

ISTC Project #3345

Source Term Assessment at Ex-vessel Stage of Severe Accident

Final Project Technical Report

on the work performed from 01.01.2007 to 31.12.2007

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January 2006

This work is supported financially by ISTC and performed under the contract to the International Science and Technology Center (ISTC), Moscow.

Title of the Project:	Source Term Assessment at Ex-vessel Stage of Severe Accident
Commencement Date:	01.01.2007
Duration:	12 months
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Keywords:

AES-91, VVER-1000, NUCLEAR REACTOR, SEVERE ACCIDENT, FISSION PRODUCTS, IMITATIONS, CORIUM, CORE MELT, INDUCTION HEATING, OXIDATION DEGREE, AEROSOL, GENERATOR, AMMONIA CHLORIDE, TURBOPHORESIS, DEPOSITION, RESUSPENSION, LES-MODEL, IODINE MODULE, CONTAINMENT, SLUDGE, SUMP, DOSE RATE, CALCULATION, BOUNDARY CONDITIONS, EXPERIMENT.

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1. Objectives of the Project, Scope of Work and Technical Approach

Objectives of the Project

The “Ex-vessel Source Term Analysis” (EVAN) ISTC project includes theoretical and experimental research of the processes affecting the late phase fission product release to the PWR containment atmosphere at the late stage of the hypothetical severe accident with core meltdown. The main results expected to be obtained within the framework of the proposed project are listed below:

1. Experimental data and numerical analysis of core melt fission product release, applicable to different designs of power units, taking into account corium transition from sub-oxidized to fully-oxidized state and water supply onto the melt surface.
2. Experimental data and numerical analysis of processes of modeling aerosol transport and deposition in primary circuit pipelines at different flow conditions of the medium-carrier, aerosol species characteristics.
3. Experimental data and numerical analysis on how composition of containment sump solution and sludge affects content and partitioning of volatile iodine species in the containment atmosphere at different temperature, irradiation dose, ambient parameters and covering characteristics.

The scope of the project may be extended by additional adaptation and validation of computer models, continuation of experimental programmes and radiological consequences calculation for selected severe accident scenarios.

This research is a sort of applied investigations. Theoretical and experimental research results can be used for computer codes validation for application to new designs, for safety assessment of both existing Russian and foreign NPP designs, including probabilistic safety analyses of 2 and 3 level, for development of severe accident management strategies and for emergency planning analysis. The project features possibility to obtain data directly applicable to new designs of NPP with VVER-type reactors providing severe accident management measures.

Scope of Work and Technical Approach

Task 1. Assessment of results of severe accident sequences modeling

Task description and main milestones	Participating Institutions
<p>Assessment of results of severe accident sequences modeling</p> <p>Task Stages: 1) Assessment of results of severe accident sequences modeling for different NPP designs with PWR or VVER reactors. Justification of initial data for experimental investigations. 2) Justification of applicability to be obtained experimental data for computer codes validation. Preparation of experimental programs.</p> <p>Tools to solve the task: 1) Results of severe accidents modeling. 2) Computer codes RATEG/SVECHA/HEFEST, CORCAT, DINCOR, KUPOL-M, SCDAP/RELAP, MELCOR. Assessment of results of numerical modeling.</p>	<p>1- SPAEP 2- IBRAE</p>
Description of deliverables	
1	Report on initial data for experimental programs for tasks 2, 4, 6 (stage 2)

The goal of the analysis is to determine the parameter ranges within the reactor plant and containment, core melt parameters, FP aerosol characteristics, surface boundary conditions at structures and equipment, chemical content of containment sump solution, dose rate ranges in the equipment and

containment, and other parameters, necessary to develop the final test specification for Tasks 2, 4, and 6. Participants will also analyze the capabilities of physical models for aerosol generation and transport for RATEG/SVECHA/GEFEST.

Task 2. Experimental research on FP release from molten pool/core melt catcher

Task description and main milestones		Participating Institutions
<p>Experimental research on FP release from molten pool Work stages: 1) Experimental investigations of low-volatile fission products release during molten corium oxidation. Melt characteristics and composition of the FP in question are determined from the results of Task 1. 2) Experimental investigations of low-volatile fission products release during water supply onto the melt surface.</p> <p>Tools for solving the task: 1) A set of “Rasplav” experimental installations, equipped with aerosol sampling system, for studying high-temperature phenomena in molten corium. 2) A complex of instruments and equipment for tests preparation, conducting posttest analyses and processing experimental results. The complex includes XRF spectrometers, microsizer, mass spectrometer, spectrophotometer, auxiliary equipment (crusher, microgrinder, laboratory balance, etc.)</p>		1- NITI
Description of deliverables		
1	Report on experimental study of low-volatile fission products release during molten corium oxidation	
2	Report on experimental study of low-volatile fission products release from molten corium pool covered by water layer	

A calculated prediction of the main radiation safety objectives during a severe accident requires substantiated data, e.g. coefficients of the radiologically significant radionuclides release from molten fuel into the environment. At present such data are available for the majority of volatile radioactive FP (radioactive noble gases, iodine, Cs, Rb, etc.), which evaporate during the core degradation and molten fuel pool formation. The release of such low-volatile FP as Ba, Sr, La, Ce, isotopes of the platinum group elements (Ru), lanthanoids and actinoids from a high-temperature molten core pool still has not been studied sufficiently due to extremely difficult engineering aspects of experimental investigations. For suppressing the FP release, protecting the superstructures from heat radiation from the melt surface and increasing the efficiency of corium pool cooling, the core melt catcher (CMC) envisages water feeding onto the melt surface after inversion of the oxidic and metallic layers. The resulting decrease in radionuclides release into the containment is mainly achieved owing to:

- capture of a significant part of aerosols by the water layer at bubbling the gas-aerosol flow through it,
- melt surface temperature decrease at the film boiling of water,
- crust formation at the melt surface.

