

ISTC 833-2003

# Final Project Technical Report of ISTC 833-2003

# INVESTIGATION OF CORIUM MELT INTERACTION WITH NPP REACTOR VESSEL STEEL (METCOR2)

(01.01.2003 - 31.05.2006)

**Project manager** 

Vladimir B. Khabensky

Authors: S.V. Bechta., V.S. Granovsky, S.A. Vitol, E.V. Krushinov, A.A. Sulatskiy,
V.V. Gusarov, V.I. Almiashev., Yu.B., Petrov., D.B. Lopukh, A.Yu.
Petchenkov, I.V. Pozniak., S.Yu. Kotova., E.K. Kaliago., I.V. Kulaghin.,
V.G. Blizniuk., S.A. Smirnov., V,V, Martinov., A,P, Martinov, A.V.
Lisenko, E.V. Shevchenko, V.R. Bulighin, N.E. Kamensky, A.A. Chertkov,
E.M. Beliaeva.

Sosnovy Bor May 2006

### ABSTRACT

The project objective is to study experimentally physicochemical phenomena taking place at the interaction of uranium-bearing melt and VVER vessel steel.

Experiments have been performed on the 'Rasplav-2' and 'Rasplav-3 tests facilities based on the technique of induction meting in a cold crucible.

Steel behavior has been examined for coria  $UO_2$ - $ZrO_2$  and suboxidized  $UO_2$ - $ZrO_2$ -ZrO

Experimental data on steel corrosion depth and kinetics have been produced. Correlations for corrosion calculation have been proposed.

Key words: nuclear reactor, severe accident, uranium-bearing corium, vessel steel, corrosion, experiment, induction melting, cold crucible.

Project title	Investigation of corium melt interaction with NPP reactor vessel steel (METCOR 2)
Annual report No	№ 833-2003
Contractor	Aleksandrov Research Institute of Technologies of the RF Federal Agency for Atomic Energy
	Russia, 188540, Sosnovy Bor of Leningrad Oblast, NITI
Project manager	Professor V.B. Khabensky, Head Researcher of Aleksandrov Research Institute of Technologies of the RF Ministry for Atomic Energy
	Russia, 188540, Sosnovy Bor of Leningrad Oblast, ul. 50let Oktiabria, 19, ap. 50
	<u>tel:</u> 7(812-69) 60-625
	<u>fax:</u> 7(812-69) 63-672
	E-mail: vbkh-npc@sbor.net
Date of project start	01 January 2003
Project duration	41 months
Collaborators and their organizations	<u>Dr David Bottomley</u> , European Commission, Directorate General Joint Research Centre, Institute for Transuranium Elements, Postfach 2340, Hermann-von- Helmholtz Pl. 1, 76125 KARLSRUHE; Germany
	<u>Dr Sieghard Hellmann</u> , AREVA NP, Dept. NGTR,Freyeslebenstrasse 1, D-91058 Erlangen, Germany
	Manfred Fischer, AREVA NP, Freyeslebenstrasse 1, D-91058 Erlangen, Germany
	Dr Eberhard ALTSTADT, FZ-Rossendorf, Germany
	<u>Dr.Walter Tromm</u> , Institut fur Kern- und Energitechnik (IKET), Germany
	<u>Dr Pascal PILUSO</u> , Commissariat a l'énergie atomique direction de l'énergie nucléaire/dtN/stRI/laboratoire pour la maîtrise des accidents graves, France
	Florian FICHOT, IRSN / DRS / SEMAR / CEN, France
	Olli Kymäläinen, Fortum Nuclear Services Ltd, Finland

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#### **1 INTRODUCTION**

Main objective of the current project is to enhance the nuclear reactor safety at severe accidents involving the core degradation. The specific subject of the project is the in-depth investigation of physicochemical phenomena taking place at the interaction of oxidic corium melt and NPP reactor vessel steel.

The 1<sup>st</sup> Project Phase studied the interaction of steel with fully oxidized oxidic corium  $UO2_{+x}$ -ZrO<sub>2</sub>-FeO<sub>y</sub> in the neutral and air atmospheres above the melt. It has been found that the interaction causes steel corrosion controlled by the diffusion resistance of corrosion layer. The provided experimental data were used for developing correlations for calculating VVER steel corrosion kinetics in the conditions of external vessel cooling.

The reported  $2^{nd}$  stage of the Project has studied the interaction of vessel steel with fully oxidized coria  $UO_{2+x}$ -Zr $O_2$  and  $UO_{2+x}$ -Zr $O_2$ -Fe $O_y$  in steam and suboxidized oxidic and metal-oxidic corium  $UO_2$ -Zr $O_2$ -Zr $O_2$ -Zr $O_2$ -Se $O_y$ .

Similarly to the 1<sup>st</sup> Project Phase the 2<sup>nd</sup> Phase studies phenomena related to the melt retention inside the reactor vessel (IVR). The project was aimed to provide the following information:

- 1. Qualitative and quantitative characteristics of vessel steel corrosion depending on
  - melt composition and above-melt atmosphere;
  - heat flux temperature and density on the interaction interface.
- 2. Interaction model based on the analysis of structure and elemental composition of the impact zone.
- 3. Correlations for calculating vessel steel corrosion.

Results of the project can be used:

- to improve numerical modeling of phenomena taking place at a severe accident stage when a molten pool is formed on the bottom of reactor vessel;
- to prove and increase the safety of operating and designed VVER, PWR and BWR reactors.

In accordance with the Work Plan the scope of work included experimental studies of the following aspects of the vessel steel – molten corium interaction:

- interaction of corium C-100 in the neutral above-melt atmosphere at different temperatures on the steel specimen surface;
- interaction of suboxidized corium having different oxidation degree in the neutral above-melt atmosphere at different temperatures on the steel specimen surface;
- interaction of suboxidized corium in the neutral above-melt atmosphere at a maximum possible temperature on the steel specimen surface;
- interaction of corium  $UO_{2+x}$ -ZrO<sub>2</sub> (C-100) and  $UO_{2+x}$ -ZrO<sub>2</sub>-FeO<sub>y</sub> in steam at different temperatures on the steel specimen surface.

Table 1 presents the experimental matrix.

Experim ental series	Description	Test №	Melt composition, mass.%	Melt temperature, °C	Temperature on the specimen surface, °C	Atmosphere
1	Interaction between suboxidized/oxidized (oxidic phase) melt and vessel steel	MC 5	~C-100 UO <sub>2</sub> /ZrO <sub>2</sub>		~ 1073 ~ 1180 ~ 1315 ~ 1434	
		MC 6	~C-30 UO <sub>2</sub> /ZrO <sub>2</sub> /Zr		~ 1400	
		MC 7	~C-30 UO <sub>2</sub> /ZrO <sub>2</sub> /Zr		~ 1150	
		MC 8	~C-70 UO <sub>2</sub> /ZrO <sub>2</sub> /Zr	up to	~ 1420	argon
2	Interaction of suboxidized metal- oxidic corium and vessel steel (oxidic + metallic phases)	MC 9	~C-30 UO <sub>2</sub> //ZrO <sub>2</sub> //Zr +steel MC <sub>T</sub> /M <sub>kop</sub> =0.1	2600	~ 1440	
3	Interaction of molten corium and vessel steel in steam	MC10	UO <sub>2+x</sub> /ZrO <sub>2</sub>	up to 2800	~ 1035 ~ 1185 ~ 1235 specimen melting	steam
		MC11	UO <sub>2+x</sub> /ZrO <sub>2</sub> /FeO <sub>y</sub>	up to 2200	~ 950 ~ 1050 ~ 1130 ~ 1200	

 Table 1 – Experimental matrix of Project 833.2

Additionally to experiments given in Table 1 auxiliary tests were performed in order to optimize the HF generator parameters for the melting of suboxidized corium and prepare charge having the composition required for the main experimental series.

### 2 METHODOLOGY

Experimental studies of the interaction between molten uranium-bearing corium and vessel steel were carried out on the "Rasplav-2" and "Rasplav-3" test facilities using the technique of induction melting in a cold crucible. Main advantages of this approach are as follows:

- melting and superheating of refractory materials;
- power deposition in the melt possessing a high chemical purity comparable to the initial composition of charge components;
- no restrictions on the test duration;
- experiments in the neutral and oxidizing atmosphere above the melt.

'Rasplav' test facilities are provided with the data acquisition system for measurements, registration and the following primary data processing during the experiments:

- temperature on the melt surface;
- temperature in the steel specimen;
- specimen corrosion depth;
- temperature and flow rate of coolants;
- electrical parameters of the generator.

Differently from "Rasplav-2" used in the 1<sup>st</sup> stage of Project implementation "Rasplav-3" having a lower generator frequency ensured the melting of suboxidized corium, which has a higher electric conductivity in comparison with fully oxidized corium. During preparation of the experimental series 'Rasplav-3" was adjusted for the purpose. The generator frequency was optimized and power deposition in the melt was increased. Along with that, the system of gas-aerosol sampling was improved in order to monitor gas flow parameters, oxygen concentration in particular.

