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ON THE PROGRESS MADE IN SAFETY ASSESSMENT AND SEVERE ACCIDENT MANAGEMENT AS PART OF THE FRENCH FUKUSHIMA POST-ACCIDENT RESEARCH PROGRAMME

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ABSTRACT

The Fukushima-Daiichi accidents in 2011 underlined the importance of severe accident management (SAM), including external events, in nuclear power plants (NPP) and the need of deepen safety research implementing efficient mitigation strategies. For these reasons, the French General Commissariat for Investment has released significant additional resources to strengthen research in this area. Within this framework, several projects have been launched on the highest priority issues, in order to fill the knowledge gap and reduce the uncertainties still hanging over the main phenomena governing severe accidents and their management.

Among them, the PERFROI, DENOPI, ICE, MITHYGENE and MIRE projects, led by IRSN, addressed remaining issues related respectively to fuel degradation and reflooding, spent fuel loss of cooling, fuelcoolant interaction, hydrogen risk and source term mitigation. For each of the projects mentioned, experimental and analytical activities were carried out to help understand possible accident scenarios and related phenomena, and to contribute to improving the safety of existing and future reactors.

This article gives an overview of the main results of these projects. The achievements in terms of modelling severe accidents and improving severe accident management are presented and their applicability to innovative reactors, including SMRs, is discussed.

KEYWORDS

Reflooding, spent fuel, ICE, hydrogen, source term

1. INTRODUCTION

Despite accident prevention measures, including design modification and operating procedures, used in the nuclear power plants (NPP) under operation, some accidents, within very low probability, may evolve into severe accidents with core melting and plant damage and lead to release and dispersion of radioactive materials into the environment, thus constituting a danger for the public health and for the environment. This risk was unfortunately evidenced by the Fukushima Daiichi accidents in Japan in March 2011, which

underlined the importance of severe accident management and the need to implement and to improve the corresponding mitigation strategies and systems. To this end, the French General Commissariat for Investment has released, in 2012, significant additional resources to strengthen research in this area. Within this framework, the PERFROI, DENOPI, ICE, MITHYGENE and MIRE projects, led by IRSN, were carried out with the objective to address remaining issues related respectively to fuel degradation and reflooding, spent fuel loss of cooling, fuel-coolant interaction, hydrogen risk and source term mitigation.

For each of the projects mentioned, experimental and analytical activities have been carried out, leading to a better understanding of the phenomena addressed and improvements to the corresponding models. These projects have also led to the development of new instruments, reliable under severe accident conditions, and the development of a coherent experimental database for the validation of LP (Lumped Parameter) and CFD (computational Fluid Dynamic) codes.

In the following, each of the mentioned projects is briefly described and main achievements presented. The last section discusses the relevance of the obtained results to the advanced reactors as water cooled SMRs.

2. PERFROI project

During a loss of coolant accident (LOCA), the fuel assemblies may dry out partially or even completely. Under these conditions, the rise in fuel temperature and drop in coolant pressure in the reactor core can lead to significant deformation and breakage of the fuel rod cladding. Fuel fragmentation can occur, causing the fuel to move within the ballooned zone. Depending on the size and distribution of the ballooned areas in the fuel bundle, the cooling flow can be disrupted, which can compromise the cooling capacity of the fuel assembly and, by extension, the cooling capacity of the reactor core. The PERFROI [1] general objective is to study the coolability of a partially deformed core in the event of a loss of coolant accident (LOCA) addressing the two following objectives:

- **Objective 1.** concerns the study of the thermomechanical deformation mechanism and fuel rods burst under LOCA conditions. To this end, separate effects and integral were carried out respectively in the ELFE¹ and the COCAGNE² facilities (Figure 1. Illustration of the COCAGNE facility.Figure 1)
- **Objective 2.** aims to characterize the thermal hydraulics behavior in the ballooned area, during cooling phase by water injection considering the effect of pressure and the water flow rate. To this end, dedicated experimental campaign was carried in the COAL facility (Figure 3).

In the ELFE facility, several creep tests were carried out over a wide temperature range from 700 to 1030°C. The samples used were manufactured either in Zircaloy or in M5 alloy. The tests carried out enabled new strain rate values to be obtained for intact, pre-oxidized and pre-hydrated Zircaloy samples. The results obtained are in line with those reported in the literature [2]. The tests carried out showed that the creep rate decreases with the presence of a pre-oxide layer for low stresses, and it increases slightly with the hydrogen content [3]. However, the effect remains weak for stresses that are representative of the LOCA conditions. Tests were also carried out in a frame of a PhD [4] to get additional information on thermal mechanical properties on cladding including Hydrogen.

