature on the interaction interface rises above a certain threshold value, which corresponds to approx. 1050°C on the steel surface, due to the formation of a liquid inter-grain phase in the corrosion layer, and in the crust uninterrupted liquid-phase percolation channels, which accelerate the diffusion transport of iron ions.

Within Task 5 the experimental data, posttest physicochemical, metallographic and numerical analyses of MCP-0 (MC-12) and results of MC-1, MC-2, MC-11 tests provided within the ISTC #833.2 METCOR Project were integrated to develop a correlation for calculating vessel steel corrosion rate at its interaction with  $UO_{2+x}$ -ZrO<sub>2</sub>-FeO<sub>y</sub> corium in the oxidizing atmosphere.

#### Subtask 2.3

The experimental study of vessel steel corrosion at its interaction with C-100 melt  $(UO_{2+x}-ZrO_2)$  in air (Test MCP-2).

The experimental objectives of the test: get experimental data on the vessel steel corrosion rate in a wide temperature range on the interaction interface; determine the influence of oxidizing above-melt atmosphere (steam, air); identify corrosion mechanism and find its difference from the corrosion taking place during the interaction of steel and corium having  $UO_{2+x}$ -ZrO<sub>2</sub>-FeO<sub>y</sub> composition in the oxidizing atmosphere; develop corrosion model and correlation, which would summarize the data of MCP-2 and MC-10 tests, provided by previous Project #833.2 METCOR on interaction with C-100 melt in the oxidizing atmosphere.

Using the data of MCP-2 experiment, posttest physicochemical and numerical analyses the following has been done:

- New experimental data on the vessel steel corrosion at its interaction with  $UO_{2+x}$ -ZrO<sub>2</sub> melt in air at 870÷1370°C on the interaction interface have substantially extended the data of MC-10 within the ISTC #833.2 METCOR.
- No influence from oxidizing atmosphere (steam, air) on the vessel steel corrosion kinetics has been proved.
- It has been found that in case of high-temperature corium  $UO_{2+x}$ -ZrO<sub>2</sub>, differently from  $UO_{2+x}$ -ZrO<sub>2</sub>-FeO<sub>y</sub> corium, corrosion does not intensify up to T<sub>S</sub>=1370°C, the maximum reached on the interface during tests.
- The physicochemical and numerical analyses of the IZ and corium ingot have shown that for UO<sub>2+x</sub>-ZrO<sub>2</sub> corium steel corrosion is determined by the diffusion processes in the corium crust on the steel surface; and the crust thickness is considerably higher than the thickness of corrosion layer. The crust remains solid and has no liquid-phase channels, which are typical of experiments with UO<sub>2+x</sub>-ZrO<sub>2</sub>-FeO<sub>y</sub> corium.

Within Task 5 the integrated analysis of experimental data and posttest analyses of MCP-2, and MC-10 (ISTC #833.2 experimental series) was followed by the development of a corrosion model and a correlation, which summarizes experimental data on the vessel steel corrosion at its interaction with C-100 corium ( $UO_{2+x}$ -ZrO<sub>2</sub>) in the oxidizing atmosphere. All experimental points within the Arrhenius coordinates are described by the proposed linear correlation with a satisfactory accuracy.

#### 3. Technical progress during the year of reference (second year) Task 1

In order to determine the characteristics of the interaction between the suboxidized corium melt and a vertical vessel steel specimen and for providing the required initial experimental conditions the following has been done:

- The generator was modified in order to increase power deposition in the melt. A scoping test was performed. A 30 % power increase was registered, which enables to increase the crucible diameter, a gap between the crucible and vertical specimen, power in the melt and facilitates the experimental procedure by avoiding a crust on the melt surface.
- The furnace and cooling system design have been adjusted for the 2<sup>nd</sup> experiment with a vertically positioned steel specimen. A crucible having an inner diameter of 100 mm, a quartz tube with the outer diameter of 154 mm, inductor and furnace lid were designed and manufactured.
- The vertically positioned vessel steel specimen was designed to make corrosion rate measurements with a USS possible. A special 'defect' was arranged, which served as a 'mirror' for the ultrasonic signal. A specimen with the 'mirror defect' was manufactured for 'cold' (without heating) tests and such tests were conducted. A specimen with the 'mirror defect' was manufactured for tests with heating in the inductor without melt, such tests were conducted and temperature correction coefficient were provided. A program for processing the USS signal was adjusted to take into account the mirror defect and temperature coefficients.
- Experimental units for tests with European vessel steel (steel 20 MnMoNi5-5) were fabricated and equipped with thermocouples.