At present, there does not exist a unified theory that would describe the mechanisms of evaporation from oxidic melts. The complexity of oxidic systems evaporation is that evaporation of few oxides follows one chemical pattern (congruently). The majority of data on evaporation have been obtained by the classical Knudsen's method (effusion into vacuum), as well as by high-temperature mass spectrometry (a combination of Knudsen's method with mass spectrometry of the evaporated products) and are available for individual oxides. These data are hardly applicable to the multicomponent oxidic melts and, correspondingly, to the severe accident conditions.

The study FP evaporation from molten corium (Task 2) envisages application of the flow method which belongs to the dynamic methods of steam pressure determination. The essence of the method is in saturation of the carrier gas passing at a constant rate above the melt (water-flooded, too) with vapors of the substance in question. The amount of the transported substance will be determined by means of physicochemical analyses.

The high temperature and chemical activity of the molten ceramic corium place limitations on the application of conventional heating methods and crucible materials. The method of induction melting in the cold crucible (IMCC) in the RF band has been chosen for producing ceramic melts and achieving objectives of the project.

Characteristic features of the method:

- internal power in the melt;
- the presence of a crystallized melt layer (crust) between the melt and the crucible cold wall, which prevents mass transfer of the crucible material to the melt.

This combination of the non-contact heating and the non-contaminating method of oxides melting ensures:

- melt purity as high as that of the initial products;
- melt superheating above T_{liq} , chemically active oxidic materials included;
- melting and long-term maintenance of an oxidic system in molten state in both neutral and oxidizing atmospheres;
- universality and compactness of the melting device.

The level of experimental investigations is determined to a considerable degree by the available material and technical basis. The backbone of the facility are three experimental installations for the induction melting of corium in which the IMCC technology is realized at different frequencies of the heating current. They allow experimenting with a broad range of corium compositions which differ greatly in electrical conductivity when in molten state. The specification of the experimental installations are given in Table 1.

Table 1. Specification of experimental installations.

Specification	Experimental installation		
	Rasplav-2	Rasplav-2/C	Rasplav-3
Molten corium preparation method	Induction melting in the cold crucible (IMCC)		
Installed capacity, kVA	250	250	100
Melt mass in crucible, kg	Up to 5	Up to 10	Up to 2
Melt temperature, °C	Up to 3000		
Above-melt atmosphere	Air, nitrogen, helium, argon	Air	Air, nitrogen, helium, argon
Melt composition	Oxidized corium		Unoxidized corium and steel
Possible manipulations with melt	Ingot production	Spreading	Ingot production
Commissioned, year	1988	1995	2002

The installations are equipped with modern monitoring instruments, including those for measuring the melt temperature, electrical characteristics of melting, calorimetry of heat fluxes and the process video monitoring. Acquisition, processing and storage of the results of measurements is done using the data acquisition and measuring system IIS-R designed and produced by the NITI specialists. IIS-R incorporates modern software controlled by the Lab View 5.5 software package.

The experimental installations are supported by the laboratory equipped with modern instruments for physicochemical analyses.

Table 2 NITI contribution to the EVAN project: suggested experimental. Stage 1.

Test	Test objective	Specifications	Notes	Time
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S t a g e 1	EVAN-FP1	Study of low-volatile fission products release during molten corium oxidation	Corium composition: $UO_2 - ZrO_2 - Zr$, $U/Zr(at)=1.2$, C-32 (specified according to the results of Tasks 1 and 2) >C-100, Corium mass: 1-2 kg, Melt temperature: $T_{liq}+(50-100)C$, FP: Ru, Mo, Ce, La, Sr, Ba	Oxidation in air	1-4 quarter
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Notes:

- Determination of the FP release coefficients is carried out using stable FP simulators by the flow method that envisages measurements of FP concentrations in the melt, gas (steam) flow and water layer on the melt surface.

Task 3. Analytical investigation of fission products release from molten pool or core catcher.

Task description and main milestones	Participating Institutions
Analytical investigation of fission products release from molten pool. Task Stages: 1) Review of modern codes capabilities to simulate FP release from molten pool. Justification of applicability to be obtained experimental data for computer codes validation. 2) Pre tests/post tests simulation (in connection with experimental program performance). Tools to solve the task: High performance computers, analytical and numerical calculation models	1- IBRAE
Description of deliverables	
1	Report on modeling results of fission products release from molten pool

By now, many physical models of FP release from the molten pool have been implemented in the computer codes. They are characterized by the various levels of sophistication, from simplest correlation models to mechanistic ones. To do the calculations, it is supposed to use codes RATEG/SVECHA (code RELOS, Germany, if provided by counterparts). Experimental data from EU Framework Programme LPP project provided by collaborators could be used for additional analysis.