In the COCAGNE facility, the aim of the test campaign was to study the behavior of sections of Zircaloy-4 rods during ballooning of the cladding, considering contacts between rods. To this end, various conditions were considered: internal pressure varying from 20 to 100 bar; temperature increase ramp of 1 to 5°C/s or state of the sample: single or double-sided oxidation.

The main outcomes of these experimental campaigns [5] can be summarized as follow:

¹ (Etablissement des Lois de FluagE) device on IRSN's MAESTRO experimental platform at Cadarache

² (COmportement d'un Crayon en APRP soumis à un GradieNt de températurE) device on IRSN's MAESTRO experimental platform at Cadarache

- Increasing the internal pressure increases the deformation at rupture, as well as the temperature at the moment of bursting. On the other hand, double-sided oxidation leads to less deformation of the rod;
- The observed opening formed on the burst balloons have the shape of an "elongated fish mouth" for tests performed at 20 and 50 bar and a rectangular shape for tests performed at 100 bar;
- There is a linear dependence between the size of the fishmouth-shaped openings formed in the test carried out at 5 and 10°C/s.
- The axial deformation profiles of the rods revealed balloon lengths with radial deformations (example in Figure 2) greater than 41%, corresponding to a clogging rate of the passage section of 80%, and deformation peaks of up to 65 and 80%. A radial deformation of 50% corresponds to a clogging rate of 90%.

These COCAGNE thermomechanical tests therefore validate the choices of clogging rates and cluster rod balloon lengths for the COAL experimental campaign.



 Figure 1. Illustration of the COCAGNE facility.
 Figure 2. typical results of rod deformation during COCAGNE experiments.

In the COAL facility (Figure 3), the objective followed was to investigate the antagonistic influences that may have the presence of a blockage induced by the rods ballooning on the flow. On one hand, the reduction of the section between the deformed fuel rods will increase the flow velocity [6]. On the other hand, the flow bypass [7] from the deformed fuel rods (Figure 4) in the blocked region to the peripheral intact rods (crossflow phenomena), will lead to a reduction of mass flowrate.

82 experiments were performed (28 tests for B0 reference campaign in 2020, 26 tests for B1 campaign in 2021 and 28 tests for B2 campaign in 2022) to investigate the different physical main inlet parameters considering a wide range of parameter pressure (2, 3, 5, 10, 15, 20 and 30 bar), water injection speed (17, 20, 25, 30, 36, 50 and 80 kg/m2/s) and rod power (1.5, 2, 2.4 and 3.3 kW per rod). B1 and B2 campaigns were performed with bundles including a partially blocked by ballooned rods at a blockage ratio respectively 80% and 90% (length of 100 mm and 300 mm), including a local power in the balloons of factor 1.5 with respect to the normal cosine profile in order to take into account the fuel relocation in the balloons, more representative to irradiated fuel rods with fragmented pellets.

These COAL experiments [8] highlighted very important results concerning the duration of reflooding and the impact on the local cladding temperature peak due to the presence of a plug in a fuel assembly (see Figure 4 a). The maximum temperature (1105° C) was obtained during the test (Figure 4b) from [9] performed at low pressure (0.3 MPa – 3 bar) with the maximum power per rod (3.3 kW) and a medium water flow rate (25 kg/m²/s, i.e 2.5 cm/s) for the B1 configuration.

We succeed to perform high pressure tests from 1 up to 3 MPa which was one of the major objectives of the COAL experiments to investigate the coolability for Medium Break size LOCA conditions.



These COAL experiments outlined very important results regarding the duration of the reflooding and the impact on the PCT (increase of peak cladding temperature) due to the presence of a blockage inside the fuel assembly. These experiments will provide thermal hydraulic information needed to improve the codes modelling during the reflooding under LOCA sequence, with the thermal behavior of the rods in a well define boundary conditions (heated shroud at well controlled temperature) and all the hydraulic parameters such as the water (as droplets) and the steam flow at the outlet of the test section. Once the simulation tools validated in such complex configuration, there may be used for safety study in real nuclear reactor conditions.