Posttest analysis included numeric modeling of steel specimen temperature conditions aimed at the determination of temperature and heat flux density on the interaction interface, it also included the following physicochemical analyses:

- X-ray fluorescence (XRF);
- chemical;
- energy dispersion X-ray (EDX);
- scanning electron microscopy (SEM);
- differential thermal (DTA);
- optical microscopy.

The above-listed analyses were performed in order to determine, specify or confirm the compositions (initial and final) of interaction materials, check material balance, which proves the credibility of experimental data, to identify side effects accompanying studied process (vessel steel corrosion) and, eventually, get information necessary for determining nature and mechanism of corrosion .

In all experiments vessel steel 15Kh2NMFA-A specimens had the same design. K-type thermocouples were embedded into them. Acoustic defect was made close to the specimen top, on which the melt was produced; it was used for a better accuracy of ultrasonic

measurements of corrosion depth. Calorimeters were located in the specimen bottom. In order to shield specimen from the direct heating in the electromagnetic induction field the crucible sections were welded together in the zone of specimen position.

The experimental procedure was as follows. After the molten pool was produced, the power deposition and temperature of the melt were varied by adjusting the electrical parameters and by changing the position of crucible in relation to the inductor and electromagnetic screens. This was done in order to get a required temperature on the specimen top. After that stable temperature conditions of the specimen were maintained during a predetermined period of time, which was specified in the course of the experiment. During this corrosion depth and other relevant parameters were measured. Within an experiment these stages were repeated for a certain number of temperature plateaus. Melt samples were taken periodically, during long-term experiments corrective additions were made to compensate for melt evaporation. Experiments were completed by the generator disconnection and corium crystallization.

#### **3 RESULTS OF INVESTIGATIONS**

Work within the project was performed in strict compliance with the schedule. The Project prolongation for 5 months (until 31 May, 2006) without additional budgeting was explained by the necessity to complete the analysis and systematize results for preparing the Final report and publications.

The following studies have been completed in the framework of Project № 833.2-2003.

# 3.1 Interaction of corium C-100 with vessel steel in the neutral above-melt atmosphere. Test MC 5

Test MC 5 studied the kinetics of molten corium – vessel steel interaction in the conditions of assumably minimum influence of chemical processes. For that a fully oxidized composition of corium C-100 was chosen, (mass %): 70 % UO<sub>2</sub> – 30 % ZrO<sub>2</sub>; the experiment was conducted in high-purity argon (partial pressure of oxygen  $P(O_2) < 2 \cdot 10^4$  bar).

MC 5 is presented in detail in Appendix 2 to the Annual report  $N_{21}$ -833.2-2003. This section briefly gives the main results.

The startup molten pool was produced from the charge having the mass of 1794.7 g (of it 300.9 g was the powder of C-100 corium produced in the preliminary Test).

In MC 5 one melting session included 4 steady-state regimes of interaction, at which different temperature levels were maintained on the specimen top. Table 2 gives maximum temperatures in the top center, average density of heat flux into the specimen and its value in the sighting spot of the ultra-sonic sensor ( $\emptyset$  15 mm) for all four regimes.

Regime	Heat flux	Heat flux	Power into the	Power into the top	Maximum
Regime	density	density	ton calorimeter	calorimeter	temperature of
	average	$\alpha$ 15	calculated	experimental	steel surface near
	average,	$\bigcirc$ 15,			
	MW/m <sup>2</sup>	MW/m <sup>2</sup>	KW	ĸw	the specimen
					axis,
					°C
1	0.66	0.95	1.28	1.40	1073
2	0.73	1.05	1.42	1.46	1180
3	0.81	1.20	1.60	1.58	1315
4	0.89	1.30	1.75	1.75	1434

Table 2 – Calculated and experimental values of power, heat flux and temperature

The uninterrupted duration of Test MC 5 was over 36 hours.

In accordance with ultrasonic measurements the vessel steel corrosion kinetics was as follows:

- During time interval 4600-22500 s. at the maximum temperature of the specimen top (in the center) T<sub>s.</sub> ≈ 1075°C no corrosion is observed.
- During time interval 23200-60300 s. at T<sub>s.</sub> ≈1180°C specimen corrosion is ≈ 70 μm.
- During time interval 65900-96000 s. at  $T_{s.} \approx 1315^{\circ}C$  specimen corrosion is  $\approx 100 \mu m$ .

• B During time interval 98500-133900 s.  $T_{s.} \approx 1435^{\circ}C$ . Due to unstable and erroneous indications of the ultra-sonic sensor corrosion depth was not measured.

In the beginning of  $2^{nd}$  and  $3^{rd}$  temperature plateaus corrosion goes intensively, after that it gradually slows down and reaches saturation.

Posttest studies of the steel specimen showed that the total corrosion depth was between 20 - 200  $\mu$ m. The affected steel surface had both uniformly corroded parts and locations with corrosion pits.

Analysis of the oxidic ingot showed that the top, bottom, central and side parts of it practically had no difference from the initial corium composition.

At the boundary between oxidic and metallic parts a layered oxidic structure was observed. The external bottom layer of the ingot (about 0.8 mm) had a homogeneous composition and structure close to the average composition of the oxidic ingot. This region is likely to consist of the sintered initial powder simulating the molten layer, which was placed on the specimen. This was confirmed by the porous structure of the bottom zone. Adjacent to it was a thin layer mostly consisting of uranium oxides. Above it - the layer of oxidic mixture having the molar ratio of  $UO_2:ZrO_2 \approx 1:2$ . In accordance with available data such ratio corresponds to the point of liquidus-solidus contact from the refractory side. It should be noted that neither crust nor the melt bulk almost did not contain iron oxides. Therefore it can be assumed that steel corrosion products are localized in a narrow zone between steel and sintered crust layer.

The SEM/EDX analysis of the metal-oxide interaction boundary showed its irregularity both in terms of microstructure and phase composition depending on the radial position of the analyzed location (on its temperature during the interaction).

Closer to the periphery, approximately one third from the edge, a distinct iron oxide layer (FeO) is observed next to the steel specimen. Above this is the oxidic layer consisting of  $UO_2$ ,  $ZrO_2$  and small quantities of FeO – this indicates he diffusion mixing of corrosion layer and sintered crust. The FeO layer has a rather high porosity.

Closer to the specimen top center there is a higher temperature region. Fe<sub>3</sub>O<sub>4</sub> fractions are observed on the boundary between oxidic corrosion layer (FeO) oxidic crust, which interacted with iron oxide. The presence of a rather high content of uranium oxide in this region should also be noted. This zone has a rather distinct microstructure and phase composition different from the FeO-based zone adjacent to steel and the zone of sintered corium crust. It can be assumed that the intermediate zone of  $UO_2$ -ZrO<sub>2</sub>-FeO was in the eutectic temperature region (~ 1320°C), and a liquid phase was formed in the mentioned interlayer.

At a maximum temperature in the center of the specimen top the molten oxidic phase propagated as far as the steel specimen surface; a partial melting of metal was observed on the boundary. The melting of metal is confirmed by the presence of uranium oxide inclusions inside the metal in the near-boundary region. It is evident that at the temperature level  $\sim$ 1420°C a eutectics was formed in the UO<sub>2</sub>-ZrO<sub>2</sub>-FeO-Fe system, which resulted in the steel specimen melting.

The metallographic analysis of steel has shown that the long-term exposure of steel to the high-temperature thermal gradient conditions resulted in the microstructural transformation of the surface layer to the depth up to 10 mm, which was detected as grain coarsening and pore formation. The deterioration of steel mechanic properties can be expected in this layer.

The MC5 experimental data on the interaction of molten corium C-100 and vessel steel in the inert atmosphere and posttest analysis have shown the following:

- 1. At  $T_s < 1100^{\circ}$ C on the steel specimen top there is no noticeable interaction between melt and steel. In these conditions the crust provides a reliable protection against melt impact.
- 2. In the temperature range  $T_{s.}=1150\div1250^{\circ}C$  steel gets oxidized and the FeO layer is formed. Steel oxidation is likely to be caused by the increased non-stoichiometry of uranium oxide; oxygen liberates at the temperature increase. At a higher temperature level due to the limited oxidation resource of the sintered oxidic crust the corrosion rate at first increases then decreases, and after 3 hours it stops. In this temperature range the corrosion regime has a solid-phase character; there are distinct boundaries between steel, FeO layer and sintered oxidic crust. The depth of steel corrosion is < 0,1 mm.
- 3. In the temperature range T<sub>s</sub>=1270÷1400°C the oxidation pattern and corrosion rate are similar to those registered during the previous temperature plateau. Corrosion is caused by the FeO formation resulting from the oxygen diffusion from the crust. After the new temperature level is achieved the corrosion rate first increases then slows down. But there are certain differences: Fe<sub>3</sub>O<sub>4</sub> globules have been found on the boundary between FeO and crust. Probably after the reciprocal diffusion of uranium/zirconium oxides from the crust and iron oxides, when a certain temperature is reached in the contact zone, the eutectic melting takes place and a liquid layer is formed between FeO layer and the layer of sintered oxides UO<sub>2</sub>-ZrO<sub>2</sub>. In accordance with adjusted evaluation the temperature of eutectic melting in the UO<sub>2</sub>-ZrO<sub>2</sub>-FeO system is approx. 1320°C. In this temperature range the oxidation regime is mixed solid-liquid phase.
- 4. At  $T_s>1400^{\circ}C$  the thickness of liquid layer increases; when a certain temperature is reached on the boundary with the steel specimen its eutectic melting is possible at a temperature below the normal temperature of steel melting. In accordance with approximate evaluation the eutectic temperature in the (UO<sub>2</sub>-ZrO<sub>2</sub>-FeO-Fe) system from Fe side is ~ 1420°C. The steel specimen melting was likely to distort the corrosion depth indications of ultra-sonic sensors. In this temperature range the oxidation regime has a liquid phase.