#### Task 3

#### Subtask 3.1

An experimental study of the non-stationary oxidation process of suboxidized C-30 melt  $(UO_2-ZrO_2-Zr)$  was conducted. In it the neutral above-melt atmosphere (argon) was replaced with steam (Test MCP-3).

The initial MCP-3 conditions corresponded to Test MC-6 (ISTC #833.2). At the early stage, after the molten pool was produced, C-32 corium (76 mass %  $UO_2 - 9.33$  mass %  $ZrO_2 - 14.67$  mass % Zr) interacted with cooled vessel steel specimen at  $T_s \approx 1400$ °C on the interaction interface. Duration of the interaction (following the eutectic melting mechanism) was  $\approx 11$  hours until the specimen corrosion stabilized (stopped). During this stage a liquid-phase metallic IZ was formed, which contained Fe, U and Zr.

At the second stage, the initial conditions of which corresponded to those of MC-6, the oxidation process was started by replacing Ar atmosphere with steam.

Experimental objectives were:

- Determine qualitative and quantitative phenomena in the IZ caused by oxidation.
- Evaluate thermal effects taking place during the molten pool and IZ oxidation.

Oxidation kinetics and its influence on the thermal behavior and corrosion rate was determined using the data of hydrogen sensor installed in the gas line at the furnace exit, readings of thermocouples in the specimen and ultrasonic measurement of the corrosion front and interaction zone boundaries, also by calorimetry of the heat flux into the specimen calorimeter and crucible.

The primary processing of experimental data provided the following:

- The USS data processing determined the history of specimen corrosion kinetics during the interaction with suboxidized melt and resulting oxidation. Typical corrosion stages at interaction with suboxidized melt are as follows: the incubation period, which lasted  $\approx 10000$  s, preceded the specimen destruction by corrosion, which followed the mechanism of eutectic melting; its initial rate was the highest, and it kept reducing with time. The experimentally measured corrosion rate of this period agrees well with previously developed correlation

$$\frac{dh}{dt} = -0.46 \cdot 10^{-4} \sqrt{T_{int}(t) - T_{B}} , mm/s$$

Where  $T_{int}(t)$  – temperature on the boundary between specimen and IZ at time moment t;

 $T_{\rm B}$  – final temperature on the boundary between specimen and  $\,$  IZ, at which corrosion stops.

In accordance with the USS data by the time of IZ oxidation the specimen corrosion depth was  $\approx$ 7.6 mm, and the final temperature on the boundary between specimen and IZ was  $\approx$ 1160°C.

The maximum corrosion depth by the end of experiment, in accordance with USS, was  $\approx$ 9.4 mm. Due to the oxidation the corrosion depth and specimen ablation was larger. It was also influenced by the heat produced by chemical reactions taking place in the IZ.

- A profilogram of corroded steel surface was made. Fig. 3 shows the photographed axial section of the steel specimen top and a profilogram of corroded steel surface. It is seen that the maximum corrosion depth (on the specimen axis) was approximately 9.9÷10.0 mm, which is close to the data of ultrasonic measurements. The surface profile is close to axisymmetrical.



1 – boundary of undamaged specimen, 2 – boundary of posttest specimen, 3 – boundary of decarbonized steel, 4 – boundary of temperature influence on steel macro- and microstructure (isotherm 727°C)
 Fig. 3 – Axial specimen section and profilogram

 The oxidation kinetics of molten corium and IZ was analyzed using the on-line data of hydrogen sensor. The hydrogen sensor indications amended taking into account a delay in lines and other factors at different oxidation stages are given in Fig. 4.

Hydrogen liberation at the corium melt oxidation (stage C, Fig. 4) had a practically stable rate up to nearly complete Zr oxidation and final oxidation of U. To prevent the IZ and specimen oxidation at this stage the temperature was considerably reduced.