3. DENOPI project

Following the Fukushima-Daiichi accident, the international community identified R&D needs regarding loss of cooling or coolant accident in Spent Fuel Pool (SFP) [10]. To enhance knowledge on accidental SFP conditions and to better evaluate the safety margins, the DENOPI project was launched aiming at improving the knowledge of thermal hydraulics phenomena involved in SFP accident prior to the severe accident phase, to evaluate the efficiency of water spray system as mitigation means³ and to analyze the fuel clad oxidation. For these purposes, experimental campaigns had been conducted considering different time frames of the LOCA accident and different scales of interest providing then valuable data for model development and code validation. Thus, several facilities with different scales were used at clad scale using standard laboratory equipment, at the fuel assembly scale, in ASPIC facility (see Figure 9), and at integral scale with pool, in the MIDI facility (see Figure 5).

3.1. Thermal hydraulics in a large basin: MIDI

The MIDI facility is dedicated to the study of thermal-hydraulic phenomena on the full scale of a SFP in the event of a loss-of-coolant accident by contributing to better understanding of natural convection flows and closely coupled vaporization regimes. Three main modes of water vaporization are likely to occur in a pool with a heater located at its lower end, such as spent fuel pools: evaporation at the free surface, pool flutter and nucleate boiling inside the fuel assemblies. Their appearance and potential predominance are closely linked to large-scale natural circulation in the pool.

³ Fuel degradation and fission products release are not addressed in the framework of the DENOPI project

The 11th European Review Meeting on Severe Accident Research (ERMSAR2024) KTH, Stockholm, Sweden, May 13-16, 2024

The MIDI facility is designed as a scale model of a typical French SFP, with a ratio of 1:6 for the free surface and 1:3 for the height of the pool [11] (see Figure 5) and is well instrumented to measure heat and mass transfer. This facility can be operated at different stages of the accident by varying parameters such as the initial pool level (from 0.5 to 4.5 meters above the simulated 3X7 cell storage rack, see figure Figure 6), the loading pattern into the storage rack or the overall power (from 0 to 50 kW per cell depending on the residual power distribution in an SFP) and the maximum overall heating of 300 kW.



The performed experiments confirmed the anticipated phenomenology with the competition during the transient of three modes of vaporization: evaporation at the free surface, boiling in the fuel bundle and "flashing" of superheated water above the racks. The "flashing" vaporization mode is highlighted in a 3D geometry such as a parallelepiped pool. Under MIDI test conditions, in addition to the evaporation that exists throughout the transient, "flashing" is the vaporization mode which participates the longest time to mass loss. Indeed, boiling begins to contribute to the loss of mass only at the end of the transient when the water level is approximately one meter above of the rack. The existence of a gravity flow of superheated water in a pool heated from below was demonstrated in the DENOPI experiment. The phenomenon would preferably occur on unheated immersed surfaces such as the side walls, which would develop in the mass of liquid. Secondly, it was pointed out that the degassing process is generally completed before the liquid flashes/boils-off, and that it is maintained for a short time at that point. The tests highlighted the important role of certain parameters in the onset of flashing, in particular dissolved gases when the liquid begins to overheat. or the above free surface conditions.

Based on the obtained experimental data, analytical modelling of the vapor diffusion process was proposed as well as the free surface evaporation, degassing of dissolved non-condensable species and bubbles escape processes. A vaporization regimes map as a function of the residual power and water level was also dressed (see Figure 7).

3.2. Thermal hydraulics in a spent fuel assembly: ASPIC

The second topic addressed in the framework of the DENOPI project concerns the coolability of an assembly during or after dewatering but before fuel clad degradation. The ASPIC facility is designed to study, at the assembly scale, different SFP accidents scenarios during partial or total fuel assembly uncovery. The ASPIC is a full height electrically heated 17x17 rod bundle (4 meters) in a cell (equivalent to a cell of a storage rack in SFP) (see Figure 8 and Figure 9. The assembly has the full geometrical characteristics of a standard PWR 900MWe assembly with prototypical top nozzle, spacer grids, and mixing grids [11]. Experimentation on the scale of the assembly makes it possible to study the thermal-hydraulics parametrically and to carry out different scenarios.



