PAPER • OPEN ACCESS

Analysis of Fukushima Daiichi unit 4 spent fuel pool using MELCOR

To cite this article: M. D'Onorio *et al* 2022 *J. Phys.: Conf. Ser.* **2177** 012020

Analysis of Fukushima Daiichi unit 4 spent fuel pool using MELCOR

M. D'Onorio, A. Maggiacomo, F. Giannetti, G. Caruso

Sapienza University of Rome, DIAEE, Corso Vittorio Emanuele II, 244, 00186 Rome, Italy

matteo.donorio@uniroma1.it

Abstract. A spent fuel pool is a temporary storage facility designed to store the nuclear fuel assemblies removed from the reactor core. Here, the depleted fuel is vertically arranged in racks structures, accurately spaced to ensure subcriticality safety margins. Until the events occurred at the Fukushima Daiichi power plants, severe accidents in spent fuel pools had never been considered as a major safety concern, mainly because of their slow progression. However, extremely unlikely accident events, aggravated by the loss of site power, highlighted the vulnerability of nuclear fuel stored within spent fuel pools. During these accident sequences, reactor fuel rods can generate enough decay heat to vaporize the coolant around them, causing, during unmitigated scenarios, the uncovering and melting of fuel bundles after several days. In past years, new features have been introduced in the MELCOR code for the evaluation of potential risks associated to accident progression in spent fuel pools. In this paper, the accidental sequence of a loss of cooling accident in a boiling water reactor spent fuel pool is analysed with version 2.2 of the MELCOR code. The spent fuel pool of the Fukushima Daiichi unit 4 has been selected as reference. Main accident events, such as fuel uncovering, claddings oxidation, hydrogen generation, and fission products release, have been analysed assuming the unavailability of safety systems.

1. Introduction

In nuclear power plants, during fuel replacement operations, the Fuel Assemblies (FAs) containing exhausted fuel are moved from the reactor core into a suitable storage facility known as Spent Fuel Pool (SFP). Such storage pools are also used to segregate fresh fuel assemblies before being moved into the core. Both irradiated and fresh fuel bundles are sorted in submerged steel racks to provide radiological protection from ionising radiations.

Spent fuel pools are equipped with auxiliary systems for the immediate cooling of the fuel rods. During normal operation, such cooling systems circulate subcooled water among the SFP racks ensuring the decay heat removal from depleted assemblies. However, if the main function of these systems is lost for a long time, fuel rods can generate enough decay heat to cause SFP's water evaporation and subsequent fuel assemblies uncovering and melting [1].

Prior to the event at the Fukushima Daiichi nuclear power plant, severe accidents in the spent fuel pool have never been considered a safety concern since their progression is slow enough to let operators begin mitigative actions. However, accident scenarios characterized by long-term loss of site power, such as the Fukushima one, highlighted a possible vulnerability of the nuclear fuel stored within SFPs.

After this accident which occurred in March 2011, international benchmark exercises were carried out to simulate the accident sequence and its consequences. Among them, one of the most important was in the framework of the NUGENIA+ project, carried out in 2015, in which participants investigated the Fukushima Daiichi occurrence of events by using different severe accident codes [2].

For the scope of this simulation, it has been decided to use the last available version of the MELCOR code, also focusing on phenomena related to radionuclide releases from fuel assemblies, which were not considered in the previous benchmark studies.

MELCOR is a computer code developed by SANDIA National Laboratories (SNL) for the US Nuclear Regulatory Commission (NRC) to simulate transients and the progression of severe accidents in nuclear power plants. MELCOR code comprises several major packages that let the user model the main systems of a reactor plant and the main phenomena occurring during a severe accident (e.g., zircalloy and steel oxidation, fuel degradation, core melting, and radionuclides release, transport, and deposition). These major packages include, for example, the CVH and the FL packages, which simulate the thermal hydraulics of the system; the COR package, which simulates the behaviour of structures and fuel inside the core; and the RN package, which simulates release, mobilization, and relocation of radionuclides.

Version 2.2.18019 of MELCOR has been used for this calculation [3][4]. The analysis of the progression and radionuclide release of the accident scenario is based on the layout of Fukushima Daiichi unit 4 SFP. The aim of this paper is to simulate a base case accident scenario to be used for futures uncertainty quantification analyses that will be carried out by Sapienza in the framework of the European H2020 MUSA Project [5].