Task 4. Experimental study of aerosols transport processes in the primary circuit equipment

Task description and main milestones		Participating Institutions
<p>Experimental study of aerosols transport processes in the primary circuit equipment</p> <p>Work stages: 1) Final completing and adjustment of experimental facilities. Development of research procedures. 2) Experimental studies of modeling aerosol particles deposition on pipeline surfaces from a gas flow and re-entrainment of particles.</p> <p>Tools to solve the task: 1) A test facility for studying aerodynamics of a "dust-laden flow": distributions of velocities, concentrations, and dynamics of deposition of aerosol particles (depending on disperse composition of aerosol) on the channel walls and re-entrainment of particles in the flow, equipped with an online optical monitoring system. A test facility for gas cleaning from aerosol impurity. A test facility for calibration of the measuring system, adjustment of aerosol particle generators and determination of spectral composition of the particles. 2) A complex of instruments and equipment for experiment preparation, performance of post-test analyses and processing of experimental results. The complex includes: lasers, thermoanemometer, signal analyzers, LDA-processor, PC-based workstations.</p>		1- CKTI
Description of deliverables		
1	Report on results of experimental studies of aerosol particles transport processes in the primary circuit pipelines	

Fission products deposited at the initial stage of a severe accident in the primary circuit (including heat-transfer surfaces of steam generators) can in future enter the atmosphere of the containment or the environment in case of containment bypass. Account of FP retention in the primary circuit at the initial stage of the accident may influence the evaluation of accidental release, which can be both overestimated (due to possible additional discharge of deposited FP from the primary circuit to the containment atmosphere at a later stage of the accident) and underestimated (in case of reliable FP localization within the primary circuit). To forecast the rates of FP leaving the primary circuit surfaces (due to resuspension and re-evaporation), it is important to know conditions of aerosol deposition on walls of pipelines of different diameters from the steam-gas turbulent flow. Experimental research of local problems of particles deposition and resuspension, and processes of FP re-evaporation from inside surfaces of the primary circuit will allow more precise definition of characteristics of FP entering the containment atmosphere from the primary circuit late in severe accidents.

Initial stages of the experimental research will be carried out using the available installation that was earlier used in experimental study of fluid mechanics and heat transfer in Russian Fissile Material Storage Facility cooling pipes. In particular, there is a steel cylindrical channel of 98 mm inside diameter, 6.2 m long. Optical windows for flow scanning by laser Doppler anemometers are located in four cross-sections along the length. Coordinate devices for flow scanning by wire sensors (thermal anemometers and resistance thermometers) are disposed in five cross-sections along the length. All measurements are computer-controlled.

Optical methods as applied to the task under consideration can be successfully used both for measuring kinematical characteristics of particles and for fractional analysis of a two-phase flow.

Kinematical measurements

The laser Doppler method is used for kinematical measurements. It permits to measure, with sufficient accuracy (of the order of several percent):

- carrying phase velocity profile,
- suspended phase velocity profile (for particle size larger than several microns).

The measuring method is absolute and does not require any calibration.

The measured velocity range is 0.1...100 m/s.

Measurement of both longitudinal and transverse velocity components is possible. Both average and pulsating velocity components are measured. The measurements are performed in an online mode.

The time of traversing the channel diameter is about 1 min. Simultaneous measurements in several (up to four) check cross-sections along the pipe length are possible. The tube is supplied with the protective glasses.

CKTI has main measuring equipment units, which does not exclude the necessity of adapting them for solution of the problem under consideration. At the same time, adaptation of the experimental setup for the radial velocity measurements is considered to be unnecessary complex so for the radial flow velocity component distribution and its pulsation characteristics will be measured by three-wire thermoanemometer.

Fractional analysis

Fractional analysis is understood as measurement of an average size (and, sometimes, also the size distribution function) and the volume concentration of particles. Unlike kinematical measurements, there is not any optical method today which enables measurements throughout the entire required range of particle sizes. Information available at present is not sufficient for final selection of a measuring method. As a first approximation, selection of the spectral transparency method (STM) for liquid particles and the particle counter method (PCM) for hard particles seems reasonable. A similar solution was used in experiments on the STORM test facility.

For measurements in flows with particle sizes of up to several microns, optical constants (refraction index) of particles must be known.

STM is absolute method and does not require calibrations, but it is integral (that is, it gives an averaged value along the beam path in the medium under study). The use of the method allows measurement of a certain combination of size and volume concentration. In some cases, both parameters can be measured separately. This depends on particle material and size range.

PCM has a high spatial resolution (fractions of a cubic millimeter) and allows measurement of a size distribution function of droplets. Thus, building of a profile of particle size and concentration over the channel diameter is possible. The main drawback of the method is that it is not absolute and requires provision of a calibrating apparatus. The exception is performance of relative measurements (e.g., piecewise concentration of particles).

The STM is limited by particles concentration at the lower boundary while the PCM at the upper boundary.

The STM allows online measurements, and the PCM requires a certain time (not more than a minute) for measurements at a single point.

All the above considerations are stated for an assumption that particles have a spherical shape. In case the particles have large deviations from spherical form, special calculations of light dissipation and additional experiments on a calibration apparatus will be required.

Engineering implementation of the fractional analysis methods under consideration will not require purchase of any special expensive equipment. All optical methods considered allow creation of a fully automated measurement complex including measurement, traversing and processing of results.

CKTI has considerable experience both in kinematical measurements of one- and two-phase flows and in fractional analysis of two-phase media (in particular, analysis of high-velocity steam flows with water droplets of a size from hundredth parts to hundreds microns).