Results of MC 5 show that the interaction of corium C-100 and vessel steel in the inert atmosphere has a lower corrosion rate in comparison with experiments conducted during the 1<sup>st</sup> Phase of METCOR project.

### 3.2 Interaction of suboxidized corium with vessel steel in the neutral above-melt atmosphere $(T_{st.surf.}^{max} > 1400^{\circ}C)$ . Test MC6

Test MC 6 was aimed at studying steel ablation kinetics during its interaction with suboxidized corium C 27 in the oxygen-free atmosphere (argon) at a constant temperature on the specimen top.

The initial corium was produced from the charge having 1850 g mass and the following composition, mass %: 76% UO<sub>2</sub>; 9.33 % ZrO<sub>2</sub>; 14.6 % Zr.

In MC 6 the studies of melt - specimen interaction were conducted during two steadystate regimes having similar temperatures of the specimen top. Table 3 gives maximum temperatures in the top center, average density of heat flux into the specimen, its value in the sighting spot of the ultra-sonic sensor ( $\emptyset$  15 mm), and the power into the calorimeter in the beginning of the steady-state regime.

Regime	Heat flux	Heat flux	Power into the	Power into the	Maximum
	density,	density,	top calorimeter,	top calorimeter,	temperature of
	average,	Ø 15,	calculated,	experimental	specimen
	MW/m <sup>2</sup>	$MW/m^2$	kW	kW	surface near
					the specimen
					axis,
					°C
1	0.85	1.23	1.67	1.64	1375
2	0.87	1.31	1.7	1.71	1397

Table 3 - Calculated and experimental values of power, heat flux and temperature

Duration of MC 6 was 10 hours.

Results of MC 6 and its posttest analyses are presented in Appendix 3 of the Annual report №1-833.2-2003. This section summarizes the main results.

The ultrasonic data on the steel specimen ablation kinetics showed that during ~ 3 hours steel ablation had the same character as MC 5 corrosion at the  $2^{nd}$  and  $3^{rd}$  temperature regimes. The corrosion rate decreased in time and in ~ 2.5 hours it reached saturation. But in ~ 5 hours the ultrasonic sensor started to give erroneous readings showing the specimen length increase, it lasted during ~ 3 hours and was followed by the return back to normal indications. These anomalous readings do not indicate the actual increase of specimen length, they are explained by the changes in the steel ablation mechanism.

The posttest analysis of the specimen showed that before cooling a partially liquid region containing Fe-U-Zr-O occupied space to the depth of 6 mm from the surface; and this region had a higher density in comparison to steel. These conditions caused the reduction of ultra-sonic speed signal in the mentioned region, a longer time of signal transfer through the mentioned zone and, consequently, apparent increase of specimen length in accordance with ultra-sonic measurements.

After the liquid phase is formed the previously used standard methodology of ultrasonic measurements of the steel specimen ablation rate cannot be used. It can be applied directly only for determining the start of liquid phase formation on the specimen top, also for the qualitative evaluation of the liquid phase progression kinetics and the boundary stabilization at a certain depth. But a special methodology of the posttest echogram processing enabled to determine the progression kinetics of the boundary between the solid steel specimen surface and the mushy zone of Fe-U-Zr-O.

The results of SEM/EDX analysis have shown the following:

- In the top part of the steel specimen corium and steel are separated by the metallic phase mostly consisting of iron enriched with uranium, zirconium and a small quantity of oxygen. The cross-section of this zone has the shape of the spherical segment, it extends as far as the lower boundary of the acoustic defect.
- Chemical composition and microstructure of this zone are vertically and horizontally homogeneous.
- There are credible indications that before the crystallization the mentioned zone was partially molten. This is confirmed by the microstructure, presence of iron dendrites formed at the fast crystallization after the HF heating disconnection (in the quenching conditions) and the presence of a characteristic eutectic structure between

dendrites. The dendrite composition is close to pure iron, i.e. different from the steel composition. This is another confirmation that the phase got crystallized from the melt, but was not a solid-phase inclusion into the melt.

 After crystallization the melt, which formed on the steel-corium boundary, had three phases: iron-based, containing a certain amount of chrome and admixture quantities of manganese, nickel, silicon; a phase based on the (U, Zr) Fe<sub>2</sub> compound having admixtures of chrome, nickel, manganese and silicon; UO<sub>2</sub>-ZrO<sub>2</sub>-based solid solution.

The interaction zone depth was determined from the axial section profilogram. The profilogram was measured using the object-micrometer, the error  $\pm 10 \,\mu$ M. It was evident that the eutectic melting caused steel specimen ablation to the depth up to 6.7 mm.

The non-uniformity of the boundary between the crystallized metallic (Fe-U-Zr-O) and oxidic phases can be explained by the partial floating of the metallic melt at the local breaks in the crust.

The experimental data of Test MC 6 on the interaction between suboxidized corium C-27 and vessel steel in the inert atmosphere and the posttest analysis have shown the following:

- At T<sub>s</sub>≈1400°C on the steel specimen surface two characteristic stages of steel ablation can be identified. At the first stage the ultrasonic measurements showed a slow ablation of vessel steel, which could be explained by the diffusion of uranium, zirconium and oxygen from corium through the crust to the interaction boundary. The ablation rate decreased in time and reached saturation in 3 hours. At the second stage, in ~ 5 hours after the interaction start, a rather intensive steel ablation was observed. It was caused by the eutectic interaction with the formed metallic melt mostly consisting of iron enriched with uranium, zirconium and a small amount of oxygen.
- 2. In accordance with US sensor the ablation after the first stage was ~ 0.27 mm, the maximum ablation depth after the second stage was 6.7 mm.
- 3. The steel ablation rate at the second stage decreased in time following the temperature decrease on the interaction interface. In 3 hours after the second stage start it reached saturation at 1120-1200°C.
- 4. There is a high degree of certainty that the metallic phase, which interacted with the steel specimen, was in the solid-liquid condition. It included iron, uranium, zirconium and oxygen.

Results of MC 6 show that the interaction of molten suboxidized corium C-27 and vessel steel is characterized by a considerably higher intensity and a different ablation mechanism in comparison to MC 5.

### 3.3 Interaction of suboxidized corium C-32 and vessel steel in the neutral above-melt atmosphere $(T_{st,surf.}^{max} > 1150^{\circ}C)$ . Test MC7

Test MC7 studied vessel steel ablation kinetics during the interaction with suboxidized corium C-32 in the oxygen-free atmosphere (argon) and temperature of the specimen top  $T_{st.surf.}^{max} \approx 1150^{\circ}$ C. The experiment examined the influence of steel surface temperature on the

interaction kinetics (other conditions being equal, .MC6 temperature on the interacting steel surface was  $T_{st.surf.}^{max} \approx 1400^{\circ}$ C) and evaluated the boundary temperature, at which specimen ablation having the mechanism of eutectic melting stopped.

The startup corium was prepared from the charge having the mass of 1850 g and composition (mass %):  $76\% UO_2$ -9.33%ZrO<sub>2</sub>-14.67%Zr (of it 150 g was prepared from the ingot produced in the preliminary test, this part of the charge was put on the specimen top before the experiment).

Total duration of the experiment was 10 hours. The experimental procedure, parameters of the furnace and steel specimen corresponded to Test MC 6.

Table 4 gives maximum temperatures in the top center, average density of heat flux into the specimen, its value in the sighting spot of the ultra-sonic sensor ( $\emptyset$  15 mm), and power into the calorimeter at the regime start.

Heat flux	Heat flux	Power into the	Power into the	Maximum
density,	density,	top calorimeter,	top calorimeter,	temperature of
average,	Ø15,	calculated,	experimental	steel surface near
MW/m <sup>2</sup>	$MW/m^2$	kW	kW	the specimen
				axis,
				°C
0.82	1.1	1.2	1.39	1153

Table 4 - Calculated and experimental values of power, heat flux and temperature

MC 7 experimental data and results of the posttest analysis are given in detail in Appendix 1 of the Annual report  $N_{2}$ -833.2-2003. This section summarized the main results.

After the molten pool was produced, the temperature of ~1150°C was reached on the specimen top, from this moment and approximately for ~36000 s. vessel steel ablation kinetics was studied in the stabilized temperature conditions. In the end of the test the generator was disconnected and the ingot with specimen was cooled in argon.