To assess the cooling efficiency of the spray, experimental campaign was carried out by varying the following parameters: the residual power of the assembly (10, 40 and 60 kW - 60 kW corresponding to a hot assembly according to the safety reports of the French PWR900), the height dewatered (1 m or 2 m), the mass flow rate of the sprinkler (10, 15, 20 and 25 g/s – 25 g/s to compensate for the steam production generated by a assembly of 60 kW), the sprinkling temperature (80 °C) and the criterion of spraying trigger it the water temperature (200, 300°C and 400°C). The obtained results show that on one hand the spray temperature does not have a significant impact on the temperature evolution in the assembly and on the other hand, the spray flow rate significantly modifies the kinetics of the temperature of the assembly (see Figure 10). Moreover, the test campaign outlined that for 20kW of residual power and one meter of dewatering, the spray flow rate of 10g/s maintained the temperature of the assembly below 500°C. Within, DENOPI projects provides first answers on the aspersion efficiency to cool of an assembly, further investigations are needed to determine the assembly coolability limit considering spray as mitigation means.

3.3. Oxidation of the clad in accidental condition

After assembly uncovering, the exothermic oxidation of the zirconium alloy cladding material becomes significant and contribute noticeably to the overall heating power and lead to hydrogen production. In the framework of DENOPI, Zircaloy 4 oxidation kinetics laws under mixed air-steam atmosphere had been studied with the objective to understand the protective effect (or its absence) of the pre-existent oxide layer formed during in-reactor irradiation (called pre-oxide hereafter). A particular attention was paid to the effect of a low temperature pre-oxidation scale that as far as possible simulates the corrosion scale formed in reactor during in-service operation.



The performed experiments highlighted the effect of defects or singularities of the pre-oxide [12]. Actually, it was observed that oxidation begins from the edges. To discard experiment artefact, tests were performed with Zy-4 spherical beads to eliminate edges. The protective effect is then linked to the presence of pre-existing defects. It is expected that in real fuel rods, such defects might be preexisting or created by clad swelling due to heat-up. Finally, a kinetic model considering the influence of the partial pressure of oxygen and steam has been developed for implementation in the ASTEC code developed by IRSN.

4. ICE project

The goal of the ICE project is to improve the knowledge and modeling the interaction between the fuel and the coolant during a melt relocation phase in the event of a nuclear reactor core meltdown accident. Pressure build-up, hydrogen generation though oxidation and debris bed constitution are the foreseen outputs of the models. But the associated major risk is that of the occurrence of a destructive explosive interaction. The understanding of such phenomena is crucial to identify ways to enhance the mitigation of this risk. The project main objectives follow closely the conclusions of the OECD SERENA-2 program [13], with an orientation of research towards 3 areas:

- the melt fragmentation and mixing in the coolant, being the leading phenomenon, still needing improvements in understanding and thus in modeling;
- the so-called material effect, including oxidation and solidification, being the area with most important uncertainties;
- the mechanisms of pressurization during the steam explosion; the existence of 2 general types of models with antagonist fundamental hypotheses, e.g. Micro-Interaction versus Non-Equilibrium heat transfer, highlights clearly the lack of understanding.

To achieve this goal, the strategy adopted several interconnected steps as indicated in Figure 13. The performed experiments and simulations provide then unique database to improve the FCI models the MC3D software, a digital simulation tool developed by the IRSN with the support of EDF.

Additional tasks regarding MC3D were related to the modeling of the behavior of the coolant (flow patterns) and to numerical issues.

Indeed, it was decided to extend the project with the decision to build a totally revisited version of MC3D, said version 4, with a new architecture to improve versatility and allow parallel computing (MPI process), preserving most the physical description principles and models but enhancing/clarifying where necessary.



4.1. Some results of the KROTOS experiments

KROTOS is a mid-scale facility built at JRC-Ispra to explore the energetics and propagation characteristics of steam explosion [14]. It has been transferred to CEA-Cadarache in 2000 to continue investigating the

pending FCI issues using prototypical corium melts. Apart from the numerous sensors installed along the test section, the facility comes with a high energy X-ray system to enable a detailed visualization of the flow evolution: melt progression and fragmentation, production and dynamics of the void in the mixing zone. 4 tests were performed, one of which, KA-1, was devoted to get more information about fragmentation processes and debris bed formation, using UO_2 -Zr O_2 corium, Figure 14. The test KA-4 was devoted to explosion, using a corium composition expected to be more realistic, including some fission product simulant oxides, which strongly expands the interval liquidus-solidus [14].