2. Spent Fuel Pool modelling using MELCOR

According to TEPCO data [5], Fukushima Daiichi unit 4 Spent Fuel Pool is a rectangular pool of dimensions 12.2 × 9.9 × 11.8 m, located in the upper part of a Mark I reactor building with a free volume of 25800 m^3 [6]. During normal operation, the water level inside the SFP is 11.5 m. When the Fukushima accident occurred, inside the SFP of unit 4 reactor, there were 1535 fuel assemblies divided into hot (548 FAs), cold (783 FAs), and fresh fuel (204 FAs) [2]. As shown in Figure 1, the fuel assemblies were stored in 53 different racks, with the average assembly decay heat pattern.

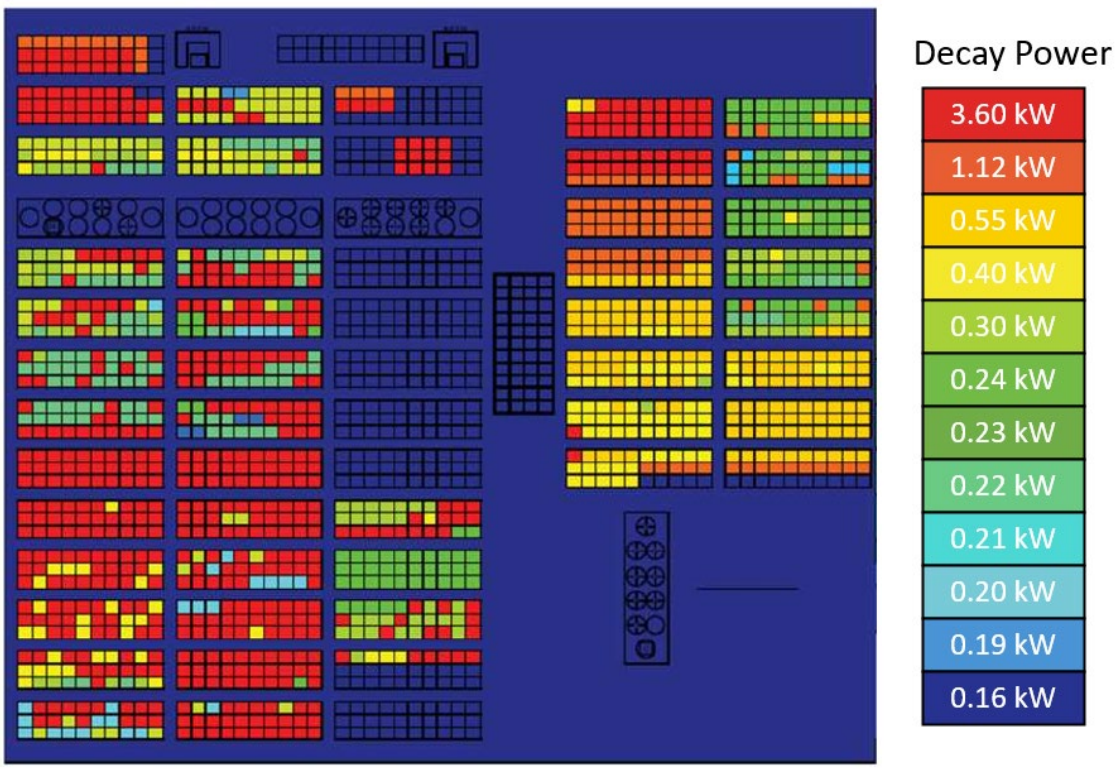

Figure 1 - Layout of spent fuel assemblies in the Fukushima unit 4 plant [2]

The dimensional data used for this analysis are referred to the BWR STEP3 fuel assembly. Each fuel assembly is composed by 72 fuel rods in a 9x9 geometry and by a central water channel. Fuel bundles are encased in a channel box. There are also seven spacers between the rods in order to keep a constant distance between them. The fuel assemblies are stored inside rectangular racks, and each rack contains 30 FAs in a 3x10 geometry for a total of 53 racks. The steel walls of the racks are double walls with some space for water flow between them. The racks are also composed of 8 legs that keep the rack suspended into water to allow natural circulation and a baffle plate to sustain the fuel rods with holes to allow water flow [2].

