Cooperation between ISTC and SARNET in the Source Term Area

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SUMMARY

The main goal of the International Science and Technology Centre (ISTC) is to provide Russian and Commonwealth of Independent States (CIS) weapon scientists with opportunities to redirect their talents to peaceful activities, e.g., non-proliferation through scientific cooperation by integration of Russian and CIS scientists into the international scientific community. The ISTC projects and proposals are tightly linked with ongoing European Framework Programmes, e.g., with SARNET in the Source Term and Corium areas. This paper considers activities relevant to the Source Term topic.

In the “VVER-QUENCH” project #1648.2 a series of small-scale tests with evaluation of the failure characteristics of VVER spent fuel rods and determination of hydrogen and fission product release into the gas phase during high-temperature quenching, is carried out by RIAR with analytical support of IBRAE.

The “Ex-vessel Source Term Analysis” (EVAN) project #3345 carried out by SPAEP includes theoretical and experimental research into the processes affecting late-phase fission product release into the PWR containment atmosphere at the ex-vessel stage of a hypothetical severe accident with core meltdown.

In the frame of the VERONIKA proposal, investigations on fission product release from high burn-up VVER fuel annealed in the temperature range between 1400 and 2300°C under various oxidising and reducing environmental conditions and development of physical models and numerical codes (e.g. MFPR), are planned by RIAR and IBRAE.

All these activities are carried out in collaboration with SARNET partners. This paper summarises the content and status of these projects, and highlights the importance of the ISTC/SARNET collaboration.

A. INTRODUCTION

The International Science and Technology Center (ISTC) [1] is an international organization dedicated to the non-proliferation of weapons and technologies of mass destruction. Founded in 1992 by an International agreement by the European Union, Japan, Russia, the USA and in 2004 by Canada. The main goal of ISTC is to provide Russian and CIS weapon scientists opportunities to redirect their talents to peaceful activities, e.g., non-proliferation through science cooperation by integration of Russian and CIS scientists into the international scientific community. The ISTC projects are often linked with ongoing European Framework Programmes and thus foster the interaction between Russian and European scientists. The expertise of the CEG-SAM (Contact Expert Group on Severe Accident Management) members supported by external EC-SARNET experts to select ISTC project proposals and to provide technical assistance as foreign collaborators is an important asset. The scope of the ISTC projects covers matters of interest in the Source Term [2] and Corium areas.

This paper illustrates three examples of this cooperation between ISTC and SARNET in source term related projects, namely #1648.2 (VVER-QUENCH), #3345 (EVAN) and the proposal VERONIKA.

B. SOURCE TERM RELATED PROJECTS IN ISTC

B.1. Project #1648.2 "Examination of VVER Fuel Behaviour under Severe Accident Conditions. Quench Stage" (VVER-QUENCH)

Leading Institute: SSC RIAR (State Scientific Center "Research Institute of Atomic Reactors")
Supporting Institutes: IBRAE (Nuclear Safety Institute of Russian Academy of Sciences), JSC MSZ (Joint Stock Company "MASHINOSTROITELNY ZAVOD")

In the VVER-QUENCH project carried out by RIAR (experiments) with analytical support of IBRAE, besides one large-scale quenching test at the Forschungszentrum Karlsruhe (FZK) QUENCH bundle facility (QUENCH-12), a series of small-scale tests with evaluation of failure character of pre-oxidized VVER spent fuel rods, determination of hydrogen and gaseous fission product (FP) release into gas phase during quenching, is carried out. The project results are of interest in both the Corium and Source Term areas of SARNET.

The project consists of three stages:

- Stage 1. Study of the irradiated fuel rod segment behaviour under reflood conditions to determine the hydrogen generation and fission product release;
- Stage 2. Conduct of one integral quench experiment with 31 VVER fuel element simulators;
- Stage 3. Development of models and codes to describe VVER core behaviour under severe accident reflood conditions.

Altogether 10 quench tests with unirradiated fuel rod simulators with cladding made from E110 alloy have been conducted in the temperature range between 1400 and 1700°C under hot-cell conditions (cladding oxidation in steam) and conditions outside of hot cells (cladding oxidation in an argon/oxygen mixture). The results of hydrogen release measurements during preliminary claddings oxidation and in the quenching stage were analysed. The test matrix was intentionally chosen similar to that used in the previous FZK tests with PWR fuel rod simulators. Post-test metallographic examinations included an estimation of the simulator cladding state, cladding and oxide layer microstructure and fuel cladding interaction area. There was good agreement with the results obtained at FZK in the tests with fuel rod simulators with Zircaloy (Zry) cladding.