Throughout the whole test the water-cooled pyrometer shaft was sparged with argon. It was used for continuous measurements of melt surface temperature using the spectral ratio pyrometer RAYTEK. The maximum temperature measured in absence of surface crust was  $\sim$ 2400°C.

After the test completion the corium ingot and steel specimen were enclosed into the epoxy resin and cut. They were used for making templates for posttest analyses.

The following results were provided by the direct measurement of interaction parameters, including the direct measurement of the steel specimen ablation by the ultra-sonic sounding, posttest physicochemical and metallographic analyses, numerical modeling of the specimen temperature conditions:

- 1. Axial section of the crystallized corium and steel specimen had three main zones: a) crystallized corium, chemical composition of which had varied vertical distribution of elements, b) interaction zone shaped as an irregular lens of crystallized metallic melt enclosed into the body of the steel specimen on its top and separated from corium by crust; c) steel specimen with a thermal impact zone located below the interaction zone.
- 2. Vessel steel ablation had three stages:

- Incubation period, which lasted ~10000 s., during which the change of chemical and phase composition on the corium-steel boundary took place.
- Slow ablation period, which lasted 8000 s., during which steel corroded at an average linear speed of  $2.86 \cdot 10^{-5}$  mm/s. to produce a liquid with eutectic composition on the interaction boundary at the end of this stage. Vessel steel corrosion at that stage was ~0.2 mm.
- Intensive ablation period, which lasted ~8000 s., during which steel ablated following the eutectic melting mechanism. At this stage the corrosion rate was ~ $10^{-4}$  mm/s., and it did not stabilize in the end of the experiment. Maximum local ablation depth was ~2.9 mm.
- 3. The interaction zone analyzed as the crystallized metallic melt had the following characteristics:
  - The integral chemical composition of the crystallized metal from the interaction zone was as follows: U/Zr/Fe/Cr/Ni/Mn/Si/O=43.95/2.15/49.63/1.58/0.72/0.45/0.4/1.11 (mass %). 80 % of the crystallized melt structure consist of the eutectics having the following composition: U/Zr/Fe/Cr/Ni/O=40.35/2.26/53.44/1.66/0.78/1.51 (mass %). The remaining 20 % of the volume consist of the grains belonging to two metallic solid solutions, which can be described as U(Zr)Fe<sub>2</sub>(O) and Fe<sub>4</sub>(O)Zr(U).
  - All physicochemical processes in the interaction zone took place within the temperature range 1150-1030° C. In accordance with calculations the temperature boundary, at which vessel steel ablation caused by the impact of C-30 corium stops, is 1030-1100°C. At this solidus temperature (eutectic temperature) of the interaction zone material determined by DTA using the SETSYS Evolution-2400 device was  $T_{sol}\approx1096^{\circ}$ C. This temperature is close to the boundary temperature, at which steel ablation stops; this indicates the mechanism of ablation by eutectic melting.
  - The interaction boundary of the steel specimen had an irregular saw teeth shape. The 'teeth' were enclosed into a spherical segment in the boundary temperature region. Such interaction zone configuration indicates that the interaction did not reach equilibrium conditions in this experiment, also that the interaction does not progress uniformly, it goes in certain directions depending on the irregularities of chemical structure. This is also confirmed by the fact that steel specimen has small locations saturated with U and Zr on the interaction boundary; they were formed by the inter-grain diffusion.
  - The interaction zone had large pores with uneven internal edges. Possible causes of the pore formation could be: hydrogen liberation during the crystallization; liberation of carbon dioxide formed during the oxidation of carbon diffused from steel; shrink cave nature of their formation. The volumetric pore formation in the MC7 interaction zone is also explained by the quick crystallization of this zone due to a low temperature level in it (1030-1150°C) and small volume of this zone, also by the presence of a comparatively thick crust between the corium melt and interaction zone, which restricts the gas evacuation.
- 4. Next to the interaction zone a thermal impact zone was formed in the steel specimen; this zone had the depth of 16 mm from the steel specimen top. It had the Widmanstatten pattern of the low-carbon steel in the zone of high-temperature impact near the interaction boundary and at a lower temperature it transformed into the region with changed grains of the initial ferrite-pearlite structure. The bottom

boundary of the thermal impact practically coincides with the 760°C isotherm calculated at the modeling of the specimen temperature field, which confirms the correctness of numerical model.

- 5. Corium ingot was separated from the interaction zone by the crust. As a result of diffusion processes on the metal-oxide boundary the steel-melt interaction products were partially transported into the oxidic melt. The crystallized corium had a distinct boundary between the corium of initial composition with admixture quantities of iron and chrome and the zone next to the crust, which was strongly enriched with uranium and depleted with zirconium in comparison with the initial corium, it also contained iron in considerable quantities. Closer to the interaction boundary U concentrations in corium grow (from 52.2 mass % to 72) and Zr concentrations decrease (from 29.8 mass % to 11). Fe content falls from the interaction zone boundary to corium bulk (from 14 mass % to 3.5 at the distance of 0.5 mm).
- 6. Experiment MC7 was different from MC6 only in the temperature level of the steel specimen top ( $T_{st.surf.}^{max}$ =1150°C in MC7 and  $T_{st.surf.}^{max}$ =1400°C in MC6). Therefore, the influence of the interface temperature on the interaction can be estimated by the comparison of the main MC6 and MC7 interaction characteristics. The comparison gave the following results:
  - In both tests the steel ablation mechanism is the same: steel dissolution in the Fe/U/Zr/O liquid phase.
  - In MC7 interaction kinetics is less intensive than in MC6.
  - The integral composition of MC7 interaction zone is substantially enriched with uranium and zirconium and inclusions of the U(Zr)Fe<sub>2</sub>(O) grains, and the integral composition of MC6 has the dendrites of doped iron as the primary crystallization phases. Both tests have similar eutectic temperatures and compositions (T<sub>sol</sub>≈1079°C in MC6 and T<sub>sol</sub>≈1096°C in MC7).
  - In accordance with calculations, the MC7 temperature, at which the interaction stops, is 1030-1100°C, and in MC6 it is 1100-1200°C.
  - The MC7 interaction zone has large pores, and the MC6 interaction zone looks solid, though it has a large pore between the interaction zone and crystallized corium.

# 3.4 Interaction of suboxidized corium C-70 with vessel steel in the neutral above-melt atmosphere at $(T_{st,suft}^{max} > 1400^{\circ}C)$ . Test MC8

The objective of MC8 was to study the vessel steel ablation kinetics during its interaction with suboxidized corium C-70 in the oxygen-free atmosphere; temperature of the specimen top  $T_{st.surf.}^{max} \approx 1400^{\circ}$ C, and to determine the influence of melt oxidation degree on the interaction kinetics (other conditions being the same as in Test MC6, its corium composition was C-32) and specify the specimen boundary temperature, at which the ablation having the mechanism of eutectic melting stops.

The startup corium was prepared from the charge having the mass of 1850 g and composition (mass %):  $UO_2$  74%-ZrO<sub>2</sub> 19.7%-Zr 6.3% (of it 150 g was prepared from the ingot produced in the preliminary experiment).

Total duration of the experiment was 13 hours. The experimental procedure, parameters of the furnace and steel specimen corresponded to Test MC6.

Table 5 gives the maximum temperatures in the specimen top center, power into the calorimeter, the average density of heat flux into the specimen and its value in the sighting spot of the ultra-sonic sensor ( $\emptyset$  15 mm) in the beginning of the regime.

			/	
Heat flux	Heat flux	Power into the top	Power into the top	Maximum
density,	density,	calorimeter,	calorimeter,	temperature of
average,	Ø15,	calculated,	experimental	steel surface
$MW/m^2$	MW/m <sup>2</sup>	kW	kW	near the
				specimen axis,
				°C
0.94	1.35	1.85	1.68	1425

Table 5 - Calculated and experimental values of power, heat flux and temperature

MC 8 experimental data and results of the posttest analysis are given in detail in Appendix 2 of the Annual report N 2-833.2-2003. This section summarized the main results.

After the molten pool was produced, the temperature of ~1400°C was reached (1350°C in accordance with thermocouple near the specimen top), from this moment and during 13 hours the vessel steel ablation kinetics at the interaction through the crust was studied in the stabilized temperature conditions. In the end of the test the generator was disconnected and ingot with specimen was cooled in argon.

Throughout the whole test the water-cooled pyrometer shaft was sparged with argon. It was used for continuous measurements of melt surface temperature using the spectral ratio pyrometer RAYTEK, it was 1700 °C. In accordance with experimental conditions during the whole interaction period the crust was present on the molten pool surface.

After cooling the ingot with specimen in argon they were taken out of the crucible. The oxidic corium ingot got separated from the specimen; they were separately enclosed into the epoxy resin, cut along the axis and used for making specimens for the posttest analyses.

