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### **Annual technical report**

### Project 833.2-2003

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

### (METCOR 2)

Phase 2. First year (01.01.2003 – 31.12.2003)

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### ISTC

### Annual technical report Project 833.2-2003

1. Project title	Investigation of corium melt interaction with NPP reactor vessel steel (METCOR 2)			
2. Annual report No	First year progress report № 1-833.2-2003			
3. Contractor	Aleksandrov Research Institute of Technologies of the RF Ministry for Atomic Energy Russia, 188540, Sosnovy Bor of Leningrad Oblast, NITI			
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5. Date of project start	01 January, 2003			

Duration

36 months

### 6. Project objective and expected results

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

In order to justify the concept of the in-vessel molten core retention it is necessary to make a numeric modeling of thermohydraulic and physico-chemical phenomena taking place at the interaction between the molten pool and inner surface of the reactor vessel cooled from the outside with water.

Previously in the concepts of in-vessel retention developed for the medium-capacity reactors (VVER-440, AP-600 and VVER-640) only thermohydraulic aspects of molten pool – reactor vessel interaction have been taken into account. The phenomena of physico-chemical interaction of oxidic melt and vessel steel are very complex, and they have not been treated with the attention they deserve. In order to refine numeric codes modeling these phenomena it is necessary to carry out experimental studies and use their outputs for code improvement. In order to determine conditions adequately representing the realistic ones let us consider possible stages of oxidic melt development.

At the early stages of molten pool formation on the bottom of a reactor vessel the oxidic melt has a composition of U-Zr-O, in which the content of suboxidized Zr may have a wide range of variations depending on the accident progression scenario. At this stage it is necessary to know quantitative characteristics of the interaction between the vessel steel and suboxidized / oxidized to stoichiometry  $UO_2$  and  $ZrO_2$  of the molten pool under an atmosphere having a low

oxygen potential. Corium oxidation degree can be described as 
$$C = \left(\frac{M_{ZrO_2}}{M_{Zr} + M_{ZrO_2}}\right) \cdot 100$$
, where

 $M_{Zr}$  and  $M_{ZrO_2}$  - molar masses of , correspondingly, suboxidized Zr and oxidic Zr (ZrO<sub>2</sub>), if we reduce melt composition to  $UO_2 + ZrO_2 + Zr$ . A possible range of melt compositions in the reactor corresponds to the changes of the oxidation factor approximately from C 30 to C 100.

At the next stage the steel from the in-vessel instrumentation and structures starts to relocate into the molten pool. If the original degree of corium oxidation is high ( $\geq$  C 70), this will result in the following: oxidic corium C $\rightarrow$ 100 will reside in the lower part of the molten pool, and its upper layers will contain molten steel with relatively small quantities of U and Zr. If the melt oxidation degree is low (C < 50), then, as it follows MASCA experimental data and theoretical evaluations, due to U and Zr extraction by the molten steel, the pool can have 3-layered structure: molten steel enriched with U and Zr in the bottom; oxidic corium C $\rightarrow$ 100 in the middle,; and molten steel with small amounts of U and Zr in the top layer. At this stage oxidic corium C $\rightarrow$ 100 interacts with reactor wall at a low oxygen potential in the system. As it has been shown by J.M. Seiler, the transient time from molten pool formation to equilibrium conditions can be several hours. Therefore, the transient from an early stage, when a molten pool having a high concentration of Zr and oxidic composition C < 50 changes to composition C $\rightarrow$ 100, does not exceed several hours under different accident development scenarios.

At the next stage of molten pool existence its top layer gets oxidized in the steam-gas atmosphere, oxygen is transported into the underlying layers of oxides and steel with U and Zr dissolved in them; most active reducers get oxidized and oxidic melt dissolves metal oxides, which have been formed in the top and middle layers of the melt. At this stage the interaction between oxidic melt UO<sub>2</sub>/ZrO<sub>2</sub>/FeO and reactor vessel steel takes place at a high oxygen potential in the system (steam-gas atmosphere above the melt). The transient time to the last stage of molten pool formation, which is determined by the kinetics of mentioned oxidation processes, is rather long and requires experimental clarification.

The interaction of oxidic melt  $UO_2/ZrO_2/FeO(Fe_3O_4)$  with reactor vessel steel in air and neutral (nitrogen) atmosphere above the melt has been experimentally studied in the 1<sup>st</sup> Phase of ISTC METCOR Project.