Task 5. Theoretical and computer modeling of aerosol transport in the primary circuit

Task description and main milestones		Participating Institutions
Theoretical and computer modeling of aerosol transport in the primary circuit. Task Stages: 1) State-of-art review of modeling capabilities of the processes in question by modern computer codes, justification of applicability of the expected experimental results for models validation. 2) Pretest/posttest calculations (according to the test schedule). Tools to solve the task: 1) 2D and 3D CFD codes coupled with mechanistic aerosol models, RATEG/SVECHA/GEFEST code with integral aerosol models. 2) High-performance computers.		1- IBRAE 2- SPAEP
Description of deliverables		
1	Short description of model for FP in reactor coolant circuit (PROFIT). Report on modeling results for aerosol particles transport in the primary pipelines	

Fission products deposited at the in-vessel stage of severe accident in the primary circuit (including heat transfer surfaces of steam generators) can be released into containment atmosphere at later stages of the accident. Taking into account FP confinement in the primary circuit at the initial stage may both increase (due to possible additional resuspension/revaporization of deposited FP from the primary circuit to the containment atmosphere at ex-vessel stage) and decrease (due to formation of stable chemical compounds with FP at surfaces of primary circuit equipment) FP release. To predict FP release rate from surfaces of primary circuit equipment (due to resuspension and revaporization), it is greatly important to know conditions of aerosol deposition from steam-gas turbulent flow onto reactor plant pipe walls of different diameters. Partly, of the principal interest are the flow characteristics (laminar and turbulent flow), fluid density and temperature gradients in the near-wall space for analysis of aerosol deposition due to diffusionphoretic and thermophoretic forces. Besides experimental investigation of these processes, it is necessary to carry out joint calculations on gas-dynamical and aerosol models. While modeling the aerosol transport in the real geometry of the primary circuit it is necessary to employ such thermal hydraulic models that allow to account for Reynolds number change for changing pipeline geometry (e.g. in elbows, and bends). Numerical and experimental investigations of local problems related to FP species deposition, resuspension and deposited FP revaporization from internal surfaces of the primary circuit will allow better prediction of FP release to the containment atmosphere at severe accident ex-vessel stage.

To model the transport and deposition processes, the hydrodynamics equations with aerosol particle dynamics equations will be solved numerically. By now many effective ways to solve hydrodynamics equations has been developed. The main problem is to correctly model the turbulent transport processes. For this two principally different ways exist. The first approach imply using the Reynolds Averaged Navier-Stokes equations (RANS) with semi-empirical constitutive models for turbulence. Using this approach, it is possible to build the effective numerical algorithm and to perform calculations for complex 3D flows. The main shortcoming of this approach is that the assumption of turbulent folws being proportional to the concentration gradients is used (which is not always correct, especially for near-wall area) along with not sufficiently accurate modeling of the turbulence parameters.

The other approach is the Direct Numerical Simulation (DNS) turbulence modelling. While using this approach, the hydrodynamics equations for turbulent flow are solved at such fine grid so it becomes possible to resolve flows for all scales up to Kolmogorov's turbulence micro-scale. Combined with the particle transport modeling, such approach can allow direct calculation of all deposition processes with the only external data necessary being aerodynamical particle characteristics and Brownian diffusion

coefficient. However, this approach is very demanding for the numerical grids used (number of nodes for such grid is about the Re number to the degree of 9/4) which limits its applicability to the relatively low Re-number flows.

The so called Large-Eddy Simulation (LES) approach holds the intermediary position between these two approaches. In this approach only relatively large, energy-retaining eddies are explicitly resolved at the grid, while flows with smaller scales (sub-grid) are modeled parametrically. This approach requires significantly less effort and allows to model the wide range of turbulent flows with minimal requirements for empirical constitutive relations. Advantage of this approach compared to RANS is that it allows to model with more accuracy the impact of turbulence on the particles behaviour and provides more detailed information about turbulence parameters necessary for modeling the particles deposition and resuspension.

SPAEP NPP safety research department and IBRAE have access to the modern high-performance computers allowing to use complex and resource-consuming computer codes for necessary calculations.

In IBRAE the aerosol transport processes are modeled with the baseline models implemented in the RATEG/PROFIT computer code. The PROFIT models are used for evaluating the fission products aerosols and vapours transport within the reactor coolant system and the in-containment source term. The PROFIT models are interfaced with RATEG thermohydraulics code. Within this project the Task 4 experimental data will be analysed with the aid of those models by specialists from SPAEP and IBRAE. The 3D models will be used to estimate the limitations of the baseline models and to gain independent insights into the modeled processes.

Thus, for Task 5 realization the hydrodynamics models coupled with aerosol models will be used. External boundary conditions necessary for calculations will be chosen based on the results of Task 1. Task 5 stages are: (I) analysis of the main processes and the physico-mathematical models for particle transport and deposition; (II) calculation and analysis of results; (III) calculations with aerosol kinetics models coupled with thermal hydraulic codes, and sensitivity study for different parameters on final results. There will be joint work of IBRAE and SPAEP on cross-validation of aerosol transport models implemented in the integral RATEG/SVECHA/GEFEST code with CFD-codes. Also analysis of experimental data for STORM (JRC Ispra) and PSAERO (VTT) installations is proposed based on data provided by collaborators.

Task 6. Experimental investigations of containment parameters impact on volatile iodine content and correlation

Task description and main milestones	Participating Institutions
<p>Experimental investigations of containment parameters impact on volatile iodine content and correlation.</p> <p>Task stages: 1) Completing and adaptation of the experimental installations, methodics perfection, experimental researches of sludges effect (Fe-oxyhydrates and silicates) on volatile iodine species content and correlation in gas and water phases.</p> <p>Tools to solve the task: Autoclave installations with sampling for researches of same iodine species and the water/gas phase iodine partitioning coefficients in the impurities present and the temperature range 20-150⁰C, pressure 0,1-0,8 MPa, γ-irradiation-0-5 kGy/h. Gamma-radiation, facility RChM-γ-20. Methods and sorbents for the determination of iodine species ratio in the water and gas phases. Analytical and chromatographic methods and techniques for the impurities radiolysis products and various iodine species detection; ion-selective and red-ox electrodes, ionometer, spectrophotometer, gas-liquid-chromatography, gamma-spectrometry, iodine sensor and other</p>	<p>1- VNIPIET 2- NITI (Department 5)</p>
Description of deliverables	
1	Reports on experimental tests (Information)
2	Report on experimental researches of sludge effects in water phase on iodine volatility

For assessment of NPP radiation safety under reactor accident conditions and for prediction of volatile radioiodine species release it is necessary to have reliable data on main containment parameters and impurities in water phase influence on iodide-ions oxidation rate, on iodine partition coefficients between water/gas phases, volatile iodine species in gas phase accumulation rate, and on the ratio of inorganic/organic gaseous iodine species, particularly during long-term containment conservation (post-accident period).