Important observations concern the post-test brittle state of the cladding. It was shown that through-wall cracks (network or single cracks) in the cladding appear under the same conditions (cladding pre-oxidation and quenching temperature) as in the FZK tests. However, unlike the experiments in FZK, no surface oxidation of the brittle cracks occurred at quenching.

One of the primary objectives of the tests with un-irradiated simulators was to check the working capacity of the test rig and create the data base for comparison with the irradiated fuel rod simulator tests. Firstly, three tests using irradiated fuel rod segments, re-fabricated from VVER fuel rods at burn-ups of 54 and 65 MWd/kgU, were performed in the temperature range of 1400 - 1700°C with the simulator surface oxide film thickness from 0 to 200 μm. The following parameters were measured during the tests:

- Hydrogen generation during preliminary oxidation and at the quenching stage;
- Gaseous fission product release from fuel;
- Concentration of caesium in the water of the cooling tank of the rig;
- Concentration of caesium in the steam condenser water.

Post-test examinations of the simulators were carried out:

- Visual inspection and photographing of simulator surfaces;
- The gamma-scanning of the simulators along the fuel stack;
- Preparation of cross-section and metallographic examinations of the fuel and cladding structure.

As a result of post-test examinations the following data were obtained:

- Integrated caesium release from the tested simulators as a function of test temperature;
- Thickness and structure of the oxide film on the external and internal clad surfaces.

Results of the tests revealed some differences with those from the unirradiated fuel rod simulator tests:

a) the increased hydrogen generation during the test, that may be attributed to the release of hydrogen accumulated in the cladding during the base irradiation;

b) intensive α-Zr(O) layer formation on the cladding inner surface due to hard fuel-cladding contact that leads to enhanced cladding embrittlement in comparison with the unirradiated simulators tested under similar conditions.

The single rod code SVECHA/QUENCH (S/Q) with advanced mechanistic models for the cladding physico-chemical interactions and thermo-mechanical behaviour, which was designed for modelling of the previous FZK quenching tests with Zry cladding, was adapted to simulate the RIAR tests. The database for Zr-1%Nb material properties was obtained from the previous RIAR tests on cladding oxidation kinetics and mechanical deformation behaviour at high temperatures. The modified S/Q code well predicts the temperature evolution during quenching, the maximum extent of cladding oxidation and the final mechanical state of the oxidized Zr-1%Nb cladding in the tests with fresh uranium fuel rods.

The S/Q code was extended for simulation of the RIAR quenching tests with irradiated fuel rods. Analysis and simulations of the tests show that the gap collapse and chemical interaction between the irradiated fuel pellets and the cladding due to swelling leads to additional embrittlement of the cladding. This effect along with a possible starvation in the tests prevent irradiated VVER fuel rods from formation of a network of through-wall cracks, however, this does not prevent failure of the quenched specimens under handling.

For analysis of the fission products release in the RIAR quenching tests the MFPR code (developed by IBRAE in collaboration with IRSN) was applied. The code was successfully validated against earlier RIAR tests on data on release of various fission products (basically, Xe, Kr and Cs) from irradiated VVER fuel with various burnups during high-temperature annealing. The first results of MFPR calculations of Cs release from the new RIAR quenching tests are in reasonable agreement with the test measurements.

B.2. Project #3345 “Ex-Vessel Source Term Analysis” (EVAN)

Leading Institute:
SPAEP (Saint Peterburg Research and Design Institute ATOMENERGOPROEKT)
Supporting Institutes:
VNIPIET (All-Russian Research and Designing Institute of Complex Energetic Technology),
IBRAE (Nuclear Safety Institute of Russian Academy of Sciences),
NPO CKTI (Joint Stock Company «I.I.Polzunov Scientific and Design Association on Research and Design of Power Equipment»),
NITI (A.P. Alexandrov Research Institute of Technology).

The project includes theoretical and experimental research on the processes affecting the late phase fission product release into the containment atmosphere. This stage is characterised by corium melt release from the reactor pressure vessel into the containment compartment. At this stage, the fission products are released into the containment atmosphere from the core melt located in the reactor cavity/lower containment compartment, along with various secondary sources like contaminated solution in the containment sump and FPs deposited at the surfaces of process equipment and building structures. Ex-vessel core melt fission product release is affected by design features and accident management strategy. The assessment of radiological consequences for severe accidents includes the determination of fission product release into the containment atmosphere and time-dependent and physico-chemical composition of the source term to the environment. The project benefited from review in the SARNET source term area, communicated through the CEG-SAM group.