The project enabled to determine:

1. Quantitative characteristics of vessel steel ablation depending on

• oxygen potential of the melt, which depends on melt composition and atmosphere,

- steel temperature on the interaction interface.
- 2. Microstructure and elemental composition of the intermediate zone between corium and steel specimen for developing the model of uranium-bearing oxidic melt vessel steel interaction process.
- 3. Structural characteristics of vessel steel after its interaction with corium.

The Project results can be used:

- to improve numeric models describing the interaction of uranium-bearing oxidic melt and reactor vessel steel;
- to justify concepts and improve safety of operated and designed reactors VVER, PWR and BWR.

### 7. Scientific approach and techniques used

The investigations have been carried out on the "Rasplav-3" test facility, in which corium melt is produced by the induction melting in the cold crucible (IMCC) technology. Its schematics is presented in Appendices 1, 2, 3. The test facility was developed on the basis of previously used "Rasplav-2" facility. Prior to the current investigations "Rasplav-3" underwent modernization, which improved its operational characteristics and capabilities (Appendices 1, 2).

Steel specimens for the interaction investigations were manufactured from a part of VVER-1000 reactor vessel (steel 15Kh2NMFA-A), which had K-type thermocouples embedded into it.

An acoustic defect was made near the specimen top – a  $\emptyset$  2 mm borehole for the ultrasonic measurements of specimen ablation kinetics.

The following on line measurements have been carried out during tests:

- coolant temperature and flow-rate;
- corium melt surface temperature;
- temperature distribution in the steel specimen;
- steel specimen corrosion depth;
- electrical characteristics of the IMCC generator.

Surface temperature of the melt was measured continuously by the «RAYTEK» spectral ratio pyrometer. The surface was periodically recorded on video, melt samples were taken, molten pool depth and bottom crust thickness were measured by the tungsten probe (only in MC5). The samples were subjected to the spot (immediate) analysis for the content of basic components. The analysis helped to determine the composition and mass of corrective additions, which were periodically introduced into the melt to compensate for released components.

A special technique was used for putting the charge into the crucible in the argon atmosphere. At first ~ 5 mm-thick layer of molten corium was put on the specimen top. The corium was prepared in separate tests (Pr1-MC5 and Pr-MC6). The charge composed of oxidic powder and zirconium pins was put on top of the molten corium layer. The melt was produced and heated to the planned temperature level.

After the system had approached a steady state, which was indicated by stabilized thermocouple readings and temperatures of the cooling water after calorimeters at its fixed flowrate, it was kept in it. The steady state was followed by a new transient condition and another steady state. When the experimental program was completed the HF-heating was disconnected. The melt and specimen were cooled in argon. When the furnace was disassembled the specimen and corium ingot were taken from the crucible and embedded in epoxy for the subsequent cutting.

To study the corium-steel interaction zone templates of corium ingot and steel specimen were prepared, they and melt samples, which had been taken during tests were subjected to posttest analyses, in which the elemental and phase composition, microstructure and material properties were determined.

The following methods and equipment were used for physico-chemical analyses:

- Elemental composition.
  - X-ray fluorescence (XRF) –spectrometer with PLE device.
  - Chemical analysis (ChA) spectrophotometer-SPh2000.

Phase composition.

• EDX.

Metallo-and ceramography.

- Optical microscopy.
- SEM.

Corium oxidation degree and free Zr in the samples were calculated from the data of

- photocolorimetry of U (IV) and U (VI) with reagent arsenazo III,
- gas-volumometry of metallic Zr from the volume of hydrogen, which was released at the interaction with phosphoric acid.

The quality control of all procured materials was carried out before the tests, it included elemental analysis and material studies.

#### 8. Experimental activities and main results

In accordance with Work Plant of METCOR2 and its experimental matrix the experimental investigation of the interaction between vessel steel and suboxidized and oxidized corium melts having final compositions ~ C 30 and C 100 has been carried out. The studies have been performed in argon atmosphere and at several temperature levels on the interacting specimen top (tests MC 5, MC 6 and MC 7).

In the course of "Rasplav-3" modernization and improvement of experimental techniques auxiliary (preliminary) tests were performed (Pr1-MC5, Pr2-MC5, Pr-MC6 and Pr-MC7).

## 8.1. Investigation of the suboxidized molten corium oxidation by air oxygen. Test Pr1-MC5

The main experimental objectives of Pr1-MC 5 were:

- 1. To make an experimental check of "Rasplav-3" modernization arrangements and to determine the furnace electrical and power characteristics.
- 2. To study the possibility of molten corium superheating in relation to liquidus temperature at its oxidation from C 32 to C 100.
- 3. To study the process of suboxidized molten corium oxidation by air oxygen, when the inert atmosphere is replaced by air within the same melting session.