The following results were provided by the direct measurement of interaction parameters, including the direct measurement of the steel specimen ablation by the ultra-sonic sounding, posttest physicochemical and metallographic analyses, numerical modeling of the specimen temperature conditions:

- 1. The axial section of the crystallized corium and steel specimen had three main zones: a) crystallized corium ~65 mm high having no vertical difference in the microstructure; b) interaction zone consisting of the crystallized metallic melt shaped as a spherical segment located in the steel specimen body under the top and separated from corium by the ~1 mm-thick crust; c) steel specimen with a thermal impact zone below the interaction zone.
- 2. Similar to Test MC6 the vessel steel ablation has two stages.
  - At the initial stage an insignificant steel corrosion to the depth of ~0.2 mm is observed, by the end of this stage a liquid layer having eutectic composition is formed. The initial stage, which included the incubation period, lasted ~9000 s., and this is much shorter than in experiments MC6 and MC7 (~18000 s.). The shorter incubation period in comparison to MC6 and MC7 can be explained by the reduction of the  $\alpha$ -Zr volume fraction in the crust having C-70 composition, which makes a barrier for the transport of uranium to the interaction boundary. This contributed to a faster (in comparison to MC6, MC7) accumulation of eutectic liquid necessary for the transition to the fast corrosion stage.

- The intensive ablation stage lasted ~30000 s.. At first the specimen ablation rate was ~1.17 mm/h, it gradually fell to 0.32 mm/h and stopped by the moment of heating termination. In accordance with the posttest analysis the final ablation depth in the center of interaction surface was ~6.7 mm, in accordance with direct ultrasonic sounding it was ~7 mm, which shows their good agreement.
- 3. The interaction zone had the form of a crystallized metallic melt and possessed the following specific features.
  - It had the dendrite microstructure, and iron dendrites included chrome, nickel, and manganese, there were eutectic zones between dendrites. There occurred dendrites with very fine structure. Eutectic zones had similar compositions in different interaction locations (~35 mass. % U; ~3.5 mass. % Zr; ~60 mass. % Fe , the rest is Cr, Ni, Mn).
  - Physicochemical processes in the interaction zone took place in the temperature range of 1425-1200°C. At the first stage uranium diffused from the crust, compound UFe<sub>2</sub> was formed on the crust-steel boundary, after that a liquid-phase region was formed at this the interaction process entered a faster phase. The key role of uranium in the eutectic melt formation can be explained by the two following factors: first, as it is evident from the experimental data, the inter-grain diffusion rate considerably exceeds Zr diffusion rate; second, eutectic temperature UFe<sub>2</sub>-Fe 1055°C is considerably lower than eutectic temperature of ZrFe<sub>2</sub>-Fe 1337°C.
  - In accordance with calculations the boundary temperature, at which ablation of steel interacting with C-70 corium stopped, corresponded to isotherm 1200°C.
  - The interaction zone was the crystallized homogeneous monolithic melt, which included cubical crystals of intermetallide  $Zr(U)Fe_2$ ; most of them were located close to the 'interaction zone steel' boundary. The interaction zone and corium ingot were separated by a large pore space. Chains of gas pores are found along the boundary with steel, in the center the interaction zone is separated from steel.
- 4. The interaction zone was separated from corium ingot by the crust; its composition is considerably different from the initial. The crust microstructure is layered, it consists of alternating layers of the U(Zr)O<sub>2</sub> solid solution and layers including three phases: iron-based, intermetallide Zr(U)Fe<sub>2</sub>-based and intermetallide U(Zr)Fe<sub>2</sub>-based. In accordance with EDX the crust contains 17 mass % of steel components. In some parts the Zr(U)Fe<sub>2</sub>-based phase is present in the matrix of the U(Zr)O<sub>2-x</sub> solid solution as drop-like inclusions. In such regions the content of iron is ~12 mass %.
- 5. Under the interaction zone to the depth of 3 mm steel microstructure consists of large ferrite formations having carbon content of 0,02 %. Multiple pores are found at the 2 mm depth from the boundary. Further to the depth of 25 mm from the initial surface of the specimen top the metal has a ferrite-pearlite microstructure, which was formed during the cooling of steel heated above the critical point from the austenite condition. Below 25 mm and temperature ~750°C the initial pearlite structure did not undergo changes.

### 3.5 Interaction of molten steel and suboxidized corium with vessel steel in the neutral above-melt atmosphere. Test MC9

Experiment MC9 studied the ablation of vessel steel during its interaction with molten metal formed in the molten pool bottom as a result of interaction between molten corium and stainless steel in the oxygen-free atmosphere (argon).

Initial corium was produced from the charge having the mass of 1800 g and composition  $76.2\% UO_2$ - $9.3\% ZrO_2$ -14.5% Zr (mass.%). After the production of molten pool 200 g of stainless steel was added into it. The melt – steel interaction lasted 12 hours. Table 6 gives maximum temperatures in the top center, power into the calorimeter, average density of heat flux into the specimen when the interaction boundary reaches the final position.

	Heat flux density, MW/m <sup>2</sup>	Power into the top calorimeter, calculated, kW	Power into the top calorimeter, experimental kW	Maximum temperature of steel surface near the specimen axis, °C
-	1.1	2.19	2.03	1440*

Table 6 - Calculated and experimental values of power, heat flux and temperature

\* - interaction start.

The experimental data of MC9 and summarized results of all tests with suboxidized coria are given in Appendix 1 of the current report.

After the temperature of the specimen top was stabilized at  $\sim 1400^{\circ}$ C (it had a drop when the oxides of stainless steel were added into the melt) the melt-steel system was subjected to the long-term exposition. The maximum depth of steel surface ablation was  $\sim 15$  mm.

The following results were provided by direct measurements and posttest analysis.

- During the whole test the steel specimen was in the interaction with metallic melt formed as a result of repartitioning of components between the C-30 melt and stainless steel in the molten pool bottom.
- By the end of the test a high temperature caused the volatilization of metallic components, mostly Fe; for this reason the corium ingot did not have a metallic component.
- Specimen corrosion started immediately after the relocation of metallic part of the melt to the pool bottom.
- By the end of the test the corrosion rate dropped from the maximum value to zero.
- The oxidation degree of corium increased considerably from the initial C-30; the final ingot was C-45 due to the transport of a large U and Zr mass to the interaction zone.
- The average composition of the interaction zone: 33.3 % U- 8.3% Zr-53%Fe (mass.%) with a small content of Gr, Ni, O,...
- The zone of thermal impact extended to the depth of 26 mm from the specimen top. In this zone the initial pearlite structure was replaced by the pearlite-ferrite. Carbon was depleted in steel near the interaction boundary.

The integrated analysis of MC6...MC9 has established the following:

- The interaction of suboxidized corium melt with cooled vessel steel is similar to the interaction between suboxidized corium melt and molten steel: it results in the establishment of a system consisting of oxidic and metallic parts. In the thermogradient conditions of MC series the metallic part of the system is the mushy zone U-Zr-Fe(Cr,Ni)-O.
- Vessel steel corrosion is caused by its eutectic melting (dissolution) in the interaction zone.
- The boundary temperature, at which corrosion stops, is the solidus temperature of the interaction zone material. For MC series and realistic compositions of the interaction zone in the VVER severe accident conditions the minimum temperature, at which the vessel steel corrosion stops, is 1090°C.
- In terms of minimum temperature for the vessel steel corrosion termination its interaction with the metallic part of the melt (Test MC9) is not different from the steel interaction with suboxidized melt (Tests MC6...8).
- Steel corrosion goes in three stages: incubation period solid phase interaction/diffusion of crust components, suboxidized oxides and steel components until the liquid phase is formed; transition period continuation of the liquid phase formation and its penetration into the crust; fast corrosion period steel dissolution in the liquid phase of the interaction zone.
- Duration of the incubation period in case of interaction with C-30 melt is ~16000 s.; with C-70 melt ~8000 s., in case of the metallic melt it is negligibly small.
- A the stage of fast ablation the corrosion rate is described by the following correlation

 $\frac{dh}{dt} = 0.46 \cdot 10^{-4} \sqrt{T_{int} - T_B}, \text{ mm/s},$ 

 $1090 \le T_{int} \le 1440^{\circ}C$ ;  $T_{int}$  – temperature on the corrosion front;

 $T_{B}$  – boundary temperature of corrosion – solidus temperature of the interaction zone composition.

The derived correlation adequately describes the results of all MC6...MC9 experiments.

# 3.6 Interaction of corium C-100 with vessel steel in steam. Test MC10

The studies were aimed at determining the characteristics of vessel steel corrosion in the experimental conditions at different temperatures on the interaction interface and their comparison with steel corrosion at the interaction with corium containing both uranium/ zirconium and iron oxides.

Molten corium was produced from the charge having the mass of ~1700 g and composition  $70\% UO_{2+x}$ - $30\% ZrO_2$  (mass.%) (C-100).

Interaction with the steel specimen was studied at three temperature plateaus; the total test duration was more than 5 hours.

Table 7 gives temperatures and heat flux densities on the corrosion front, also power into the top calorimeter.

	Temperature Heat flux density		Power into the top calorimeter, kW		
Regime №	corrosion front, °C	front, MW/m <sup>2</sup>	calculated	experiment	
1	1035	0.95	1.37	1.38	
2	1185	1.05	1.56	1.58	
3	1235	1.10	1.70	1.73	

 Table 7 – Temperature and thermal parameters of the specimen

Results of MC 10 are presented in detail in Appendix 2 to this report.