The debris of the tests were collected and examined with different other CEA installations to analyze the composition, the thermos-physical properties and the internal structure. Of particular interest was the extent and role of oxidation of a stoichiometric or sub-stoichiometric (UZr)O_{2-x} composition.

4.2. Fragmentation studies

The largest part of the research ICE program was devoted to study fragmentation. Small-scale to mid-scale experiments have been built by LEMTA/CNRS to investigate the fragmentation of a liquid metal in water, without considering boiling aspects, thanks to advanced visualization and post-treatment methods. One particular issue was to get information on drop fragmentation in water, a configuration for which very limited information is available in the literature, in contrast with drop/gas case. The GaLaD experiments [16] provided important information on drop fragmentation in liquids, highlighting important differences compared to the fragmentation in gas. These data were complemented by Direct Numerical Simulations performed with the Gerris and Basilik softwares [17-18]. A particularly important finding is that the fragmentation occurs without dispersion of the fragments (as in gas), these ones being trapped in the wake (see Figure 17). This gives strength of the so-called "Micro-interaction" process during the explosion, a concept popularized by Theofanous.

JaLaD is an upgrade of GaLaD to allow small jet studies. However, a larger-scale facility called JeDi has also been built. Information on the fragmentation process could be obtained from post-processing methods, (see e.g. Figure 16) An important conclusion is that the fragmentation occurs through a 2-step cascading mechanism, with an integral scale mechanism driven by tangential Kelvin-Helmholtz like destabilization process followed by a local-scale fragmentation mechanism. This hypothesis has been translated to a model for MC3D involving both primary large-scale and secondary small-scale mechanisms.



4.3. Outcomes of studies related to oxidation.

Despite the expected scenario related to ex-vessel FCI, where the melt is expected to be fist a metallic mixture (SS-Zr mainly), oxidation is incorporated only in few FCI codes, with poor verification, very parametrically. Although the general mechanisms are well-known, the very high temperatures involved, and the liquid nature of the fuel leads to strong uncertainties. The theoretical works were focus on the elaboration of a modeling for MC3D, based on a scarce and quite old experimental database, although recent studies could also be considered [20]. In principle, oxidation is limited by external diffusion processes in the gas boundary layer (in the film in the situation of film boiling) and internal ones in the melt. Attention has been focused first on the external processes. No data has been found for the drop in gas configuration and numerical simulations allowed to build and verify a proposed correlation. As for the oxidation in water, though film boiling, the new model (not published yet) allows to retrieve the dynamics and strength of oxidation in most cases without the need for a precise modeling of the internal diffusion. This conclusion needs to be strengthened but it would be quite fortunate as internal diffusion is a pure matter of material properties, which represents a particular difficulty.

4.4. On the pressurization process during explosion

This issue was tackled though direct numerical situations. As already mentioned, fragmentation process could be largely clarified [18]. Note that these simulations did not account for the boiling. It is believed that this is not a too strong hypothesis for a large part of the process, which occurs at very high pressure mostly before the expansion process with large void production [21]. Meanwhile, other DNS studies related to the boiling process, made with a specific application of MC3D code, concluded on the existence of a film even at very highly supercritical conditions, so that film boiling correlations are still usable during this process. Although there is not real interface, a film composed of hot light water (quasi-vapor) separates the melt fragment and the cold heavy liquid with a strong thermal disequilibrium. The overall conclusion is that the principles of the two "opposite" general models of 'Micro-Interaction' and 'Non-Equilibrium heat transfer' should be joined to a 'Non-Equilibrium Micro-Interaction model'. Non equilibrium transfers occur very locally, and micro-interaction occurs dynamically at the scale of the fragmenting drop.

4.5. Status of MC3-V4

The analyses made during the project led to numerous improvements of the MC3D code in the latest version 3.10. However, some of the foreseen improvements were found difficult to implement and subject to instability. Also, discussion with partners led to the need to launch the development of a totally revisited version of the code, with a new application for FCI as a first goal. The version is based on several innovative features:

- New architecture allowing a step-by-step construction of an model through the deployment of intermediate but possibility autonomous applications (see Figure 18);
- Multi-Domain structure to allow fast building of data sets with complex geometries.
- Parallel computing based the multi-Domain capabilities (MPI);
- Simplified interface for the user with a new notion of "meta-models", collections of coherent models and parameter setting with different levels of complexity.

The complete version of the FCI application is expected end of 2024.



