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ACCIDENTS in nuclear facilities

Nuclear facilities are complex and constantly changing, whether they involve electrical power generating reactors, experimental reactors, or fuel cycle laboratories. To assess measures taken by facility operators to prevent accidents or limit their consequences requires specialized skills and in-depth knowledge of all the phenomena involved.

That is why the Institute conducts research, particularly experimental research, and develops computer codes, usually through scientific cooperation with other national or international research laboratories, universities, or foreign organizations providing technical support to nuclear safety authorities.

The following articles take a brief look at the areas in which the Institute has invested considerable research resources.

- An outbreak of fire in a nuclear facility, as in many other facilities, is a serious event that must be brought under control quickly before equipment items important for facility safety or for containment of radioactive substances are no longer operational. For this purpose, the Institute conducts full-scale experiments to validate the advanced numerical simulation tools that it develops and uses to assess the consequences of a fire. Two articles have been dedicated to this topic.

- The first presents an overview of experimental studies carried out by IRSN in the last twenty years on enclosed fires in mechanically ventilated compartments, characteristic of nuclear facilities. This question is examined by IRSN from two complementary aspects: conducting full-scale tests and developing calculation tools.

- The second article illustrates basic research conducted with other laboratories, in this case Ineris, to advance towards a more precise understanding of mechanisms used to calculate the heat release rate of a fire.

- The probability of a major accident leading to core meltdown is very small, since operators take the necessary measures to avoid this situation. Nonetheless, any radioactive release that could occur in this case is studied in research work conducted throughout the world, especially in France. Certain studies aim to validate the ASTEC numerical simulation computer code, developed in close cooperation with GRS and now a reference in Europe. Five articles illustrate the important work conducted by the Institute in this area.

- The first article presents an overview of results from the FPT2 test conducted in 2004 in CEA’s PHÉBUS test reactor. This test is referred to as an integral test, since it represents all phenomena occurring during a water-reactor core meltdown on a reduced scale. The lessons learned are extremely important and have contributed significantly to advancing knowledge on this subject.

- The second article takes a glance at studies conducted at IRSN to collect data required to reliably predict the progression of core meltdown in a reactor, which requires accurate knowledge of basic thermodynamic data on reactor components and the systems featuring these components.
The third article describes the success story of the European network of excellence on core meltdown accidents that the Institute has supervised for the last four years, which set out to coordinate all research work conducted in this area across Europe.

The fourth article illustrates research conducted on iodine behavior by IRSN using an irradiator installed at Cadarache. Iodine-131 can present a considerable health risk in the event of accidental release. The Institute has conducted many studies and participates in several international programs relevant to this topic.

The fifth article takes inventory of knowledge developed on the physicochemical behavior of ruthenium, a fission product that may lead to significant risks if released in large quantities. Physical chemistry plays a major role in determining the chemical state of a radioactive substance after it has been released by fuel in a major accident situation. The chemical state determines volatility, or the propensity of the substance to reach the containment in a gas form and then escape to the environment if leakage is present.

The reactor containment provides the last barrier capable of preventing release in the event of failure of the two previous barriers, i.e. the metal cladding on fuel assemblies and the reactor coolant system. The last article illustrates work carried out by the Institute to assess vulnerability to impact on civil engineering structures.
INFLUENCE OF ENCLOSURE ON A POOL FIRE in a mechanically ventilated compartment

For over twenty years, IRSN has been closely investigating fire development in the enclosed, mechanically ventilated spaces characteristic of nuclear facilities. For fuel cycle facilities, these studies aim to provide a more accurate assessment of conditions that could jeopardize containment of radioactive materials should a fire occur. There are different ways in which fire can affect containment: by suspending radioactive materials in the air inside the building; by disturbing flow in the ventilation system that normally creates a vacuum in rooms so that air circulates from the less contaminated areas to the highly contaminated areas; by damaging the HEPA filters (heated by warm gases, clogged with soot) that constitute the last filtration level before release to the environment; or by damaging the physical containment items (fire doors, fire dampers) that establish fire compartments inside the facility. To correctly simulate these phenomena, it is crucial to establish a model that accurately represents the heat released by the fire and its duration. Aside from size effects, one of the most important points is the close interaction between the development of fire in an under-ventilated environment and the air-flow behavior of the ventilation system. For several years now, this question has been approached by IRSN through two complementary lines of research: conducting full-scale tests, such as those carried out through the FLIP program (in cooperation with Areva) and the Prisme program (supported by the OECD), and the development of calculation tools such as ISIS (3D software) and the SYLVIA code, which covers the entire facility, including the ventilation system.

The heat released by fuel can usually be determined in a relatively direct manner in well-ventilated conditions using small-, medium-, or large-scale calorimeters(1). In most codes designed to study compartment fires, entering an experimentally measured heat release rate in the data set provides an acceptable agreement between calculations and experiment results for the main parameters that characterize the consequences of a fire, such as the temperature and pressure of gases in the compartment. In actual fire situations inside a facility, however, knowing the heat release rate of the relevant fire type in an open atmosphere is not enough to predict the course it will take in an enclosed space, due to the

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(1) The calorimeter is designed to estimate the combustion heat release rate by simultaneously measuring the suction flow rate of the hood and the concentrations of the various combustion products and oxygen.
significant interaction between combustion and ventilation in the room where the fire is developing. Confinement significantly changes certain key physicochemical parameters surrounding the fire, such as the oxygen rate, the temperature of walls and gases and, in general, heat flux on the fuel surface. In return, these parameters affect both the fuel pyrolysis rate and the combustion heat, used to determine the heat release rate. This feedback effect complicates attempts to model these phenomena. Furthermore, by influencing the pyrolysis rate, fire confinement also has an effect on fire duration, an important parameter, since safety-related items such as fire doors are designed to withstand a thermal hazard for a limited time period.

In the nuclear industry, compartments are generally hermetically separated from each other, but remain interconnected by a ventilation network. The mechanical ventilation system provides a cascade of vacuum zones designed to prevent any accidental leakage of radioactive substances to the environment. A vacuum is maintained in the rooms containing radioactive materials, with the exhaust air passing through HEPA filters. This particular configuration plays an important role in a fire situation, since safety-related items such as fire doors are designed to withstand a thermal hazard for a limited time period.

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Fuel pyrolysis

For over twenty-five years, numerous researchers have studied the influence of confinement on the heat released by a pool fire. Experiments conducted by Takeda, Fleischman and more recently by Quintiere [Utiskul et al., 2005] focus on small-sized compartments (a maximum of 1 m³), with natural ventilation, usually consisting of vertical openings that simulate an open door. This configuration was used to identify various combustion conditions, depending on the size of the openings, and whether they were non-oscillating, steadily oscillating, or unsteadily oscillating. Pyrolysis rates up to seven times greater than those observed in the open air were reported, the influence of the additional radiation due to confinement perhaps explaining these observations. On a larger scale [Peatross and Beyler, 1997] or in configurations where the oxygen rate was controlled using nitrogen dilution [Tewarson et al., 1981], a linear decrease in the pyrolysis rate in parallel with the oxygen concentration was, however, observed. Fuel pyrolysis is the result of fuel vaporization, an endothermic process. It is based on the heat flux balance at the surface of the liquid pool: the surface pyrolysis rate is related to incident fluxes in the gas phase by conduction, radiation, and convection, respectively, and to conduction/convection flux towards the center of the fuel liquid by vaporization heat.

\[
\dot{m}^p_{\text{L}_{\text{vap}}} = \dot{m}_{\text{cond}} + \dot{m}_{\text{rad}} + \dot{m}_{\text{conv}} - \dot{m}_{\text{liq}}
\]

Figure 1 illustrates the main phenomena that affect this energy balance as well the related modeling issues. For fuel arranged horizontally, beyond a certain size, flame radiation becomes much greater than exchanges through convection and conduction in the gas phase. The radiated flux is closely related to soot formation; precursors of soot are produced above a certain temperature (around 1,350 K) and the aggregates that constitute the complete soot formation contribute primarily to the flame radiation properties. Therefore as soot formation decreases, so does the fuel pyrolysis rate. Detailed modeling of soot formation and oxidation must take into account an extremely complex chemical process, which is often slow compared to the time characteristic of flows in the flame, and must also provide an accurate representation of the temperature fields in the flame, while calculating the associated aggregate formation and radiation; this is why the phenomena that influence the pyrolysis rate, such as soot formation, are often modeled analytically or empirically.

Size effect

For fires measuring more than 10 cm, an increase in size entails an increase in the zone containing soot. The flame is then more opaque and radiates more towards the fire. This also explains why large fires are more sensitive to radiation from the ambient environment. According to Hottel and Babrauskas [Babrauskas, 1983], the size of the soot formation depends linearly on the size of the fire (D), and a correlation links the pyrolysis rate per unit of surface area to that of very large fires, through a coefficient \( \kappa \), which takes into account both the propensity of the fuel to produce soot and the optical properties of the soot aggregates.

\[
\frac{\dot{m}^p_{\text{L}_{\text{vap}}}(D)}{\dot{m}^p_{\text{L}_{\text{vap}}}} = 1 - e^{-\kappa D}
\]

Figure 2 shows the good agreement between this correlation and measurements taken on TPH(T) under the SATURNE calorimeter hood at IRSN, for different combustion tank sizes [Pretrel, 2007].

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(2) The fire heat release rate is given by the equation \( Q = m \times \Delta H_c \), where \( \Delta H_c \) is combustion heat, i.e. the heat released by the reaction of one kilogram of fuel, and \( m \) is the pyrolysis rate, more commonly expressed as the product of the fire surface area and the surface pyrolysis rate.\(^2\)

(3) Hydrogenated tetrapropylene (TPH) is a dodecane (C\(_{12}\)H\(_{26}\)) used as a solvent in the nuclear industry.
Effect of decreasing oxygen in the ambient environment

The technical literature indicates a linear dependence between the pyrolysis rate \( (m^p) \) and the oxygen concentration in the ambient environment \( (Y) \).

\[
\frac{m^p(Y, D)}{m^p_{ref}(D)} = (1 + \alpha) \frac{Y}{Y_{ref}} - \alpha
\]

This rate is expressed in terms of the corresponding rate representing well-ventilated conditions (indicated by \( Y_{ref} \) where \( Y_{ref} = 0.233 \) for standard conditions) with a linear dependence coefficient \( \alpha \) that varies depending on the author: Peatross and Beyler [1997] propose \( \alpha = 1.1 \) while others [Utiskul et al., 2005] set \( \alpha = 0 \), but add an ambient environment temperature function to this correlation. This dependence is illustrated in Figure 3 for a test conducted through the PRISME SOURCE program: at any given instant, the experimental measurements of the pyrolysis rate and the oxygen concentration near the fire corroborate the Peatross and Beyler correlation for this configuration.

Transient effects on fire ignition

In experiments, the fire is ignited artificially using either a radiating panel or a small pilot flame aimed at the fuel tank. The experiment itself does not begin until after this startup phase, which lasts a few minutes. When the fire ignites, two phenomena tend to gradually increase the fuel pyrolysis rate. First, the flame propagates to the entire combustion tank at a relatively high spreading rate (a few cm/s), so that the tank is quickly completely in flames. Second, the fuel liquid, which is initially cold, thereby forming a heat sink, is gradually heated to its center through conduction by the radiated heat received at the surface, which prompts vaporization. Assuming a constant incident heat flux and observing that the thermal behavior of the liquid in the tank is similar to that displayed in a semi-infinite environment, it is possible to establish an analytical solution of the liquid warming conditions that expresses the increase in pyrolysis rate as a function of time and the thermophysical properties of the fuel, its initial temperature and the tank dimensions.
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Figure 2: How size affects pyrolysis rate for different tanks containing TPH and the corresponding Babrauskas laws.

Figure 3: How decreasing the oxygen concentration affects the pyrolysis rate in Test 1 (0.4 m² of TPH in the DIVA facility, PRISME SOURCE program), with the corresponding correlations.

Figure 4: Modeling pyrolysis rate increase on ignition for TPH tanks of three different sizes burning in the open air.

Fire extinction due to oxygen starvation

As the oxygen concentration in the ambient environment decreases, extinction phenomena appear, first limited to certain areas within the flame, then leading to complete extinction of the fire. Two approaches are currently proposed to model these phenomena. The first (see Beyler, 2005) for a bibliography review) is based on the fact that the flame temperature decreases as the oxygen concentration drops due to dilution of the comburant, until the flame is extinguished. The second approach [Delichatsios, 2007] models heat exchange between the flame and the fuel, and infers that there is a critical mass transfer number that determines flame extinction. At present, it is difficult to choose between these two approaches. An engineer’s solution would consist of applying an extinction criteria based on a threshold value of the surface pyrolysis rate (about 10 g/m²/s), deduced from a large set of liquid pool fire test results. It would then be necessary to establish the theoretical principles that justify this value.
The test results provided very interesting data on how confinement affects combustion, frequently revealing a relatively long quasi-steady state. Certain experiments were selected to validate the models, and their main characteristics are listed in Table 1. All the experiments were instrumented with type K thermocouples positioned on the vertical measuring axes to provide a precise map of the gas temperatures inside the compartment. Oxygen concentration was measured using Servomex 4100 analyzers at three points in the compartment where the fire developed: one near the fire, 35 cm above the floor, another located 80 cm above the floor, and the last one placed 70 cm below the ceiling. An "average" oxygen concentration in the compartment was estimated, based on measurements from the second and third analyzers. Table 2 summarizes the main results from these experiments.
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All tests showed a temperature rise that was limited compared to the temperatures leading to flashover\(^{(4)}\), while the oxygen concentration often decreased significantly. This made it possible to study the influence of the oxygen concentration on the pyrolysis rate, while ignoring the temperature effect.

Two experiments (Figures 6 and 7) led to early extinction of the fire due to oxygen starvation, while the other tests displayed a phase in a quasi-steady state characterized by a pyrolysis rate much lower than that observed in the open air for the same type of tank.

\(^{(4)}\) Flashover occurs when all combustible materials in the room ignite simultaneously. It is usually assumed that a temperature of 875 K (approx. 600°C) in the upper part of the room will lead to flashover.
Validation of the "Well-mixed Reactor approach"

The "Well-mixed Reactor" approach consists of considering the fire compartment as a reactor where properties are homogeneous. Simple mass and energy balances are therefore enough to determine the general characteristics of the gases and walls in the compartment. This approach is used to characterize fires after flashover, and also to conduct dimensional studies aimed at establishing the primary parameters for modeling. Applying this approach to the case of a fire confined in a ventilated compartment leads to an equation that gives the pyrolysis rate in a quasi-steady state [Melis and Audouin, 2008]. By considering that the admission and exhaust branches follow a Bernoulli law, the conservation of mass, oxygen, and energy in the compartment makes it possible to determine the oxygen fraction during the quasi-steady state, and thus the pyrolysis rate.

\[
\frac{\dot{m}_p}{\dot{m}_{p,\text{av}}} = \frac{1}{1 + \frac{q_v}{\dot{q}_\text{av}} (1 + \alpha) \Phi} \cdot \frac{1}{q_v \sqrt{1 + \frac{q_v}{\dot{q}_\text{av}} (1 + \alpha) \Phi}}
\]

This equation establishes a relationship between the fuel pyrolysis rate during the quasi-steady phase (\(\dot{m}_p\)) and the pyrolysis rate in a well-ventilated environment (\(\dot{m}_{p,\text{av}}\)). It displays three terms:

- the oxygen concentration dependence coefficient \(\alpha\);
- the ratio \(q_v/\dot{q}_\text{av}\) representing the ventilation rate in the quasi-steady state over the nominal ventilation rate of the facility. This ratio depends to a slight degree on the compartment temperature and the cascade of vacuum zones operational in the normal state, such that \(q_v/\dot{q}_\text{av} = 0.9\) is a satisfactory approximation;
- the global equivalence ratio \((5) \Phi = A_t \dot{m}_{p,\text{av}} \frac{s}{\dot{q}_\text{av}}\), where \(A_t\) is the fire surface area and \(s\) the oxygen mass consumed by the fuel mass burned in stoichiometry. This ratio expresses the relationship between the oxygen required for burning and the oxygen supplied in the normally-ventilated state.

*Figure 7* suggests two types of combustion behavior in the quasi-steady state: the tests where the air inlet is in the upper part of the compartment reveal a remarkable degree of agreement with the well-mixed reactor theory, whereas a significant difference is observed when the inlet is located near the floor:

- when the air inlet is near the ceiling, fresh air is convected towards the floor of the compartment, mixing the gases so that gas layering is attenuated;
- in opposition, enclosed fires where the air inlet is near the floor of the compartment behave similarly to fires naturally ventilated by an open door, where significant thermal layering of gases is observed. The fire and the lower part of the compartment are immersed in a practically non-vitiated environment favorable to combustion and supplied with fresh air, while the combustion products accumulate in the upper part of the compartment. The difference between the mean oxygen concentration and the oxygen concentration near the fire (*Table 2*) confirms this analysis.

(5) The Global Equivalence Ratio (GER) was introduced by Tewarson and characterizes under-ventilation of a fire in comparison to stoichiometry.
Yet in both cases, the Peatross and Beyler correlation ($\alpha = 1.1$) can be applied satisfactorily (Figure 8).

The “Well-mixed Reactor” approach therefore cannot be used to predict fire behavior for all configurations. It nonetheless reveals the global equivalence ratio as a dimensionless value characteristic of the fire: this number is used to theoretically classify fire experiments and scenarios and plays an important role in describing the qualification limits of computer codes.

**Validation of the Lumped-Parameter Approach (SYLVIA code)**

**Description of the Lumped-Parameter Approach**

“The Well-mixed Reactor” approach shows that it is essential to correctly estimate the oxygen available for combustion in order to correctly model the fire heat release rate. Models based on the Lumped-Parameter Approach take into account thermal gas layering in an enclosed fire in a simplified manner. The space inside the compartment is divided into two zones, the height of the interface between these zones changing over time. In each zone, the thermodynamic properties (gas pressure and temperature, concentration of the various species) are assumed to be uniform. The SYLVIA code, developed by IRSN, was inspired by this simplified compartment model that nonetheless gives a detailed description of the entire ventilation system.

The ventilation system is modeled as a network of branches and nodes. The branches represent the ventilation ducts, fans, filters, etc. The main unknown is the gas flow rate, the relevant equation being a general form of the Bernoulli equation that takes into account the inertia of momentum. Nodes represent either the compartments or the connection points between branches. The conservation of mass for each specie and the conservation of energy make it possible to determine the thermodynamic properties of the gas in the branches and nodes.