Among the main factors providing the iodine safety there are the following: suppression of gaseous iodine species generation, stability of processes of volatile iodine species fixation and trapping, accounting for containment parameters and impurities effects on iodine state and volatility.

Effect of containment parameters, such as temperature, pH and red-ox-potential of water phase, dose rate of gamma-irradiation, was researched enough and correctly interpreted. So iodide-ion oxidation to molecular iodine (volatile species) under gamma-irradiation of aqueous low-acidic solutions depends on iodide-ion oxidation by radiolysis water products (mainly OH-radicals), and oxidation rate depends on dose rate, solution pH, temperature and iodide concentration. Essential dependence iodide oxidation rate from pH was stipulated by participation of H⁺-ions in competing reaction of radiolytical oxidation of ion I⁻ and reduction of I₂ by radicals of hydrogen peroxide. Limitation of I₂ formation would lead to reducing of generation organic iodides RI (R- alkyl-, aryl), thus mainly two processes influence the volatile iodine compounds generation and the ratio of iodine volatile/unvolatile species: water radiolysis, molecular iodine and organic iodide generation. Binding or trapping of elemental iodine are more effective processes in comparison with methyl iodide where trapping is ineffective.

The interaction with surfaces in the containment can influence the iodine volatility due to its temporary hold-up at the surfaces or due to iodine chemical species change-over owing to reactions with surface material components (polymeric paints, steel, sludges).

In realistic severe accident conditions sludge including ferric oxides and hydroxides, silicates and gels of silicium acid, perhaps boron carbide will be accumulated in the containment sump. These impurities can influence the iodine content in the water phase, as they can adsorb and retain the iodide-ions. So sludges can act as effective iodine “sinks” and reduce iodine concentration in water phase; accordingly, the possibility of iodide-ions oxidation and accumulation of volatile iodine species in the gas phase can decrease. Ferric ions (Fe^{3+}) can oxidize iodide ions in solution (at low pH), therefore iodine behaviour in presence of ferric sludges would be predicted with difficulty.

These processes have hardly been studied, data on iodine adsorption are scarce, data on iodine sorbing behavior are in general not available. It is noteworthy to mention that iodine chemistry and behavior under increased temperature (up to 150-200⁰C) have not been studied enough and there were problems in predicting iodine behavior in realistic accident condition.

Experimental programme includes the investigations of influence on iodine volatility ferric and silicate sludges, at varied proportions m/V, in temperature interval from 25 to 100⁰C, pH – from 7-8 to 5, iodine concentration – 10⁻⁵-10⁻⁸ g/l, under γ -irradiation and otherwise.

Ferric oxides sludges impurities are representative for sump water under accident conditions. Ferric ions come into water phase by the various types of steel corrosion (mainly carbon steel) and in a state of aerosols from corium molten pool (core catcher) (sources – “sacrificial” ferric oxide and structure core materials); silicium also can come with aerosols from core catcher (refractory facing) and as impurity from construction materials. It is necessary to determine kinetics and sorption degree of various iodine species for temperature exposure (20-25; 50; 100⁰C) and gamma-irradiation (0; 3 kGy/h).

Proposing matrix of investigations is given in the Table 3. Experiments are performed in autoclave apparatus for researches of iodine mass transfer. Solutions are prepared with high purity water (“nanopure”) and addition of boric acid, KOH and HNO₃ (for pH regulation), CsI (KI). Inner volume of autoclave – up to 2 dm³.

Table 3. Proposed matrix of tests on EVAN-project, Task 6

Test code	Test purpose	System investigated			Medium parameters				Analyzed parameters	Type of technique	Comment
		Water phase	Sludge mass	CsI concentration	T, °C	pH	γ -dose rate	Atmosph.			
I6-B1	Determination of effect of γ -irradiation on sorption by sludge and on iodine volatility in Ar and air atmosphere; investigation of process kinetics, impact of γ -dose	Water – 10 g/l H ₃ BO ₃	0 2.5 g/l	10 ⁻⁵ mole/l	50-60	~5	0 ~1 kGy/h	Argon	pH; E _h ; concentration Γ^- , IO ₃ ⁻ , I ₂ , Fe in water phase; I ₂ in gas phase	pre-test and post- test	Blank-tests with ¹³¹ I; ampoule technique
I6-B2			2.5 g/l				~1 kGy/h Σ 5-10 kGy				
I6-1	Investigation of process kinetics; determination of sorption by sludge and of iodine volatility depending on pH, sludge mass, T, C ₁ .	Water – 10 g/l H ₃ BO ₃	0 2.5 g/l	10 ⁻⁵ mole/l	50-60	~5	~1 kGy/h	Air	pH; E _h ; concentration Γ^- , IO ₃ ⁻ , I ₂ , Fe in water phase; I ₂ in gas phase	pre-test and post- test	Kinetics 1-24 h with ¹³¹ I; ampoule technique
I6-2			0 2.5 g/l	10 ⁻⁵ mole/l	25; 95- 100; 150	~5					
I6-3			2.5 g/l 5.0 g/l	10 ⁻⁷ -10 ⁻⁸ mole/l	50-60	4; 5; 6-7; 8					
I6-4	Determination of iodine sorption by sludge depending on water iodine species, pH and T (no γ -irradiation). Comparison of ampoule and autoclave tests	Water – 10 g/l H ₃ BO ₃	2.5 g/l	10 ⁻⁴ mole/l (CsI, CsI+I ₂)	50-60 95- 100	5; 6- 7; 8	0	Air	pH; E _h ; concentration Γ^- , IO ₃ ⁻ , I ₂ , Fe in water phase; I ₂ in gas phase	pre-test and post- test	Ampoule technique
I6-5	Investigation of iodine sorption by sludge and sludge components depending on content and parameters of water phase	Water – 10 g/l H ₃ BO ₃ (+5·10 ⁻⁵ mole/l H ₂ O ₂)	0 2.5 g/l 5.0 g/l (H ₂ SiO ₃ , FeOOH)	10 ⁻⁴ mole/l (CsI, CsI+I ₂)	25 50-60 95- 100	4; 5; 6-7; 8			pH; E _h ; pI-on- line; concentration Γ^- , IO ₃ ⁻ , I ₂ , Fe in water phase; indication of I ₂ in gas phase	on-line pre-test and post- test	Autoclave technique