5. MITHYGENE project

After the Fukushima Daichi accidents, the European stress tests highlighted the need for further research to practically eliminate containment failures due to hydrogen combustion. To this end, the MITHYGENE project explored ways of strengthening the capacity of safety tools and the management of hydrogen-related risks in nuclear power plants [24]. Fundamental and applied research activities were carried out on the remaining safety issues related to hydrogen distribution and combustion, as well as flame-structure interaction. In addition, in situ gas monitoring prototype, reliable with severe accidents have been developed, with the objective to provide information on containment atmosphere flammability in emergency conditions. The updated models and codes were then used to analyze the hydrogen explosion occurred in the Fukushima Daïchi NPP. In the following and for each of the mentioned area, selected achievements are presented and discussed.

5.1. Hydrogen distribution: effect of passive autocatalytic recombiners location

To prevent the risk of hydrogen explosions and limit their consequences, most mitigation strategies adopted in Western countries are based on the use of passive autocatalytic recombiners (PARs). PAR operation converts hydrogen to steam in an exothermic chemical reaction that activates the natural convection flow which brings hydrogen to the PAR inlet ensuring then its passive operation. This convection flow affects the distribution of hydrogen in the surrounding area and, consequently, the efficiency of recombination. As the PAR implementation design was mainly carried out using LP codes, the effect of the convection flow on PARs efficiency is not explicitly addressed. To answer the question of PAR location effect on its efficiency and consequently derive recommendation for optimal PARs implementation design, experimental campaigns were carried out in the MISTRA facility.



The obtained experimental data were used validate the CFD models [25]. The analysis of the experimental and numerical results help deducing recommendations on the PAR optimal locations that allow atmosphere mixing and high recombination rate.

5.2. Hydrogen combustion

For combustion, experience feedback and lessons learned from the various benchmarks (HYCOM, SARNET, SARNET2 and ISP49) [22][23] show that the calculation codes can predict reasonably well the combustion pressure peaks. Hower, most of the combustion models were not able to predict the flame regimes observed experimentally in particular the transition to detonation, as was a priori the case for the explosion that occurred in reactor 3 at the Fukushima Daïchi NPP. In the framework of the MITHYGENE project, the new highly instrumented ENACCEF2 facility had been constructed with the objective to provide relevant data to fill the knowledge gap regarding: 1) water vapor effect on hydrogen flame propagation, 2) initial temperature effect on hydrogen flame propagation, 3) initial mixture heterogeneities on hydrogen flame regimes and 4) water spray actuation on hydrogen flame propagation regimes. To this end, several experimental campaigns were performed providing then unique data for combustion modelling improvement [26].



The experiments thus carried out contribute to filling the lack of data on the hydrogen flames propagation in wet medium and to the combustion modeling improvement. Moreover, the obtained results highlight the promotional effect of the initial temperature on flame acceleration and permit the flame acceleration criteria extension to representative gaseous containment atmosphere temperature. This revised acceleration criterion allows a better identification of potentially dangerous mixtures and improves consequently, the relevance of the risk assessment.

5.3. Flame structure interaction

As pointed out previously, in case of severe accident, the containment of a nuclear reactor is the last barrier to avoid radioactive materials release into the environment. Therefore, excessive loads should not jeopardize its integrity. Moreover, the availability of different safety components important for accident management

The 11th European Review Meeting on Severe Accident Research (ERMSAR2024) KTH, Stockholm, Sweden, May 13-16, 2024

and mitigation should be ensured over the entire duration of the accident. The hydrodynamic force acting on a freestanding object depends not only on the pressure field but also on the flow velocity via the drag force and inertia. Hence, the importance of a fine characterization of flame and shock wave propagation in confined geometries. To this end, experimental campaign was carried out at the SSEXHY facility to build up a database on the deformations of simple structures following an internal hydrogen explosion [27]. Different explosion propagation regimes were studied, ranging from slow deflagration to detonation. Different targets were tested, such as cylinders and plates of varying thickness and diameter. Detailed instrumentation was used to obtain data to validate the coupled CFD models of combustion and structural dynamics. These improvements are an asset for predicting the response of the equipment needed to manage the accident to loads induced by hydrogen combustion.



