Chapter 6
Research on Spent Fuel Pool Uncovery Accidents

Nuclear power reactors have a spent fuel pool where spent fuel assemblies are stored until they are ready to be transported to the reprocessing plant, in other words, when they have lost enough of their residual heat. During reactor outages, the spent fuel pool can also be used to temporarily accommodate fresh or already irradiated fuel assemblies for loading into the reactor core before it is restarted. The spent fuel pool is about 12 m deep. The fuel assemblies are stored in racks placed on the bottom of the pool, under about seven meters of water, thus providing workers with sufficient protection against ionizing radiation under normal operating conditions. The pool also serves as a major source of water for cooling fuel assemblies in the event of a cooling system failure.

A spent fuel pool can accommodate a large number of spent fuel assemblies (from 300 to 600 depending on the type of reactor) containing radioactive substances. It is located in a fuel building (BK) with a dynamic confinement system ensured by the ventilation system.

The reactor cavity in the reactor building (RB) and the spent fuel pool in the fuel building (BK) are shown in Figure 6.1.

It was considered in the safety demonstration carried out at the time of design that, in the event of pool cooling system failure, and given the low residual heat of the spent fuel assemblies, there would be enough time to implement safety measures before the fuel was uncovered. That is why, in the event of loss of fuel rod integrity, the spent fuel pool building does not have the same radioactive confinement capability as the reactor containment building.

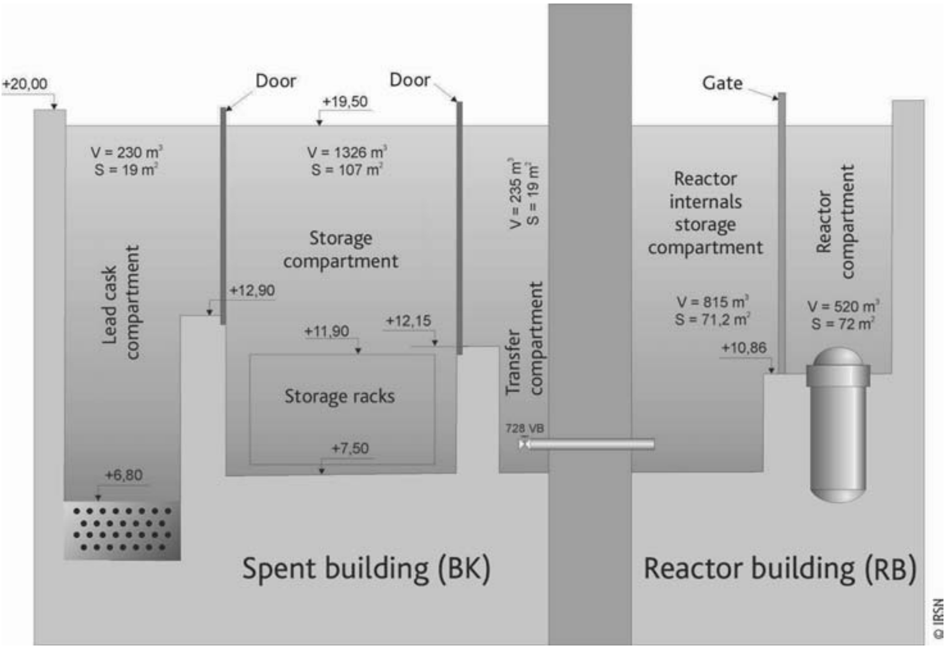

Figure 6.1 Cutaway view of the reactor cavity in the reactor building (RB) and the spent fuel pool in the fuel building (BK) in a 900 MWe CPY[48]-series plant.

Various design features, such as vacuum relief valves on the fuel pool cooling and purification systems (FPCPS), were implemented to preclude the risk of the pools emptying. These features were significantly reinforced following several incidents (loss of tightness of doors and gates, drainage induced by lineup errors, unsatisfactory performance in vacuum relief valves, foreign matter blocking these valves) or in connection with safety reviews.

The Fukushima Daiichi accident confirmed the importance of examining all the possible strategies for guaranteeing fuel assembly cooling, especially now that fuel and fuel pool operating conditions have changed considerably since design (fuels more highly irradiated, existence of MOX fuels).

If it becomes impossible to ensure sufficient fuel assembly cooling, the dreaded cliff edge effect is a runaway exothermic oxidation reaction in fuel cladding due to air and steam, leading to cladding deterioration and significant radioactive release. In this event, very little radioactive iodine-131 should be released, given the time the fuel is stored after removal from the reactor core. On the other hand, a very large quantity of ruthenium should be released. This is a particularly radiotoxic element, even if its radioactive half-life is considerably shorter than that of cesium (see Section 9.4).