Walls are meshed in two dimensions, but heat conduction is only resolved for thickness. A gradual representation where meshing is tighter near the surface is used to take into account significant temperature gradients in the walls. Heat transfer takes place through natural convection in compartments and through forced convection in branches. Heat transfer coefficients are obtained using conventional correlations from the technical literature. Heat exchanges through radiation involve the flame, walls, and gases. The model used is a hot-spot model where the fire is represented as a hot spot emitting radiated heat that is a constant fraction of the fire heat release rate. Absorption and emission by gases take into account carbon dioxide, steam, and soot content.

One of the unique features of SYLVIA is its ability to calculate the pyrolysis rate, which must be provided as input for most computer codes used in fire modeling. The various phenomena that affect the pyrolysis rate are processed using correlations: the size effect is calculated using the Babrauskas correlation; the impact of decreasing oxygen concentration is processed by the Peatross and Beyler correlation, where the oxygen...
An example of this configuration is Test 5, illustrated in Figures 13 and 14. The slight deviation with respect to the Peatross and Beyler correlation (Figure 7) is not enough to explain the gap between calculation results and experimental results. In this case, it is the chemical process in the reaction that varied significantly from well-ventilated conditions: when the upper part of the flame was surrounded by a vitiated atmosphere, combustion became less exothermic, consumed less oxygen, and produced more carbon monoxide and soot. This phenomenon (not modeled) was revealed by observing the \( \text{CO}/\text{CO}_2 \) ratio, which was ten times greater in Test 5 than in Test 1 (2\% and 0.2\% respectively), the only difference between these two tests being the position of the air inlet.

In more general terms, confinement can have two effects with opposite consequences, depending on the value of the global equivalence ratio:
- for slightly under-ventilated fires, the decrease in the pyrolysis rate entails an increase in the fire duration in comparison to the same fire in the open air, thereby aggravating the thermal impact of the fire;
- in the opposite case, when under-ventilation is significant, i.e. when the global equivalence ratio is high, fuel pyrolysis is not sufficient to sustain combustion and the fire is extinguished before all the fuel has been consumed, thus attenuating the thermal impact of the fire. Tests 6 and 7 are representative of this early extinction effect.

Validation of the SYLVIA code

Systematic comparison between predictions obtained using the SYLVIA code and results from full-scale tests conducted at IRSN led to broad validation of this computer application. Figures 9 to 14 illustrate the enclosed pool-fire dynamics. Two examples representative of tests where the air inlet duct was located in the upper part of the compartment are given for Tests 1 and 2 (Figures 9 to 12). The pyrolysis rate as well as the oxygen and carbon dioxide concentrations were calculated quite satisfactorily in the quasi-steady state, although the representation of pyrolysis behavior at fire ignition was rather approximate. Agreement was not as good when the air inlet duct was located in the lower part of the compartment, the lumped-parameter approach providing better results than the “Well-mixed Reactor” approach.

In more general terms, confinement can have two effects with opposite consequences, depending on the value of the global equivalence ratio:
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accurately quantify the effect of vitiation on the pyrolysis rate of liquid fuels. Work in this field must be pursued to study the validity of these models for carbonaceous fuels and vertical fires.

As shown in Figure 15, these two effects were correctly predicted using a simple lumped-parameter approach coupled with the Peatross and Beyler correlation. The model nonetheless greatly overestimates fire duration for Test 5.

**Conclusion**

Modeling an enclosed fire requires, above all, correctly predicting the heat released through combustion, therefore the fuel pyrolysis rate. For well-ventilated environments, the technical literature proposes correlations that take into account the various phenomena that affect pyrolysis. The size effect and transient aspects of fire ignition were studied on liquid fuels of interest with regards to nuclear safety in fire studies conducted using the SATURNE calorimeter hood. For slightly ventilated environments, several experiments that studied the effect of fire confinement were conducted at IRSN using the PLUTON and DIVA facilities, consisting of closed compartments connected to a ventilation system. The main effect of enclosure can be expressed as a linear decrease in the pyrolysis rate occurring simultaneously with the decrease in the oxygen concentration. The technical literature, however, reports different values for the linearity coefficient, the one proposed by Peatross and Beyler being in closest agreement with experiments conducted at IRSN. It was therefore possible to more accurately quantify the effect of vitiation on the pyrolysis rate of liquid fuels. Work in this field must be pursued to study the validity of these models for carbonaceous fuels and vertical fires.

Furthermore, vitiation of the environment near the fire may also entail extinction of the fire. An extinction criteria based on the fuel surface pyrolysis rate** would make it possible to obtain a satisfactory model of this phenomenon.

Figure 14 Oxygen and CO₂ concentrations (Test 5).

Figure 15 Comparison of calculated and experimental fire duration periods from all tests conducted.
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2.2

CHARACTERIZING COMBUSTION of a liquid material

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As a fire burns, the heat released by vaporized fuel combustion determines how the fire will develop, along with fire characteristics such as the flame height above the fire, the energy radiated from the flame to the immediate environment, the gas temperature and velocity in the flame, and the smoke plume. In practice, this thermal power can be estimated by multiplying the fuel mass flow per unit area by the fire surface area and the combustion heat, a thermodynamic property of the material. The mass flow per unit area of a liquid fuel depends on the size of the tank containing the fuel, as illustrated in Figure 1 for hydrogenated tetrapropylene (TPH), a liquid hydrocarbon used in the nuclear fuel recycling process.

The mass flow per unit area of a fuel is measured using a calorimeter cone for tank diameters ranging from roughly 10 cm (Figure 2a) to several meters (Figure 2b). Experimental results obtained in this way can be used to determine the parameters of a correlation that gives the mass flow per unit area as a function of the liquid pool diameter (see example in Figure 1). The database containing mass flow per unit area for combustible materials serves to run fire scenario simulations using computer codes.

Since it is easier, faster, and less costly to run tests on a small scale rather than a large one, in the last few years researchers have been developing an experimental protocol to estimate the mass flow per unit area of a large-sized fire (with a diameter greater than one meter) based only on small-scale tests. Ineris(1) and IRSN, working in partnership on this research subject, have contributed to achieving the first step by proposing a method that determines the asymptotic value of the mass flow per unit area for liquid fuels on a large scale (noted $m'_{\text{TPH}}$) based on tests conducted in a Tewarson calorimeter. The method consists of measuring the mass flow per unit area on a material burning in an over-ventilated environment (i.e. where the volume fraction of oxygen is

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(1) Ineris is the French national institute for the study of industrial environments and risks.
greater than 21%), as proposed in the FRST technique\(^{(2)}\).

The study conducted with Ineris showed that the level of over-ventilation required to reach the asymptotic value \(\dot{m}^*\) of the mass flow per unit area is correlated to the adimensional parameter \(c_s\). This parameter represents the relationship between gasification heat and combustion heat, which are thermodynamic properties of the material. In practice, the value \(\dot{m}^*\) of a material with an \(c_s\) equal to 0.018 (such as TPH, for example) is obtained by burning a sample of this material in a Tewarson chamber in an atmosphere containing 47% oxygen by volume (Figure 1).

To date, this experimental technique for determining mass flow per unit area on a large scale has been verified successfully on nine liquid fuels. The next step consists of extending the scope of validity of this new empirical approach to other liquid fuels and developing a model of the physicochemical phenomena involved. More long-term research goals aim to verify whether this approach can be extended to solid fuels, where thermal degradation is more complex (pyrolysis with surface oxidation, carbonaceous residues, etc.).

The first results obtained through this scientific partnership between Ineris and IRSN appeared in a joint publication in the 11\(^{th}\) Proceedings of the International Interflam Conference held in London in September 2007.

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\(^{(2)}\) FRST: Flame Radiation Scaling Technique. Approach proposed in the 1980s by Tewarson, based on the empirical observation that a material burning in an over-ventilated environment manifests a mass flow per unit area that is higher than in an ambient environment.
The PHÉBUS-FP program consisted of five tests successfully conducted from 1993 to 2004 [Clément et al., 2006]. FPT2 was the fourth test in the program, carried out from October 12 to 16, 2000. The study focused on core meltdown under steam-starvation conditions, where the steam contained boric acid, and the sump was basic and evaporating, whereas in previous tests steam was in excess and the sump was acid and non-evaporating (2).

The experimental data collected during the test and subsequent destructive and non-destructive test campaigns was processed and overall data consistency analyzed. Findings from the FPT2 test results [Clément et al., 2006; March, 2008; Grégoire et al., 2008] are presented in this article.

Experimental facility

The PHÉBUS facility is designed to create experimental conditions representative of a core meltdown accident in a pressurized water reactor [Schwarz et al., 1999; Clément et al., 2003b], in order to investigate the degradation of fuel rods and a neutron-absorbing rod (3) up to the formation of a molten pool. It is also used to study the release and transport of materials (fuel fission products, vapors, or aerosols from fuel bundle degradation) resulting from deterioration of the primary circuit and the

(1) Studies conducted on a limited number of phenomena.
(2) These conditions are chosen according to the accident sequence to be simulated.
(3) Used in nuclear reactors to control reactor power.
These three systems are simulated at a scale of approximately 1:5000 with respect to a 900 MWe pressurized water reactor (Figure 1, left-hand side). The test facility is equipped with various instruments to measure flow rate, temperature, radiation (high-count gamma spectrometry), concentrations of hydrogen and oxygen, and to take sequential samples of circuit fluid, containment atmosphere, and sump liquid. Three thermal gradient tubes (TGT) with a wall temperature profile set from 700°C to 150°C were installed on the hot leg to determine the element condensation temperatures and thus deduce their chemical speciation. Non-destructive post-test measurements were performed in the facility to quantify the gamma emitters retained in the experimental circuit and vessel samples, and to characterize fuel degradation (using X-ray radiography, computed tomography, and gamma spectrometry to establish the γ-emitter distribution profile of the bundle). Destructive measurements were also carried out on the fuel bundle [Bottomley et al., 2007] and on a selection of sample.

FPT2 investigated physical phenomena occurring in the following plant systems (see Figure 1, right-hand side):

- the reactor core, simulated by a fuel bundle containing 18 UO₂ fuel rods previously irradiated in the BR3 reactor with a burnup of 32 GWD/tU, two instrumented rods containing fresh UO₂ fuel, and a neutron-absorber silver-indium-cadmium (SIC) rod. The fuel rods have Zircaloy cladding(5) and the absorber rod has steel cladding with a Zircaloy guide tube (4);
- the primary circuit, represented by a hot leg where the wall temperature is controlled to 700°C (2), and a cold leg controlled to 150°C (4), interconnected by an inverted U-shaped tube four meters high, simulating the steam generators (3);
- the containment building, simulated by a 10 m³ vessel (6) with an electropolished surface, a 120-liter tank at its base filled with a pH 9 alkaline buffer solution simulating the reactor sump (6), a gas containment volume (7) and, in the upper part, condensing, cooled, painted surfaces (7) (6). The cold leg discharges into the containment, simulating a break downstream from the steam generator.

Figure 1

(1) Bundle consisting of fuel rods and the absorber rod. (2) Experimental circuit hot leg. (3) Inverted U-shaped tube simulating the steam generator. (4) Experimental circuit cold leg. (5) Gas containment volume in the vessel simulating the reactor containment. (6) Tank containing liquid to simulate the sump. (7) Painted surfaces.

(4) Reactor operated by Belgonucléaire in Mol, Belgium.
(5) Zirconium alloy.
(6) Referred to as the “vessel” in the rest of this article.
(7) Referred to as “condensers” in the rest of this article.
(8) Cross-sectional views of the experimental cell, where each pixel is assigned a specific density.
Accidents in nuclear facilities

2.3

FPT2 test objectives

FPT2 set out to achieve the specific objectives described below.

On the bundle

☑ The test aimed to achieve significant degradation of the fuel rods and the absorber rod by melting up to 20% of the fuel, or roughly 2 kg.
☑ It also set out to observe the consequences of the release of fission products and structural materials by the bundle, in a hydrogen-rich, low-pressure atmosphere (0.2 MPa) containing boric acid.

In the experimental circuit

The main goal was to study the transport and retention of fission products and structural materials in the primary circuit at low pressure (0.2 MPa) in a highly reducing atmosphere. Data was to be obtained on fission product chemistry (particularly on the influence of boric acid), and the interaction between fission products and the circuit walls at high temperature, especially in places where significant temperature changes were expected (at the outlet of the bundle and at the inlet of the steam generator tube).

In the containment vessel

The main objective was to study fission product chemistry, especially iodine, in the hours and days following their release from the bundle, and the effect of the presence of boric acid.

Researchers focused on iodine radiochemistry in the sump water and the containment atmosphere, using several dedicated instruments. Painted surfaces installed in both the sump and the containment atmosphere (condensers) provided a source of organic compounds capable of interacting with iodine. The temperatures of the various vessel components were imposed to favor water evaporation from the sump and promote volatile iodine transfer from the sump to the gas containment.

In addition to the objectives indicated previously, the tests also set out to characterize:

☑ the size of aerosols received in the vessel and the aerosol deposition processes (gravitational settling to vessel bottom, diffusiophoresis to the condensers, and deposition on electropolished vessel walls);
☑ any potential fission product poisoning effect on the catalytic hydrogen recombiners used in nuclear reactor containments to recombine hydrogen into water, thereby avoiding the risk of explosion.

Most of these objectives were reached. However, since most of the flow meters associated with samplings on the hot leg did not operate, the release flux curves of the various elements in the hot leg could not be established. In return, on-line gamma spectrometry measurements taken on the vessel and cold leg flux curves established using post-test gamma spectrometry measurements taken on samples generally showed good consistency. They provided reliable quantitative data on kinetics and the element quantities recovered in the vessel. It should be noted that boron was not measured on the samples.

FPT2 test scenario

Prior to the actual experimental phase, the bundle was “re-irradiated” in the PHÉBUS reactor for eight days to obtain a representative inventory of short-lived fission products (such as iodine-131, with a half-life of around eight days). Then the bundle dried for about 37 hours and boundary conditions were adjusted. During the ensuing degradation phase, which lasted about six hours, pressure in the experimental circuit was held at 0.2 MPa and the steam injection flow rate in the lower portion of the bundle was maintained at 0.5 g/s (boric acid with a boron mass concentration of 1,000 ppm was added to the steam). Reactor power was increased in a stepwise manner, leading to gradual degradation of the test bundle (Figure 2). Once the degradation targets had been reached, the reactor was shut down. After this transient, the fuel bundle was then cooled for about one hour, then the containment vessel was isolated.

It was initially planned to introduce the hydrogen recombiner coupons device in the containment atmosphere for 30 minutes between the reactor shutdown and the end of steam injection, but mechanical problems prevented this operation from being performed.

The experimental phase then moved into a long-term phase lasting four days, with three successive stages:

☑ an “aerosol” stage lasting around 37 hours, aimed at analyzing aerosol deposition mechanisms in the containment vessel. During this stage, the vessel thermohydraulic conditions remained unchanged;

☐ Representative of situations where steam is almost completely transformed into hydrogen by the Zircaloy oxidation reaction on the fuel cladding.
☐ Aerosols are entrained by vapor condensation on the cooled parts of the condensers.
☑ For PHÉBUS-FPT3, the holder for the hydrogen recombiner coupons was improved and introduced successfully [Biard et al., 2007].
a 23-minute washing phase, in which aerosols deposited by gravitational settling on the hemispheric vessel bottom were transferred to the sump. This was carried out by spraying the sump water on the hemispheric vessel bottom through a loop provided for this purpose;

- a two-day chemistry stage, dedicated to analyzing iodine chemistry in the sump and containment atmosphere, with particular focus on iodine speciation. The water temperature during this chemistry stage was brought from 90°C up to 120°C to favor a 0.98 g/s evaporation/condensation cycle between sump and condensers.

Main results on bundle degradation

On-line measurements from the bundle, experimental circuit, and containment vessel during the reactor power increase detected the following events:

- the first **cladding failure**, which took place around the bundle midplane (roughly at the 406 mm elevation) at a temperature close to 800°C, as in previous tests;
- **absorber rod failure** occurred around the 500 mm elevation. The peak temperature measured on the guide tube at that time was about 1,350°C, close to the steel melting point (1,425°C, the temperature recorded a little later on the guide tube at 500 mm);
- the **main oxidation phase** occurred shortly after absorber rod failure in the bundle midplane, with a maximum bundle temperature of about 1,590°C at 500 mm. No significant material displacement was detected during the main oxidation phase, in contrast to FPT1, where oxidation was more acute;
- **displacement of materials** from the center of the bundle downwards was observed starting from the heating phase at a temperature of about 2,000°C and led to the formation of a molten pool in the lower portion of the bundle, as in previous tests;
- a chimney formed in the molten pool, allowing steam to escape. The chimney then plugged up, the molten pool spread throughout the entire cross-section of the test bundle, and a significant temperature excursion was recorded, leading to reactor shutdown.

During FPT2, the hydrogen generation kinetics (**Figure 2**) were slower than in FPT0 and FPT1: the lower steam injection rate, approximately 0.5 g/s in FPT2 instead of about 2 g/s in FPT0 and FPT1, led to a slower progression of Zircaloy oxidation on the cladding. The oxidation front moved axially, first in the lower portion at a rate of 1.7 mm/s, then in the upper portion of the
bundle. Nonetheless, hydrogen generation lasted longer (an H₂ concentration greater than 10% in volume is measured for 43 minutes) and led to a period of 18 minutes during which the hydrogen volume concentration in the circuit was between 75% and 97%.

As in earlier tests, a second, less-significant oxidation stage occurred in the final bundle degradation phase, when materials flowed into the lower portion of the bundle. During this late oxidation phase, which lasted about 18 minutes (with a hydrogen volume concentration greater than 10%), a maximum hydrogen concentration of 17% was reached in the circuit for three to four minutes.

Based on the amount of hydrogen generated during degradation, it was estimated that 81% of the Zircaloy in the cladding was oxidized.

The final state of the bundle was observed on a post-test X-ray image. Figure 3 shows the presence of a cavity at the center, with a molten pool in the lower portion consisting of a mixture of relocated, melted materials, with deformed rods in the upper and lower portions. As expected, significant degradation of the bundle was obtained during FPT2 (to approximately the same degree as that observed during FPT0, and to a greater extent than that observed during FPT1).

Total mass gain in the lower portion was evaluated at 3.9 kg through non-destructive testing, 1.1 kg being attributed to Zircaloy cladding oxidation, the rest corresponding to relocation of the materials. These observations were consistent with the initial test objective in terms of degradation.

Main results from experimental circuit and containment vessel

Fission product release, transport, and deposition in the primary circuit

During FPT2, as in the previous tests, release and transport of materials in the primary circuit correlated closely to events occurring on the bundle. Three main phases of release were identified:

- A third significant period of release (of less volatile materials) was measured in the vessel during the second oxidation phase, corresponding to oxidation of the lower portion of the bundle, associated with the beginning of fuel relocation and liquefaction.