Appendix 1 gives a detailed report about Pr1-MC 5 results. Here the main results are presented in brief.

"Rasplav-3" was developed using the operational experience of "Rasplav-2" test facility, the advantages of which it inherited. The test facility upgrading measures improved its operational characteristics.

Main differences between "Rasplav-2" and "Rasplav-3" are as follows:

- The electrotechnical part of "Rasplav-3" has been modernized, which enabled to increase power deposition in the cold crucible and melt by a factor of two (from 34 kW to 68 kW).
- The current frequency of HF-generator VChG -60/0,066 has been changed, and this enabled to melt oxidic and metal-oxidic corium compositions in the cold crucible.
- To exclude induction heating of the specimen the crucible sections around it were welded together, they served as an electromagnetic shield. A movable electromagnetic screen provided an additional shielding of the specimen.
- The gas flow parameter control, gas drying and aerosol removal in the gas-aerosol sampling system have been improved. The electrochemical sensor enables to measure the partial pressure in the off gases.
- The system of the contact-free ultrasonic measurement of steel corrosion has been improved by introducing a more adequate sensor and optimization of its resonance frequency, computer processing of sensor readings, identification of the useful signal and taking into account the sensitivity to specimen temperature conditions. The completed improvement of the ultrasonic measurement system has enabled to monitor changes in the specimen length in relation its temperature conditions.

The initial charge used in the "Rasplav-3" experimental program consisted of mixed oxides and metallic zirconium powder, mass %: 76UO<sub>2</sub>+9ZrO<sub>2</sub>+15Zr.

In order to evaluate electromagnetic losses into the induction furnace and load circuit the blank runs have been performed, the parameters measured and the calorimetry of the elements made.

To make the calorimetry measurements of heat fluxes from the molten pool to the crucible bottom the latter was cooled with water. The experiment was carried out in argon, which later was replaced with air.

After the steady-state regime of the molten pool had been achieved, all electrical and thermal characteristics measured, the inert atmosphere was replaced with air. The air was supplied in stages, at each stage the flow-rate was kept steady, but it was increased at each next stage.

Table 1 presents the air flow-rate and duration of each stage; the calculation of melt oxidation degree from the averaged readings of oxygen sensor and gas flow-rate meters.

#### Table 1

Regime №	Duration Δt, h	Air flow- rate l/min	Mass of oxygen for oxidation Zr, g	Mass of oxidized Zr by the stage end, g	Mass of unoxidized Zr by the stage end, g	С
1	0,44	2,5	9,57	147,21	242,72	37,75
2	0,56	4	13,34	185,25	204,68	47,51
3	0,76	10	19,22	240,03	149,89	61,56
4	1,1	15	27,58	318,64	71,28	81,72
5	0,67	24,5	27,44	369,85	0	100,00

Corium melt oxidation degree increase calculated from the averaged readings of oxygen sensor and gas flow-rate meter At the 1<sup>st</sup> and 2<sup>nd</sup> steady regimes melt turbulence, droplet discharges and sharp increase of anode current were observed. Anode voltage was lowered to exclude the activation of electronic safety system. Further on, approximately until the middle of the 4<sup>th</sup> regime, an increase of anode current, voltage being kept stable, accompanied by the melt temperature growth up to 2700°C was observed. This happened due to the decrease in the melt electric conductivity and free Zr oxidation, which resulted in additional power in the melt. Due to the spontaneous changes in the generator mode the grid current achieved the safety limit, for this reason the inductor voltage had to be lowered, which resulted in a melt temperature drop.

In the end of the 5<sup>th</sup> regime the melt was sampled and, using readings of the oxygen sensor, an assumption was made about the equilibrium state of the gas-melt system. After this argon was supplied into the furnace and after another equilibrium state was achieved the melt sample was taken in order to determine the newly-established ratio between oxygen and total metals. Further on the HF heating was disconnected and corium was cooled in argon.

Using the data of oxygen sensor and flow-rate meters the calculations were made for determining Zr oxidation kinetics during the test. Oxygen sensor data of different stages were averaged. For the calculations of C (Zr oxidation degree) it was assumed that oxygen is spent only on the oxidation of metallic Zr. Oxygen spent on the  $UO_2$  oxidation to  $U_3O_8$  in aerosols was not taken into account, therefore the calculated C value is an overestimation. The results of calculations show, that by the end of the 5<sup>th</sup> stage the corium oxidation degree was 100 %, i.e. all metallic Zr was oxidized. The chemical analysis confirmed this, e.g. in a sample, which was taken before air was replaced with argon, metallic Zr was absent. Table 1 gives the calculations of corium oxidation degree.