Since the MELCOR COR package modelling is based on a cylindrical geometry, the fuel assemblies contained in the SFP have been modelled using concentric rings. As shown in Figure 3, the SFP is divided into 3 radial rings and 13 axial levels. The total number of core cells is 39. Hot fuel assemblies have been modelled in the central radial ring, which is surrounded by a second ring modelling cold FAs, and by a third ring modelling fresh FAs. With respect to the axial position of the rods, the active fuel is 3.71 m long, starting from a height of 0.3689 m to 4.0789 m. Fuel assemblies in each ring are surrounded by the stainless steel of the racks and by the water of the pool. All the masses for stainless steel, Zircalloy and fuel are evaluated from available TEPCO data.

Considering the low heat flux and the large amount of water, a detailed nodalization that considers each fuel assemblies' correct position is not required for our simulation purposes. Moreover, as a conservative assumption, the hot FAs are placed in the central ring of the COR model.

The cells modelled in the COR package are coupled with hydrodynamic package control volumes. In particular, COR cells containing nuclear fuel are coupled two by two with a control volume. In Figure 2, the thermal-hydraulic nodalization developed for this simulation is shown. It is based on the modelling of 26 control volumes and 49 flow paths. Each core channel (1 for each COR radial ring), modelling the fluid region inside the canister, has been axially split into 6 different control volumes. The three associated bypass control volumes, modelling the interstitial space between the channel box and the rack cell walls, have been simulated with three different control volumes (CV210, CV220, CV230 in Figure 2). The flow blockage model is activated for each core channel flow path.

Moreover, logically controlled flow paths connecting channels and bypass CVs are opened if the canister wall fails. The remaining water inside the pool is modeled with two large control volumes, one surrounding the core and one representing the upper part of the pool. Two time-independent control volumes have been used to model the reactor containment and the environment.

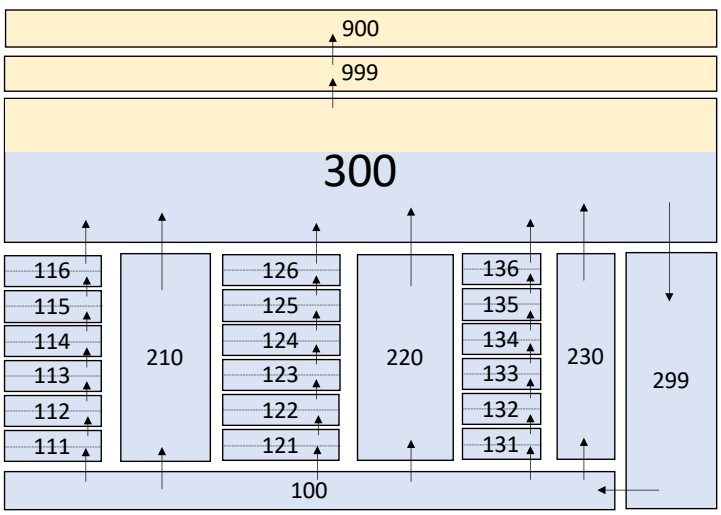

Figure 2. Fukushima Unit 4 SFP - Thermal-Hydraulic model

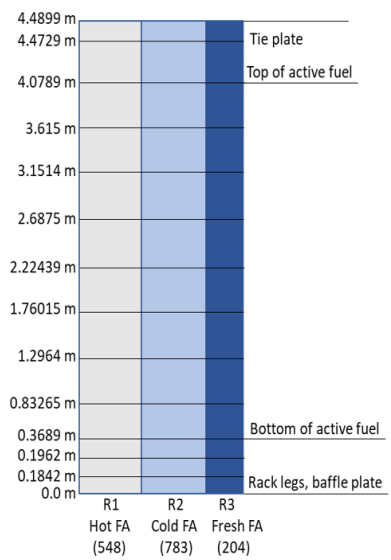

Figure 3. Axial COR nodalization

Structures surrounding the SFP have been modelled with 4 different 2-layer HSs composed of a steel liner of 0.635 cm and a 2 m thick layer of concrete. As a conservative assumption, to consider the pool as an adiabatic system, no heat exchange between the reactor building and the SFP wall structures has been assumed.

With the use of two different packages, namely the DCH (decay heat) and the RN (radionuclides) packages, MELCOR allows the user to input the decay heat power and the masses of radionuclides by defining radionuclide classes. MELCOR divides radionuclides in 17 chemical classes, indicating them with the most relevant element of the class itself. Through the DCH package, the user can define a decay power for each of these classes. Moreover, with the RN package, the user can define more specifics about radionuclides, and it is possible to define for each core cell the exact mass of present radionuclides for class. For the current analysis, radionuclides mass inventory calculated by ENEA and reported in [7] has been considered. Summing all the isotopes present for each class, an initial decay heat and radionuclides inventory have been calculated for both hot and cold fuel assemblies. As a conservative assumption, any radionuclide and decay power has been assumed for the third ring, containing only fresh fuel. The radionuclides masses for each class have been uniformly divided for the 8 cells (from axial level 4 to 11) containing active fuel, assuming a uniform distribution inside the rings.