After formation of the molten pool, no more significant amounts of fission products were received in the vessel, which does not imply, however, that release had stopped, since part of the released materials could have been deposited in the circuit.

The elements measured were classified according to the kinetics that led them to the vessel:

- Release of noble gases started from the beginning of the first oxidation phase. They accumulated in the vessel much more quickly than the volatile fission products;
- Volatile and semi-volatile fission products (Cs, I, Rb, followed slightly later by Te and Mo) arrived in the vessel at an almost steady rate from the end of the first oxidation phase to the end of the second oxidation phase, which is consistent with a gradual degradation of the FPT2 bundle (Figure 4). This is also the case for indium, released by the absorber rod. Molybdenum was only measurable in the vessel after the first oxidation phase, suggesting that release of this element was limited during the hydrogen-rich phase and only became significant in the steam-rich phase that followed. This is consistent with the fact that oxidized molybdenum is more volatile than the metal;
- The low-volatility fission products (Ba, La, Y, Sr), silver (mainly from the absorber rod), elements from the structural components and instrumentation (Zr, Re, W) and the fuel were mainly released during the end of the transient phase (second oxidation phase and final heating period).

The elements measured can be grouped together according to their fraction of global release from the fuel bundle:

- Elements released in substantial amounts (around 80% of the bundle’s initial inventory [hereafter noted i.i.]) such as noble gases (e.g. Xe);
- Volatile fission products (I, Te, and Cs) with a released fraction between 67% and 81% i.i. (Table 1);
- Elements released in small or very small amounts, such as Ba or Zr, which reached about 1.2% i.i. and 0.015% i.i., or even less for other elements.

The elements were also classified according to the amounts received in the vessel (referred to as the “vessel inventory”):

- Noble gases such as Xe and Kr, which did not react with surfaces inside the circuit, reached the vessel without being retained in the circuit, so that the vessel inventory coincided with the amount
The mass composition (Figure 5) of aerosols transported to the vessel consisted predominantly of fission products (19% Cs, 20% Mo), elements from the absorber rod (16% Ag, 15% Cd, 11% In), and an element contained in the cladding (7% Sn). Total mass was estimated at 55 g, taking into account material oxidation. This value was low compared to the 125 g measured in FPT1 [Jacquemain et al., 2000; Dubourg et al., 2005]. This significant difference can be explained partially by the lower steam injection rate used in FPT2 (four times less than FPT1). This factor not only changed the degradation phenomena, hence the mass and composition of released elements, but also increased residence time in the bundle and the circuit, thereby favoring deposition of certain elements in these areas.

The processes whereby elements were deposited on the surface of the circuit lines include condensation of steam species in thermal transition zones, impaction, thermophoresis, and aerosol settling. Deposition was probably followed by chemical interaction with the surfaces or with other deposited elements. For most of these elements, the amounts in the vessel were significantly less than those measured during FPT1.

As in the previous tests, low amounts of barium were released, while separate effects tests seemed to indicate that the reducing conditions and high temperatures would increase the released fraction of this element. FPT2 results therefore confirm the low volatility of barium, probably due to reactions between barium and the cladding materials [Dubourg and Taylor, 2001].

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Deposition

The processes whereby elements were deposited on the surface of the circuit lines include condensation of steam species in thermal transition zones, impaction, thermophoresis, and aerosol settling. Deposition was probably followed by chemical interaction with the surfaces or with other deposited elements. In FPT2, the low steam injection rate (0.5 g/s) resulted in significant deposition of volatile fission products (Cs, I, and Mo) in the upper portion of the bundle (Figure 6). This behavior was

Figure 4 Volatile fission products received in the vessel and hydrogen concentration in the circuit.

Released by fuel (about 79% i.i. and 60% i.i., respectively):

- highly volatile fission products, particularly iodine (57% i.i.) and cesium (41% i.i.), behaved in a manner similar to that observed in FPT1;
- for certain elements, a fraction ranging between 10% and 40% of the initial inventory was received in the vessel (Rb (32% i.i.), Mo (31% i.i.), Te (28% i.i.), Cd (23% i.i.), etc.). But significantly smaller amounts of Rb, Te, and Cd were transported to the vessel in comparison to FPT1, where they were more volatile, with a vessel inventory of about 50% i.i.;
- small amounts of the other elements were also received in the vessel, for example Sn (6.8% i.i.), In (5.7% i.i.), W (3.7% i.i.), Ag (1.5% i.i.) and Tc (0.92% i.i.). Among these elements, the amounts of Sn, Ag, and Tc were considerably less than those measured during FPT1, where the vessel inventory was 33% i.i., 6.7% i.i. and 21% i.i., respectively;
- negligible amounts (less than 0.5% i.i.) of a few elements were transported to the vessel, such as Sr, Ba, Ru, La, Li, Zr, and Re. For most of these elements, the amounts in the vessel were significantly less than those measured during FPT1.

As in the previous tests, low amounts of barium were released, while separate effects tests seemed to indicate that the reducing conditions and high temperatures would increase the released fraction of this element. FPT2 results therefore confirm the low volatility of barium, probably due to reactions between barium and the cladding materials [Dubourg and Taylor, 2001].

(12) Taking into account xenon and krypton, the mass received in the vessel was 150 g for FPT1 and 90 g for FPT2.
In the hot leg, significant fractions of cesium and iodine were detected in the form of vapors (14) and aerosols. Measurements taken on circuit samples after the FPT2 test showed that, first, vapor speciation changed throughout the different degradation phases, and second, cesium iodide was not the only iodine species, contrary to common opinion.

The condensed materials were either deposited on the walls of the steam generator tube, or transported to the cold leg as mixed aerosols. They were sized between 0.5 and 1 µm and were sometimes agglomerated.

The total transported aerosol mass was about 60 g in the hot leg and 45 g in the cold leg (without taking into consideration the oxygen contribution in the oxides formed). The aerosol concentration in the circuit reached its maximum during the last oxidation phase and its mass composition changed as the various degradation phases took place: approximately 12% of the total mass went to the cold leg during the first oxidation phase (predominantly the absorber rod materials, especially cadmium), 17% during the phase that led to fuel melting and relocation (mostly Cs and Mo volatile fission products, silver coming primarily from the absorber rod and tin from the structural components), and 71% during the second oxidation phase (mostly silver, with significant amounts of volatile fission products (Cs and Mo), absorbent materials such as Cd, and structural materials such as Sn).

Not observed in FPT0 and FPT1, in which a higher steam injection rate (2 g/s) led to significant deposits downstream (in the upper plenum above the bundle, and in the steam generator tube).

Aside from tellurium, deposited in very large amounts in the upper plenum above the bundle and in the hot leg, and iodine, deposited in the upper plenum and the vertical line in quantities similar to those of FPT1 (although the proportions were inverted), deposits in the upper plenum, the vertical line, and the steam generator tube were less significant in FPT2 than in FPT1 and FPT0. The total amounts deposited in the circuit are shown in Table 1.

After reactor shutdown, about 45% of the cesium deposited in the upper plenum and in the vertical line was revolatized and deposited mainly on the surfaces of the steam generator tube. These revaporization phenomena may be explained by a drop in the partial cesium pressure in the gas flow(13). Moreover, radioactive tellurium deposited in the hot leg was transformed into iodine through radioactive decay. The volatile iodine was also transported to the steam generator tube. These observations indicate that in the event of a hot leg break, tellurium and cesium deposits in the hot leg could lead to additional release of volatile forms of iodine and cesium, respectively.

<table>
<thead>
<tr>
<th>Release from bundle (% i.i.)</th>
<th>Vessel inventory (% i.i.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cs 67</td>
<td>41</td>
</tr>
<tr>
<td>I  72</td>
<td>57</td>
</tr>
<tr>
<td>Te 81</td>
<td>28</td>
</tr>
<tr>
<td>Mo 52</td>
<td>31</td>
</tr>
</tbody>
</table>

Table 1 Comparison of fractions of various fission products released by the fuel bundle that reach the vessel.

**Physicochemical properties of materials released in the circuit**

In the hot leg, significant fractions of cesium and iodine were detected in the form of vapors (14) and aerosols. Measurements taken on circuit samples after the FPT2 test showed that, first, vapor speciation changed throughout the different degradation phases, and second, cesium iodide was not the only iodine species, contrary to common opinion.

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(13) Corresponds to steam injection, maintained for one hour after reactor shutdown to cool the fuel bundle.

(14) In this context, vapors refer to the compounds that condense between 150°C and 700°C, as opposed to gases, which do not condense.
Aerosol behavior in the vessel

Inside the vessel, aerosols were subject to three deposition phenomena (March et al., 2007; March 2008):
- diffusiophoresis, corresponding to aerosol entrainment through steam condensation on the condensers\(^{(15)}\);
- gravitational settling at the bottom of the vessel (in the sump and on the hemispherical vessel bottom);
- deposition on all vessel walls.

Most of the isotopes detected using gamma spectrometry showed the same overall behavior. Gravitational settling was the predominant mechanism in the vessel: on an average, 74% of the vessel inventory was deposited at the bottom of the vessel. Diffusiophoresis entrained 12% of the vessel inventory to the condensers. The aerosol deposition rate on the condensers was, like the condensation rate, four times less than that observed in FPT0 and FPT1, which is consistent with a steam injection/condensation rate that was four times lower in FPT2 (0.5 g/s instead of 2 g/s in FPT0 and FPT1). Measurements showed non-negligible deposits on the vessel walls (about 11% of the vessel inventory), higher than FPT1 (2% at most). These differences in deposition can be partially explained by the aerosol size, which was lower in FPT2 than in FPT1.

At the end of the aerosol phase, the vessel inventory fractions deposited on the vessel walls and bottom were practically the same for all the elements, whereas the distribution between the sump and the condensers depended on element solubility. The soluble elements (mainly cesium and rubidium) were almost all found in the sump, while insoluble elements were found in both the sump and on the condensers.

\(^{(15)}\) Condensates are recovered at the bottom in a capsule that empties into the sump as soon as the level reaches a given threshold.
During the degradation phase and the beginning of the aerosol phase, aerosol solubility led to three main categories:

- elements found mainly in the aqueous phase and considered as soluble (Cs, Rb, and I);
- elements partially soluble in water (Ba, Mo, Cd, Re, and Tc);
- elements that remain practically insoluble (Ce, Te, Zr, Ru, Sn, In, Ag, W, and U).

This behavior was equivalent to that observed during FPT1 (with an acid sump), except for iodine, which, in FPT1, behaved like a soluble element during degradation, then like an insoluble element during the aerosol phase.

In the containment atmosphere, aerosol size showed a tendency to increase during the degradation phase, with a mean mass aerodynamic diameter that increased from 1.4 μm in the first oxidation phase to 3.68 μm at the beginning of the aerosol phase.

**Iodine release and behavior in the circuit**

Iodine released from fuel during the degradation phase was estimated at about 73% i.i., which was lower than FPT1 and FPT0 (about 87% i.i.). This difference can be explained by the significant deposits observed in the upper portion of the bundle. This retention phenomenon was related to the lower temperatures measured in this zone, due to the lower steam injection rate (four times lower than in FPT1 and FPT0). Moreover, in the upper plenum and the vertical line, iodine probably reacted with fission products (Cs) and absorber rod materials (Ag, In, and Cd) to form metal iodine vapors, leading to significant deposition: 4.1% i.i. in the upper plenum and 0.4% in the vertical line, respectively (in comparison, in FPT1 deposition was 1.5% and 3.8%, respectively), where large temperature drops were measured on the walls. In the hot leg, as in previous tests, iodine was found mainly in the vapor form, at 700°C, gaseous iodine representing only 0.1% i.i. and iodine in aerosol form representing between 10% i.i. and 44% i.i., depending on the degradation phase, which was higher than FPT1 results (less than 15% i.i.).

During the degradation phase, various chemical forms of the iodine transported as vapor through the hot leg of the primary circuit were revealed, during analysis of the condensates deposited on the walls of the sampling lines and the thermal gradient tube in a zone where the temperature dropped from 700°C to 150°C. At least two species were distinguished: cesium iodide, detected only after the main oxidation phase, and a second, more volatile specie that remains unidentified at this time. During FPT2, about 6.2% i.i. was deposited on the steam generator tube, with an exponential deposition profile characteristic of aerosol deposition through thermophoresis. This behavior was different from that observed during FPT0 and FPT1, where vapor condensation and aerosol thermophoresis were both observed, leading to larger deposits, i.e. 23.5% i.i. for FPT0 and 19.2% i.i. for FPT1.

In FPT2, the lower steam injection rate may have led to deposition that was not measured, through condensation upstream from the steam generator tube.

In the cold leg, most of the iodine was transported as aerosols and about 57% i.i. reached the vessel, which is slightly less than in FPT0/FPT1 (about 63% i.i.) [Clément et al., 2006; Girault et al., 2006; Jacquemain et al., 1999].

**Iodine behavior in the containment vessel**

During the degradation phase, iodine that reached the vessel was deposited mainly through gravitational settling on the bottom of the vessel and in the sump (74% of vessel inventory), while the rest was either entrained through diffusiophoresis to the condensers (11% of vessel inventory), or deposited on the vessel walls (11% of vessel inventory). This behavior is typical of aerosols. The solubility of iodine in the sump (approx. pH 9) changed during the course of the test. Before the hemispheric bottom of the sump was washed, the iodine collected in the sump appeared primarily as soluble species or colloidal suspensions. The iodine brought in during the washing phase generates an insoluble iodine fraction, reducing the soluble iodine fraction to 66%. This phenomenon can be explained by two mechanisms:

- during the washing phase, soluble iodine and insoluble elements (silver particles) were added simultaneously to the sump; iodine was then able to react with these elements to form insoluble iodine species;
- the iodine species deposited on the hemispherical vessel bottom may then have reacted chemically with other elements present in the containment atmosphere to form insoluble iodine species, which were then entrained to the sump during the washing phase.

In the chemistry phase of FPT2, the soluble form of the iodine fraction was much higher than in the previous tests (iodine was almost always insoluble in FPT1). This difference can be explained by the lower Ag/I molar ratio (only 10, whereas it was 50 in FPT1 and 2,000 in FPT0), slowing down the kinetics of the reaction between Ag and I, and by the alkaline state, which did not favor the formation of the insoluble specie AgI [Funke et al., 1996].

In FPT2, the gaseous fraction of iodine measured in the vessel reached its peak (about 0.3% i.i.) of the iodine mass initially present in the bundle), during the second oxidation phase, whereas this peak was reached in the first oxidation phase in
FPT0 and FPT1. Changes in the gaseous iodine fraction in the containment atmosphere over the long term are shown in Figure 7.

As in the previous tests, a sharp drop in the gaseous iodine fraction in the containment atmosphere was observed after the second oxidation phase (from 0.3% i.i. to 0.1% i.i.), suggesting that iodine was efficiently trapped on the cooled painted surfaces. During the aerosol phase, the gaseous iodine fraction reached a plateau of about 0.1 to 0.15% i.i., probably corresponding to a physicochemical iodine equilibrium reached inside the vessel. After the washing phase, the gaseous iodine fraction decreased by about one order of magnitude, reaching 0.01% i.i. at the end of the chemistry phase. This decrease was not observed in FPT1, where the gaseous iodine fraction was doubled after the washing phase, reaching equilibrium at about 0.2% i.i. The difference is attributed to alkalinity in the sump (approx. pH 9) and the evaporating conditions chosen for FPT2, whereas FPT1 took place with a non-evaporating, acid sump (approx. pH 5).

During the chemistry phase, a decrease in the iodine deposits on vessel surfaces was observed, where about 0.9% i.i. and 0.2% i.i. of the iodine was desorbed from the vessel walls and the condenser surfaces, respectively. In the latter case, the desorption mechanism was not related to gaseous desorption, but was most likely the result of condensation washing the condensing surfaces, since this decrease in deposition was also observed for Mo and Te, present only in aerosol form. In FPT0, FPT1, and FPT2, the gaseous iodine fraction during the chemistry phase was low (less than 0.1% i.i.) because it was retained in the sump.

In FPT0 and FPT1, the trapping mechanism was explained by the presence of silver (mainly from the absorber rod), whose effect was enhanced by the acid pH. In FPT2, consumption of gaseous iodine was enhanced, first, by the increase in mass transfer between the sump and the containment atmosphere (favored by an evaporation/condensation cycle of 0.98 g/s), and second, by the hydrolysis reactions in the sump, under alkaline (approx. pH 9) and high-temperature (120°C) conditions.

Throughout FPT2, the gaseous iodine fraction was predominantly inorganic (80 to 90% of total gaseous iodine), in contrast to FPT0 and FPT1, where organic iodine was the major component.

### Outlook

On an overall basis, the FPT2 objectives were reached satisfactorily [Gregoire et al., 2008] and the numerous experimental results obtained either confirmed experimental observations made during previous tests (for example, the iodine trapping effect produced by silver in the sump due to the formation of insoluble AgI), or contributed new findings (such as the discovery that CsI is not the primary iodine species transported through the circuit). FPT3 [Biard et al., 2007], conducted successfully from November 18 to 22, 2004, brought the PHÉBUS-FP experimental program to a close. The special feature of the FPT3 test was that boron carbide was used as the boron absorber, instead of the silver-indium-cadmium material. Experimental studies on severe accidents in nuclear water reactors will continue at IRSN with the international Source Term program [Clément and Zeyen, 2005b], consisting of separate-effect testing.

To assess environmental release in the event of a severe accident, these results must be transposed to a power reactor scenario investigated using numerical simulation tools. This work is currently under way.
References

Mephista: A Thermodynamic Database on Nuclear Fuel developed for and applied to nuclear safety

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Nuclear fuels are complex materials, subjected to specific environmental constraints (irradiation in normal operation, oxidizing environment and high temperature in accident conditions, etc.). To understand their behavior in normal operation and in accident conditions, it is necessary to call on several disciplines that include thermodynamics, solid physics, mechanics, and others.

Among these fields, thermodynamics plays a very particular role. It provides knowledge on the arrangement of the atoms of a material (in other words, its phases at the thermodynamic equilibrium), the manner in which this arrangement is modified depending on parameters, such as composition and temperature, and subsequently, the conditions in which a transformation may occur in a given direction. Of course, it does not say anything about the transformation mechanisms themselves, or their duration, and thus says nothing about kinetics of this transformation. But it is important to make a distinction between those transformations that are possible, and those that are not, which is what thermodynamics is capable of achieving with certainty. From this point of view, thermodynamics is the focal point of all the disciplines called on to describe fuel behavior.