The metallographic analysis of the surface and longitudinal section of the bottom specimen has shown:

- The specimen surface is uneven, it has pits, which are covered by the layer of U and Fe oxides. The pits in the metal can be 2,5 3,0 mm deep.
- Not only the specimen top, but also its lateral surfaces have been affected by corrosion.
- The layer of the oxidized metal on the specimen top is 0,3 3,1 mm.

During the regime of corium oxidation the specimen top had temperatures about 950÷1050°C.

The analysis of specimen top microstructure near its surface has shown, that the thermal impact zone, within which the initial austenite structure underwent structural and phase changes, was 6-9 mm thick.

# 8.2 Investigation of the interaction between molten corium C 100 and reactor vessel steel under neutral atmosphere above the melt (Test MC 5)

The experimental objective of MC 5 was the study of corium melt – vessel steel interaction kinetics in the conditions of assumed minimal influence of chemical phenomena. A fully oxidized molten corium C 100 (mass % 70 %  $UO_2 - 30$  %  $ZrO_2$ ) was taken for the purpose, and the test was conducted in high-purity argon (partial pressure of oxygen  $P(O_2) < 2 \cdot 10^4$  bar).

The MC 5 test results are presented in detail in Appendix 2. Brief information about its main results is given in this chapter.

The initial molten pool was produced from the 1794,7 g charge (of it 300,9 g was the powder of C 100, which had been produced in the preliminary test Pr2-MC 5).

In MC 5 during one melting session the melt-steel interaction was investigated at 4 different steady temperature regimes on the specimen top. The values of maximum temperatures in the top center, the average density of heat flux to the top and its magnitude in the sighting spot of the ultrasonic sensor ( $\emptyset$  15mm) for all four regimes are given in Table 2.

### Table 2

Regime	Heat flux	Heat flux density,	Power into the top	Power into the top	Maximum
	density,	Ø15,	calorimeter,	calorimeter, measured,	temperature on the
	average,	$MW/m^2$	calc.,	kW	steel surface near the
	$MW/m^2$		kW		center,
					°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

Calculated and measured value of power, heat flux and temperature

The uninterrupted duration of MC 5 was over 36 hours.

In accordance with ultrasonic measurements the steel specimen corrosion kinetics has the following behavior:

- In the interval 4600-22500 sec, at maximum temperature in the specimen top center  $T_s \approx 1075^{\circ}C$  corrosion is not observed.
- In the interval 23200-60300 sec, at  $T_s \approx 1180^{\circ}$ C, the specimen corrosion depth is  $\approx 70 \,\mu$ m.
- In the interval 65900-96000 sec, at  $T_s \approx 1315^{\circ}$ C, the specimen corrosion depth is  $\approx 100 \,\mu$ m.
- In the interval 98500-133900 sec., at  $T_s \approx 1435^{\circ}$ C, due to the unstable and anomalous indications of the ultrasonic sensor the corrosion depth was not determined.

In the beginning of the  $2^{nd}$  and  $3^{rd}$  temperature regimes the corrosion was intensive, later it gradually slowed down and reached saturation level.

The posttest measurement of the steel specimen gave an overall corrosion depth varying from 20 to 200  $\mu$ m. The corroded specimen surface had parts with uniform corrosion depth and local pits.

The analysis of different parts of oxidic ingot composition has shown that the ingot top, bottom, central and lateral parts practically do not differ from the original corium composition.

A layered oxide structure has been found on the boundary between the oxidic and metallic parts. The ingot bottom, about 0,8 mm, had a homogeneous structure and composition close to the average for oxidic ingot. The bottom part is likely to consist of the sintered initial powder of the molten layer simulator, which was put on the specimen top, it is also confirmed by the porous structure of the bottom layer. This region borders on a thin layer mostly consisting of uranium oxides. It underlies the layer of mixed oxides with mole ratio of  $UO_2:ZrO_2\approx1:2$ . In accordance with references such ratio corresponds to the liquidus-solidus point of contact from the side of a more refractory phase. It should be noted that neither crust, nor the melt bulk contain any iron oxides. This gives reasons to assume that steel corrosion products are localized in a narrow region between steel and a layer of sintered crust.

SEM/EDX analysis of the metal-oxide interaction border showed the inhomogeneity of the border both in terms of microstructure and in phase composition depending on the radial position of the analyzed place (and on its temperature).