This was followed by a period without hydrogen generation, and the IZ and specimen temperature grew with a partial dissolution of oxidic crust, which separated the IZ from molten corium (Stage D, Fig. 4). With temperature growth the IZ oxidation took place, which was accompanied by the hydrogen generation at a constant rate (Stage E, Fig. 4).

Using the mentioned MCP-3 data and experimental results of MASCA, METCOR and EVAN it was determined that oxidation rate does not depend on the content of reducing agents in the melt and IZ; it is sensitive only to the conditions of oxidant supply. I.e. at the stable oxidant availability the reaction rate is practically steady up to nearly complete depletion of reducing agents inventory. The oxidation rate also does not depend on the oxidant type. As shown by MASCA results (experiment MA7), the oxygen and nitrogen absorbed from the supplied air during the melt oxidation were in proportion to their volumetric fractions (in absence of melt surface crust, which has a different permeability for oxygen and nitrogen).



#### Subtask 3.2

The physicochemical posttest analysis of MCP-3 specimens and samples was made. A template of the axial ingot section was prepared for the SEM/EDX analysis, as well as the polished section with crystallized melt samples. The microstructure of studied zones was determined; also their total and phase compositions.

The SEM/EDX analyses provided the following:

- Average composition of all samples gives the following data (with small divergence), mass %: 37.2±1.6 U; 31.8±1.1 Zr; 29.7±1.4 Fe, which agrees well with the XRF data. Three phases have been identified in the samples: one based on solid solution from the UO<sub>2</sub> side; one based on the solid solution from the ZrO<sub>2</sub> side and Fe(Gr, Ni)O<sub>y</sub>-based phase, in which U and Zr oxides do not melt.
- Chemical analysis of molten products. Using the methodologies tested in the project the content of U, Zr, Fe ions was determined (U<sup>+4</sup>, U<sup>+6</sup>, Zr<sup>+4</sup>, Fe<sup>+2</sup>, Fe<sup>+3</sup>). The sensitivity of U<sup>+4</sup> and U<sup>+6</sup> determination is ≈0.04 µg/ml. The method enables to determine separately the content of tetravalent uranium and total uranium. The amount of U<sup>+6</sup> is calculated from the difference of total and U<sup>+4</sup>. Fe<sup>+2</sup>, Fe<sup>+3</sup> are determined by the method tested in the project, which evaluates their amount in samples without uranium separation. The method determines Fe<sup>+2</sup> and total iron in the melt separately. Fe<sup>+3</sup> is evaluated from the difference. The evaluation error is ≤5 rel.%. A peculiarity of MCP-3 is a large amount of iron, which was transported into the melt during its interaction with steel. The method enabled a more accurate phase analysis of samples. Chemical analysis enabled to conclude that in these conditions iron oxide is close to the Fe<sub>3</sub>O<sub>4</sub> stoichiometry.

The SEM/EDX identification of metallic droplets in the melt (they had no time to get oxidized) confirms the melting of steel specimen at the final stage of IZ oxidation in the experiment.

- To study the microstructure of the steel ingot top templates and polished sections were prepared from the ingot and specimen axial sections. The studies of microstructure showed the presence of ferrite structure with a minimal carbon content in the top surface layer of steel specimen. This zone is  $\approx 11.5 \div 11.7$  mm deep in the central part of the specimen (Fig. 3). Macro- and microstructure of the specimen below this depth had changes caused by the carbon and chrome redistribution and grain coarsening. The formation of 'new' ferrite-pearlite structure took place in this zone at a fast cooling of austenite steel heated above the critical point. The formed needle-shaped pearlite can increase steel strength and hardness with a simultaneous reduction of shock viscosity and relative lengthening. This layer goes to the depth of 32 mm from the specimen top surface.