Investigating the transformations of a nuclear fuel in terms of thermodynamics only makes sense if the object of the study covers both the elements contained in the fuel (or fuel matrix, for example uranium and oxygen for the uranium dioxide fuel used in pressurized water reactors, PWRs) and the fission products generated by irradiation. It is essential to understand the thermodynamics of the entire system for various reasons related not only to fuel safety (as in the case of cladding damage caused by a chemical interaction between cladding and the irradiated fuel in a PWR component), but also fuel performance (such as the impact of the different phases of irradiation on fuel properties).

Under normal irradiation conditions in a PWR, fission products are found inside the fuel matrix in different phases and different physical states (condensed or gaseous) [Kleykamp, 1985]:
- in the form of oxides dissolved in the matrix for nearly half of them: Sr, Y, Zr, La, Ce, Nd, etc.;
- as oxide phases: Ba and Nb;
- as metallic phases: Mo, Ru, Tc, Pd, Rh;
- in gaseous form: Br, Rb, Te, I, Cs;
- in the form of atoms dissolved in the matrix or fission gas bubbles: Xe and Kr.
Difficulties arise due to the fact that the chemical nature of certain fission products, especially those belonging to the first three categories, is not permanent and can change depending on the operating temperature, the oxygen potential in the fuel, and the burnup.

In a severe accident (involving partial melting of fuel matrix components), the role of fission products must be examined from a different angle. The most volatile (Xe, I, Cs, Te) are released from the reactor core when it is degraded, and form part of the radioactive substances that may potentially be released from the facility. Other less volatile fission products (Zr, La), some of which release heat, may remain trapped inside the fuel and contribute to heating the core, thereby aggravating core degradation. In this situation, a thermodynamic study of the fuel must include not only the chemical reactions between the fuel and fission products, but also any possible interaction between the degraded fuel and its cladding (and certain structural materials, as necessary), for various gaseous atmospheres representative of accident scenarios, and for temperatures that may range up to the fuel melting point.

It quickly becomes apparent that this presents an extremely complex chemical system, especially in terms of scope (i.e. the number of chemical elements to be considered), and the possible associations that may be formed between the various chemical elements (through the formation of compounds, for example). It is therefore necessary to work with a thermodynamic model that covers a sufficient number of chemical elements to accurately describe the thermodynamic behavior of each one.

**Approach to building thermodynamic databases**

Conventionally, thermodynamic knowledge of a material is ascertained by establishing a phase diagram, i.e. a graphic representation of the arrangement of the atoms of the material, which generally depends on its composition and temperature. The phase diagram is determined experimentally by measuring different properties (phase-change temperature, phase composition after quenching, etc.). Compendiums list these diagrams, established experimentally for simple materials, binary systems (i.e. compounds formed by two chemical elements, as in [Hansen and Anderko, 1991]), and certain ternary systems (three elements). For fuel, the task is much more complex, given the large number of chemical elements to be considered and the wide temperature range to be covered. It is easy to understand that an experimental approach alone cannot meet this challenge, even if it remains essential, as will be seen.

Another approach consists of building a phase diagram through calculation. The most fundamental approach consists of writing the thermodynamic partition function (Z) representative of the microscopic properties of the material, and studying the singularities of Z to determine the phase boundaries. Z provides access to all the thermodynamic quantities (for example, the Gibbs energy is written \( G = -kT \ln Z \) at the thermodynamic limit).

Calculation (even approximate) of a partition function is certainly the most difficult problem to solve in thermodynamics. It requires having the appropriate model to express the internal energy of the system (generally based on its electronic structure) and an approximation to calculate entropy (mean field, Monte Carlo simulation, etc.) [Finel, 1987; Barrachin, 1993]. This methodology, already quite complex for a binary system, quickly becomes unworkable when considering materials consisting of more than three chemical elements.
For complex materials, the selected approach consists of determining the macroscopic thermodynamic behavior of each phase that may appear in the phase diagram. For a constant pressure phase diagram, this behavior may be described using the Gibbs energy, \( G \). The mathematical form of \( G \) for a given phase is generally a function of composition and temperature. To write the function explicitly, it is usually necessary to call on models designed to describe the microscopic reality of the material as closely as possible, while being careful to express \( G \) in a relatively simple manner.

The coefficients used in these models are determined from experimental data, or based on assumptions when data is not available. Given the Gibbs energies for the different phases, it is possible to determine the "Gibbs energy function" for all the composition and temperature domains, and by minimizing this functional, to obtain the phase stability domains, i.e. the phases that may occur for a given composition and temperature.

This method, called the CALPHAD method (CALculation of PHAse Diagram), described for the first time by [Kaufman and Bernstein, 1970] (Figure 1), at the end of the 1960’s, can be used to merge the phase equilibrium thermodynamics of J. W. Gibbs and numerical calculation. This mixed approach, based on both calculation and experimentation and widely used to describe complex materials, was employed to describe fuel thermodynamics in the MEPHISTA database.

Contrary to the "fundamental" approach described previously, the CALPHAD method is only partially predictive, since construction of the Gibbs energies for the various phases is based on adjusting models to reproduce experimental results. It nonetheless offers the advantage of being able to predict the thermodynamics of a complex material based on modeling of the binary and ternary systems alone, making it a very powerful method. It can also be used to integrate experimental data deduced from phase diagrams (mainly stability domain boundaries and transition temperatures) and data obtained by measuring thermodynamic quantities (enthalpy of formation, activity, chemical potential, etc.) in a single function (\( G \)), thereby ensuring consistency between these various sources of information. It is nonetheless important to remember that these thermodynamic quantities unequivocally determine the energy level in the Gibbs energies of the various phases, which cannot be achieved using only the topology data from the phase diagram. This is a very important point to be considered when attempting to model the thermodynamics of a complex material. From this point of view, it is the abundance of experimental data, especially measured thermodynamic quantities, that determines the significance level assigned to the thermodynamic model of the material in question. For the MEPHISTA database, this significance level translates into a quality level associated with the model of each binary and ternary system.

**Models to describe Gibbs energy in different phases**

To set the CALPHAD method to work, it is first necessary to describe how Gibbs energy, \( G \), in the different phases depends on temperature and composition. This dependence is a function of the type of phase: a distinction is made between pure elements (U, O, etc.), stoichiometric compounds (\( U_3O_8 \), \( ZrO_2 \), etc.) and solutions (solids, for example \( UO_2^{±x} \), or liquids).

For elements and stoichiometric compounds, the fundamental thermodynamic quantities are the standard enthalpy of formation (\( \Delta H^f_0 \)), entropy at ambient temperature (\( S^0_{298,15k} \)) and the heat capacity as a function of temperature at constant pressure (\( C_p(T) \)). Based on this data, \( G \) can be calculated using the following classical equations:

\[
G(T) = \Delta H(T) - TS(T)
\]

where:

\[
S(T) = S^0_{298,15k} + \int_{298,15k}^T \frac{C_p(T)}{T} dT
\]

\[
\Delta H(T) = \Delta H^f_0 + \int_{298,15k}^T C_p(T) dT
\]

The analytical form used to express heat capacity at constant pressure (\( C_p(T) \)) leads to a general equation for \( G(T) \) expressed as:

\[
G(T) = a_0 + a_1 T + b_1 T \ln T + a_2 T^2 + a_3 T^3 + a_4 T^4 + ...
\]

For solutions that may be stable over a wide concentration domain, \( G \) is developed as a function of temperature \( T \) and composition \( c = (c_i)_{i=1,N} \) (\( N \) being the number of constituents in the solution).

The double dependence of \( G \) is generally taken into account by calculating the sum of two terms:

\[
G(c,T) = G^\text{ref}(c,T) + G^\text{mix}(c,T)
\]

The first term, \( G^\text{ref} \), is the weighted sum of the Gibbs energies of the solution constituents. \( G^\text{mix}(c,T) \) is the sum of the Gibbs energies of the solution constituents.
2.4

Nonetheless, each of these contributions can take a noticeably different form, depending on the model.

To describe the thermodynamics of oxide fuels, it is particularly important to carefully choose the models relative to the solution phases present in the O-U binary system (the principal system to be modeled). Uranium dioxide, UO₂, crystalizes in the fluorite-type cubic structure (CaF₂) (Figure 2).

Atom ordering in the non-stoichiometric solid solution UO₂±ₓ can be described using a three-sublattice model with defects (see [Sundman and Agren, 1981]), the defects making it possible to cover the entire solution composition domain using the same model. The first sublattice contains the uranium atoms, the second the oxygen atoms and vacancy-type defects used to describe the sub-stoichiometric composition domain (UO₂-x), and the last sublattice contains interstitial oxygen atoms for UO₂+x. This representation does not correspond strictly to microscopic reality as described by [Willis, 1964], but it makes it possible to represent the experimental data obtained to establish the phase diagram (Figure 3) and the measured thermodynamic quantities (Figure 4), which are quite abundant for the U-O system.

For the liquid solution, Gibbs energy is calculated using the associated model, developed by [Prigogine, 1950], which assumes non-ideal mixing between the uranium and oxygen atoms and the uranium dioxide molecules. This description covers the entire composition domain, from the liquid solution of the metal (pure uranium) to the over-stoichiometric oxide (UO₂+x).

The second term, \( G_{\text{mix}}(c,T) \), corresponds to the solution’s Gibbs energy of mixing. The difficulty lies in estimating this mixing term, for which there are different models, each new model being developed to improve the reliability of the extrapolation of \( G \) to temperature and composition domains for which no experimental data is available.

In general, the Gibbs energy of mixing is expressed as the sum of two terms, the first corresponding to the Gibbs energy of ideal mixing (mixing “without excess interaction”\(^{(1)}\) between the different constituents in the solution, i.e. the perfect gas hypothesis), the second expressing the difference between the Gibbs energy of mixing and this hypothetical ideal (referred to as the “excess Gibbs energy of mixing”):

\[
G_{\text{mix}}(c,T) = G_{\text{mix}}^\text{ref}(c,T) + G_{\text{ex}}^\text{mix}(c,T)
\]

where:

\[
G_{\text{mix}}^\text{ref}(c,T) = RT \ln c
\]

The excess Gibbs energy of mixing is usually approached using polynomials given by [Redlich and Kister, 1948]:

\[
G_{\text{ex}}^\text{mix}(c,T) = \frac{1}{2} \sum_{i=1}^{n} \sum_{j=1}^{n} c_i c_j L_{ij}(T)(c_i - c_j)^2
\]

The above expressions are very general, making it possible to separate the different contributions (reference, ideal, and excess).

\(^{(1)}\) “Without excess interaction” means that the interaction force between atoms of a different type is about half the sum of the interaction forces between atoms of the same type.
To give an idea of the enormous task accomplished, building the MEPHISTA database required modeling the Gibbs energy of 49 solution phases, 219 pure elements or condensed stoichiometric compounds, and 151 gaseous compounds.

The model can process irradiated fuel thermodynamics for products as varied as uranium dioxide (\(\text{UO}_2\)) and the MOX fuels used in PWRs, or TRISO particles (\(\text{UO}_2\) or \(\text{UO}_2\)-\(\text{UC}_2\) fuel) which are expected to fuel 4th generation high-temperature reactors.

The MEPHISTA database also serves to develop applications for carbide fuels that may be used in gas-cooled fast reactors (GFR) and for oxide fuels used in sodium-cooled fast reactors (SFR).

In the coming years, the MEPHISTA database will be enriched with chemical elements to complete the list of fission products, resulting in a more detailed description of oxide fuel thermodynamics, especially with regards to fission products involved in what is referred to as the metal phase of Pd-Tc-Ru-Mo-Rh.

The database may also be extended to include doping elements currently planned for use in oxide fuels, such as gadolinium and chromium.

MEPHISTA thermodynamic database

The MEPHISTA database (Multiphase Equilibria in Fuels via Standard Thermodynamic Analysis) [Cheynet and Fischer, 2006] has been under development since 2000, and aims to describe fuel thermodynamics. This work is being conducted through a collaborative project between Thermodata, INPG (French National Physics Institute of Grenoble), CNRS (French National Scientific Research Center), and IRSN. The database contains the Gibbs energy coefficients of phases that may be formed in the O-U-Pu-Zr-Fe-Si-Cs-La-Mo-Sr-Ru chemical system. Oxygen (O), uranium (U) and plutonium (Pu) are the elements used to produce oxide fuels; zirconium (Zr, used for fuel cladding, but also considered as a fission product), barium (Ba), cesium (Cs), lanthane (La), molybdenum (Mo), strontium (Sr), and ruthenium (Ru) are the fission products that are important in describing the thermodynamics of irradiated fuel.

In the MEPHISTA database, iron (Fe) is included to describe interactions between the fuel rod and certain structural materials (reactor internals, control rod cladding, etc.) in accident conditions. The chemical elements silicium (Si) and carbon (C) were added to study the thermodynamics of TRISO fuel on high-temperature reactors [Phélip et al., 2006]. Lastly, argon and hydrogen are included only in modeling the Gibbs energy of gaseous compounds.

To give an idea of the enormous task accomplished, building the MEPHISTA database required modeling the Gibbs energy of 49 solution phases, 219 pure elements or condensed stoichiometric compounds, and 151 gaseous compounds.

The model can process irradiated fuel thermodynamics for products as varied as uranium dioxide (\(\text{UO}_2\)) and the MOX fuels used in PWRs, or TRISO particles (\(\text{UO}_2\) or \(\text{UO}_2\)-\(\text{UC}_2\) fuel) which are expected to fuel 4th generation high-temperature reactors. The MEPHISTA database also serves to develop applications for carbide fuels that may be used in gas-cooled fast reactors (GFR) and for oxide fuels used in sodium-cooled fast reactors (SFR).

In the coming years, the MEPHISTA database will be enriched with chemical elements to complete the list of fission products, resulting in a more detailed description of oxide fuel thermodynamics, especially with regards to fission products involved in what is referred to as the metal phase of Pd-Tc-Ru-Mo-Rh.

The database may also be extended to include doping elements currently planned for use in oxide fuels, such as gadolinium and chromium.
Programs to support MEPHISTA database development

As mentioned previously, the quality of the CALPHAD approach depends primarily on the experimental measurement data available to determine the Gibbs energy coefficients. Most metallic binary systems have been studied experimentally, but data on oxide binary systems, particularly those containing uranium and plutonium oxides, are not as extensive. The amount of experimental data on ternary and quaternary systems is even less.

For more than ten years, a vast experimental effort has been undertaken through various national and international programs to fill in these gaps. IRSN is extensively involved in these programs, which include CIT (a European Commission (EC) project), COLOSS (EC) (example of determination of liquidus and solidus temperatures in the UO$_2$-ZrO$_2$ system, Figure 5), RASPLAV (an OECD/CSNI$^{(2)}$ project), MASCA (OECD/CSNI), MASCA2 (OECD/CSNI), ENTHALPY (EC) and CORPHAD (an ISTC$^{(3)}$ project). Particular focus has been given to the U-O-Zr-Fe quaternary system [Chevalier et al., 2004; Bechta et al., 2006; Bechta et al., 2007], a key system for describing fuel rod degradation in accident conditions.

This effort is being pursued today in the ISTC PRECOS program, in which IRSN plays an important role. The MEPHISTA database is gradually incorporating new experimental data obtained through the above programs, as well as information published in open technical literature, by performing regular updates on coefficients used to express Gibbs energy, thus contributing to a constant improvement of the significance level. In this aspect, it is a unique tool for building knowledge on nuclear fuel thermodynamics.

In spite of these continuous efforts, there are composition and temperature domains for which there is no experimental data. Phase-equilibrium thermodynamics being a relatively old discipline (dating back to the 19th century), whatever has not been accomplished so far is not easy to achieve. Calling on modeling hypotheses to build Gibbs energy data for certain phases can open a new road to this information. In this context, it is important to confront these hypotheses with other alternatives. This confrontation can take place by comparing thermodynamic databases. In 2003, as part of the ENTHALPY program [Debremaecker et al., 2003], the MEPHISTA database was compared with the THMO database established by AEA-Technology (Great Britain), with the aim of reaching a consensus on modeling hypotheses [Cheynet et al., 2004]. There are plans to pursue this exercise in the context of the SAMANThA$^{(4)}$ collaborative network (currently being organized), with the database developed at the Royal Military College of Canada.

Another source of information consists of the results obtained from ab initio calculations, which involve resolving quantum mechanics equations. Without entering into a detailed theoretical discussion, it should be noted that this approach can be applied to obtain values on the enthalpy of formation ($\Delta H_{f,0}$) for stoichiometric compounds to a satisfactory degree of accuracy. These values can contribute to the determination of coefficients used to express Gibbs energy (equations (a) and (b)). For example, Table 1 shows a comparison of the standard enthalpies of formation obtained by calculating the electron structure and measured enthalpy values for certain stoichiometric compounds of cesium.

### Table 1: Comparison of standard enthalpies of formation obtained by calculating the electron structure (F. Gupta, 2008) and measured enthalpy (Cordfunke and Konings, 1990) (in kJ/mol).

<table>
<thead>
<tr>
<th>Compound</th>
<th>Calculated</th>
<th>Measured</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cs$_2$MoO$_4$</td>
<td>-1664,9</td>
<td>-1514,5 ± 1.0</td>
</tr>
<tr>
<td>Cs$_2$UO$_4$</td>
<td>-1879,9</td>
<td>-1928,2 ± 1.0</td>
</tr>
<tr>
<td>Cs$_2$ZrO$_3$</td>
<td>-1587,9</td>
<td>-1584,8 ± 1.0</td>
</tr>
</tbody>
</table>

Applications in nuclear safety

The MEPHISTA database and the NUCLEA database, specifically dedicated to corium thermodynamics, are used by a large number of institutes, industrial partners, and universities, including EDF, CEA, Areva, VTT (Finland), AEAT-T (Great Britain), Alexandrov RIT (Russia), KAERI (South Korea), AECL (Canada), Risø National Laboratory (Denmark), Boise State University (USA), and others.

At IRSN they are used either coupled to applications developed at the Institute to simulate core degradation in accident conditions (for example, the ICARE/CATHARE code [Salay and Fichot, 2005]), or to support interpretation of various test results (such as those from tests conducted in the OECD MASCA programs [Barrachin and Defoort, 2004]).

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$^{(2)}$ CSNI: Committee on the Safety of Nuclear Installations.

$^{(3)}$ ISTC: International Science and Technology Center.