Closer to the specimen periphery, approximately 1/3 from the edge, a layer of FeO was separated next to the steel specimen. The overlying oxidic layer consisted of UO<sub>2</sub>, ZrO<sub>2</sub> and small quantities of FeO, which proves the diffusion mixing of corrosion and sintered crust layers. The FeO layer has a rather high porosity.

Closer to the center of the specimen surface there is a region with a higher temperature on the top, on the boundary between the oxidic corrosion layer (FeO) and oxidic crust, which had interacted with iron oxide, a  $Fe_3O_4$  phase was isolated. It should be noted that this region has a rather considerable presence of UO<sub>2</sub>. In terms of microstructure and phase composition this zone has a rather distinct boundary with a FeO-based zone adjacent to steel and the zone of sintered crust corium. It can be assumed that by composition this intermediate zone UO<sub>2</sub>-ZrO<sub>2</sub>-FeO is in the eutectic melting region (~ 1270°C) and this interlayer used to be in a liquid state.

In the top center, which had maximum temperatures on the surface, the molten oxidic part was thicker up to the metallic ingot, which included a partial melting of metal on the boundary. The molten state of metal on the boundary is confirmed by the presence of  $UO_2$  inclusions into metal in the near-boundary region. Evidently at ~1420°C the formation of eutectics in the  $UO_2$ -ZrO<sub>2</sub>-FeO-Fe system causing the steel melting is possible.

The metallographic analysis of the steel specimen has shown that at a long exposure of steel to high-temperature thermal gradient conditions the microstructure of the surface layer has changed to the depth up to10 mm, which is observed as a grain coarsening and pore formation. The deterioration of mechanic properties in this layer can be expected.

The MC 5 experimental data on the interaction of corium C 100 with vessel steel and posttest analyses have shown the following:

- 1. At  $T_s < 1100^{\circ}$ C on the specimen top there is no noticeable interaction between the melt and vessel steel. I.e. in these conditions the crust is a reliable protector of steel from melt attack.
- 2. In the temperature range of  $T_s=1150\div1250^{\circ}C$  on the specimen top its oxidation followed by FeO layer formation takes place. Steel oxidation is likely to take place due to the UO<sub>2</sub> non-stoichiometry growth accompanied by oxygen liberation at temperature increase. At this, due to the limited oxidation resources of the sintered oxidic crust during the transient toward a higher temperature level, the corrosion rate, which originally accelerated, starts to decrease and approximately after 3 hours corrosion stops. The corrosion regime in this temperature range is of a solid-phase type, there are distinct boundaries between steel, FeO layer and sintered oxidic crust, the depth of steel ablation by corrosion is < 0,1 mm.
- 3. In the temperature range T<sub>s</sub>=1270÷1400°C on the specimen top the oxidation pattern and corrosion rate became close to those observed during the previous temperature regime. Corrosion is sustained due to the FeO formation because of oxygen diffusion from crust. The original increase of corrosion at a higher temperature level goes down with time. But certain differences have also been observed. Fe<sub>3</sub>O<sub>4</sub> globules were found on the boundary between the corrosion layer FeO and crust. In all probability during the inter-diffusion of U and Zr oxides of the crust and iron oxides in the interaction zone after a certain temperature is achieved, the eutectic melting takes place and a liquid layer is formed between FeO and sintered oxides UO<sub>2</sub>-ZrO<sub>2</sub>. The approximate evaluations of eutectics melting temperature in the UO<sub>2</sub>-ZrO<sub>2</sub>-FeO system is about 1270°C. The oxidation regime in this range is mixed solid- and liquid-phase.

4. At T<sub>s</sub>>1400°C on the specimen surface the thickness of liquid layer increases, and when a certain temperature on the steel specimen boundary is achieved the eutectics melting of the steel specimen below the steel melting temperature is possible. In accordance with estimates the eutectics temperature in the (UO<sub>2</sub>-ZrO<sub>2</sub>-FeO-Fe) system from the side of Fe is ~ 1420°C. The beginning of the steel specimen surface ablation was likely to distort the readings of the ultrasonic sensor. The oxidation regime in this range is a liquid-phase.

The MC 5 results have shown that the interaction of molten corium C 100 and vessel steel in the inert atmosphere is characterized by a lower corrosion rate as compared to tests performed in the 1<sup>st</sup> Phase of METCOR project.

# 8.3 The investigation of suboxidized corium melt interaction with reactor vessel steel under inert atmosphere (T<sub>steel surf.</sub> »1400°C). Test MC 6

The experimental objective of Test MC 6 was to study the vessel steel ablation kinetics during its interaction with suboxidized corium C 27 in oxygen-free (argon) atmosphere and constant temperature on the specimen top.