#### Task 5

The MCP-3 experimental data and posttest analysis were used in the modeling of temperature distribution on the specimen surface and in its bulk, heat flux from melt into the specimen, heat effects from redox reactions taking place during the melt oxidation, when neutral atmosphere is replaced by the oxidizing one. An integrated analysis of data provided by MCP-3 and experimental series of METCOR, MASCA, EVAN was made. Deliverables and publications were also prepared in the framework of Task 5. During the second year the following outputs were provided within Task 5:

- Temperature conditions of the steel specimen during the quasistationary 10-hour interaction between the melt and specimen in the neutral atmosphere. The calculation of temperature conditions used the finite-element program, in which the equation of stationary heat conductivity in the axi-symmetrical formulation was solved. The boundary conditions were: temperature on the internal surface of the top calorimeter, which was estimated as  $\approx 100^{\circ}$ C; temperature on the internal surface of the bottom calorimeter ( $\approx 20^{\circ}$ C) and temperature on the external surface of the periphery heat insulation, which was assumed to be equal to the average temperature of cooling water. In the calculations the average density of heat flux into the specimen top and thermal conductivity of the periphery heat insulation layer were varied in order to get the best convergence of calculations and experimental temperature values in the specimen, where thermocouple junctions were located. At this the distribution of the heat flux density on the specimen top radius was assumed to be the same as in MC-6 (project ISTC #833, METCOR).

The following reactions having temperature effects take place during the corium melt oxidation:

$$\label{eq:2} \begin{split} Zr + 2H_2O &= ZrO_2 + 2H_2 + 530 \text{ kJ/mol}\\ UO_2 + H_2O &= UO_3 + H_2 - 160 \text{ kJ/mol}\\ Fe + H_2O &= FeO + H_2 - 160 \text{ kJ/mol}\\ 2Fe + 3H_2O &= Fe_2O_3 + 3H_2 - 120 \text{ kJ/mol} \end{split}$$

Only the first of the above-listed reactions is exothermal.

Taking into account oxygen content in the exit gases for the numerical model it was assumed that the reaction is heterogeneous, i.e. takes place on the melt surface. Therefore, the oxygen dissolution heat in the suboxidized melt can be neglected. Note that the oxygen dissolution heat in the molten metallic uranium or zirconium is quite insignificant – about  $60 \div 70$  % of the heat of oxidation reaction with the corresponding element. And the process is exothermal.

Assuming that the melt cation composition after stage C is the same as the cation composition of molten products and knowing the melt composition and energy effects of reactions, we can determine the total power deposition from the melt oxidation at stage C, which is:

Then, knowing the oxidation time, the average heat input from redox reactions at stage C is:

$$Q^{(C)}=0.53 \text{ kW} (\pm 7\%)$$

By determining the melt mass  $(m_{melt})$  and its density  $(\rho_{melt})$  we find the average specific heat deposition per a unit of volume

$$q^{(C)} = \frac{Q^{(C)}\rho_{melt}}{m_{melt}} = 3.3 \text{ MW/m}^3 (\pm 8\%)$$

Heat is released during the IZ and steel oxidation at redox reactions, when oxygen dissolved in the melt is supplied to the IZ by the melt convection. Possible redox reactions at such interaction are as follows:

$$Zr + O_2 = ZrO_2 + 1090 \text{ kJ/mol}$$
  

$$U + O_2 = UO_2 + 1060 \text{ kJ/mol}$$
  

$$2U + 3O_2 = 2UO_3 + 2360 \text{ kJ/mol}$$
  

$$2Fe + O_2 = 2FeO + 500 \text{ kJ/mol}$$
  

$$4Fe + 3O_2 = 2Fe_2O_3 + 1440 \text{ kJ/mol}$$

At this all possible reactions are exothermal.

By determining masses of reducing agents (Fe, U and Zr) before stage E (Fig. 4) in the IZ and the mass of molten steel during oxidation we can find the total power deposition from the IZ oxidation at stage E, which is:

The average power input from the redox reactions in the IZ at stage E is

#### $O^{(E)} \approx 0.55 \text{ kW} (\pm 14\%),$

recalculation for the unit of specimen surface area and its 64 mm diameter gives the average heat flux on the surface

 $q^{(E)} \approx 140 \text{ kW/m}^2 (\pm 14\%).$ Comparing the calculated value  $q^{(E)}$  with a characteristic heat flux value on the external reactor vessel surface in the IVR conditions:  $0.5\div1.0 \text{ MW/m}^2$  we get  $q^{(E)}$  of approx. 14÷28% of this value, which can be important for the IVR case, if similar IZ oxidation rates are reached in reactor conditions. Here it should be noted that in case of IVR a part of IZ is close to the melt surface.