5.4. In situ gas monitoring system reliable with severe accidental conditions

As part of the MITHYGENE project, a prototypical device was developed based on Raman spectrometry measurement technology for in situ gas monitoring [28]. Several qualification campaigns were carried out to check the effect of conditions representative of a severe accident, including radiation, on the response of the device. To this end, qualification tests were carried out in the IRSN-IRMA facility, for resistance to irradiation, and in ENACCEF2, for the simultaneous measurement of 6 gases (H₂, O₂, CO, CO₂, H₂O, N₂) and the impact of initial temperature. The technological maturity level (TRL) of the Raman probe is estimated at 6 (demonstration of a prototype in a representative environment), while that of the measurement unit (qualifiable) is estimated at 3 (proof-of-concept analytical functions). A patent has been filed for this development.



At the same time, a study of the optimal location of the probes in the containment vessel was carried out. This study identified the minimum number of probes and determined their locations. Secondly, a simple algorithm was developed to use point measurements to reproduce the distribution of gases in the containment and thus provide information on the risks of ignition and flame acceleration.

6. MIRE project

The MIRE (Mitigation of Releases to the Environment in the event of a nuclear accident) project aimed to study and improve the mitigation of gaseous radioactive releases during a core melt accident on a nuclear reactor (referred to as a severe accident). When a core melt accident occurs in a nuclear reactor, the radioactive elements contained in the fuel rods can be released to the containment in the form of particles (aerosols) or gases. To mitigate outside releases in the event of an excessive increase in pressure in the containment, filtering containment venting system (FCVS) [29] are commonly used. In France, the FCVS used is made up of metallic prefilters and sand filters arranged inside and outside of the containment, respectively. This system, which is relatively effective for the filtration of aerosols, cannot retain gaseous forms. Among gaseous fission products, iodine and ruthenium may form volatile species with potential high radiological consequences. The main objective of the MIRE project is to improve mitigation means in case of the containment venting by increasing trapping capacity of gaseous fission products (I₂, CH₃I and RuO₄). To achieve this objective, the following three main topics were addressed by performing both experimentation and modelling activities.

6.1. Topic 1: Source term studies

This topic aimed to improve existing knowledge concerning the resuspension or revolatilization of radioactive elements that could occur during venting or subsequently, over the longer term (delayed releases). Two main issues were then addressed:

- The remobilization of radioactive elements deposited on, or adsorbed by, wall and equipment surface inside the containment. These processes were analyzed using tests involving revolatilization (caesium, iodine and ruthenium) from surfaces of the reactor coolant system. The main parameters studied were the type of carrier gas and thermal conditions. It was shown that in some oxidizing conditions, deposits can be remobilized with the formation of gases and aerosols. This issue is linked to experimental observations during the Fukushima-Daiichi accident corresponding to releases that occurred few days/weeks after the accident starts.
- The characterization of iodine oxides particles issued from the oxidation of volatile iodine by air radiolysis products (ozone formed under radiation, for example). Although these particles can in certain cases represent a large portion of the iodine particles suspended in the containment. Only few data is available on their features (size, composition, stability, etc.). To fill the gap of knowledge, experiments were conducted to obtain qualitative and quantitative information (size, mass, distribution, composition, etc.) on the interactions between iodine oxides and air radiolysis products, as well as on the chemical stability of these oxides in the medium term [30-31]. These studies concluded that iodine oxides are formed by (UV and radiolytic) oxidation of volatile iodine species, and are found in the form of small particles (several hundreds of nanometres) whose chemical composition can vary as a function of temperature and humidity. These particles are also unstable and can be partially redecomposed into gaseous iodine (I₂), depending on the chemical form of the oxides.

6.2. Evaluation of the effectiveness of existing filtration systems

The studies performed aimed to evaluate and compare the effectiveness of filtration systems currently available worldwide [29]. Research conducted in this area focused on determining experimentally the

effectiveness of trapping molecular iodine and organic iodide, as well as ruthenium in gaseous form but in lesser extent. The filters tested were those already installed in nuclear facilities in France and elsewhere in the world: sand filters, liquid filters (sparge filters), metal filters. Adsorption tests for the gaseous species considered were performed on at lab scale (SAFERI experimental device) for each type of filter. The parameters studied included adsorption rate, maximum trapping capacity, temperature and humidity effect in some cases. The irreversibility of this capture under radiation conditions has also been checked. These tests were complemented by tests on a new IRSN facility, the PERSEE test bench, designed for studying filtration and purification systems with potentially the use of labeled iodine. The data acquired confirmed that for volatile iodine species, metallic, sand-bed or liquid filters do not effectively trap organic iodides, and CH₃I in particular [32].