The initial corium was produced from the 1850 g charge, with composition, mass %: 76%  $UO_2$ ; 9,33 %  $ZrO_2$ ; 14,6 % Zr.

In MC 6 the melt-specimen interaction was examined at two steady states, which had similar temperature on the specimen top. Table 3 gives maximum temperatures in the center of the specimen top, average density of the heat flux to the specimen, its value in the ultrasonic sensor sighting spot ( $\emptyset$  15mm) in the beginning of the regime.

#### Table 3

Regime	Heat flux density, average, MW/m <sup>2</sup>	Heat flux density, Ø15, MW/m <sup>2</sup>	Power into the top calorimeter, calc. kW	Power into the top calorimeter, measured kW	Maximum temperature on the steel surface near the center, °C
1	0.85	1.23	1.67	1.64	1375
2	0.87	1.31	1.7	1.71	1397

Calculated and measured values of power, hear flux and temperature

The duration of MC 6 was over 10 hours.

MC 6 experimental results and post-experimental data are given in Appendix 3. This section presents main results in brief.

In accordance with ultrasonic measurements of steel specimen ablation kinetics during the first 3 hours the ablation character was similar to the steel corrosion of MC 5 at the  $2^{nd}$  and  $3^{rd}$  temperature regimes. Corrosion rate decreased with time and in ~ 2,5 hours it reached saturation level. But in another ~ 5 hours the ultrasonic sensor started to give spurious data - indicated the lengthening of specimen to the value exceeding the original length, after ~ 3 hours the indications got stabilized. The anomalous character of ultrasonic measurements probably does not reflect the actual change in the specimen length, it is explained by the changes in the signal transfer conditions and a different steel ablation mechanism.

The post-test analysis of the specimen has shown that at ~6 mm depth from the top surface there is a region containing Fe-U-Zr-O, which was in molten condition before cooling and had a higher density than that of steel. These circumstances caused the reduction of speed, at which the ultrasonic signal passed through the mentioned region, increase of the total signal transmission

time and, consequently, the apparent specimen length increase in accordance with ultrasonic measurements.

The previously used standard methodology of the ultrasonic signal processing for determining the steel specimen ablation rate is not applicable, if the liquid phase is formed. It can be used only for the diagnostics of the liquid phase establishment, qualitative evaluation of the liquid phase boundary progression into the specimen bulk and its stabilization at a certain point. To make up for the ultrasonic measurements inadequacy a special methodology of the posttest echogram massif processing enabled to give an approximate evaluation of the boundary between the solid specimen surface and liquid alloy Fe-U-Zr-O progression kinetics.

The SEM/EDX analysis results have shown:

- In the specimen top zone, between corium and steel, there is a metallic phase zone mostly consisting of iron enriched with U, Zr and a small amount of O. In cross section this zone looks like a spherical segment and it propagates as far as the acoustic defect bottom.
- As it follows from SEM/EDX analysis, the chemical composition and microstructure of this zone is uniform in height and radius.
- It can be asserted with a high degree of assurance, that before the crystallization this region was in a molten state. This is confirmed by the character of its microstructure: dendrites of iron, which were formed at a fast crystallization of the melt after the HF heating was disconnected (in hardening conditions) and by a typical eutectics structure between the dendrites. The composition of dendrites is close to pure iron, i.e. is different from steel composition. This is another confirmation that the phase has crystallized from the melt rather than existed as a solid-phase inclusion into it.
- After crystallization the melt, which had been produced on the boundary between corium and steel, formed three phases – iron-based, containing a certain amount of chrome, also Mn, Ni, and Si in admixture quantities; compound (U, Zr) Fe<sub>2</sub> –based phase with a certain amount of Cr, Ni, Mn and Si admixtures; and UO<sub>2</sub>-ZrO<sub>2</sub> -based solid solution.

As it can be concluded from the analysis of regions in the corium ingot, a part of metallic melt broke off from the interaction zone and rose to the melt surface. It changed its composition because of the contact with corium components (it was enriched by them).

The depth of the interaction zone was determined from the profilogram of the specimen axial section. The profilogram was measured by the stage micrometer having the error of  $\pm 10 \ \mu m$ . The steel specimen ablation caused by eutectics melting was up to 6,7 mm deep.

The irregularity of border between the crystallized metallic (Fe-U-Zr-O) and oxidic phases can be explained by the metallic melt part going up through a local rupture in the crust.