The decay power for the 40 most relevant radionuclides is given in [7] in terms of kW/FA both for hot and cold fuel assemblies. By multiplying the given decay power for the FAs present in rings 1 and 2, a total decay power of 2.4 MW has been obtained. This has been divided into 1.9 MW and 0.5 MW for hot (ring 1) and cold (ring 2) fuel assemblies. As a conservative assumption, the decay power has been assumed to be constant throughout all the simulation time. The card for the combination of Iodine has been activated to simulate the formation of CsI and CsM in class 16 and 17, respectively.

3. Accident scenario and assumption

The postulated initiating event for the accident scenario is a loss of cooling accident occurring together with a total station blackout. The analysis has been performed assuming the unavailability of all emergency and mitigation systems. Because of the relatively low amount of decay power and the large water inventory, SFP postulated accident progression would result in a large time-consuming process. The accident sequence is assumed to start when the water level is just above the racks, in order to reduce the simulation time. For this initial condition, the total mass of water in the spent fuel pool has been evaluated to be 450 t (instead of the 1320 t of the full pool). A preliminary analysis highlighted that around 10 days are needed to reach this initial condition. Thus, for all the results described in the next session, it should be considered that for a complete accident sequence, including water evaporation from the top of SFP to rack levels, the beginning of simulation corresponds to 10 days. Since the top of the unit 4 reactor building of the Fukushima Daichi NPP was damaged by the hydrogen explosion, occurred around four days after the reactor SCRAM, its effect on mitigating radiological releases can be, conservatively, not considered.

4. Results

The predicted collapsed water level of the pool after the loss of cooling is shown in Figure 4. Since the initial bulk temperature of water has been set to saturation conditions, the water level decreases quite fast. The uncover of fuel starts after around 24 hours, reaching the end of active fuel after 6.6 days and the dry-out after around 9 days.

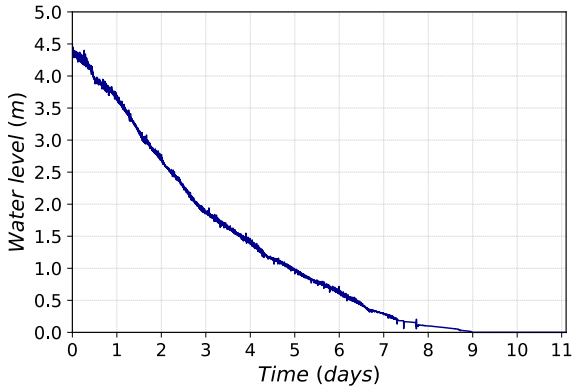

Figure 4. Water level

With the ongoing water level decrease and the consequent fuel uncovery, temperatures of core components start to increase. Figure 5 and Figure 6 illustrate the predicted cladding temperature and stainless-steel rack temperature at various axial levels for the central COR ring, respectively. The temperature of top-level cladding elements at the uppermost axial level begins to heat-up after around 36 h, when the active fuel is fully uncovered. The ability to remove decay heat generated by the spent fuel is reduced as the water level continuously drops, especially when most of the active fuels are exposed to air without any emergency mitigation available. From both Figure 5 and Figure 6, it is also possible to study the failure sequence and cladding, and rack supporting structure. The top-axial level rack cell failure occurs at 4.62 days when the temperature of the rack is 1700 K. Fuel clad failure occurs after around 6.8 days when a temperature of 2500 K is reached. The MELCOR interactive (INT) model was activated to simulate the eutectic temperature of the ZrO2–UO2 binary mixture. Two unique materials were introduced, UO2-INT and ZRO2-INT, with a eutectic temperature of 2500 K, as suggested by the Phébus experiment [8].