$^{(4)}$ The SAMANThA collaborative network (Simulation by Advanced Mechanistic and Thermodynamic Approaches to nuclear fuels), coordinated by IRSN, aims mainly to develop detailed models describing fission product behavior in nuclear fuel.
The following discussion presents two recent applications of the MEPHISTA database, the first involving interpretation of test results from the PHÉBUS-FP program, focusing on estimation of the temperature at which loss of integrity occurs on fuel rods under accident conditions in a pressurized water reactor [Barrachin et al., 2008], the second having a more predictive objective, since it is dedicated to fission product behavior in TRISO fuel, which may be used in 4th generation very-high temperature reactors (VHTR) [Barrachin et al., 2008].

Fuel failure temperature under severe accident conditions

In a hypothetical core meltdown accident in a pressurized water reactor caused by loss of coolant and unavailable safeguard systems, interaction between the Zircaloy-4 cladding and the fuel (UO₂) is the main mechanism leading to fuel rod degradation, thus core degradation. Fuel can be subjected to various gaseous environments (steam and hydrogen), which determine the type of interaction between the cladding and the fuel, and the temperature at which fuel integrity will be lost. Fuel exposure conditions change throughout the course of an accident scenario: fuel may be exposed to temperatures ranging from 900 K to 2,800 K, and to varying atmospheres, from a high-oxidation environment (steam in excess), to a high-reduction environment (rich in hydrogen).

In a reducing environment, the chemical affinity of Zircaloy for oxygen will extract oxygen from the fuel, partially dissolving the fuel through reduction. This interaction between cladding and fuel begins at 1,273 K, when the materials are in contact, which is generally the case for an irradiated fuel rod. At 2,023 K, liquefaction of the cladding accelerates the interaction process which ultimately leads to loss of fuel integrity at a temperature of approximately 1,000 K, below the UO₂ melt point (3,120 K).

In a high-oxidation environment, the phenomenology is considerably different. Cladding exposure to this type of atmosphere reduces the interaction process described previously, since the cladding is quickly oxidized by the ambient environment, and interaction between the oxidized cladding (ZrO₂) and the fuel (UO₂) only begins at very high temperature (2,800 K, according to the UO₂-ZrO₂ phase diagram), leading to “late-phase” fuel liquefaction.

This understanding of fuel degradation phenomena as a function of the ambient atmospheric conditions prevailed for a long time, until the conclusions established after the first two tests conducted in the PHÉBUS-FP Program (FPT0 and FPT1), dedicated to studying core meltdown in an accident situation on a PWR [Clément et al., 2003; Jacquemain et al., 2000]. These tests, conducted in an oxidizing atmosphere (at a pressure of 0.2 MPa) with UO₂ fuel, unirradiated for FPT0, irradiated to 2.4 at % for FPT1, revealed that fuel collapse occurred near 2,500 K, i.e. 300 K lower than what was generally expected.

Analysis of fuel composition (solid solution composed of \(\text{U}_{0.86}\text{Zr}_{0.12}\text{Fe}_{0.05}\text{Cr}_{0.01}\text{Nd}_{0.006}\text{Pu}_{0.004}\text{Ce}_{0.004}\text{O}_{2.08-2.11}\)) after FPT1 showed that the fuel had, in fact, reacted with the oxidized cladding, as well as with structural materials (such as iron in an oxidized state), and that even the fuel itself had oxidized. To determine the liquefaction temperature of this material, and the relative influence of the various effects (irradiation, cladding and structural materials, fuel oxidation) on this temperature, the MEPHISTA database naturally appeared as an appropriate tool.

All the experiments conducted on irradiated uranium dioxide show that fission products have only a slight effect on its liquefaction for burnup fractions up to 5 at % [Bates, 1970; Yamanouchi et al., 1988]. The presence of fission products therefore cannot be put forward to explain why the fuel collapse temperature was lower in FPT0 and FPT1.
2.4

Accidents in nuclear facilities

Evaluations reveal the role that fuel oxidation could play in fuel liquefaction under core meltdown accident conditions, and the need to introduce this mechanism in computer codes that simulate core degradation to improve quantification of the material mass that flows to the bottom of the reactor as a function of time, and thereby assess the risks of ex-vessel progression.

The very-high temperature reactor (VHTR) is one of the technological options being studied in the development of 4th generation nuclear reactors. This type of reactor uses thermal neutrons, is cooled with gaseous helium, and has a graphite moderator. The fuel, which reaches temperatures ranging from 1,273 to 1,473 K in normal operation, is based on a special technology employing UO₂ particles (called “TRISO particles”) less than one millimeter in diameter, coated with different layers of material, each having its own specific function.

In the calculations, occurrence of the liquid phase for the above composition is related to modeling of the various phases (liquid [L], overstoichiometric solution [F] and gas [G]) at high temperature in the U-O binary system (Figure 6) and extrapolation of this model to the composition domain bounded by UO₂, ZrO₂ and O in the U-O-Zr ternary system (Figure 7). Modeling of the U-O binary system takes into account the oxygen potential of the UO₂+x overstoichiometric solution [Chevalier et al., 2004], and recent measurements performed by [Manara et al., 2005] on liquefaction of UO₂+x solid solutions. These two sources of information, which unequivocally set the Gibbs energies of the various phases at high temperature, support the validity of the described thermodynamic calculations. These evaluations reveal the role that fuel oxidation could play in fuel liquefaction under core meltdown accident conditions, and the need to introduce this mechanism in computer codes that simulate core degradation to improve quantification of the material mass that flows to the bottom of the reactor as a function of time, and thereby assess the risks of ex-vessel progression.

Behavior of fission products in TRISO fuels planned for use on high-temperature reactors

The very-high temperature reactor (VHTR) is one of the technological options being studied in the development of 4th generation nuclear reactors. This type of reactor uses thermal neutrons, is cooled with gaseous helium, and has a graphite moderator. The fuel, which reaches temperatures ranging from 1,273 to 1,473 K in normal operation, is based on a special technology employing UO₂ particles (called “TRISO particles”) less than one millimeter in diameter, coated with different layers of material (Figure 8), each having its own specific function.

![Figure 7](image.png)

**Figure 7** UO₂-ZrO₂-O phase diagram at Ptot = 0.1 MPa at 2,573 K (left) and 2,673 K (right).

<table>
<thead>
<tr>
<th>Composition</th>
<th>T_L(K)</th>
<th>T_S(K)</th>
<th>Composition</th>
<th>T_L(K)</th>
<th>T_S(K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(U₀.8₈Zr₀.1₂)O₂₀₀₀</td>
<td>3,080</td>
<td>3,020</td>
<td>(U₀.8₈Zr₀.1₂)O₁₀₆₂₁</td>
<td>2,980 - 2,960</td>
<td>2,760 - 2,660</td>
</tr>
<tr>
<td>(U₀.8₇Zr₀.1₂F₀.₇₀)O₂₀₀₀</td>
<td>3,060</td>
<td>2,860</td>
<td>(U₀.₈₇Zr₀.₁₂F₀.₀₁)O₂₀₈₂₁</td>
<td>2,960 - 2,920</td>
<td>2,560 - 2,460</td>
</tr>
</tbody>
</table>

**Table 2** Liquidus (T_L) and solidus (T_S) temperatures, calculated by MEPHISTA for compositions representative of those measured after FPT1.
and only a few measurements are available. Estimates were made by [Lindemer and Nordwall, 1974], for particles irradiated to 6 at %, that indicate values between –480 and –380 kJ/mol at 1,450 K, and between –500 and –380 kJ/mol at 1,873 K.

Thermodynamic calculation of CO and CO₂ generation depends on modeling of the U-C-O ternary system. In MEPHISTA, this model is based on modeling of both the U-O binary system (Figure 3) and U-C binary system (Figure 9), as well as modeling of the ternary phases, based on a bibliographic review [Potter, 1972]. MEPHISTA calculates the CO pressures measured above the different composition domains of the U-C-O ternary system [Gossé et al., 2006] without any preliminary adjustments, thereby correctly validating the thermodynamic model.

Figure 10 shows the change in carbon monoxide and carbon dioxide (CO and CO₂) pressures in the free space of the TRISO particle buffer at 1,450 K and 1,873 K as a function of the oxygen potential. The temperatures selected correspond to current VHTR design-basis data for normal operating conditions and the maximum admissible temperature in accident situations. Calculations show that the pressure leading to failure of the SiC layer (estimated at 100 MPa) is obtained for an oxygen potential between –330 and –325 kJ/mol.

This value, measured on UO₂ fuel of a PWR reactor for a burnup fraction of about 9 at % [Walker et al., 2005], should be reached by VHTR fuel at higher burnup, given the respective conditions.
these stoichiometric compounds come from measurements taken between 670 K and 1,070 K \cite{Salzano and Aronson, 1965; Salzano and Aronson, 1966}.

At higher temperatures (corresponding to an accident situation), decomposition of these compounds could result in cesium release from the buffer. Based on the thermodynamic data and current understanding of the type of chemical bonds in these compounds, an approach was developed by \cite{Salzano and Aronson, 1967} to predict the conditions in which cesium carbides may be formed or broken down.

Using this approach shows that at a temperature of 1,873 K, cesium should not bind with carbon. It therefore should contribute to gaseous release from the TRISO particle. This point is currently being studied within the context of the European RAPHAEL Program \cite{Phélip et al., 2006}.

The MEPHISTA database can serve to study the chemical form of cesium in the TRISO particle, which has a crucial influence on cesium release in normal operation and in accident conditions. Thermodynamic calculations show that in the presence of carbon (from the buffer), for an oxygen potential less than –385 kJ/mol, the formation of cesium carbides $C_nCs$ ($n = 8, 10, 24, 36, 48, 60$) may occur at 1,450 K, consequently trapping cesium in the buffer (Figure 11).

The stability of these compounds, however, was only studied at relatively low temperatures. The thermodynamic data from the MEPHISTA database used to build the Gibbs energies of these stoichiometric compounds come from measurements taken between 670 K and 1,070 K \cite{Salzano and Aronson, 1965; Salzano and Aronson, 1966}.

The contribution of the other volatile fission products to total pressure is much smaller. Nonetheless, examination of TRISO particles after irradiation \cite{Minato et al., 1994} showed that while the fissile kernel generally retained rare earth elements (La, Nd, etc.) and part of the alkaline earth elements (Ba, Sr), certain fission products were capable of diffusing to the buffer and becoming trapped (Cs), even migrating outside the particle (for certain metal fission products such as Ag), with no explanation found for these migration mechanisms.

The MEPHISTA database can serve to study the chemical form of cesium in the TRISO particle, which has a crucial influence on cesium release in normal operation and in accident conditions. Thermodynamic calculations show that in the presence of carbon (from the buffer), for an oxygen potential less than –385 kJ/mol, the formation of cesium carbides $C_nCs$ ($n = 8, 10, 24, 36, 48, 60$) may occur at 1,450 K, consequently trapping cesium in the buffer (Figure 11).

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Conclusions and outlook

Initiated by IRSN, development of the MEPHISTA thermodynamic database began eight years ago, in cooperation with Thermodata, INPG and CNRS. Today it contains the model of a relatively vast chemical system that can be used to study a large number of thermodynamic issues involving either oxide or carbide irradiated fuels. Future modeling work aims to explore these subjects in two main directions:

- extension of the MEPHISTA database to include other elements, such as fuel doping elements, to obtain a more precise description of how oxygen potential changes as a function of irradiation, a key thermodynamic parameter for determining the chemical form of fission products;
- analysis and incorporation of experimental data resulting from various experimental programs in progress (such as ISTC PRECOS).

Lastly, researchers envisage using the MEPHISTA database to support the development of thermodynamic models adapted to the MFPR (Module for Fission Product Release) computer code, covering the behavior of fission products and the microstructure evolution in irradiated fuels, developed at IRSN in cooperation with IBRAE (Russia).

Acknowledgements

The author would like to thank B. Cheynet (Thermodata) and E. Fischer (INPG) for the thermodynamic modeling work they have conducted over the years, including the MEPHISTA database and NUCLEA database, dedicated to corium thermodynamics. He also thanks his colleagues at IRSN, R. Dubourg and M. Kissane, for their cooperation on work involving the behavior of fission products in TRISO fuel, and F. Gupta for contributing to this overview.
Studies examining the impact of an aircraft crash on the safety of a nuclear facility are carried out by facility operators and are assessed by IRSN. They focus mainly on the mechanical behavior of civil engineering structures subjected to impacts, and seek to assess the consequences in terms of facility safety requirements.

Up until the 1990’s, these studies primarily covered specific hazards involving moderate degrees of energy (a light touring airplane, military aircraft) and used empirical methods to study structural behavior (slab perforation).

In the last few years, the definition of the hazards to be considered has changed (more extensive and powerful impacts), and the computing power of software and servers has advanced considerably, making it feasible to take into account interaction between the missile and the target in a transient state through fast dynamic analysis.

Nonetheless, these new computing capabilities must be accompanied by simple and easy-to-model tests to provide an in-depth understanding of the phenomena at work and to validate the simulation tools.

To investigate the impact of commercial aircraft on an EPR reactor, in 2004, the Finnish safety authority (STUK) asked the public research institute VTT (Valtion Teknillinen Tutkimuskeskus), to build a facility for testing missile impact tests on targets (metal plates and concrete slabs). In 2006, IRSN joined this program, in which the first phase being completed in early 2009, and the second phase scheduled from 2009 to 2011.

The purpose of the program is to conduct test analyses using conventional materials resistance methods in order to improve understanding of the physical phenomena observed and enhance numerical simulation capability in this area. The main test parameters have therefore been scaled appropriately for the study of aircraft impact on reinforced concrete structures.

The first test program phase covers the two essential subjects in assessment of civil engineering structural behavior in reaction to impact: first, the missile deformation mechanisms and the corresponding impact loads, and second, the mechanical behavior of the “target” structure subjected to these impact loads.

The first subject was studied through impact tests on deformable metal tubes on “rigid” steel plates instrumented to measure contact loads, as close as possible to the impacted surface. These tests aimed to col-
lect data on a point that is essential in assessing a deformable missile impact on a target, i.e., the contact load exchanged between the missile and the target throughout the duration of the impact.

Since the publication of the method described by Dr. Riera (1) in 1968, this load is generally assessed through calculations, taking into consideration the distribution of mass and crash load along the centerline of the missile. Part of the VTT tests validated the practical choice of these distributions and identified limits in applying the method. The initial results suggest that, under comparable experimental conditions, the missiles filled with water would convey to the target a momentum greater than that conveyed by missiles that do not contain water, which would convey momentum comparable to their momentum drag before the impact.

Consequently, the Riera method would not be strictly applicable to the case of missiles containing a significant amount of liquid. This phenomena, which can significantly affect safety analyses on certain external hazards, must be confirmed by dedicated tests, which will be performed in the second phase of the experimental program.

The second topic was studied in impact tests using the same metal tubes on square reinforced concrete slabs with 2-meter sides, from 15 cm to 25 cm thick. One of the main objectives of these tests was to predict displacement and structural damage in the reinforced concrete structures loaded beyond their yield strength.

These tests revealed the main operating modes of reinforced concrete (bending and shear strength) and contributed to improvements in numerical simulation of structural elements subjected to loads leading to deformation of steel and concrete materials in the non-linear domain, even up to failure. Numerical simulations using a computer code to resolve fast-dynamic mechanical problems (LS-DYNA) had been conducted previously and gave satisfactory results in the case of slabs subjected to bending.

Based on an experimental program to qualify the VTT tests concrete specimens, IRSN would like to see the development of a behavior law specific to concrete subjected to high dynamic loads, to be used in the LS-DYNA and the CAST3M finite-element codes. Moreover, simulations conducted using a simplified slab model can be used to quantify the macroscopic parameters that drive dynamic bending behavior, i.e. slab stiffness, which determines slab vibration frequency and the plastic moment of reinforced concrete sections that determine its final displacement.

Continued pursuit of work to define and interpret these tests should lead to a method for simulating the behavior of civil engineering structures subjected to impact. This method will be applicable to actual structures and will serve to support safety assessments conducted by IRSN.

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SARNET: major achievements and outlook

To achieve these goals, all organizations belonging to the SARNET network contribute to a joint program of activities with the following objectives:

- implement an Advanced Communication Tool to access all project information, promote information exchange, and manage documentation;
- harmonize and re-orient existing research programs and define new programs;
- analyze experiment results generated by research programs to develop a shared view of the corresponding phenomena;
- develop the ASTEC European reference code (an “integral” code used to predict the behavior of nuclear power plants in a severe accident condition).

Fifty-one organizations have pooled their research capacity in SARNET (Severe Accident Research NETwork of Excellence), to work on what are considered as the most important topics for improving safety in existing and future nuclear power plants in terms of severe accident conditions. Co-funded by the European Commission (EC) through the 6th Framework Program, this program was established to optimize the use of available resources and build sustainable research groups in the European Union. SARNET has set out to reverse the fragmented state of the various existing national R&D efforts by defining joint research programs and developing shared methodologies and computer tools for safety assessment. SARNET includes almost all organizations participating in severe accident research in Europe and Canada.
postulated severe accident situation), taking advantage of knowledge generated on physical models within SARNET;
- develop scientific databases that store all research program results in a shared format (DATANET);
- develop a shared methodology for conducting probabilistic safety assessments on nuclear power plants;
- prepare training courses and a textbook on severe accidents aimed at students and researchers;
- promote mobility of personnel between the different European organizations.

This article presents major achievements accomplished in the last four and a half years of networking new knowledge, along with improvements on the ASTEC European reference code, dissemination of results, and integration of research programs conducted by the different partners.

Following this initial period (2004-2008), co-funded by the EC, another four-year contract for the coming years is being negotiated with the EC as part of the 7th Framework Program. During this period, the network’s activities will focus mainly on problems considered as top-priority during the first period; experimental activities will be included directly in collaborative work and the network will evolve towards complete self-sufficiency. The principles of this development plan are presented in the last section of this article.

SARNET background and goals

Nuclear power plants operated in Europe are designed according to the principles of defense-in-depth. They feature robust engineering and containment systems conceived to protect the population from radioactive release as depicted in a complete series of postulated accident scenarios.

Nonetheless, in certain highly improbable circumstances, severe accident conditions may cause core meltdown, leading to plant damage and dispersion of radioactive substances in the environment, thereby creating a risk for public health.

In 2004, several studies had already been conducted on severe accidents in water reactors, particularly through numerous European efforts engaged in the European Commission’s 4th and 5th Framework Programs. In spite of these projects, there were still several topics requiring research work to reduce what was considered as significant uncertainty, and consolidate severe accident management plans [Magallon et al., 2005].

Severe accident research programs were (and still are) initiated at the national level. Cooperation agreements are often built around national programs, but on a case-by-case basis. Faced with an inevitable need to reduce national budgets in this area, it was necessary to coordinate research more effectively so as to optimize the human and experimental resources employed towards the improvement of safety on existing and future nuclear power plants.