The project is divided into four work packages consisting of altogether seven tasks:

- WP1. Analysis of severe accident scenarios (SPAEP, IBRAE);
- WP2. Experimental and theoretical investigations on fission product release from molten corium pools (NITI, IBRAE);
- WP3. Experimental and theoretical investigation of the aerosol transport, deposition and re-vaporisation in the primary circuit pipes (NPO CKTI, SPAEP, IBRAE);
- WP4. Experimental and theoretical investigations on the impact of containment parameters on the behaviour of iodine species (VNIPIET, SPAEP).

The overall project is divided into two phases:

- Phase 1 (1 year) comprises the mounting, installation and conduct of pilot experiments;
- Phase 2 (2 years) covers a more detailed testing and analysis of phenomena based on the evaluation of the results obtained in phase 1.

WP1: The goal of the severe accident analysis for various designs of nuclear power plant (NPP) of PWR and VVER types is to determine the representative boundary ranges for the fluid parameters in the reactor and containment, the core melt parameters, FP aerosol characteristics, boundary conditions at the surfaces of structures and equipment, sump solution chemical composition, dose rate level, and other parameters necessary for specification of the experimental conditions in the three other Work Packages.

WP2: Experimental investigations of fission product release from the molten pool/core catcher are to be carried out in the tests with model corium compositions, to determine:

1) Low-volatile fission product release from the molten pool during its transition in inert and oxidizing atmospheres;

2) Fission product release from the molten pool with water supply onto the melt surface (for pure water and water contaminated with FP species).

The extent of melt oxidation is important for the low-volatile oxidizing FP release, especially Ru, Ba, and Mo, whereas water supply onto the melt can effectively reduce FP transport from the melt to the atmosphere, though, it can generate a secondary source of soluble FP (Cs, I, Ru) from contaminated solutions boiling at the melt surface.

The goal of theoretical and numerical analysis of fission product release from the molten pool/core catcher is to confirm applicability of the obtained experimental results for computer model validation.

WP3: Fission products deposited at the in-vessel stage of an accident in the primary circuit (vertical, horizontal sections, tube bends and so on, including heat transfer surfaces of steam generators) can be resuspended/revaporised and present an important source in the ex-vessel stage.

Experimental investigations of aerosol transport in the primary circuit pipes are to be carried out in the tests with different aerosol types. Deposition and resuspension behaviour of a typical aerosol (e.g. CsI) and an inert reference monodisperse aerosol (e.g. Cu) will be examined in the reference case (horizontal or vertical tubes), and then in various options for chosen tube diameters (ranging from about 10 to 100 mm) and flow rates in a wide Reynolds number (Re) range 4000-10000, in an inert atmosphere under normal conditions. Investigations of deposition under normal conditions as well as revaporisation of Cs-bearing compounds (at heating of pipe surfaces up to 600-700 °C) in a steam atmosphere may be carried out in the second stage of the project.

Theoretical and numerical modelling of deposition, transport and revaporisation of aerosols in the primary circuit pipes includes: 1) analysis of the main processes of aerosol spectra formation and formation of fission products in the flow taking into account of vaporization, condensation and turbulent mass transfer; 2) pre-test calculations using integral thermo-hydraulic and CFD-codes, with and without aerosol transport models; 3) post-test calculations with consideration of deposition and revaporisation of Cs compounds; 4) analysis of calculation results and development of recommendations for models of aerosol and fission products transport in the primary circuit under severe accident conditions for implementation in integral severe accident codes.

Aerosol transport calculations are performed using the base models implemented in the SOCRAT (or RATEG/SVECHA) code. Cross-verification calculations with CFD codes are additionally carried out.

WP4: For various severe accident management strategies, the containment sump is often used for long-term heat removal from the melt and from the containment. Investigation on how different chemical species in the sump solution (boric acid, Fe oxides and organic forms) affect concentrations and partitioning of the iodine species is very important for radioiodine source term predictions.

Experimental investigations include tests on the effect of impurities coming out into the containment water under emergency conditions, on the content of I_2 in the solution and on volatile iodine species in the gas phase. Ferric/ferrous ions and silicates can adsorb ions of I^- and O_3^-, keeping them in the compound and affecting the rate of I_2 -formation in the solution as well as concentration and partition of organic and inorganic volatile iodine species in the gas phase. Organic impurities, polymeric coating and cable insulation materials present in the water also substantially affect the volatile iodine species production, since thermo-radiolysis of these materials results in formation of organic acids, chloride ions and chlorine derivatives, as well as in reduction of pH-level and volatile and non-volatile organic iodide formation.