Therefore, the experimental data of MC 6 on the interaction of suboxidized corium C 27 and vessel steel in the inert atmosphere and posttest analysis have shown:

- 1. At Ts≈1400°C on the specimen top two specific stages of vessel steel ablation can be identified. At the first stage the ultrasonic measurements showed a slow ablation of the specimen, which was probably explained by the diffusion of oxygen, uranium and zirconium through the crust to the interaction boundary. The ablation rate decreased with time and reached saturation after 3 hours. At the second stage, in ~ 5 hours after the interaction start a rather intensive specimen ablation caused by the eutectics interaction with produced metallic melt was observed. The melt consisted mostly of iron enriched with uranium, zirconium and a small amount of oxygen.
- 2. In accordance with ultrasonic measurements the ablation depth at the first stage was ~ 0,27 mm, the maximum ablation depth at the 2<sup>nd</sup> stage was 6,7 mm.

3. At the  $2^{nd}$  stage the steel specimen ablation rate decreased with time following the reduction of temperature on the interaction interface; after 3 hours from the  $2^{nd}$  stage start it reached saturation at ~ 1100-1200°C (in accordance with preliminary evaluation). Additional studies are necessary for specifying this temperature.

With a high degree of certainty it can be asserted that the metallic phase, with which the melt interacts, is the melt consisting of iron enriched with U, Zr and O.

MC 6 results show that the interaction of suboxidized corium melt C 27 with vessel steel is characterized by a much more extensive ablation and a different mechanism of it as compared to MC 5.

# 8.4 Investigation of the interaction between the suboxidized corium melt and vessel steel under the inert atmosphere above the melt (Tst.surf»1150°C). Test MC 7

The experimental objective of MC 7 was to study vessel steel ablation kinetics during its interaction with suboxidized corium ~ C 27 in the oxygen-free atmosphere and temperature on the specimen top  $T_{st.surf} \approx 1150^{\circ}$ C and to specify the boundary temperature of steel on the interaction interface, at which the ablation following the eutectics mechanism starts. The initial corium was prepared from 1850 g charge having the composition of mass %: 76 % UQ : 0.33 % ZrQ : 14.67 % Zr (of it 150 g was prepared from the inget produced in test Pr MC

UO<sub>2</sub>; 9,33 % ZrO<sub>2</sub>; 14,67 % Zr (of it 150 g was prepared from the ingot produced in test Pr-MC 6).

The total test duration was over 10 hours. The experimental procedure, furnace and vessel steel specimen characteristics completely corresponded to those of MC 6. The processing of MC 7 results and posttest studies is still in progress; the report, in which the

test results will be presented in full detail, will be prepared in the first quarter of 2004. Here only preliminary results are mentioned.

- 1. At  $T_s \approx 1150^{\circ}$ C on the specimen top only two specific stages of steel ablation could be recognized. At the first stage the ultrasonic measurements showed a slow ablation of the specimen, which was probably explained by the diffusion of oxygen, uranium and zirconium through the crust to the interaction boundary. The ablation rate decreased with time and reached saturation after 3 hours. At the second stage it happened in ~ 4 hours after the interaction start.
- 2. In accordance with ultrasonic measurements the steel specimen ablation depth at the 1<sup>st</sup> stage was ~ 0,18 mm, a maximum ablation dept at the 2<sup>nd</sup> stage determined from the axial section profilogram was ~ 2,9 mm.
- 3. The axial section of the steel ablation caused by eutectics melting looks like a segment located in the central part of the specimen, the chord of which is 29 mm long.
- 4. The boundary temperature, at which the intensive eutectics melting does not take place, in accordance with approximate evaluation, is ~ 1020÷1100°C, which is about 100°C lower than a boundary temperature of MC 6 evaluations.

### 9. Current stage of the project progress

The activities of the 1st year of the Project have been performed in full compliance with Work Plan, experimental matrix and specified conditions of certain tests, which were developed during the discussion of experimental matrix with collaborators and recorded in the minutes of meetings (Appendix 4). Additionally to the tests listed in the Work matrix preliminary tests (Pr1-MC5, Pr2-MC5, Pr-MC6, Pr-MC7) have been performed, which were aimed at improving experimental methodologies, capacities of the "Rasplav-3" test facility and ablation mechanism analysis.