Experiments in presence impurities and under γ -irradiation can be performed with ampoule methodic. VNIPIET is performing experiments without irradiation, NITI – with irradiation. At iodine low concentrations ($<10^{-8}$ mol/l) test are performed with ^{131}I . Solution samples are analyzed on iodine total content and ratio of inorganic/organic species. Iodine partition coefficients are calculated from experimental results. Experimental autoclave apparatus would be reconstructed and adapted to experimental matrix conditions. Techniques and analytical instrumental equipment would be checked and adapted to necessary measurement ranges and increased temperatures. Used experimental methods and equipment are given in the Table 4. New experimental results on volatility and iodine speciation would be used to iodine behavior model for its approximation to realistic accident regimes, also for the assessment of iodine environmental source terms for selected accident scenarios.

Table 4.- Methods of iodine analysis

Phase	Component	Analytical technique	Equipment	Sensitivity, mole/l
Water	I^-	I-selective electrode Photometry	Ion meter ANION-4110 photocolorimeter KFK-2MP	$3 \cdot 10^{-7}$ 10^{-6}
	IO_3^-	Photometry	Photocolorimeter	10^{-6}
	I_2	Extraction, photometry	Photocolorimeter, spectrophotometer	10^{-6}
	^{131}I (I^- , IO_3^- , IO_4^-)	Radiochromatography (frontal chromatography at non-organic sorbent) Gamma-spectrometry	Gamma-spectrometer	10^{-8} - 10^{-11}
	pH, E_h	Potentiometry	Ion meter ANION-4110	pH=0.01 $E_h = \pm 1$ mV
	electrical conductance	Conductometry	Conductometers UPK HJ 98309 PWT HJ 98308	0.001 $\mu\text{Sm/cm}$ 0.1 $\mu\text{Sm/cm}$
Water	ΣFe , Fe(III)/Fe(II) soluble forms	Analytical techniques, photometry, membrane filtration	Photocolorimeter, spectrophotometer SF-2,6	10^{-6}
Gas	I_2 (RI)	Sorption at filters assembly, extraction, photometry	Spectrophotometer SF-2,6	1-5 μg of I_2 in sample
	I_2 (RI)	Gas chromatography	Gas chromatograph	10^{-9}
	^{131}I (R ^{131}I)	Collecting at selective sorbents, gamma-spectrometry	Gamma-spectrometer	$1 \cdot 10^{-5}$ Bq/l
	I_2 , HI, CH_3I , HIO_3	Ion Mobility Spectrometry (IMS)	Ion Mobility Spectrometer (prototype)	1 μg in sample

January 2006

This work is supported financially by ISTC and performed under the contract to the International Science and Technology Center (ISTC), Moscow.

Task 7. Numerical and theoretical modeling of containment parameters impact on volatile iodine species content and correlation

Task description and main milestones		Participating Institutions
Numerical and theoretical modeling of containment parameters impact on volatile iodine species content and correlation. Task stages: 1) Iodine model analyses and adaptation, choice of parameters and process constants. 2) Pre-test/post-tests calculation (according to test schedule). Tools to solve the task: 1) Containment iodine behavior model under accident conditions, developed by VNIPIET/SPAEP. 2) Computer codes for calculations of pH and E_{red-ox} in solution; databases on iodine rate and equilibrium constants; published experimental and calculation data; proprietary unpublished data.		1- SPAEP 2- VNIPIET
Description of deliverables		
1	Preliminary report on iodine model adaptation	
2	Pre-test/post-tests calculation results (information)	

The main computer code is based on developed computer model of iodine mass transfer and chemical iodine species (I , I_2 , IO_3^- , CH_3I) permanent partition in systems of /water phase/steam-gas phase/rooms/equipment surfaces/. Calculations are carried out with the available database on rate and equilibrium constants of radiolytical, chemical reactions and iodine mass transfer.

The uncertainty study of the results on volatile iodine species release into containment atmosphere is usually performed on the base of mathematical statistics and probability theory methods. Uncertainty of results is connected with uncertainty of the values used for constants of processes and medium physico-chemical parameters, and also with incompleteness of processes and contaminating substances accounted for. Completeness of considered processes and contaminating substances in gas/water containment phases is to be estimated. It is necessary to range the processes with iodine participation in the containment by their significance for iodine release into gas phase, to determine the confidence intervals for values used and final calculated results, to carry out the correlation analysis of dependence of volatile iodine species release to the containment atmosphere against radiolytical processes with iodine participation and iodine mass transfer rate in the containment.