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[48] Second serie of French power reactors of 900 MWe (EDF).

Boiling water in the storage racks could also cause the fuel assemblies in the rack to return to criticality. This could lead to rod failure, human radiation exposure and radioactive release.

Assessing the risks associated with loss-of-coolant or loss-of-cooling accidents entails modeling convection phenomena that are mixed (forced and natural); three-dimensional (owing to the shape of the storage racks and the non-uniform distribution of residual heat in the spent fuel assemblies in the pool); and involving multiple fluids (water, steam and air during the uncovery phase). Oxidation phenomena of zirconium alloys in contact with mixtures of steam, oxygen and nitrogen must also be considered [1]. These oxidation reactions generate heat and, possibly, hydrogen, with a related risk of explosion. Research work also focuses on the effectiveness of engineered safety systems according to the scenarios considered (for example, risk of pump cavitation if steam or air is entrained in the systems, effectiveness of spray systems for cooling uncovered fuel assemblies).

Although tests carried out in the 1950s already showed that the kinetics of zirconium oxidation by air at high temperatures differed considerably from that of oxidation by steam, air-induced oxidation of zirconium alloys only recently became the focus of detailed studies.

The zirconium oxidation reaction due to oxygen releases about twice as much heat (1101 kJ/mole compared with 528 kJ/mole when steam is the oxidizing agent). Zirconium also reacts with nitrogen. The resulting nitride can then react with oxygen and also release considerable heat (736 kJ/mole). If the conditions are suitable for initiating these chemical reactions (air in the storage racks and temperature around 600 °C), the heat generated could lead to runaway temperatures and damage to the fuel rods.

In France, IRSN carried out air-induced oxidation tests on Zircaloy-4 and M5^{TM} cladding samples, a few centimeters in length, as part of the International Source Term Program (ISTP, 2005–2013, MOZART[49] test series). The studies were carried out in a temperature range between 600 °C and 1100 °C. A thermogravity system was used to continuously monitor the increase in sample weight, and thereby define oxidation kinetics laws. These were then incorporated in the ASTEC[50] (see chapter 9 on severe accidents) simulation code used to calculate the different stages of the accident.

Other analytical tests on different alloys were performed at more or less the same time in the United States and Germany. These differed in terms of the type of alloys studied, sample geometry (cladding sections with open or closed ends), the initial state of the materials (fresh or pre-oxidized and pre-hydrided to simulate the effects of in-service oxidation), and the type of oxidizing fluid studied (pure nitrogen, mixture of nitrogen and steam, or mixture of air and steam).

These tests, and those performed by the Karlsruhe Institute of Technology (KIT) in Germany, confirmed that cladding oxidation in air could lead to more serious consequences than oxidation by steam. This is because the kinetics of the oxidation reaction

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[49] Measurement of Zirconium Oxidation by Air at Temperature.

[50] Accident Source Term Evaluation Code.

accelerates as from a certain oxidation rate. First, a dense layer of zirconium oxide is formed that limits oxygen diffusion to the metal. Its thickness increases with the square root of time (see Section 3.2). The kinetics then accelerates further when this dense oxide layer cracks. The air then reaches new areas of unoxidized metal, and the oxidation reaction proceeds in a linear manner with time. This phenomenon – known as breakaway – is not very well understood at the present time. One explanation might be the change in the zirconia crystal system (from monoclinic to tetragonal) at around 1000 °C (see Section 2.2). However, another hypothesis was put forward to explain the specific role of nitrogen and the formation of dense nitrided compounds (Zr-O-N or ZrN), the oxidation of which could cause the zirconium oxide layer to crack at temperatures below 1000 °C.

Integral tests were also carried out by Sandia National Laboratories in the United States. These were part of a program called "Spent Fuel Pool Heatup and Propagation Project" (2004–2006), set up to study BWR fuel assembly damage[51]. This was followed by the OECD/NEA "Sandia Fuel Project" (2009–2013), devoted to the study of PWR fuel assembly damage, bringing together 13 countries, including France. The purpose was to obtain thermal-hydraulic data on natural convection cooling of fuel rods by ambient air in the event of loss of cooling, in order to validate accident computer codes (such as ASTEC and a dedicated version of DRACCAR).