### 10. Cooperation with foreign collaborators:

The foreign collaborators within this project are:

- 1. Dr. Walter Tromm, Germany Institut für Kern-und Energitechnik (IKET), Karsruhe.
- 2. Dr. David Bottomley, Germany EUROPÄISCHE KOMISSION, Joint Research Center Institut für Transurane (ITU), Karsruhe.
- 3. Dr. Manfred Fischer, Germany Framatome ANP GmbH, Erlangen.
- 4. Dr. Gerard Cognet, France CEA/DEN/DSNI, Saclay.
- 5. Dr. Florian Fichot, France IRSN/DRS/SEMAR/CEN, Cadarache.
- 6. Dr. Ole Kymäläinen, Finland Fortum, Vantaa.

During the first year of project implementation the close cooperation with foreign collaborators included:

- the detailed discussion and approval of Work Plan of the Project 2<sup>nd</sup> stage, its experimental matrix;
- the analysis and evaluation of results of each experiment carried out during the first year;
- making required modifications in the specifications of conducted tests.

In 2003 after the joint analysis of studies completed within the 1<sup>st</sup> Phase of METCOR project two papers have been prepared jointly with collaborators and sent to Nuclear Engineering and Design . The results of the 2<sup>nd</sup> METCOR Phase were used in preparing the presentation at the ICAPP-04 conference in Pittsburgh, USA.

The discussion of experimental results and exchange of opinions during the 2<sup>nd</sup> phase of METCOR took place both at joint meetings and by e-mail. Along with that the results of studies carried out during the first 8 months were presented on 19 September 2003 at a CEG-CM meeting.

The first meeting of the METCOR 2 Steering committee together with European collaborators took place on October 3, 2002 in FZK-FTU, Karlsruhe, Germany. At the meeting the experimental matrix was finalized, a reference test on the interaction with fully oxidized corium  $UO_2/ZrO_2$  in argon was added.

The second meeting of the METCOR 2 Steering committee took place on January 29, 2003 in Aix-en-Provence, France. The following presentations on the work preformed during the 2<sup>nd</sup> Phase of METCOR were made a) Preparation of technical means and test Pr-MC5 results and b)

Numeric modeling of the molten pool thermohydraulics. The specifications of the first experimental series were finalized and an agreement was reached about the parallel calculations of MC 4 molten pool by project contractors and collaborators (G. Cognet).

The third meeting of the METCOR 2 Steering committee together with collaborators was organized on September 16, 2003 in NITI, St. Petersburg, Russia. The results of work during 9 months of the project were reported in 6 presentations. Project contractors and collaborators presented the results of calculations of MC 4 free convection. The collaborators approved the scope of carried out research and noted the novelty of experimental results, which provided a better insight into the qualitative and quantitative characteristics of the interaction process. In the discussion of the test completing the 1<sup>st</sup> experimental series in the 4<sup>th</sup> quarter of the project it was decided to conduct test MC 7 with ~1100°C on the specimen interaction interface and to use its results for amending experimental matrix of the 2<sup>nd</sup> year at a next meeting. The completed research enabled to take a decision about preparing a joint presentation at the conference ICAPP-04 in Pittsburgh.

### 11. Perspectives of further investigation

The work plan and experimental matrix foreseen for the 1<sup>st</sup> year of the METCOR 2 implementation have been fully implemented. Additional experiments have been performed (Pr1-MC 5, Pr2-MC 5, Pr-MC 6, Pr-MC7), which helped to improve numeric methodologies, extend the experimental capabilities of the "Rasplav-2" test facility and have a more detailed studies of the vessel steel ablation mechanisms.

But new data on the ablation mechanisms, which have been produced in the  $1^{st}$  experimental series, may require additional experiments for getting information important for the numeric model development. The necessity of additional experiments will be discussed at the  $4^{th}$  meeting with collaborators. In case of a joint decision about experiment, which should complement the 1st experimental series, the  $2^{nd}$  year matrix is to be amended – the second series program reduced. The specifications of the second experimental matrix, in which the studies of vessel steel corrosion by the melt at an additional introduction of steel into the suboxidized corium and measurements of the bottom specimen ablation will be discussed with collaborators at the  $4^{th}$  meeting in February 2004 in Paris.

Project manager, Professor General Director of Aleksandrov RIT V.B. Khabensky V.A. Vasilenko

### Appendix A

Category	Employment per year (work days)	Incremental employment	Total
Category I	2898	2898	6180
Category II	1062	1062	2279
Category III	180	180	360
Category IV	31	31	150

### Personnel involvement during the 1<sup>st</sup> year

### Appendix B

Main procured equipment The following equipment has been procured in accordance with Work Plan:

- Laptop 5205-S703 (Toshiba 5205 Portable Personal Computers)

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