The influence of thermal gradient conditions on the repartitioning of components between the oxidic and metallic melts has been analyzed using the results of METCOR, METCOR-P and MASCA projects. Knowledge of these characteristics is essential not only for determining the interface temperature, at which the corrosion of steel interacting with suboxidized corium stops; it also can be used as the basis for modeling the structure of molten pool formed on the reactor vessel bottom under thermal gradient conditions.

The analysis has confirmed that the two-liquid molten pool structure is not critical for the IVR, if the metallic melt is located only on the bottom or in the top layer (and if it is thick enough). The least favorable IVR conditions can result from the formation of three-liquid pool with a metallic layer in the bottom and top in case of thermal gradient conditions and repartitioning of components through the crust at established equilibrium, which is very well illustrated by the METCOR data. It was shown that for the realistic modeling of three-liquid pool it is sufficient to develop a realistic twophase model with thermal gradient conditions and component repartitioning between oxidic and metallic melts through the crust. By now the development of such model has not been completed. Some METCOR and METCOR-P data can be used for the verification and updates of such model, but the data are still limited and heterogeneous. Therefore, taking into account the importance of these data for the IVR, an issue of replacing one of the remaining METCOR-P experiments with an experiment aimed at the model verification is discussed with collaborators.

- The following Task 5 activities have been completed, which are related to the integrated analysis of experimental series and preparation of the 2<sup>nd</sup> year deliverables.
  - Final versions of technical reports on experiments MCP-0 (MC-12), MCP-1, MCP-2 and MCP-3 have been completed in Russian and in English.
  - All experimental data of METCOR and METCOR-P on the interaction of steel • with coria having  $UO_{2+x}$ -ZrO<sub>2</sub>-FeO<sub>y</sub> and  $UO_{2+x}$ -ZrO<sub>2</sub> compositions in the oxidizing atmosphere have been analyzed. It was established that the corrosion rate depends on the melt composition, but practically is not influenced by the atmosphere (steam, air). It has been shown that at the vessel steel interaction with molten corium in the oxidizing atmosphere the corrosion rate is modified by the resistance to Fe ions diffusion of the corium crust adjacent to the specimen surface. At this the corrosion rate is stable at the steady temperature on the interaction interface. For 'low-temperature' melt (UO<sub>2+x</sub>-ZrO<sub>2</sub>-FeO<sub>y</sub>) a corrosion intensification is possible in case of temperature growth on the interaction interface, when the temperature rises above certain level ( $T_s > 1050^\circ$ ). This effect does not take place in case of 'high-temperature' corium (UO<sub>2+x</sub>-ZrO<sub>2</sub>).

Correlations have been produced for calculating vessel steel corrosion rate at the interaction with molten corium in the severe accident conditions; they can be used in the IVR analyses.

A publication has been prepared on the summarized experimental data.

• Using the correlations the numerical analysis of corrosion influence on the reactor vessel strength has been prepared jointly with collaborators. The finiteelement calculations, which used the developed model of heat exchange in corium, modeled temperature and stress-and-strain conditions of reactor vessel. It was shown that in the DNB absence on the external water-cooled surface of reactor vessel the influence of corrosion on the vessel strength characteristics is insignificant. A paper was published on the results of completed numerical study.

## 4. Current technical status

During two years of the Project three experiments (MCP-1, MCP-2, MCP-3) out of eight foreseen by the Work plan matrix have been completed. In agreement with collaborators an additional experiment MCP-0 (MC-12) was made, which provided important information for determining vessel steel corrosion patterns at the IVR. It was decided with collaborators that in the beginning of the third project year an experiment will be made, which will follow MCP-3. It will study the interaction of molten corium and European reactor vessel steel in the oxidizing conditions. This experiment combines two planned tests on the steel interaction with two coria compositions:  $UO_{2+x}$ -ZrO<sub>2</sub>-FeO<sub>y</sub> and  $UO_{2+x}$ -ZrO<sub>2</sub>.