Consequently, in April 2004, 51 organizations, including technical safety organizations (TSOs), industry, research institutes and universities, decided to take the opportunity offered by the EC in the context of the 6th Framework Program to group their severe accident research capabilities together in a lasting structure that became SARNET (Severe Accident Research NETwork of Excellence). These organizations come from 18 Member States of the European Union (Austria, Belgium, Bulgaria, the Czech Republic, Finland, France, Germany, Greece, Hungary, Italy, Lithuania, the Netherlands, Romania, Slovakia, Slovenia, Spain, Sweden, and the United Kingdom), as well as Switzerland, Canada and joint EU research centers (Figure 1).

![Figure 1 General features of SARNET.](image)
The network benefits from the complementarity that exists between the various partners (experts in fission product materials and chemistry, small-scale/large-scale testing, simulated and real materials, experimenters, model developers, code developers) and reinforces their strengths through synergy [Micaelli et al., 2005].

SARNET aims to achieve broad general goals:
- to counter the fragmented state that exists between the various R&D organizations, in particular by defining joint research programs, and by developing and validating IT tools;
- to harmonize methodologies applied in risk assessment and improve Level 2 probabilistic safety assessment (PSA) tools;
- to disseminate knowledge to newcomers in the European Union more efficiently and promote their involvement in the definition and accomplishment of research programs;
- to bring together preeminent scientists specialized in severe accident research to establish world leadership in IT tools used in severe accident risk assessment;
- to train the next generation of severe accident experts.

This article presents major achievements accomplished in the last four and a half years of networking new knowledge, as well as improvements on the ASTEC European reference code for severe accidents, dissemination of results, and integration of research programs conducted by the different partners.

**Organization**

The SARNET network is organized in a two-level structure. The first level consists of a Governing Board, in charge of strategic decisions, supported by an Advisory Committee. The second level is occupied by the Management Team, responsible for managing the network on an everyday basis.

The Governing Board examines progress made by the network, especially with regards to gradual integration, and makes recommendations on future orientations. It decides how the EC financial contribution will be apportioned and approves the research activity program.

The Governing Board is made up of one delegate from each party to the agreement and an EC representative, who participates as an observer. In general, the members are ranking executives in their home organization and may call up resources from their organization to execute SARNET activities.

The Advisory Committee advises the Governing Board on strategic orientations for SARNET research activities. Its members are composed of executives from the “end-user” organizations, including industry, electrical power operators, and regulatory bodies (who are not necessarily members of SARNET), appointed by the Governing Board.

The Management Team, operating on behalf of the Governing Board, is in charge of the daily administration of SARNET. In keeping with the Joint Program of Activities (see section below), the Management Team consists of the SARNET Coordinator (the team leader), the Advanced Communication Tool Leader, the ASTEC Coordinator, the PSA2 Leader, the Experimental Database Leader, the Severe Accident Research Priorities Leader, three Scientific Coordinators (for corium, containment, and source term research), the Excellence Spreading Coordinator, and a Secretary.

The Management Team monitors progress made in the various activities, works with the relevant project leaders to examine what action can be taken to overcome any problems, reviews new projects, promotes cooperation within the network and with external organizations (OECD, ISTC, IAEA, etc.), makes proposals to the Governing Board to update the program of activities, and circulates information inside and outside the network, particularly by regularly organizing specialized conferences and seminars via the SARNET public website. The Coordinator operates under the supervision of the Governing Board and reports to this authority. He/she presents technical and financial reports to the Governing Board and implements its decisions, in particular by updating the program of activities. The Coordinator is also responsible for relations with the EC which, once a year, organizes a review on the project’s state of progress, carried out by a panel of independent experts.

**Joint Program of Activities**

To meet SARNET goals, a Joint Program of Activities (JPA) is established and updated every year. All members of the SARNET network contribute to the JPA, divided into 20 internal projects (*Figure 2*) that can be grouped into three categories:
integration activities to reinforce relationships between organizations;
- joint research activities to advance knowledge;
- activities to spread excellence and knowledge.

Main accomplishments

After four and a half years of operation, the network can proudly claim the achievements described below.

Integration activities

The integration aspect of the program is given utmost importance and projects in this area are considered as key items in the JPA.

The Advanced Communication Tool (ACT) was implemented to promote communication between project partners and ensure documentation management. ACT is a key step towards meeting SARNET goals: the current technology based on portals establishes a unified system for efficient cooperation across the network by providing:

- access to documents and computer codes, search functionality, and publication (the knowledge base concept);
- contact and communication between partners (interactive and collaborative services);
- coordination of action and programs (cooperative network management);
- links to projects carried out by satellite communities (R&D projects, related sites).

The ACT platform was built based on the MS Sharepoint Portal Server. Access is provided through web browsers, via any Internet connection. Access to this tool has been granted to about 250 SARNET users. ACT is accessed intensively and efficiently, with 500 to 1,000 connections per month and 8,000 items in storage (Figure 3). A public website (www.sar-net.org) also provides essential information on SARNET.

DATANET, the SARNET experiment database network, was developed to preserve, exchange, and process experimental data on severe accidents, including any related documentation. Data comes from existing experimental data that SARNET partners wish to share within the network, and any new data generated under the SARNET program.

WP 9, 10, 11: CORIUM
- Early-phase core degradation
- Late-phase core degradation
- Ex-vessel corium recovery

WP 12, 13: CONTAINMENT
- Hydrogen behavior
- Fast interaction in containment

WP 14, 15, 16: SOURCE TERM
- FP Release and Transport
- Impact of Aerosol Behavior on Source Term
- Impact of Containment Chemistry on Source Term

WP 17: ET
- Education and Training

WP 18: BOOK
- Book on severe accident phenomenology

WP 19: MOB
- Mobility program

WP 20: MANAGEMENT
- Spreading knowledge

**Figure 2** SARNET Joint Program of Activities.
Based on these detailed models, simplified models are then established and introduced in the ASTEC code. Twenty-eight organizations cooperate with IRSN and GRS in developing and assessing this code. ASTEC describes the behavior of an entire nuclear power plant in severe accident conditions, including management of the severe accident situation through procedures and engineering systems. It is used intensively by IRSN to establish Level 2 PSAs for 900 and 1,300 MWe PWRs, and by GRS in consolidating Level 2 PSAs for 1,300 MWe Konvoi reactors. A growing number of partners also call on this code to calculate complete accident scenarios.

Efficient and close cooperation between ASTEC users and developers has been implemented using ACT and the MARCUS web tool for code maintenance. Training courses on using the code have been organized. Three main versions of the code have been released to partners: V1.1 mid-2004, V1.2 mid-2005 and V1.3 at the end of 2006 (with update V1.3 rev2 released at the end of 2007). Code documentation has been improved considerably.
since SARNET began. Three meetings of the ASTEC Users Club, the last one taking place in April 2008, gave users and developers the opportunity to exchange views in straightforward and constructive discussions.

CEA has advanced development of models on corium behavior in the vessel lower head and ex-vessel cooling, including validation through experiments such as LIVE (FZK), SULTAN (CEA) and ULPU (University of Santa Barbara). Other models were proposed or improved after discussion between partners, such as silver-indium-cadmium and ruthenium release from the core, boron carbide (B$_4$C) oxidation, mass transfer of iodine and radiolysis oxidation in containment (see details and other examples in the section below on joint research activities). New improvements have been planned for the near future, such as work on the mechanical resuspension of aerosols in system lines. ASTEC has also been used to prepare and interpret several experiments, including QUENCH and LIVE at FZK. Preliminary calculations of accident scenarios for boiling water reactors and CANDU reactors, performed using current versions of the code, have given promising results. They provided the opportunity to prioritize and specify necessary adaptations in the models.

An advanced code validation process was carried out by the partners on 65 analytical and integral experiments, often involving OECD/NEA/CSNI international standard problems. In general, the results can be considered as satisfactory, and even very satisfactory for plant system thermohydraulics, core degradation, and the behavior of aerosols and fission products. The code has been applied to a large number of severe accident scenarios for different types of nuclear power plants (PWR, Konvoi 1300, Westinghouse 1000, VVER-440, VVER-1000 and RBMK), and has frequently been compared with other codes. Good agreement has been shown with integral codes MELCOR and MAAP4 with regards to trends and orders of magnitude in the results, and very good agreement with results of detailed codes such as ATHLET-CD and ICARE/CATHARE for core degradation. The numerical robustness of ASTEC has improved greatly since SARNET began.

IRSN and GRS now take into consideration all requests expressed by users for upgrading ASTEC as they prepare the new ASTEC V2 series. The first in the series, V2.0, scheduled for release in June 2009, will apply to the EPR plant (especially the corium catcher) and will include most of the detailed models from ICARE2, IRSN’s reference code for core degradation. Harmonizing methodology for Level 2 PSAs also comes under the integration activities. Level 2 PSA is a powerful tool for assessing the specific vulnerability of nuclear power plants to severe accidents. It assesses possible scenarios in terms of frequency, loss of containment integrity, and release of radioactive substances to the environment, and quantifies the contribution of prevention and attenuation measures to risk reduction. Various approaches are used in Europe, derived more or less from those followed in the USA. The principle methods used by the various partners to establish PSAs have been described and compared in a document. For several problems involving Level 2 PSA, a survey was taken and the answers were analyzed to define the next steps to be taken towards harmonization. Case studies were then carried out on specific problems, such as hydrogen combustion, iodine chemistry, corium cooling and corium/concrete interaction, significant early-phase release, the definition of the final stabilized states of the reactor, and the Level 1 and Level 2 PSA interface methods. A certain degree of harmonization has been achieved and recommendations written.

A report on the current state of knowledge on dynamic reliability methods was written and the limits of classical methods that could be overcome by using these reliability methods were identified. The advantages of one of the possible methods (Monte Carlo dynamic simulation of event trees) were examined for a station blackout. Considerable work was carried out in a benchmark exercise (quantification of the risk of containment failure caused by activation of a safety system during the core degradation phase in the reactor vessel). This exercise included a comparison between dynamic reliability methods and classical methods. Work is currently under way to determine exactly what is required in ASTEC to meet the needs of Level 2 PSAs and to couple ASTEC with probabilistic tools.

The internal Severe Accident Research Priority (SARP) project identified research priorities to gradually re-orient existing national programs and contribute to initiation of new programs in a coordinated manner, by eliminating redundancy and developing complementarity. This activity was developed in close cooperation between participants representing technical safety organizations, industry, and public institutions. In the same manner as the Phenomena Identification and Ranking Table (PIRT) established within the European EURSAFE project [Magallon et al., 2005], this activity took into account the complete range of severe accident situations, from core uncovering to long-term corium stabilization, long-term containment integrity, and environmental release of fission products. Special
emphasis was placed on a risk-oriented approach to truly focus on the most pertinent pending problems. Consensus was reached in the final phase and 18 major problems were classified in four categories.

Six subjects were considered as requiring in-depth analysis and were given high priority:
- core cooling by reflooding and debris cooling;
- configuration of the corium pool in the reactor pit during corium/concrete interaction, and top-cooling of corium;
- relocation of the molten pool in water, ex-vessel Fuel/Coolant Interaction (FCI);
- hydrogen mixing and combustion in containment;
- impact of oxidation (Ru oxidation conditions and air penetration for high burnup and MOX fuel elements) on the source term;
- iodine chemistry in the reactor coolant system and the containment.

Four subjects were given medium priority, implying that work will be pursued as planned in the different research programs involved. Risk assessment has been defined through considerable advances in knowledge, but certain issues remain open:
- core cooling in the vessel lower head;
- vessel integrity after ex-vessel cooling;
- direct heating of containment through ejection of pressurized corium after reactor vessel failure.

For five subjects, current knowledge was considered sufficient with regards to risk and safety, and given research in progress. These subjects were given low priority and will most likely be closed once current projects have been completed:
- corium cooling in a core-catcher with ex-vessel cooling;
- corium spreading in the reactor pit after reactor vessel failure;
- formation of cracks and leaks in the concrete containment structure;
- impact of aerosol behavior on the source term (in steam generator tubes and in containment cracks);
- impact of core reflooding on the source term.

Three subjects were considered as ready to be closed. Given the current state of knowledge and the low level of risk in comparison to other subjects involving greater risk and uncertainty, no other experimental programs are required in the following areas:
- reactor coolant system integrity and heat distribution;
- interaction between corium and ceramics in an ex-vessel core-catcher, cooling through bottom flooding;
- fuel/coolant interaction with a vapor explosion in a vulnerable reactor vessel.

This prioritization of subjects will serve to reapportion competencies and human resources to high-priority studies, for both the EC 7th Framework Program and other international (OECD) projects.

**Joint research activities**

These activities form the foundation of SARNET R&D. In connection with the research priorities described above, their goal is to tackle pending problems that have been given precedence. They have been divided into three areas: corium behavior, containment integrity, and source term.

The same method was adopted for all three fields: review and selection of available, pertinent experiments with synthesis of analyses and interpretation of data provided by these experiments; review and synthesis of physical models with proposals for new or improved models for ASTEC.

**Corium**

Corium behavior is a vast subject involved in more than half the problems selected in the PIRT of the EURSAFE program [Magallon et al., 2005]. The study of corium behavior extends from early-phase core degradation to late-phase corium formation in the reactor core and its relocation outside the vessel.

Significant efforts are currently being devoted to the development and improvement of databases on the physical properties of corium materials and corium thermodynamics.

Joint projects have been conducted by 22 SARNET partners [Journeau et al., 2008], involving contributions to the definition and interpretation of tests, benchmark exercises, and improvement of the associated models: CCI-OECD tests on corium/concrete interaction; the QUENCH-10 test on air ingress in a fuel bundle; the QUENCH-11 test on fuel bundle degradation and reflooding; QUENCH-12 test on a VVER bundle; COMET-L1 and L2 tests on corium/concrete interaction in 2D geometry; LIVE tests on corium in the vessel lower head; VULCANO test on corium/concrete interaction on real materials.

Similar activities were led for the International Science and Technology Center (ISTC): PARAMETER on top flooding, METCOR on the impact of thermomechanical interaction on vessel behavior, and CORPHAD on corium thermodynamics (and enhancement of the NUCLEA database). For the international Source Term project (see Source Term section below), FZK and IRSN harmonized their test matrices on Zircaloy oxidation by air/steam mixtures, and boron carbide (B4C) oxidation and degradation.
Some of the main achievements in corium research [Journeau et al., 2008] include the following:

- understanding of oxidation phenomena in steam and air atmospheres was improved, and oxidation correlations were validated. The importance of material composition was demonstrated. Research on new cladding materials, and on hydrogen generation and fission product release in particular, are necessary;
- data on B₄C boron carbide oxidation collected from experiments conducted by FZK and IRSN have led to a shared interpretation of the PHÉBUS-FPT3 integral test;
- launching of LIVE experiments (late-phase in-vessel degradation) started a new series on modeling molten pool behavior in the reactor vessel;
- the various reactor vessel creep failure models were compared and a shared understanding of the OECD test OLHF-1 was obtained. Modeling of crack propagation is in progress;
- analyses on cooling heterogeneous 2D debris beds showed increased cooling capability as compared to 1D particle beds. These results renewed interest in this problem, from both the experimental and numerical point of view, including research on debris bed formation;
- recent tests on corium/concrete interaction gave unexpected results with marked ablation anisotropy for silicon-rich materials. Interpretation and modeling of this behavior will be pursued;
- the core-catcher concepts based on spreading (EPR) and bottom flooding (COMET, downcomers) are under study and progress has been made in modeling;
- validation of the NUCLEA thermodynamic and chemical database was extended through experiment result analyses funded by the EC and an inter-laboratory exercise assessed the uncertainty of these analyses using energy dispersive X-ray spectrometry.

**Containment**

Research work on energy phenomena that could jeopardize containment integrity involves hydrogen behavior and fast interactions in the containment. With regards to hydrogen behavior, hydrogen combustion and attenuation of the associated risk focus on the formation of combustible gas mixtures, the local composition of gases, and potential combustion modes, including the reaction kinetics inside recombiners. The hydrogen distribution inside the containment is studied to assess the risk of high concentrations. Experimental programs on combustion with concentration gradients (ENACCEF at IRSN) and recombiner kinetics (REKO-3 at FZJ) have been conducted and/or are under way. ENACCEF experimental results were used for benchmarking with three different 3D Computational Fluid Dynamic (CFD) codes (FLUENT, TONUS-3D and REACFLOW). Results revealed certain weaknesses in the combustion models.

New tests are planned on the ENACCEF facility to study the interaction between hydrogen flames and spraying, and on the REKO-3 facility to investigate gas ignition on hot catalytic surfaces. The results will be used to validate computer codes.

The main achievement in containment atmosphere mixing is the containment spraying experiment simulation carried out on a small-scale facility (TOSQAN at IRSN) and on a large scale (MISTRA at CEA). This work covered atmosphere depressurization and destruction of atmosphere stratification. Numerical simulations of these experiments were conducted using CFD codes and lumped-parameter codes. Results demonstrated the feasibility of these simulations, which could also be applied to real containment structures within the next few years, once adequate computational capacity is available.

At present, however, the resolution achieved when applying CFD tools to the containment is insufficient, due to the wide range of turbulence scales involved and the computing power required. CFD simulations of Passive Autocatalytic Recombiner (PAR) operation with simplified 2D containment models represent an important first step towards complete simulation of PARRatmosphere interaction in actual power plants. Simulations of condensation experiments performed on the CONAN facility at the University of Pisa successfully assessed the different steam condensation models used in the CFD codes, steam condensation having a significant influence on atmosphere behavior. As regards research on fast interactions, Fuel/Coolant Interaction (FCI) was studied to further knowledge on parameters that affect steam explosion energetics during corium relocation in water and to determine the risk of vessel or containment failure. Investigations focused on specific processes such as pre-mixing, corium fragmentation, and particle thermal transfer. FCI research was closely tied to Phase 1 of the OECD-SERENA program, which set out to assess the ability of the current generation of FCI codes to predict reactor structural loads induced by a steam explosion, and to reach a consensus on the understanding of important FCI phenomena in different reactor situations [Buck et al., 2008].