Experiments were carried out on a full-length, finely instrumented assembly comprising 17 x 17 Zircaloy-clad, magnesium oxide[52] heater rods. First, nondestructive tests were carried out for natural convection in air. These were followed by a destructive test that was carried out until the onset of runaway oxidation (also called zirconium fire) and its axial propagation within the assembly. In a second test campaign, the heated assembly was surrounded by four unheated assemblies to study the radial propagation of runaway oxidation to surrounding, lower power assemblies. Some of the rods in these unheated assemblies were also pressurized to simulate ballooning effects (possibility of oxidation accelerating after spalling of the oxide layers formed). It was seen that runaway oxidation did propagate to the surrounding assemblies. Following this research, core melt accident simulation codes, such as ASTEC, benefited from improved models for describing this phenomenon more accurately.

► Planned programs

In 2013, IRSN launched a research program, called DENOPI[53], to learn more about these phenomena. The following CNRS research laboratories also participate in the program: PROMES – Processes, Materials and Solar Energy, laboratory in Perpignan –, LEPMI – Laboratory of Electrochemistry and Physico-chemistry of Materials and Interfaces in Grenoble –, ARMINES-SPIN – Carnot M.I.N.E.S Institute – Center for Industrial and Natural Processes that is part of the École des Mines in Saint-Etienne; and LVEEM – Vellave Laboratory on the Development and Study of Materials in Le Puy-en-Velay.

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[51] Unlike PWR fuel rod assemblies, BWR assemblies are arranged inside Zircaloy channels.

[52] Magnesium oxide is not radiotoxic and its thermal properties (conductivity, specific heat, etc.) are similar to those of uranium oxide.

[53] Spent Fuel Pool Water Uncovering.

Scheduled to last six years, the program is jointly funded by the French National Research Agency (ANR), as part of the "Investment in the Future" program and, more specifically, the call for research projects in the field of nuclear safety and radiation protection (RSNR), issued in the wake of the Fukushima Daiichi accident in 2012.

Work scheduled as part of DENOPI includes experiments, modeling and simulation code validation, with the aim of learning more about the different phases of accidents of loss of heat removal or uncovery of fuel assemblies stored in the spent fuel pool. The program adopts an analytical approach seeking to expand knowledge in the following three areas:

- natural convection cooling at pool scale,
- thermal-hydraulic behavior at fuel assembly scale in the event of fuel uncovery, and effectiveness of spraying,
- acceleration mechanisms observed in fuel cladding oxidation in contact with an air and steam mixture.

Natural convection in the spent fuel pool will be studied (2015–2017) using a 1/5 model of a spent fuel pool, storage racks and the fuel assemblies they may contain. Results will also be interpreted using computational fluid dynamics tools designed to solve equations governing fluid movement.

In 2018–2019, the study of thermal-hydraulic behavior in a fuel assembly will use a full-length model of a fuel assembly and its storage rack, under conditions that represent the various phases of the accident, namely loss of cooling, pool water boiling, fuel uncovery, and resumption of cooling. Fluid velocities in the gaps between rods and oxygen concentrations will be measured to obtain very precise experimental data that will be fed into a database to validate the models used in accident calculations.

Cladding oxidation and the related runaway phenomena will be studied, together with the presumed role of nitrides, using high-performance laboratory technology including:

- *in situ* X-ray diffraction (XRD) in a steam-rich gas flow will be used for real-time monitoring, at reaction temperature, of changes in compounds formed at high temperatures (500 °C to 1200 °C);
- Raman micro-imaging will be used to study the microstructure and composition of the zirconium oxide layer formed.

Integral tests are also planned using simulated fuel assemblies in KIT's QUENCH facility as part of the Severe Accident Facilities for European Safety Targets (SAFEST) program, which is partly funded by the European Union.

Lastly, IRSN coordinates the AIR-SFP project (2015–2017), which is jointly funded by the European Union, following a call for research projects as part of NUGENIA+[54]. The

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[54] NUGENIA+ manages research for the development and safety of Generation II and III nuclear power reactors on behalf of NUGENIA (see Chapter 1). This association has more than a hundred members, including industrial companies, nuclear licensees, technical safety organizations, and research laboratories, for the most part in Europe.

purpose of this project is twofold: to assess the ability of different codes used in simulating reactor core melt accidents, such as ASTEC, to predict the different phases of an accident caused by fuel uncovery in a spent fuel pool; and to prepare a research program to fill the most significant gaps in knowledge. Fourteen countries are involved in this project, the results of which will lead to a better understanding of these accidents, and help to prevent them and mitigate their consequences.

References

[1] Status Report on Spent Fuel Pools under Loss-of-Coolant Accident Conditions. Nuclear Safety NEA/CSNI/R(2015)2, May 2015.