A certain delay in the Work plan implementation is explained by the following reasons:

- complexity in developing a new system for direct corrosion measurement by ultrasonic techniques on a vertical surface,

- required generator upgrade for increasing power deposition in the melt,

- delay in the European vessel steel shipment.

During two years of implementation 45 % of the total scope of work has been done.

## 5. Cooperation with foreign collaborators

Foreign collaborators within the Project are well-known experts representing scientific research centers of the European Union:

- 1. Dr. Walter Tromm, Germany
  - Forschungzentrum Karlsruhe GmbH, NUCLEAR
- 2. Dr. Alexei Miassoedov, Germany Forschungszentrum Karlsruhe GmbH, IKET
- 3. Dr. David Bottomley, Germany EUROPAISCHE KOMISSION, Institut fur Transurane (ITU)
- 4. Dr. Pascal Piluso, France CEA Cadarache – DEN/DTN/STRI
- 5. Olli Kymalainen, Finland FORTUM Nuclear Services Ltd.
- 6. Manfred Fischer, Germany AREVA NP GmbH
- 7. Dr. Sieghard Hellmann, Germany AREVA NP GmbH
- 8. Dr. Eberhard Altstadt, Germany Forschungzentrum Dresden-Rossendor (FZD)

Close cooperation with Western collaborators was maintained during the second year of Project implementation. It included:

- detailed discussion and approval of Work plan and experimental matrix,

- analysis and evaluation of results of each experiment made during the reported period,

- update of the planned tests specification in parallel with numerical studies,

- joint preparation pf papers and conference presentations.

Discussion of the METCOR-P scope of work and results was made at meetings with collaborators and during CEG-SAM sessions.

Second meeting of the METCOR-P Steering committee took place on the 9<sup>th</sup> of July, 2008 in St. Petersburg. Detailed attention was paid to the following:

- Results of MCP-1 on the interaction of suboxidized melt and vertically positioned vessel steel specimen. A decision was taken to conduct a second experiment with a vertical steel specimen after a modification of the RASPLAV test facility and corrosion rate measurement system including ultrasonic techniques.
- Results of MCP-2 on the studies of vessel steel corrosion rate during its interaction with UO<sub>2+x</sub>-ZrO<sub>2</sub> corium in a wide range of temperatures on the interaction interface in air.

Advisability of combining experiments on the studies of corrosion kinetics at corium interaction with European vessel steel in air for the following compositions  $UO_{2+x}$ -ZrO<sub>2</sub> and  $UO_{2+x}$ -ZrO<sub>2</sub>-FeO<sub>y</sub> by adding Fe into the melt.

 Calculations made by collaborators using the developed corrosion correlations on the influence of melt-induced vessel steel corrosion on the stress-and-strain condition of the reactor vessel

During the year Project contractors have taken part in the session of the CEG-SAM expert group: at the 14<sup>th</sup> CEG-SAM on 9-11 September 2008 in Kiev, Ukraine, and at the 15<sup>th</sup> CEG-SAM on 9-13 March 2009, in Villigen, Switzerland. At these meetings the participants were briefed about the progress of studies within the Project.

Two papers and a presentation for an international conference were prepared jointly with collaborators.

#### 6. Problems encountered and suggestions to remedy

Due to a delay in the Work plan implementation explained by the required modifications in the system of direct corrosion rate measurements on the vertical specimen surface using ultrasonic techniques, by additional requirements on increasing the generator power, by a delay in the European vessel steel shipment by collaborators, a necessity of project period extension for 5 months without an increase of project budget has become evident.

#### 7. Perspectives of future developments of the research/technology developed

Due to the above-mentioned reasons the scope of work planned for the first two years of METCOR-P implementation has been completed by 70 %. By the end of the 2<sup>nd</sup> year the project collaborators have shipped the European vessel steel specimens and the system for corrosion rate measurement with an ultrasonic sensor for the vertical specimen surface has been developed. In the beginning of the third Project year an experiment on the corium melt interaction with a European vessel steel specimen in the oxidizing atmosphere is planned.