This section summarizes the main results.

Steam atmosphere caused increased aerosol generation. Beside that steam excluded the possibility of corium surface monitoring and its temperature measurements. They were performed only during the short-term replacement of steam with air.

The steel specimen corrosion rate was measured at each temperature plateau using ultrasonic sounding. The average rate for regimes  $N \ge N \ge 1-3$  given in Table 7 was 0.55, 1.07 and 2.07 mm/h respectively; it increased as the temperature grew.

At all established regimes the specimen temperature fluctuations were registered, they can be attributed to the oscillations of electrical parameters in the induction system.

At the attempt to reach the 4<sup>th</sup> temperature plateau the specimen surface melting started, and the experiment was terminated. The posttest analysis showed that melting took place on the whole specimen surface, and this distorted the corrosion layer structure and composition.

At its extraction from the crucible the corium ingot broke into several parts, for this reason its fragments were used in the posttest analysis.

The following results were provided by direct measurements and posttest analysis.

- Steel corrosion is not time-dependent, if the corrosion front temperature is constant.
- In comparison with experiments in air (MC2) the corrosion rate of steel is much higher at its interaction with corium containing iron oxides (MC10), other conditions being comparable.
- For the temperature range  $T_s \le 1230^{\circ}C$  the corrosion rate correlation is

$$\frac{W}{q} = 5870 \exp\left(-\frac{E_a}{RT_S}\right)$$

and for the temperature range  $1340>T_S \ge 1230^{\circ}C$  it is

$$\frac{W}{q} = -13.3 + \frac{1.59 \cdot 10^3}{1613 - T_S},$$

where W – corrosion rate, mm/h;

q – heat flux density,  $MW/m^2$ ;

 $E_a=1.02 \cdot 10^5$  J/mol. – activation energy;

R=8.314 J/(mol.·K) – universal gas constant;

 $T_S$  – temperature on the corrosion front, K.

# 3.7 Interaction of corium melt $UO_{2+x}$ -ZrO<sub>2</sub>-FeO<sub>y</sub> with vessel steel in steam. Test MC11

The experiment was aimed at determining vessel steel corrosion characteristics in the experimental conditions at different temperatures on the interaction interface, and their further comparison with data on steel corrosion in air and corium C-100 – steel interaction.

Initial corium was prepared from the charge having the mass of ~1440 g and composition  $57.54\% UO_{2+x}-24.69\% ZrO_2-17.77 FeO_y$  (mass.%).

Corium interaction with steel specimen was studied at 4 temperature plateaus. The total duration of the experiment was  $\sim$  4 hours.

Table 8 gives temperatures, heat flux densities, power into the top calorimeter of the specimen.

Regime	Temperature of the corrosion front, °C		Heat flux density on the corrosion front, MW/m <sup>2</sup>			Power into the top calorimeter, kW				
N⊵	start	final	average	start	final	average	sta	rt	fin	al
							calc.	exp.	calc.	exp.
1			950			0.99				
2	1060	1040	1050	1.18	1.15	1.16	0.99	1.24	0.98	1.11
3	1125	1135	1130	1.25	1.21	1.23	1.07	1.13	1.17	1.20
4	1220	1180	1200	1.34	1.24	1.29	1.26	1.27	1.28	1.30

 Table 8 – Temperature and thermal parameters of the specimen

Results of Test MC11 are given in detail in Appendix 2 of the current report.

This section presents the main results.

Corrosion rates of each temperature plateau (regimes  $N \ge N \ge 1...4$ , Table 8) measured by ultrasonic sounding were 0.17; 0.25; 2.8; 7.8 mm/h respectively. The final specimen corrosion depth was ~ 3.7 mm.

As the heat flux into the steel specimen top was increased, the specimen temperature conditions became more and more unstable. In the end of the 4<sup>th</sup> plateau a spontaneous uncontrollable specimen temperature growth took place. It was stabilized by reducing power deposition in the melt. Following that the experiment was terminated. This behavior was likely to be caused by the instability of oxidic crust and crust on the specimen surface.

The posttest analysis showed that corium ingot contained many pores, some of them were large, and that made it different from ingots produced during similar tests in air (MC2).

The corrosion layer on the steel surface consisted of extremely thin layer of iron oxides lying under a layer of compounds mostly based on U. This structure probably corresponds to a short period of ablation after the corrosion layer was broken in the end of the 4<sup>th</sup> plateau.

The following basic results were provided by direct measurements, MC11 posttest analysis, and the integrated analysis of all tests conducted in the oxidizing atmosphere:

- In the oxidizing atmosphere above the melt steel corrosion can have two kinetically different regimes. The change of corrosion regime takes place when the external surface of corrosion layer reaches  $T_{\delta}=1340^{\circ}C$  the eutectic temperature of  $UO_{2+x}$ -ZrO<sub>2</sub>-FeO<sub>y</sub> system. For corium containing iron oxides this condition is met at  $T_{s}=1050^{\circ}C$  temperature of the specimen surface.
- At T<sub>s</sub>≤1340°C and assumed constant temperature gradient in the corrosion layer the experimental data are very well described by the Tamman equation. In this case data provided by experiments MC1, MC2 (in air) and MC11 (steam) are generalized by the same correlation.

$$\frac{W}{q} = 2230 \exp\left(-\frac{E_a}{RT_S}\right)$$
, mm/h,

at T<sub>s</sub>≤1050°C.

• At  $T_s$  growth above 1050°C the eutectic melting starts on the corrosion layer boundary, and temperature gradient on the corrosion layer is reduced. For the temperature range 1340>T<sub>s</sub>≥1050°C the experimental data are described by correlation

$$\frac{W}{q} = -5.28 + \frac{1.59 \cdot 10^3}{1613 - T_S}$$
, mm/h.

In the temperature range T<sub>δ</sub>≤1340°C during the interaction with corium C-100 the corrosion rate is ~ 2.5 times higher than at the interaction with corium containing iron oxides. A possible reason for this is the smaller thickness of the corrosion layer due to a more intensive diffusion of iron oxides into corium in the conditions of experiments with corium C-100.

#### 4 CONCLUSIONS

The following has been established by the experimental studies of the interaction between uranium-bearing corium melt and cooled VVER vessel steel, the analysis and integration of produced data.

- 1. In case of the in-vessel melt retention the vessel wall thinning can be caused not only by the steel melting, but also by its corrosion during the physicochemical interaction with corium melt.
- 2. There are two types of corrosion, which depend on the chemical potential of the melt:
  - In case of the superstoichiometric melt UO<sub>2+x</sub>-ZrO<sub>2</sub>-(FeO<sub>y</sub>) and the oxidizing above-melt atmosphere steel corrosion is caused by its oxidation; the process kinetics is controlled by the diffusion resistance of corrosion layer on the steel surface;
  - In case of the pre-stoichiometric (suboxidized) melt UO<sub>2</sub>-ZrO<sub>2</sub>-Zr-(SS) in the neutral above-melt atmosphere steel corrosion follows the mechanism of eutectic melting (dissolution) in the liquid-solid interaction zone formed on its surface U-Zr-Fe(Cr, Ni, ...)-O;
  - Other conditions being the same, the minimum effect of the physicochemical interaction corresponds to the stoichiometric melt UO<sub>2</sub>-ZrO<sub>2</sub> (in the neutral above-melt atmosphere).
- During the interaction of steel with molten superstoichiometric corium two corrosion regimes of different kinetics are possible, one replaces another when the corrosion layer boundary reaches T<sub>δ</sub>=1340°C – eutectic temperature of the UO<sub>2+x</sub>-ZrO<sub>2</sub>-FeO<sub>y</sub> system. Foe corium UO<sub>2+x</sub>-ZrO<sub>2</sub> this condition corresponds to the steel surface temperature of 1230°C, and for corium UO<sub>2+x</sub>-ZrO<sub>2</sub>-FeO<sub>y</sub> – 1050°C.
- 4. Irrespective of the type of oxidizing atmosphere (steam or air) the experimental data on the corrosion rate for the  $T_{\delta} \leq 1340^{\circ}$ C temperature region can be described by a single correlation

$$\frac{W}{q} = A \exp\left(-\frac{E_a}{RT_S}\right),$$

where W – corrosion rate, mm/h;

q – heat flux density, MW/m<sup>2</sup>;

 $E_a - 1.02 \cdot 10^5$ , J/Mol. – activation energy;

R – 8.314 J/(mol.·K) – universal gas constant;

 $T_S$  – temperature of the steel surface, K;

A – coefficient; for corium UO<sub>2+x</sub>-ZrO<sub>2</sub> – A=5870; for corium UO<sub>2+x</sub>-ZrO<sub>2</sub>-FeO<sub>y</sub> – A=2330.