Temperature, γ-irradiation dose-rate, pH-level and iodine concentration in the water phase also affect the iodine volatility. The main purpose of investigations is to determine the complex effect of the above-mentioned factors on concentration and partition of the volatile iodine species in the gas and water phases, partitioning coefficient for different iodine phases and accumulation rate of different iodine species in the containment atmosphere.

Theoretical activities include adaptation of the earlier developed model for iodine species behaviour in the containment atmosphere under emergency conditions to the conditions of the new tests and development of a model for accident environmental radioiodine source term assessment. Using the developed iodine species behaviour model and the computer code, pre-test and post-test calculations of iodine behaviour and volatility are to be carried out, and the significance of the different factors will be determined. The model and the computer code will be adapted to the test conditions, applicability of the obtained results to iodine dynamics computer modelling has to be justified, adequacy of the iodine species behaviour model will be analyzed and uncertainties of the used constants will be estimated.

B.3. Project proposal "VVER Experiments on Release due to Over-heating: Normalization and Knowledge Augmentation" (VERONIKA)

Leading Institute: SSC RIAR (State Scientific Center "Research Institute of Atomic Reactors")

Supporting Institute: IBRAE (Nuclear Safety Institute of Russian Academy of Sciences)

In the frame of the VERONIKA proposal (by RIAR and IBRAE), investigations on fission product release from high burn-up fuel annealed under steam, steam-hydrogen and hydrogen atmosphere conditions are planned. The objective of the proposed project is to obtain experimental data on the release of fission products (Kr, Xe, I, Cs, Ru, Ce, Mo, Ba, Zr) from highly-irradiated VVER fuel of 60MWd/kgU in the temperature range between 1400 and 2300°C. The results will be used to develop, validate and improve physical models and numerical codes to describe the high burn-up fuel behaviour and fission product release under severe accident conditions (e.g. Model for Fission Products Release (MFPR)). In contrast to earlier similar tests (VERCORS), it is planned to perform comparative tests with and without cladding, as well as turning off the heating at intermediate temperatures before fuel collapse, in order to analyze thoroughly the fuel microstructure and fission product distribution at each stage. Both pre- and post-test examinations will be performed for detailed fuel microstructure characterization, including:

- Optical metallography (grain size, porosity, gas swelling);
- EPMA and SEM analysis (local content and radial distribution of FPs, composition of solid precipitates in the fuel).

It is planned to conduct the proposed project in 2 phases of altogether 5.5 years duration. In the first phase of the project (3 years) altogether 10 experiments in pure steam and in an inert-reducing atmosphere will be performed. In the second phase (2.5 years) 10 tests in hydrogen/argon and air/argon mixtures are planned. The experimental matrix may be corrected, if necessary, on the basis of the first phase's results and results of other related European projects (the completed VERCORS series, and the future VERDON experiments, performed by CEA). This approach will provide the important feedback necessary for long-term projects.

An extensive review by EC-SARNET partners in the Source Term area was conducted on the VERONIKA proposal resulting in several recommendations regarding the work programme. The essential recommendations have been to address in the experimental investigations the whole range of possible oxygen potentials, to measure the oxidation state of the clad before the test, to characterize well the fuel properties and the pre- and re-irradiation histories, and to use B_4C and steel next to the fuel sample to study the possible enhancement of released fission products. The results will contribute valuable source term information concerning air-ingress scenarios. VERONIKA will supplement the completed VERCORS and future VERDON programmes interpreted and planned in the SARNET frame.

C. Concluding Remarks

This paper has summarised the main results of the VVER-QUENCH and EVAN projects along with their forward planning, and also the VERONIKA proposal at present under detailed consideration for execution in the near future. The projects promotes the ISTC goals to let Russian weapon scientists and specialists to apply their knowledge to peaceful applications, to facilitate their integration to the global NPP accident analysis community, and also to support the applied research in the field of environment protection, energy generation, and nuclear safety. The work on the projects is carried out in close cooperation with foreign collaborators from EC (JRC), Germany (FZK, GRS), France (CEA, IRSN), Switzerland (PSI) and other countries. The results of all these activities are and will be of considerable mutual benefit to ISTC and SARNET members. It is foreseen that this fruitful cooperation will continue into the future.

References

[1] http://www.istc.ru/ .

[2] T. Haste, P. Giordano, L. Herranz and J.-C. Micaelli, "SARNET: Integrating Severe Accident Research in Europe - Safety Issues in the Source Term Area", International Conference on Advances in Nuclear Power Plants (ICAPP'06), Reno, 4-8 June 2006.