6.3. Research and development of new filtration systems based on the use of dedicated porous materials

Some porous materials have the advantage to be quite selective and can be implemented in passive systems. The potential of "innovative" porous materials for trapping volatile iodine (I₂ and CH₃I) and gaseous ruthenium as ruthenium tetroxide (RuO₄) had been investigated. Two types of porous materials were examined: zeolites and MOFs (Metal Organic Framework). Zeolites are aluminosilicate species with high porosity and thermally resistant. MOFs are porous and crystalline materials created by combining metal ions and organic linkers, they are very tunable due to this hybrid configuration. After synthesis and complete characterizations of the studied porous materials, trapping of different iodine species was studied along with associated mechanisms (modelling at molecular scale by DFT and/or *ab initio* methods and experimental studies). Analytical tests in conditions as close as possible to severe accident conditions (radiation, temperature, steam, presence of gaseous inhibitors, etc.) were performed using the SAFARI (measuring the filtration efficiency of materials on ruthenium and iodine) test bench and PERSEE test bench (cf. **Figure 29**). The most promising materials are silver exchanged zeolites for trapping methyl iodide [33] with some difference in performance linked to their structure and silver mass content (see Figure **28**. Relative stability of methyl iodide trapping). For ruthenium tetroxide, MOF appears to be more suitable due to their higher tunability than zeolite type [30].

To help interpretation/understanding experiments, theoretical simulations were performed to study impact of metal exchanged zeolites, gas contaminants (H_2 , Chlorine, organic compounds, H_2O ...) [34]. For gaseous iodine, density functional theory (DFT) molecular dynamic calculations confirmed that silver dispersed in zeolite is the best candidate for trapping iodine. For gaseous pollutants, carbon monoxide (CO) may have an inhibiting effect on the adsorption of iodine species, even at low concentrations. This point has been checked by co-adsorption tests of CO/ CH₃I mixtures in steam/air carrier gas and not confirmed experimentally. There was high water content in the gas, which can also have a significant effect. From an experimental perspective, of the zeolites tested, adsorbents prepared by ion exchange with silver in Yfaujasite (particularly with around 20% of silver by mass) showed the highest performance for CH_3I and I_2 trapping. A commercial Ag-X zeolite with 35% silver by mass also showed good trapping capacities. For MOFs, some materials offer good radiation resistance, but at this stage, we have not been able to find a structure that can efficiently trap iodomethane. For gaseous ruthenium, the results obtained show variable trapping capacities depending on the porous materials studied. The series of tests performed provides an initial ranking of existing porous solid materials. MOFs and organically modified silica had the highest trapping capacities of the compounds tested. Additional tests are required to explore their performance in conditions that are as representative as possible of severe accidents. To sump-up, the most advanced

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materials/capable of trapping volatile iodine and in particular methyl iodide are zeolites "exchanged" with silver to promote chemical adsorption and form silver iodide (AgI), compound with very low volatility and thermodynamic stability. Two suitable zeolites studied are, an AgY type faujasite with 25% in mass of Ag and a natural zeolite, with approximately 9%/w of Ag. Depending of the design of the filter implemented on the venting line of the containment, such able can efficiently trap volatile iodine and thus should reduce the radiological consequences in the event of containment venting. The knowledge developed within the framework of the MiRE program has been translated into different criteria to guide the choice of the filtering material and possibly improve the efficiency of trapping volatile iodine in the French U5 line.



7. Conclusion

The Fukushima Daiichi accidents claimed the crucial need to improve the safety equipment and the mitigation strategies for severe accident. To achieve this ambitious goal, several projects were launched in the severe accidents field of endeavor to address the topics considered of highest priority and reduce the still pending uncertainties on several selected main phenomena. The PERFROI, DENOPI, ICE, MITHYGENE and MIRE projects, led by IRSN, addressed remaining issues related respectively to fuel degradation and reflooding, spent fuel loss of cooling, fuel-coolant interaction, hydrogen risk and source term mitigation. The achievements of these projects allow getting a better understanding of the above phenomena and providing unique data base for LP and CFD codes validation. Moreover, these projects allow the development of gas monitoring systems and support the development of novel mitigation equipment for source term reduction. The lessons learnt enable recommendations to be made for improving the SAMG for water-cooled reactors and provide an asset for assessing the safety of water-cooled SMR reactors.

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