The codes used for this purpose were mainly MC3D and IEMIX/IDEMO. A certain number of experiments were performed in the MISTEE and DEFOR facilities at KTH, and KROTOOS at CEA Cadarache, which was restarted after being moved from the JRC Ispra center. These experiments aimed to further understanding of the fragmentation and explosion of corium pools. The
The second problem concerning fast interactions is Direct Containment Heating (DCH) after vessel failure. This includes corium dispersion in various reactor compartments, thermal transfer processes, and chemical processes such as hydrogen generation and combustion. Since the consequences of DCH are essentially related to reactor pit geometry, a database was built for different power plant designs, including EPR, the French PWR-1300, VVER-1000 and the German Konvoi reactor, through an experimental program run on the DISCO facilities (FZK). For the EPR and VVER-1000 plants, the DCH issue can be considered as closed due to the reactor pit geometry. Efforts will continue to improve the prediction capability of the MC3D CFD code on one hand, and the COCOSYS and ASTEC codes on the other. Benchmark exercises revealed serious deficiencies in current models: correlations on debris dispersion must be more closely adapted to reactor pit geometry, and adequate models for hydrogen oxidation and combustion are required, since these phenomena seriously jeopardize containment integrity (experimental results have shown that direct thermal effects alone should not jeopardize containment integrity). Based on available experimental data, extrapolation to the reactor scale under an air-steam-hydrogen atmosphere needs to be established using combustion codes (COM3D, REACFLOW), then the modeling parameters must be transferred to codes with DCH models (CFD codes or COCOSYS), and in the final phase be adapted to ASTEC.

Source term
In the source term area of study, release, transport and deposition of fission products were investigated, including situations with air ingress to the reactor vessel, i.e., in an oxidizing environment. For transport and deposition, one study focused on iodine, analyzing both volatility in the reactor coolant system (for interaction with concomitant Ag-In-Cd release) and the proportion of iodine gas and liquid forms inside containment (also extended to ruthenium). Another study involved aerosol behavior in scenarios where risk is of utmost importance: containment bypass sequences (steam generator tube rupture, SGTR), containment through-wall cracks, and thermal and mechanical revolatilization. The main accomplishments are described in detail in [Haste et al., 2006].

An important effort was devoted to pursuing the experimental International Source Term Program (ISTP), initiated by IRSN, CEA, and EDF, with support from the EC [Clément and Zeyen, 2005]. Interpretation of available AECL and RUSON test results (the latter at AEKI) showed that Ru release occurs in the form of oxide, after an incubation period in which complete oxidation of the fuel and cladding occurs. The RUSON and VTT tests showed that the forms of oxide can remain volatile in the lower temperature range, and then be transported to the reactor containment in a stable gaseous form, a very important finding. These tests are still in progress. Test conditions for the future VERDION program (CEA) [Clément and Zeyen, 2005] were defined through simulations of air ingress scenarios. These new data will be used to validate ASTEC modeling. The proposed VERONIKA experiment, in the ISTC context, on the release of fission products from a highly irradiated VVER fuel was examined and SARNET proposals for a test matrix were adopted. A similar exercise was conducted on iodine chemistry through the EVAN test project, the results of which are currently being studied. FIPRED experiments (INR) provided data on self-disintegration of UO₂ pellets.

With regards to fission product transport, IRSN interpreted the iodine chemistry in the system based on data generated through the PHÉBUS-FP and VERCORS HT tests (the latter conducted at CEA). In reducing conditions and with no control rod materials, it appears relatively clear that iodine is transported mainly in the form of cesium iodide (and rubidium). In oxidizing conditions, the question is more complicated: iodine may take the form of CsI or, in the presence of control rod materials, other metal iodides or, if no control rod materials are present, conditions tend to favor HI formation. These points have not yet been confirmed, The CHIP experimental program (IRSN) [Clément and Zeyen, 2005], in which VTT provides experimental support, has produced kinetic and thermodynamic data on iodine transport through the reactor coolant system, especially for key systems such as (I-Cs-O-H).

This data will be used to improve ASTEC models and extrapolate experimental results to reactor conditions. In the QUENCH-13 fuel bundle test (FZK), carried out using a powerful experimen-
Several facilities have been used to study aerosol retention in the steam generator under SGTR conditions: PSAERO/HORIZON (VTT), PECA/GSSTR (CIEMAT) and ARTIST (PSI). On an overall basis, these tests showed that the “wet” scenarios (where rupture occurs below the secondary water level) seem to provide effective particle retention and that the same “dry” scenarios may capture a fraction of the particles that ingress to the secondary side, but in much smaller amounts. The VTT tests showed that resuspension is significant for aerosol retention in the horizontal tubes, and that it increases for sudden variations in flow rate. All available resuspension models are being assessed through intercomparison of data (STORM at EC/JRC Ispra and new VTT results). The VTT data will serve to develop a new empirical model. Revolatization tests in the REVAP small-scale facility (JRC/ITU) on PHÉBUS-FP samples show that Cs revaporation can be very high (approx. 95%) on flat metal substrates. CsOH deposits on stainless steel showed the same behavior as PHÉBUS-FP deposits. VTT is starting new tests on the speciation of revaporization aerosols, to complement the previously mentioned CHIP program tests.

Aerosol retention in containment cracks can be effective, especially in the presence of steam (SIMIBE tests at IRSN). Independent models or models incorporated in developed codes will be tested with respect to data taken from COLIMA experiments (CEA), which will be performed on the experimental PLINIUS platform. A SARNET team will provide assistance before testing.

Facilities designed to study containment chemistry include PHÉBUS-FP, CAIMAN, SISYPHE, CHALMERS, PARIS and EPICUR (Clément and Zeyen, 2005, as well as RTF (AECL), which recently produced data from the P9T3 test. The Iodine Data Book, compiled by Waste Management Technology, provides a critical review of chemical data and models. CAIMAN results showed that in the presence of paint, under irradiation and at high temperature, organic iodine may be the predominant form of gaseous iodine; in alkaline conditions, gaseous iodine concentrations decrease by several orders of magnitude. Mass transfer between the sump and the gas phase was studied on the SISYPHE facility; evaporation conditions increase the rate of transfer from the liquid phase to the gas phase and change the iodine equilibrium concentrations, as the sump iodine concentration is reduced. The well-known two-film model was extended to these conditions and entered in ASTEC. Validation of a more mechanistic model is currently in progress [Herranz et al., 2007].

The effect of radiation on the containment atmosphere and the effect of metal impurities in the sump were studied in the PARIS and CHALMERS experimental programs, respectively, while CHALMERS and VTT studied speciation of the oxides and iodine nitroxides formed in the atmosphere through radiolysis. Organic iodine formation models take into account the thermal and radiolytic mechanisms in gaseous and liquid phases. There are nonetheless deviations in the aqueous model, essentially with regards to the organic sources. The data provided by the EPICUR tests are now being interpreted using codes such as ASTEC/IODE, INSPECT, and LIIRIC.

The behavior of ruthenium inside containment was studied experimentally and theoretically by IRSN (EPICUR tests) and CHALMERS, along with the effects of heating fission products on the passive autocatalytic recombiners, successfully modeled by ASTEC/SOPHAEROS based on the RECI laboratory tests (IRSN). Scaling effects are currently being studied. Lastly, a benchmark exercise using several codes was performed in the context of the ThAI-Iod9 integral test on containment iodine chemistry, coordinated by GRS.

Integrating research activities

The previous sections have demonstrated that effective integration of research work has been achieved through:

- collaborative work on calculations to prepare and interpret experiments, for example PSI on the QUENCH tests at FZK;
- joint experimental achievements, such as the specific facility installed and operated by VTT experimenters for CHIP experiments at IRSN;
- collaborative definition and interpretation of experiments (several “interpretation circles” have been created and are truly active);
- comparisons between computer code results;
- dissemination of codes among the partners to reach a shared vision of experimental phenomena (primarily ASTEC modules);
- exchanges on the application of R&D results at the reactor scale;
Extension of the SARNET experience

The SARNET contract with the EC covered a period of four and a half years, from April 2004 to September 2008. In 2006, a special working group, made up of nine representatives of the Governing Board, was created to prepare the network’s continuation. A positive response was obtained in the context of the second request for proposals of the EC’s 7th Framework Program and contract negotiations are now in progress. The network is therefore expected to be co-funded by the EC for another four years.

In the course of this period, network activities will focus mainly on high-priority subjects (see section on integration activities) and experimental activities will be included directly in the partially financed common program. The aim is to evolve towards complete self-sufficiency. The basic principles defined to reach this goal are explained below.

The 41 partners from Europe, Canada, Korea, and the USA will network their research capacities in the SARNET2 project. Following through with SARNET’s initial objectives, the project was defined to rationalize the use of available resources and build a sustainable consortium, where severe accident studies form the basis for development of joint research programs and a shared computer tool (the integral ASTEC code).

In the second contract period, network organization will be similar to that of the first contract. To enhance decision-making efficiency, the Governing Board will be replaced by a Steering Committee of 10 members in charge of strategy, supported by an Advisory Committee made up of executives from end-user organizations (for example, electricity companies).

A general assembly, composed of a representative from each of the parties to the consortium and an EC delegate, will be convened once a year to report on and discuss network activity progress, the detailed implementation plan, and decisions taken by the Steering Committee. At the second level, as for SARNET, a Management Team (with its Coordinator and seven Project Leaders) will be tasked with daily management of the network.

The principle activities to promote integration and excellence will be pursued: collecting available experimental data in a scientific database in a format common to all, integration of

- interlaboratory exercises for EDX analyses of prototype corium samples by three laboratories (Cadarache in France, Karlsruhe in Germany, Rez in the Czech Republic);
- annual technical meetings in all three areas (corium behavior, containment integrity, and source term), in addition to a large number of specialist meetings.

Spreading excellence

The third major activity of SARNET involves spreading knowledge and excellence. The more experienced organizations began spreading excellence by preparing a training course on severe accident phenomenology, intended for doctoral students and junior researchers, with five-day sessions organized in June 2006. A five-day training course on accident progression (data, analyses, and uncertainty) for senior nuclear safety specialists was organized in March 2007. A third course covering both phenomenology and codes to simulate severe accident scenarios was organized in Budapest in April 2008, where 40 to 100 people attended each course.

A book on severe accident phenomenology is currently being written. It will cover the historical aspects of water reactor operating basics and safety principles, the phenomena involved in accident progression inside the reactor vessel, containment failure, and the release and transport of fission products. It will present a description of analysis tools and codes, management and limitation of the consequences of severe accidents, and environmental management. It will also provide information on third-generation reactors. The partners who have agreed to work together to prepare these courses and the book are universities, TSOs(1), national laboratories, and representatives of industry who share their vast experience and talent through SARNET.

A mobility program, allowing students and researchers to work in different SARNET laboratories to further their training, is another aspect of spreading excellence. There have been 33 transfers funded by SARNET, for periods lasting an average of three months.

Three European Review Meetings on Severe Accident Research (ERMSAR) were held in France, Germany, and Bulgaria, providing forums for the severe accident research community. They have become major world events on the subject.

Lastly, about 300 talks or papers have been presented by SARNET members in conferences or scientific journals. The complete list is given in the SARNET final summary report (open to the public) [Albiol et al., 2008].

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(1) Technical safety organizations.
knowledge acquired through network projects in ASTEC (with special attention given to applicability to all types of nuclear power plant found in Europe), dissemination of knowledge through the SARNET public website and the ACT tool, organization of conferences and seminars, organization of teaching and training sessions, encouraging student and researcher exchanges. Research priorities will also be reviewed periodically. Level 2 PSA activities will not be continued in the network, since a specific EC contract on harmonization of Level 2 PSA studies was established and launched in 2008 (www.asampsas2.eu).

Contrary to the first contract period, joint research activities under the second contract will include experimental programs. In keeping with EC recommendations, among the high-priority issues defined in SARNET (see section on integration activities), experimental work will be dedicated primarily to two subjects considered of utmost importance, which are expected to advance towards closure: coolability of corium and debris, and corium/concrete interaction. The other high-priority subjects will not be neglected. Shared analyses of experimental results and comparisons of models and codes to achieve a shared view of the corresponding phenomena will continue through different technical circles. Relations with end-users and other international programs and organizations such as the OECD/NEA, ISTC and other programs co-funded by the EC (mainly SNE-TP, PHÉBUS-FP, ISTP, ASAMPSA2) will be maintained and reinforced. Plans also envisage a gradual extension of program activities to fourth-generation reactors.

Finally, the network will move towards effective financial self-sufficiency by creating a legal entity for this purpose. Further details are given in [Albiol et al., 2008].

Conclusion

The SARNET network of excellence began its activities in April 2004, with the ambitious but extremely important goal of providing the appropriate framework for lasting integration of European severe accident research capabilities.

By pooling knowledge acquired in the ASTEC European reference code and the DATANET database, SARNET has effectively set up the necessary conditions for preserving the knowledge generated by thousands of people dedicated to research, and disseminating this knowledge to a large number of end-users. The ASTEC code and DATANET are now widely used resources.

By promoting collaborative work in Level 2 PSAs, SARNET has begun to create the necessary conditions for harmonizing approaches and making Europe a leader in risk assessment methodology for severe accidents.

Through a teaching and training program (organizing classes, writing a book for junior scientists), SARNET is developing synergy with educational institutions so that this field of activity remains attractive to young people. This action has been reinforced by a successful mobility program that provides young students and researchers with exchange opportunities at various laboratories throughout Europe.

By promoting collaborative R&D work on corium behavior, containment integrity, and source term issues, SARNET has advanced towards the resolution of residual questions and provided modeling recommendations for ASTEC. Proposals have been made for various ASTEC models, for which implementation has been completed or is in progress.

Pursuing SARNET will clearly make this network a reference in terms of severe accident research priorities, with an impact on national programs and the corresponding budgets. In time, all research activities in this field will be coordinated by the SARNET, contributing to a more rational use of European resources.

Acknowledgements

We would like to extend our thanks to the following project and sub-project leaders: M. Steinbrück (FZK), G. Repetto (IRSN), C. Duriez (IRSN), W. M. Ma (KTH), V. Koundy (IRSN), M. Bürger (IKE), B. Spindler (CEA), H. Wilkening (JRC/IE), I. Kijenak (JSI), D. Magallón (CEA), P. Giordano (IRSN), L. Herranz (CIEMAT). The Management Team has strongly relied on these leaders as technical relays to other network members, too numerous to mention here, but to which the authors are grateful. Finally, and above all, the authors thank the European Commission and its representative (M. Hugon, who has shown a true, profound interest in the network) for financing SARNET through the Sixth Framework Program, in the area "Nuclear Fission: Safety of Existing Nuclear Installations", contract number Fi6O-CT-2004-509065.

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References

- T. Albiol et al. (2008). SARNET Final Activity Report (Synthesis), SARNET Deliverable No 124 (to be issued also as a EC public report with a specific number).
Iodine is one of the most radiotoxic fission products that can be released into the environment during a pressurized water reactor (PWR) accident leading to core meltdown. The amount of radioactive iodine released subsequent to a containment leak or filtered containment venting depends on the chemical form of the iodine. Changes in the concentration of volatile iodine in containment depend on the balance between reactions involving the formation of iodine compounds, their fixation on surfaces, and their destruction.

Iodine chemical reactions in the liquid phase (solution present in the reactor vessel lower head) have been studied extensively in the last thirty years and understanding of the mechanisms involved has improved greatly, particularly through tests conducted on the PHÉBUS reactor at Cadarache. Nonetheless, the data available for high-temperature and high-dose-rate conditions are not sufficient to accurately predict the amount of volatile iodine fractions that may be formed. To complete the existing database, as part of the International Source Term Program (ISTP) (1), IRSN has set up a research program using the EPICUR irradiator to conduct tests where iodine volatization under irradiation is measured on line.

During testing, liquid-phase iodine is simulated by an iodide solution (I\textsubscript{2})\textsubscript{7}, traced using radioactive iodine (I\textsubscript{131}) and placed in a 5-liter vessel simulating the containment (50,000 m\textsuperscript{3}). The vessel is connected to the test loop, which includes selective filters to trap aerosols, non-organic iodine (I\textsubscript{2}) and organic iodine species (Figure 1). The volatile iodine species formed under the effect of irradiation (at dose rates of approximately 3.5 kGy/h) and high temperature (80-120°C) were transferred by gas flow from the vessel to the filters. Nal

Figure 1 | Diagram and photograph of the EPICUR test loop.

(1) The ISTP program is funded by IRSN, EDF, CEA, the European Commission, US-NRC, AECL, Suez- Tractebel and PSI.
counters performed on-line γ spectrometry to measure activity trapped on the various filters and represent the kinetics of iodine specie formation.

Five tests dedicated to iodide radiolytic oxidation were conducted on IRSN’s EPICUR facility (Experimental Program on Iodine Chemistry Under Radiation) at Cadarache between May 2005 and July 2007. Investigations covered the influence of oxygen presence, temperature, the initial iodide concentration, and pH on molecular iodine formation.

The results obtained confirmed that iodine volatility drops quickly when the solution pH changes from 7 to 5 (Figure 2). The solution temperature and the presence of oxygen have less effect on iodine volatility. The results obtained for initial iodide concentrations between $10^{-4}$ and $10^{-5}$ mol/liter indicate that this parameter has a relatively low impact on iodine volatility. These experiment results are being used to validate iodine behavior models in the French-German ASTEC code (Accident Source Term Evaluation Code), that serves in nuclear safety studies. The purpose is to identify the strong points and weak points of this code and, if necessary, develop new models to improve assessment of the amount of volatile iodine involved in a severe accident situation.

Testing currently under way on EPICUR sets out to study the formation of organic iodides from immersed painted surfaces (simulating painted containment surfaces) or from iodine-loaded paints placed in a gaseous phase (simulating surfaces in the containment atmosphere where iodine may be deposited).

![Figure 2](image-url)  
*Figure 2*  
Iodine fraction trapped on the molecular iodine selective filter, initially contained in the solution; effect of pH and temperature on iodine volatility for an initial iodide concentration of $10^{-5}$ mol/liter.
ASSESSING THE CONSEQUENCES OF MAJOR ACCIDENTS IN THE PRESENCE OF AIR: recent advances

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Fuel rod cladding on pressurized water reactors constitutes the first containment barrier of radioactive substances present in the reactor core. Fuel cladding may be exposed to air under certain accident conditions, for example, extended uncovery of irradiated fuel assemblies during handling or storage in pools; or in a core meltdown situation, where it is exposed to air either after a reactor vessel failure, or in an accident that occurs while the reactor vessel is open. These accidents take place at "low" temperatures (600-1,000°C) and "high" temperatures (900-2,200°C), respectively.

How does air increase risk?

The probability of a core meltdown accident occurring is extremely low, since it implies the loss of fuel cooling along with a partial or total failure of safety systems. Nonetheless, when an accident of this type does occur, exposure of zirconium alloy cladding to air is one of the key physicochemical phenomena involved in accident progression.