For pre- and post-tests calculations of effect of water phase impurities on the iodine volatility and iodine speciation in water/gas phases, it is necessary to update the iodine model. Experimental tests are carried out in steady-state condition without mass transfer in other volumes, and at 25 and 50⁰C – without water evaporation and condensation. Types of impurities are also limited. Therefore it is necessary to adapt the iodine model to test conditions. Numerical simulation of reactions and iodine mass transfer under experimental test conditions with database available will be performed. Then the algorithm and computer programme for assessment of iodine concentration and its speciation in water/gas phases and partition coefficients between phases will be developed. Then pre- and post-tests calculations are carried out, from their results the applicability of experimental results for numerical modeling is justified, separate factor sensitivity effect is determined, correctness of iodine model is estimated and degree of uncertainty of used constants is obtained. Then correction of model and computer programme for every tests and for the whole total experimental programme is

carried out. Besides the computer model for assessment of radioiodine accident environmental source term is developed and used for model calculation, significance of influence of impurities and containment medium parameters on iodine concentration in gas phase and on iodine accident source term is assessed.

2. Fulfilled work

2.1 Task 1. Analysis of calculation results for severe accident scenarios.

This report presents principal results of implementation of MNTC Project #3345, Task 1, for quarters 1-X, 2007. The work is executed in FSUE "SPAEP", Saint-Petersburg. The research was focused on analyses & evaluation of beyond design basis severe accidents at NPP-91 featuring VVER-1000 reactors, and designed to be a basis for experiments underway.

2.2 Task 2. Experimental studies of the low-volatile fission products release at the oxidation of suboxidized molten corium.

The present work has been performed in the framework of the project entitled "Ex-vessel Source Term Analysis". Application of the flow method has yielded experimental data on the release of low-volatile fission products (FP), uranium and zirconium from molten corium at different temperatures and degrees of melt oxidation. Partial pressures of components have been studied using high-temperature mass spectrometry.

2.3 Task 3. Analytical investigation of fission products release from molten pool or core catcher .

Molten corium is a very complex high-temperature melt which consists of molten fuel, construction materials (CM) and fission products (FP). In order to describe releases of these substances from the molten corium pool it is necessary to describe two-phase system: melt-gas. Experiments show that in fact the melt itself can split into metal phase and oxide phase, i.e. the system has three phases.

We expect that FPs and CMs will evaporate from the open surface of the melt. In the case of corium in the core-catcher, corium can interact with concrete which may lead to appearance of gas bubbles but our models are applicable only to corium at the bottom of the reactor vessel where no bubbles expected.

At present time two models of FP and CM output are being developed. The first model is based on the assumption that liquid phase can be considered as an atomic mixture with equilibrium state described in terms of the regular solution model. Equilibrium pressures of single-atomic gases over such liquid are considered, and the release rates are calculated using phenomenological Langmuir model. In the other model, equilibrium state of the melt is considered in terms of semi-ideal molecular solution model, and two-phase («liquid-gas») equilibrium is evaluated taking into account the (given) composition of atmosphere over the melt. For simplicity, all kinetic processes,

such as condensed-phase and gas-phase diffusion are neglected, and the rates of FP releases are proportional to partial pressures of FP-bearing gases and to the (given) flow rate of the gas mixture over the melt. In both models the possibility of corium splitting into metal and oxide phases is neglected.

2.4 Task 4. Experimental study of aerosols transport processes in the primary circuit equipment

The research has been carried out within the framework of the project on Ex-vessel Source Term Analysis. Experimental data have been obtained the characteristics of aerosol deposition and resuspension in a riser with ascending air. The investigations of deposition have been performed on liquid aerosol and solid aerosol and those of resuspension – on solid aerosol. Optical methods as well as number of other measurement procedures have been used for measuring flow velocity, size and concentration of aerosol particles.

2.5 Task 5. Theoretical and computer modeling of aerosol transport in the primary circuit

This report presents principal results of implementation of MNTC Project #3345, Task 5, for quarters 1-4, 2007. The work was executed in FSUE "SPAEP", Saint-Petersburg.

The research was focused on analyses & evaluation of beyond design basis severe accidents at NPP-91 featuring VVER-1000 reactors, and designed to be a basis for experiments underway.

2.6 Task 6. Experimental investigation of containment parameters impact on volatile iodine species content and correlation

The work has been fulfilled within the framework of ISTC project #3345 "Ex-vessel Source Term Analysis". There have been received the experimental data on volatile iodine forms release in gas phase at thermal and radiolytic oxidation of iodide ions in aqueous mediums in the range pH 4-8, temperature – 30-120(150) oC, in the presence of admixture sludge containing FeOOH and in its absence. It was assessed the influence of water quality on the volatile iodine forms release.

2.7 Task 7. Numerical and theoretical modeling of containment parameters impact on volatile iodine species content and correlation

Work is executed within the limits of project ISTC #3345 "Ex-vessel Source Term Analysis". The version of iodine module are created and the calculation program for severe accident code modelling behaviour of iodine forms in containment of WWER reactors at a chemical stage of accident with destruction of core. On the basis of autoclave experiments results on influence of temperature, pH and gamma irradiations on formation of volatile iodine forms in water solution of iodide-ion and their output in gas phase verification of the created calculation code is lead, including the same at presence iron hydroxide sludge in water phase.