In the zone of eutectic melting of iron oxides on the external surface of the corrosion layer:

For corium UO<sub>2+x</sub>-ZrO<sub>2</sub> at 1340>T<sub>S</sub> $\geq$ 1230°C

$$\frac{W}{q} = -13.3 + \frac{1.59 \cdot 10^3}{1613 - T_S};$$

For corium UO<sub>2+x</sub>-ZrO<sub>2</sub>-FeO<sub>y</sub> at 1340>T<sub>S</sub>>1050°C

$$\frac{W}{q} = 5.28 + \frac{1.59 \cdot 10^3}{1613 - T_S}$$

- 5. Corrosion rate for corium  $UO_{2+x}$ -ZrO<sub>2</sub> in the temperature region  $T_{\delta} \leq 1340^{\circ}C$  is approximately 2.5 times higher than for corium  $UO_{2+x}$ -ZrO<sub>2</sub>-FeO<sub>y</sub>. It can be explained by the difference in the thickness of corrosion layer due to different diffusion rates of iron oxides from the corrosion layer into corium.
- 6. At the interaction of cooled vessel steel and suboxidized corium melt (in the neutral atmosphere) the repartitioning of components between oxidic melt and steel results in the formation of the mushy interaction zone on their boundary. This process is similar to the component repartitioning between the melts of suboxidized oxides and steel studied within the OECD MASCA program. Quantitative difference in composition and kinetics is explained by the thermal gradient conditions of the METCOR series.
- During the interaction of molten steel and suboxidized molten corium UO<sub>2</sub>-ZrO<sub>2</sub>-Zr-(SS) the minimum temperature, at which steel corrosion stops, is equal to the minimum solidus temperature of the interaction zone material U-Zr-Fe(Cr, Ni, ...)-O, it is 1090°C.
- Three vessel steel corrosion periods are distinguished: incubation, transition and fast corrosion. The duration of incubation period depends on the corium oxidation degree, it decreases from C-30 to C-70 from ~ 16000 to ~ 8000 s.
- 9. The empirical correlation for calculating the fast corrosion rate has the following formulation

W = 
$$0.46 \cdot 10^{-4} \sqrt{T_{int} - T_B}$$
,

where W – corrosion rate, mm/s.;

 $T_{int}$  – temperature in the corrosion front, °C;

 $T_B$  – solidus temperature of the interaction zone material, °C.

# 5 LIST OF PUBLICATIONS

# 5.1 Papers

N⁰	Title	Abstract
1	Corrosion of Vessel Steel During its Interaction with Molten Corium. – Part 1: Experimental. S.V. Bechta, V.B. Khabensky, S.A. Vitol, E.V. Krushinov, V.S. Grahovsky, A.P. Martinov, V.V. Martinov, G. Fieg, W. Tromm, D. Bottomley, H. Tuomisto	Results of experiments within the ISTC METCOR ( $1^{st}$ Phase) are presented. Interaction of molten corium $UO_{2+x}$ -ZrO <sub>2</sub> -FeOy and cooled vessel steel specimens in the neutral atmosphere and air is studied. The range of temperatures on the specimen surface 720-1200°C. Data on corrosion kinetics are provided. Posttest analysis has determined the composition and structure of crystallized corium ingots, corrosion zone and steel specimen.
	Nuclear Engineering and Design. 2006 (in print)	
2	Corrosion of Vessel Steel During its Interaction with Molten Corium. – Part 2: Model Development S.V. Bechta, V.B. Khabensky, S.A. Vitol, E.V. Krushinov, V.S. Grahovsky, D. B. Lopukh, I.V. Kulagin, V.V. Gusarov, A.P. Martinov, V.V. Martinov, G. Fieg, W. Tromm, D. Bottomley, H. Tuomisto Nuclear Engineering and design. 2006 (in print)	The experimental studies are used for developing a model of vessel steel corrosion. Steel ablation is caused by its surface oxidation. Corrosion kinetics is controlled by the diffusion resistance of corrosion layer. In the neutral atmosphere the corrosion rate gradually slows down. In air above the melt the corrosion rate is constant. Correlations are proposed, which generalize the produced experimental data. The steel vessel corrosion calculations are applied to the in-vessel melt retention conditions.

# 5.2 Reports

N⁰	Code	Title	Abstract
1		Oxidation of the suboxidized corium melt by oxygen from air. Test Pr1-MC5	The modernized 'Rasplav-3' test facility provides the possibility of melt superheating and its maintaining in the above-liquidus temperature state in a wide range of oxygen potentials - from C-30 to C-100. This enables to conduct studies on the interaction of suboxidized melt and vessel steel.
2		Interaction of molten corium with reactor vessel steel in the neutral above-melt atmosphere. Test MC5/03	Interaction of corium C-100 with a vessel steel specimen is studied in argon, i.e. in the conditions of minimum chemical interaction. In the experiments the maximum temperature on the specimen surface is varied from 1070 to 1435°C. The maximum corrosion depth is 0.2 mm.
3		Interaction of suboxidized molten corium with reactor vessel steel in the neutral above-melt atmosphere. Test MC6/03	Interaction of corium C-30 with a vessel steel specimen is studied in argon. Maximum temperature on the specimen surface is ~ 1400°C. Experiment duration -10 hours. Corrosion kinetics is characterized by a long incubation period with a low rate and small depth of corrosion; and a fast corrosion period when the rate decreases in time. The maximum corrosion depth is ~ 6.7  mm. The final position of the corrosion front corresponds to 1120-1200°C in the specimen. The interaction zone consists of the U-Zr-Fe(Cr, Ni,) – O system, in which the eutectic melting (dissolution) of steel takes place.
4		Interaction of suboxidized molten corium (C-32) with reactor vessel steel in the neutral above-melt atmosphere ( $T_{st.surf.}^{max} \approx$ 1150°C). Test MC7/04	The incubation period characteristics are the same as in MC6. The fast corrosion period has a lower initial rate and smaller depth, which is $\sim 2.9$ mm. The final position of the corrosion front corresponds to 1030-1100°C in the specimen. The interaction zone is porous. It has the same composition as in MC6, and the structure has a larger volumetric fraction (80%) of the eutectic phase.
5		Interaction of suboxidized molten corium (C-70) with reactor vessel steel in the neutral above-melt atmosphere $(T_{st.surf.}^{max} \approx$	The intermediate period is identified between the incubation and fast corrosion periods, during it the corrosion rate increases to the maximum. The incubation period is ~ 2 times shorter in comparison with MC6,7, this correlates with the $\alpha$ -Zr reduction on the interaction boundary, which serves

	1400°C). Test MC8/04	as a barrier to U diffusion from corium to the interaction zone and retards the formation of liquid eutectic phase, in which steel is dissolved. The maximum corrosion depth is $\sim 6.7$ mm during 13 hours of the test. Specimen temperature on the final corrosion front boundary - 1200°C.
6	Hydrodynamics and heat exchange of corium melt in the IMCC	The results of MC4 and MC5 are used for the verification of numerical code DYMELT used for modeling free convection in corium melt. The modeling has to be made in order to determine the radial distribution of melt – steel heat flux density. This is used for the subsequent calculation of the specimen temperature conditions. The calculation is a component of the posttest analysis and it enables to determine temperature and thermal conditions, in which the melt – specimen interaction take place.
7	Interaction of molten steel and suboxidized corium with vessel steel in the neutra above-melt atmosphere. Test MC9/04	The interaction of specimen with the metallic part of corium melt, which is produced by the preceding interaction of melt C-30 and stainless steel, is studied. The maximum specimen surface temperature ~ 1400°C, duration of experiment ~ 12 hours. The maximum corrosion depth is ~ 15 mm. Specimen temperature on the final boundary of corrosion front 1060÷1100°C. The analysis of all experiments with suboxidized corium determines the boundary temperature, at which corrosion stops - 1090°C; a correlation for calculating corrosion kinetics is derived.
8	Interaction of molten corium and vessel stee in the above-melt steam atmosphere. Tests MC10/05 и MC11/05	Vessel steel corrosion at its interaction with melt $UO_{2+x}$ -ZrO <sub>2</sub> (C-100) and $UO_{2+x}$ -ZrO <sub>2</sub> -FeO <sub>y</sub> in steam and 950-1235°C temperature on the specimen surface is studied. The comparison with corrosion in air proves that corrosion kinetics is not influenced by the composition of oxidizing atmosphere. In case of C-100 corium corrosion goes 2.5 times faster than in corium containing iron oxides. Two kinetically different corrosion mechanisms are identified depending on the presence or absence of eutectic melting of iron oxides on the external surface of corrosion layer. Correlations describing steel corrosion rates in the oxidizing atmosphere are derived.