When zirconium oxidizes through contact with the air, the reaction is highly exothermic (released energy representing 85% more heat than steam oxidation), and may lead to temperature runaway, with destruction of the first containment barrier and acceleration of fuel assembly degradation.

Furthermore, the air oxidation reaction can lead to two other consequences:

- formation of highly inflammable zirconium nitride, if oxygen in the air is completely consumed.

The article below presents an accident transient and the associated phenomena. This description is followed by details on recent research work carried out in international programs to further knowledge of the corresponding phenomena. The last part is dedicated to the description of the modeling enhancements realised in the ASTEC computer code.

Flooding fuel assemblies: what are the risks and how can they be assessed?

After having been irradiated in the reactor, fuel assemblies are immersed in water, first in the spent fuel pool located near the reactor, then, once they have lost enough residual power, in a storage pool at the La Hague plant while waiting to be reprocessed.
Underwater storage consists of placing each individual irradiated assembly in a compartment (integral with the pool bottom) that keeps the assembly in an upright position (Figure 1), even in the event of an earthquake.

Water drainage from a spent fuel pool or storage pool can expose the uncovered part of the stored assemblies to air in the building, depending on the accident scenario and the emergency measures taken by the operator to restore the correct water level in the pool. If the fuel assemblies are no longer cooled, the temperature in the uncovered part of the fuel rods could rise due to the decay heat from irradiation that the fuel continues to release.

The first risk associated with fuel assembly heating under accident conditions is failure of the first barrier that separates fission products from the environment. This can occur due to fuel rod cladding failure, as the cladding loses its mechanical properties under the effect of internal pressure caused by the filler gas and fission gases. Two main heat exchange mechanisms have been identified that naturally limit fuel assembly heating during the accident. The first is natural air convection produced in the storage building by cooled air located in the upper part of the building and warm air located in the lower storage area. This convection mechanism can effectively cool the uncovered part of the storage compartments and the fuel assemblies they contain. The other exchange mechanism, which occurs at higher temperatures, consists of heat radiation between the fuel assembly cladding and the compartments, on one hand, and between the compartments and the building and pool structures, on the other.

The efficiency of cooling uncovered assemblies using natural air convection and heat radiation depends in particular on the storage configuration, the distribution of the assemblies and their residual heat, and the density of the storage compartment network.

In certain storage situations involving irradiated fuel assemblies, studies conducted by IRSN showed that the risk of a highly exothermic reaction from Zircaloy cladding oxidation under air atmosphere is real, leading to fuel assembly temperature runaway, within time periods ranging from a few hours to several tens of hours. The main risk associated with this thermal runaway phenomenon involves first of all additional release of fission products or forms of uranium oxide volatilized through contact with the air. The other significant risk entailed by exothermic air-oxidation of fuel cladding is the embrittlement or even melting of certain metal compounds (Zircaloy, steel), jeopardizing mechanical strength, cooling, and post-accident handling of the fuel assemblies.

Studies on pool drainage accidents currently conducted at IRSN focus on the risks associated with air-oxidation of Zircaloy. These studies are conducted at three different levels: fuel rod cladding, fuel assembly and storage, and finally, the spent fuel building and storage pool.

The purpose of the studies is, first of all, to gain a better understanding of the physicochemical mechanisms that microscopically drive air-oxidation of Zircaloy, in which nitrogen plays a particularly important role. This knowledge is based on interpretation of MOZART(1) analytical test results, and is essential in building oxidation models that take into consideration the various kinetic conditions of the oxidation reaction, which depends mostly on temperature and the thickness of the existing oxide scale. By using experimentally validated oxidation models, it is possible to calculate the temperature thresholds at which temperature runaway may occur on uncovered assemblies more accurately, to locate the zones where runaway occurs, and to predict runaway propagation conditions in the assemblies.

The study of air-oxidation conditions on uncovered fuel rods also requires assessment of the thermal-mechanical behavior of fuel assemblies in the accident situation. The first calculations performed using the ICARE-CATHARE code(2) [Fichot et al., 2006], for a fuel assembly stored in its compartment or handled outside of the compartment, showed that fuel rod deformation induced by creep in the first temperature escalation phase could have a significant effect on the fuel assembly's mechanical integrity. Further studies have been conducted to refine these models and to better understand the complex interplay between thermal runaway and mechanical failure in irradiated fuel assemblies.

References:

1. MOZART: program to measure air-oxidation of zirconium as a function of temperature.
2. ICARE/CATHARE: coupling between the mechanistic code for core meltdown ICARE 2, developed by IRSN, and the thermohydraulic code CATHARE 2, developed by CEA, IRSN, EDF and Areva-NP.
Which research programs will improve knowledge and computer codes?

Scientific approach

A status report on the study of fuel rod cladding oxidation under air atmosphere, conducted by IRSN in 2003 [Le Dantec, 2004] highlighted, first, the considerable disparity between existing experimental results, and second, the non-representativity of the cladding used in the tests, since it had never been exposed to reactor conditions, and therefore bore no signs of corrosion (neither oxidation, nor absorption of hydrogen from water).

Nonetheless, within the limits of existing tests, parabolic laws representing zirconium air-oxidation kinetics were established (Figure 2), based on recommendations from the OPSA report [Shepherd et al., 2000] and used in computer codes to calculate accident transients. During qualification of these codes through integral tests such as CODEX-AIT1 and QUENCH-10, tests simulating an accident transient with air-oxidation on a bundle containing about twenty fuel rods clearly revealed the inadequacy of these models [Duriez et al., 2008]. More specifically, it appeared that the models based on parabolic oxidation reaction kinetics did not take into consideration all the significant physical phenomena and were not conservative.

In the context of the International Source Term Project (ISTP) coordinated by IRSN, in cooperation with various national and international partners, the analytical test program MOZART was proposed, dedicated to studying oxidation of fuel cladding under air atmosphere [Le Dantec, 2006].

In parallel, research work to improve understanding and modeling of fuel cladding air-oxidation phenomena were conducted through the SARNET network of excellence [Micaelli et al., 2006], as part of the EU 6th Framework Program. In this context, network partners FZK, INR and IRSN conducted experimental programs [Duriez et al., 2008], while ENEA, GRS, and other partners contributed to enhancement of existing models [Coindreau et al., 2008]. The purpose of this work was to enrich the experimental database required to develop and qualify the models used in the ASTEC integral code [Van Dorsselaere et al., 2005], the European reference computer code used to study core meltdown accidents in water reactors.

In a final note, IRSN has recently agreed to participate in an important experimental program directed by the US-NRC, carried out at the Sandia National Laboratory. Tests have been scheduled from 2009 to 2011 to study and measure the behavior of models representative of pressurized water reactor fuel assemblies in their storage environment when exposed to air at temperatures above 600°C. The results of these US tests will serve as the basis for validation of computer codes used at IRSN to study uncovering in spent fuel pool accidents.

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**Figure 2** Correlations for constants in the parabolic oxidation laws recommended in the OPSA report.

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(3) ASTEC: Accident Source Term Evaluation Code.
(4) CFX: Computational Fluid Dynamics.
(5) OPSA: Oxidation Phenomena in Severe Accident.
(6) SARNET: Severe Accident Research Network.
Progress in understanding physical phenomena

Within the MOZART experimental program, initiated in 2004 and pursued until the end of 2007, IRSN carried out small-scale oxidation tests on zirconium alloy cladding. The test specimens, fuel cladding sections from 1 to 2 cm long, were oxidized under air atmosphere in isothermal conditions on a thermobalance. This tool is used for thermogravimetric analysis (TGA), i.e. it monitors mass gain on the specimen in real time as it reacts to high temperature in the ambient atmosphere. The oxidation rate was obtained by taking the derivative on the mass gain signal time. Temperature in the MOZART tests ranged from 600 to 1,200°C, covering the range in which temperature runaway and reaction runaway may be expected in an accident situation. After each test, metallographic cross-sectional cuts were made on the specimens to interpret results.

The various zirconium alloys used in French PWRs were studied: Zircaloy-4 (tin-based alloy), M5 (a recent niobium-based alloy developed by Areva), and Zirlo (tin- and niobium-based, marketed by the American company Westinghouse). In the first phase, the cladding specimens were used in the new state, as received from the manufacturer. In the second phase the specimens were pre-oxidized at low temperature under steam before air-oxidation, to simulate an initial corroded state representative of used assemblies. In the final phase, a few tests were conducted to assess any influence of hydrogen dissolved in the metal, which is known to be present in the cladding at the end of its service life.

This small-scale test program made it possible to compile a detailed database on air-oxidation kinetics, while converging towards a better understanding of the phenomenology behind zirconium alloy degradation under air atmosphere, which pointed to the fundamental role played by nitrogen [Duriez et al., 2008; Duriez et al., 2009]. A typical result from thermogravimetric analysis on the new cladding specimen is shown in Figure 3. After a temperature excursion under argon, mass gain started when air sweeping began in the oven. This test ran until the metal was completely oxidized, indicated by a steady mass gain signal. Two distinct kinetic regimes were identified on the derived signal.

The first, referred to as the pre-transition regime, was characterized by an intense peak due to oxidation of the bare metal, followed by a decrease in the oxidation rate caused by a protective layer of oxide scale, which gradually grew thicker. In this case the solid-phase diffusion of oxygen through the zirconia is the layer up to the metal/oxide interface limiting step of the reaction. In an initial approach, the regime could be described by a parabolic law such as \( \Delta m/S = k t^{1/2} \), where \( k \) is a constant called the parabolic reaction constant and \( \Delta m/S \) is the mass gain over the metal unit of area. The parabolic reaction constants were determined for each of the three alloys investigated. They are presented in Figure 4, where they are compared to different recommendations available in the technical literature to describe oxidation in the parabolic regime of zirconium alloys at high temperature, under steam and air atmospheres. In the lower part of the tested temperature range, the experimental values obtained for Zircaloy-4 were noticeably lower than those of the other two alloys, corresponding to slower diffusion of oxygen in the zirconia layer. The change in gradient observed at 1,000°C corresponded to the monoclinic/quadratic phase boundary in the zirconia. On an overall basis, the data obtained were closer to the “steam” kinetic correlations than the very conservative recommendations given in the OPSA report [Shepherd et al., 2000], commonly used in major accident computer codes to describe oxidation under air atmosphere.

At a thickness that depends on temperature and type of alloy, the protective zirconia layer cracked due to the effect of high compressive stress at its center, which gave way to an acceleration of oxidation. Cracking was associated with a zirconia transformation from the quadratic phase to the monoclinic phase when stress was relieved, inducing microcracking of the oxide scale, contributing to attenuation of its protective role. This complex phenomenon of cracking and phase transformation is referred to as “breakaway”. The thermogravimetric technique determined the precise moment when kinetics changed, along with the corresponding thickness, calculated based on the minimum mass gain in the oxidation rate curve. The thickness measured at the kinetic changeover point for the three investigated alloys ranged from a few micrometers at low

![Figure 3](image-url) Example of an isothermal oxidation test on the thermobalance: oxidation at 800°C of a Zircaloy-4 specimen, pursued until complete oxidation.
Accidents in nuclear facilities

2.8

Once breakaway occurred, the post-transition regime was practically linear at low temperatures (up to 700°C). After a rapid rise, the oxidation rate stabilized at a constant value. But starting from 800°C, the oxidation rate did not stop rising after the transition. The metallography images show the formation of zirconium nitride (ZrN) particles near the metal/oxide interface, dispersed in a porous structural oxide (Figure 6). ZrN was initially formed at the bottom of the cracks in the dense oxide layer, where the oxygen was consumed and nitrogen had accumulated. This revealed local oxygen starvation at the microscopic scale. The oxidation front continued to progress towards the inside of the material and the ZrN particles were converted into oxide. The high increase in volume associated with this conversion generated oxide cracking, making the oxide scale porous and eliminating its protective role. Once it was initiated, this growth of porous oxide under the effect of nitriding was self-fed, since the nitrogen released through nitride oxidation remained trapped in the material and was available to resume formation of nitride particles from the metal. The significant metal creep generated by this nitriding/oxidation mechanism, associated with its self-feeding quality, explained the continuous growth of the cladding degradation rate observed after breakaway.

The presence of an initial layer of corrosion favored the transition to this nitro-oxidation regime, which resulted in a decrease in the duration (or even the absence) of the parabolic regime for specimens pre-oxidized at low temperature. Nonetheless, while the layer of corrosion was thick (approx. 60 to 80 µm), it played a protective role with regards to the oxygen supply, making degradation of the specimen in the final phase slower than for a new specimen.
Moreover, in an accident situation, the nitrogen-rich atmospheres that may be found locally would also encourage this transition to a nitrided regime.

The zirconium air-oxidation reaction therefore appears as a complex phenomena, where nitrogen plays a predominant role by considerably accelerating degradation. Given the highly exothermic nature of the zirconium reactions with oxygen and nitrogen, it appears easy to understand why fuel assembly exposure to the air can lead to much greater runaway than exposure to steam. It is therefore essential to take these phenomena into consideration in simulation codes designed for severe accidents (such as fuel assembly uncover in spent fuel storage pools, reactor core meltdown accidents).

**Modeling enhancements**

Initially, the ICARE code, a module of the ASTEC integral code simulating the behavior of fuel assemblies during an accident transient, partially took into account air-oxidation phenomena by using the parabolic oxidation reaction kinetics taken from OPSA recommendations [Shepherd et al., 2000], to assess growth of the zirconia (zirconium oxide) layer. Results of qualification runs of this model in the QUENCH-10 test [Mladin et al., 2006] showed that the ICARE code could reliably predict the temperature trend for the pre-oxidation phase (under a mixture of steam and argon), but did not correctly reproduce the air-oxidation phase, underestimating temperature runaway, in particular.

It therefore appeared necessary to improve this model of oxidation under air atmosphere. A study on experimental results from the MOZART program and tests carried out in the SARNET program led to a new model that simulates the phenomena occurring during the fuel rod cladding air-oxidation reaction, to differentiate oxidation kinetics before and after breakaway, and also to evaluate the instant when this transition occurs [Coindreau et al., 2008].

The new model thus assesses an oxidation regime before breakaway, through parabolic oxidation kinetics that describe total mass gain as a function of square root of time. The model can use different correlations to calculate these oxidation kinetics, taken from NUREG1-2 (7) [Powers et al., 1994], NUREGB and AEKI [Shepherd et al., 2000], as well as a correlation based on MOZART results.

![Figure 7](image-url)

*Figure 7* Comparison between MOZART experimental results (brown and pink curves) and the mass gain curves calculated using ICARE/CATHARE: standard oxidation model (parabolic law) in blue, new model taking into account the kinetic transition (breakaway) shown as a solid black line.

(7) NUREG: Nuclear Reactor Regulation.
In an evaluation on the moment of breakaway, MOZART test results showed that a transition occurred between the parabolic oxidation phase and the accelerated oxidation phase for a critical mass gain value that depends to a great extent on temperature. It also appeared that mass gain at the instant of transition could be correlated to temperature by a hyperbolic law, based on the assumption that the kinetic transition is related to a phase change in the zirconia layer from a quadratic to a monoclinic phase. After breakaway, the new model described accelerated oxidation kinetics for total mass gain using an empirical law based on the new experimental results.

\[ \frac{d (\Delta m / S)}{dt} = K_a \text{ where } K_a \left[ kg^{-0.5} m^{-1} s^{-1} \right] = 45.9 \exp (-1.325 x 10^9 / RT) \]

To appreciate the contribution of this new model, the isothermal MOZART tests were calculated using the old model and the new model. A comparison between experimental results and calculated results (Figure 7) showed that the time required for complete cladding oxidation estimated using the old model is largely overestimated compared to the new model, which predicts values very close to the experiment observations, and is therefore more conservative in terms of the oxidation rate and the temperature excursion rate.

## Conclusions and outlook

Research programs on fuel assembly cladding oxidation under air atmosphere conducted in the last few years have contributed knowledge on the physical phenomena involved. More specifically, new experimental data obtained in the ISTP and SARNET international programs have revealed the following findings:

- the commonly used parabolic oxidation kinetics taken from OPSA recommendations only describe a limited portion of the cladding degradation transient;
- the transition from a parabolic oxidation regime to an accelerated oxidation regime must be taken into consideration.

To acknowledge all of these observations, the models used in accident transient simulation codes have been enhanced and the first applications show a better predictive capability when compared with experimental results. These improvements constitute an important advance in simulation computer codes, which should now be capable of producing more realistic assessments of the oxidation kinetics and temperature runaway phenomena induced by accident situations where fuel rod cladding is exposed to air.

Further progress still lies ahead. Regarding the transition from one oxidation regime to another, it appears that additional tests are necessary to obtain a more accurate characterization of this transition. Current models process the different phenomena that influence the oxidation rate in the post-transition phase using accelerated kinetics. Analysis of experimental results has shown, however, that the zirconium alloy fuel rod cladding oxidation kinetics seems to be strongly related to the mechanical behavior of the zirconia layer. It is therefore necessary to develop a detailed model simulating the various important phenomena that impact the state of stress in this zirconia layer, such as, for example, an increase in layer volume, the presence of nitride at the metal/oxide interface, and oxidation of the ZrN formation. Furthermore, current models must be extended to cover new cladding materials such as Zirlo and M5.
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KEY EVENTS and dates

Dissertations Defended

February 14, 2008  
Elie CHALOPIN submitted a thesis entitled "Characterization of the radiative properties of a porous medium using the RDFI method – Application to a degraded reactor core" in Paris.

September 24, 2008  
Florence GUPTA submitted a thesis on the "Study of cesium behavior as a fission product in uranium dioxide using the ab initio method" in Cadarache in southern France.

October 29, 2008  
Sébastien DESTERCKE submitted a thesis in the field of information processing in the presence of uncertainty, entitled "Methods of synthesis of imprecise probabilistic information" at the Université Paul Sabatier in Toulouse.

Other Major Events

February 2008  
COPERNIC Platform  
February 5, 2008: Label awarded by the PACA region’s Territorial Risk and Vulnerability Management Cluster for the Copernic joint platform on fire knowledge, led by IRSN. This platform allows companies to call on IRSN’s competence and test facilities in areas involving fire risk.

April 2008  
Patent application submitted for a device to measure surface displacement and its application to measuring the regression of the surface of a combustible material that is optically transparent in a fire.

May 2008  
CHIP facility commissioned  
May 14, 2008 saw the commissioning of CHIP, the new facility designed to quantify and characterize radioactive iodine release in the event of core meltdown in a nuclear reactor. The experimental CHIP program (to study iodine chemistry in the reactor coolant system), with its new test facility located at Cadarache in southern France, aims to increase assessment capabilities at the Institute in the field of emergency prevention and emergency response management applied to the environmental release of radioactive iodine.