3. Attachment 1. List of published papers and reports

N ^o	Document	Confidentiality	Title
1	Report	Non-classified	Analysis of calculation results for severe accident scenarios.
2	Report	Non-classified	Experimental studies of the low-volatile fission products release at the oxidation of suboxidized molten corium.
3	Report	Non-classified	Analytical investigation of fission products release from molten pool or core catcher.
4	Report	Non-classified	Experimental study of aerosols transport processes in the primary circuit equipment.
5	Report	Non-classified	Theoretical and computer modeling of aerosol transport in the primary circuit.
6	Report	Non-classified	Experimental investigations of containment parameters impact on volatile iodine content and correlation.
7	Report	Non-classified	Numerical and theoretical modeling of containment parameters impact on volatile iodine species content and correlation.

4. Attachment 2. List of presentations at conferences and meetings

The EVAN ISTC project progress meeting, Saint Petersburg, Russia, September 10, 2007

- A. Frolov Radioactive release during severe beyond-design-basis accidents at nuclear power plants with WWER reactors
- I. Potapov Analysis of FP buildup in fuel, FP release from fuel and molten pool (review of models).
- V. Sidorov Analysis of conditions for release of radioactive sources from reactor plant during severe accidents (LOCA and primary-to-secondary circuit leaks).
- S. Tsaun
- M. Zatevakhin Analysis of aerosol behavior inside containment during severe accidents.
- I. Ivkov
- S. Beshta Experimental studies of fission product release from molten pool / core catcher
- O. Tarasov Theoretical analysis and computation of fission product release from molten pool / core catcher
- O. Krektunov Result of an experimental study of aerodynamic characteristics and processes of liquid aerosol transport in a vertical tube
- V. Alipchenkov Pre- and post-test analysis of experimental findings obtained at ATF facility
- N. Ampelogova Experimental studies of effects produced by in-containment parameters on content and ratio of volatile forms of iodine
- Yu. Bobrov Analytical modeling of effects produced by in-containment parameters on content and ratio of volatile forms of iodine. Pre- and post-test calculations.

Yu. Gorbachev “Modeling of particle carryover from a surface in turbulent flow”
A. Ignatiev “Numerical simulation of aerosol particles transport and deposition based on 3D hydrodynamic codes

The EVAN ISTC project progress meeting, Budapest, Hungary, March 04, 2008

V. Sidorov Task 1. Assessment of results of severe accident sequences modeling
S. Bechta Task 2. Experimental research on FP release from molten pool/core melt catcher
O. Tarasov Task 3. Analytical investigation of fission products release from molten pool or core catcher
M. Lebedev Task 4. Experimental study of processes aerosols deposition and resuspension in the primary circuit equipment
A. Ignatiev, Yu. Gorbachov Task 5. Theoretical and computer modeling of aerosol transport in the primary circuit
N. Ampelogova Task 6. Experimental investigations of containment parameters impact on volatile iodine content and correlation
N. Ampelogova, L. Lebedev Task 7. Numerical and theoretical modeling of containment parameters impact on volatile iodine species content and correlation

V. Bezlepkin .“Status of the ISTC project #3345 “Ex-vessel source term analysis” (EVAN)”, phase 1”. 11th Meeting of Contact Expert Group on Severe Accident Management (CEG-SAM). March 6-9, 2007 Dresden

V. Bezlepkin. Status of the ISTC project #3345 “Ex-vessel source term analysis” (EVAN)”, phase 1 (SPAEP). 12th Meeting of Contact Expert Group on Severe Accident Management (CEG-SAM), September 11-13, 2007, St. Petersburg

V. Bezlepkin. Final report on the ISTC project #3345 “Ex-vessel source term analysis” (EVAN)”, phase 1. 13th Meeting of Contact Expert Group on Severe Accident Management (CEG-SAM), Budapest, Hungary, Hungarian Academy of Sciences KFKI, Atomic Energy Research Institute AEKI, March 5-7, 2008

5. Attachment 3. Information on patents and copy rights

Requests for patents and copy rights have not been submitted.

6. Attachment 4. Final Project Technical Report for Task 1.

Source Term Assessment at Ex-vessel Stage of Severe Accident

Task 1 Final Report

Analysis of calculation results for severe accident scenarios.




1/01/07 – 31/12/07

7. Attachment 5. Final Project Technical Report for Task 2.

Source Term Assessment at Ex-vessel Stage of Severe Accident

Task 2 Final Report

Experimental studies of the low-volatile fission products release at the oxidation of suboxidized molten corium. 

1/01/07 – 31/12/07

8. Attachment 6. Final Project Technical Report for Task 3.

Source Term Assessment at Ex-vessel Stage of Severe Accident

Task 3 Final Report

“Analytical investigation of fission products release from molten pool or core catcher”




1/01/07 – 31/12/07

9. Attachment 7. Final Project Technical Report for Task 4.

Source Term Assessment at Ex-vessel Stage of Severe Accident

Task 4 Final Report

**Experimental study of aerosols transport processes
in the primary circuit equipment** 

1/01/07 – 31/12/07

10. Attachment 8. Final Project Technical Report for Task 5.**Source Term Assessment at Ex-vessel Stage of Severe Accident**

Task 5 Final Report

Numerical and theoretical investigation of aerosol transport in the primary circuit. 

01/01/07 – 31/12/07

11. Attachment 9. Final Project Technical Report for Task 6.

Source Term Assessment at Ex-vessel Stage of Severe Accident

Task 6 Final Report

**Experimental investigation of containment parameters
impact on volatile iodine species content and correlation**




1/01/07 – 31/12/07

12. Attachment 10. Final Project Technical Report for Task 7.

Source Term Assessment at Ex-vessel Stage of Severe Accident

Task 7 Final Report

Numerical and theoretical modeling of containment parameters impact on volatile iodine species content and correlation 

1/01/07 – 31/12/07