### 6 PRESENTATIONS AT CONFERENCES AND MEETINGS

# 6.1 Conferences

N⁰	Conference	Name	Abstract
1	Proceedings of ICAPP'04 Pittsburgh, PA USA, June 13-17, 2004. Paper 4114	<ul> <li>New Experimental Results on the Interaction of Molten Corium with Reactor Vessel Steel.</li> <li>S.V. Bechta, V.B. Khabensky, V.S. Granovsky, E.V. Krushinov, S.A. Vitol, V.V. Gusarov, V.I. Almiashev, D.B. Lopukh, W. Tromm, D. Bottomley, M. Fischer, G. Cognet, O. Kymalainen</li> </ul>	Results of first two tests within the ISTC METCOR2 are analyzed. Interaction of vessel steel specimens and corium C-100 (Test MC5) and C-30 (Test MC6) has been studied in the neutral atmosphere. Results of Test MC5 correspond to the conditions of minimum chemical interaction. Corrosion depth does not exceed 0.2 mm. In MC6 with suboxidized corium at the max. temperature on the specimen surface ~ 1400°C and test duration ~ 10 hours the depth of corrosion is ~ 6.7 mm. The interaction is caused by eutectic melting (dissolution) of steel in the formed mushy zone having the U-Zr-Fe-O composition.
2	Proceedings of ICAPP'06 Reno, NV USA, June 4-8, 2006. Paper 6054	<ul> <li>Experimental Study of Interaction Between Suboxidized Corium and Reactor Vessel Steel.</li> <li>S.V. Bechta, V.B. Khabensky, V.S. Granovsky, E.V. Krushinov, S.A. Vitol, V.V. Gusarov, V.I. Almiashev, D.B. Lopukh, W. Tromm, D. Bottomley, M. Fischer, P. Piluso, A. Miasoedov, E. Altstadt, H.G. Willschutz, F. Fichot</li> </ul>	Results of all experiments on the interaction of suboxidized corium and vessel steel within the ISTC METCOR2 are analyzed. Corium oxidation degree and maximum temperature of steel specimens are varied. In one of the tests stainless steel is added to corium melt. It is determined that minimum temperature on the corrosion front corresponding to the steel ablation depth is 1090°C. The empirical correlation for calculating corrosion kinetics has been developed. Similarity of the interaction zone formation in METCOR tests and metallic melt formation in the OECD MASCA program is analyzed.

# 6.2 Meetings

N⁰	Meeting	Name	Abstract
1	1 The first European Review Meeting on Severe Accident Research (ERMSAR-2005) Aix-en-Provence, France, 14-16 November 2005	CORPHAD and METCOR. ISTC projects	Two quantitatively different corrosion mechanisms of vessel steel at its interaction with corium melt are identified.
		S.V. Bechta, V.B. Khabensky, V.S. Granovsky, E.V. Krushinov, S.A. Vitol, V.V. Gusarov, V.I. Almiashev, L.P. Mezentseva, Yu. B. Petrov, D.B. Lopukh, W. Tromm, D. Bottomley, M. Fischer, M. Barrachin, P. Piluso, E. Altstadt, F. Fichot, S. Hellman, F. Defoort	In the oxidizing above-melt atmosphere the corrosion is caused by steel oxidation. Its kinetics is controlled by the diffusion resistance of corrosion layer. For suboxidized corium in the neutral above-melt atmosphere corrosion is caused by the eutectic melting of steel in the mushy zone having the composition of U-Zr-Fe-O. The minimum corrosion intensity corresponds to the stoichiometric corium $UO_2$ -Zr $O_2$ at the neutral above-melt atmosphere.
2	2 2 <sup>nd</sup> meeting with collaborators on the METCOR 2 Project Aix-en-Provence, France, 29 January 2003	S.V. Bechta METCOR, 2 <sup>nd</sup> Phase: preparation of technical facilities and results of Test Pr1-MC5	The HF generator has been modernized to produce higher-frequency modes. 'Rasplav-3' test facility has improved electric characteristics to provide higher power capacity. The furnace design is improved. In Pr1-MC5 the possibility to melt corium having a wide range of oxidation degrees is tested.
		S.V. Bechta Numerical modeling of molten pool thermohydrodynamics	The mathematical model and method of numerical solution of thermohydrodynamical equations is presented. Examples of calculations of molten pool conditions are given.

3	3 <sup>rd</sup> meeting with	S.V. Bechta	Methodology of tests on the interaction with coria C-100 and C-30 in the
	collaborators on the	Experimental procedure and results of	neutral atmosphere is presented, also an auxiliary experiment on the

	METCOR 2 Project St. Petersburg,	measurements within Tests MC5, Pr-MC6, MC6	evaluation of heat fluxes into the pool bottom and charge preparation for MC6.
	Russia		Data on corrosion kinetics and depth are presented.
	16 September 2003	V.S. Granovsky	Methodology and results of numerical modeling of the specimen temperature conditions in the process of interaction with molten corium is discussed.
		Temperature calculations using tests MC5 and MC6	
		V.V. Gusarov	SEM/EDX results are used to explain the mechanism of specimen - melt
		Results of physicochemical and metallographic analyses of tests MC5 and MC6	interaction following the model of eutectic melting of steel: Test MC5 in the $UO_2$ -ZrO <sub>2</sub> -FeO-Fe system, and MC6 – in the U-Zr-Fe-O system
		A. Yu. Petchenkov	A numerical model of electromagnetic system, method of solution and
		Calculation of power deposition in the melt, specimen and induction furnace components	methodology for calculating power deposition in molten corium are presented.
		S.A. Smirnov	Methodology and results of the DYMELT numerical code verification using
		Free convection calculations for Test MC4	MC4 experimental data are presented
		J.M. Bonnet	The boundary layer model and finite-element melt thermohydrodynamics
		Free convection calculations for Test MC4	model are presented. Comparison of calculations and MC4 experimental data is used for the analysis of possible influence of uncertainties on the numerical modeling accuracy.

4	4 <sup>th</sup> meeting with	S.V. Bechta	Results of auxiliary tests and MC7 are presented. Measured data on corrosion
	collaborators on the	Results of tests Pr-MC7 and MC7; their	kinetics and depth, and results of the posttest analysis are used for explaining

	METCOR 2 Project Paris, France 10 February 2004	posttest analyses	the interaction zone irregularity, its porosity and unsteadiness of the specimen corrosion rate in time.
		D. Knoche, D. Bottomley Experiments with irradiated materials	The experimental facilities of the Institute of Transuranium Elements (Karlsruhe), their capacities in studying characteristics and properties of irradiated materials are presented. Results of studies on the separation of high-temperature fuel compositions (irradiated $UO_2$ and $MOX$ ) are reported. Results of optical microscopy and phase analysis are given. Comparison with corresponding unirradiated fuel compositions is made.
5	5 <sup>th</sup> meeting with collaborators on the METCOR 2 Project Dimitrovgrad,	S.V. Bechta MC8 experiment and posttest analysis	Test results are presented. Final temperature on the corrosion front is evaluated as 1200°C. The interaction zone composition: U-Zr-Fe-O. A hypothesis on the causes of the incubation period reduction is put forward. The transition period in the specimen corrosion is identified.
	Russia 14 September 2004	V.S. Granovsky Comparative analysis of MC6, MC7, MC8	Analogies of phenomena studied within the OECD MASCA program and METCOR2 project are discussed. An approach is proposed for the integration of data on steel corrosion kinetics during its interaction with suboxidized corium melt.

6	6 <sup>th</sup> meeting with collaborators on the METCOR 2 Project St. Petersburg, Russia	V.S. Granovsky Results of Tests MC9, MC10	Results of the final test with suboxidized corium are presented. The absence of incubation period of steel corrosion is noted; it is explained by the specimen interaction with metallic melt, which is formed in the C-30-SS system. Corrosion kinetics has no qualitative difference with previous tests. Results of experiments with corium C-100 in steam are presented. Specimen temperature instability is noted. Comparison with experiments in air and melt
	12 July 2005		containing iron oxides is made.
		S. A. Smirnov Thermohydrodynamic modeling of the metal-oxidic melt	Calculations of the two-liquid molten pool are presented. The influence of Lorenz forces on the configuration of metallic part and peculiarities of the heat flux distribution in the specimen top are shown.
		E. Alstadt Application of METCOR results in the finite-element models of the IVR	The integrated thermo-mechanical model of the vessel in the IVR conditions is shown. Stress-strain and creep conditions are treated additively. In the considered conditions additional corrosion influence does not result in the vessel failure.
		V.B. Khabensky Continuation of research on the subjects of the Project with additional funding	<ul> <li>Proposed directions of investigation are analyzed:</li> <li>Vessel steel corrosion at its interaction with corium in steam, steel melting is reached.</li> <li>Vessel steel corrosion at its interaction with suboxidized melt, vertical position of the interaction interface.</li> <li>Corrosion during the changes in the oxidizing potential of the above-melt atmosphere.</li> <li>Interaction with vessel steel of European reactors.</li> <li>Investigation of steel properties in the interaction zone.</li> </ul>

Currently a publication is prepared on the results of experimental studies of the interaction between corium melt and vessel steel in the oxidizing above-melt atmosphere.

Project Manager, Professor

V.B. Khabensky

Director of NITI, Professor

V.A. Vasilenko

# Appendix 1

Investigation of molten steel and suboxidized molten corium (C30) interaction with vessel steel in a neutral atmosphere ( $T_{st.surf.}^{max}$ »1400°C). Test MC9

# Appendix 2

Investigation of Molten Corium Interaction with Vessel Steel in Steam Above the Melt Tests MC10/05 and MC11/05