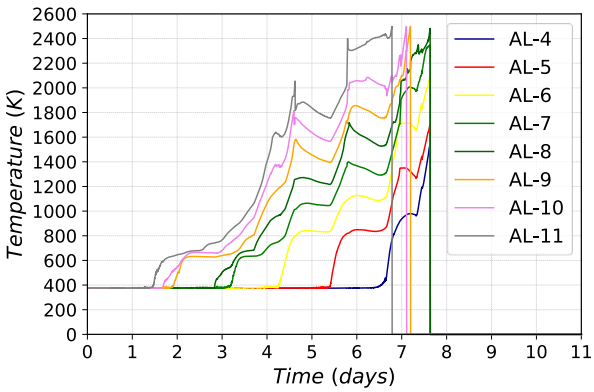

Figure 5. Fuel clad temperature in ring-1 for different axial levels

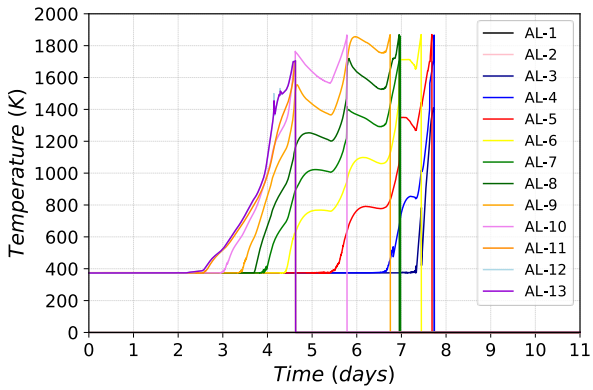

Figure 6. SFP rack temperature in ring-1 for different axial levels

The heat-up of cladding and stainless-steel structures led to the onset of oxidation reaction with steam. Zircalloy oxidation begins after a couple of days, while stainless-steel oxidation after around 4 days. In Figure 7, the waveform of oxide production is shown. After 9 days, the oxide masses are almost constant, reaching 28000 kg for ZrO2 and 26000 kg for SSOx. At the end of the simulation, around 16.0 % of Zr, and 13.7 % of SS, were oxidized, respectively. A large quantity of hydrogen gas will be generated as a product of the chemical reaction. As shown in Figure 8, hydrogen production follows the same trend of oxidation. At the same time, the hydrogen production is almost finished due to the complete evaporation of initial water, and the mass of hydrogen remains relatively constant at

1730 kg. In these conditions, it is possible to see that the progression of the accident is quite slow and let the operators have enough time to do mitigative actions. Hydrogen production results are in good agreement with the one obtained in past analyses related to loss of cooling accidents in SFP [9][10][11].

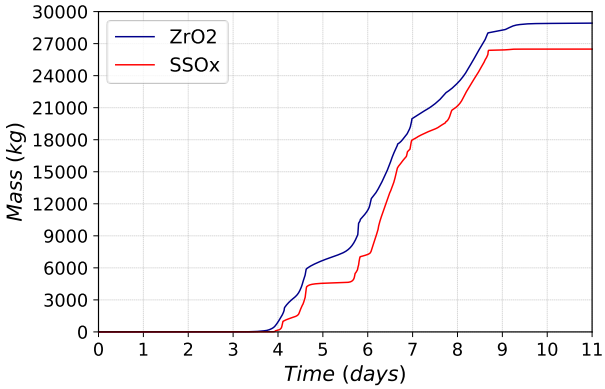

Figure 7. Zirconium and SS oxides production predicted by MELCOR

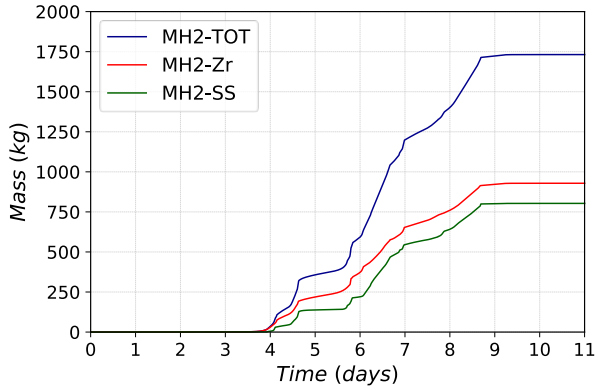

Figure 8. Mass of Hydrogen produced by oxidation reactions

When the cladding temperature reaches the oxidation temperature threshold (set to 600 K [12][13]) in the MELCOR oxidation model, the oxidation reaction in the presence of steam produces a strongly exothermic reaction that accelerates the rising of the cladding temperature. The oxidation heat generation rate in the spent fuel pool is shown in Figure 9. The heat generated is in good agreement with data obtained in the OECD/NEA Sandia Fuel Project program, conducted to study thermal-hydraulics and fire characteristics of spent fuel assemblies [14].

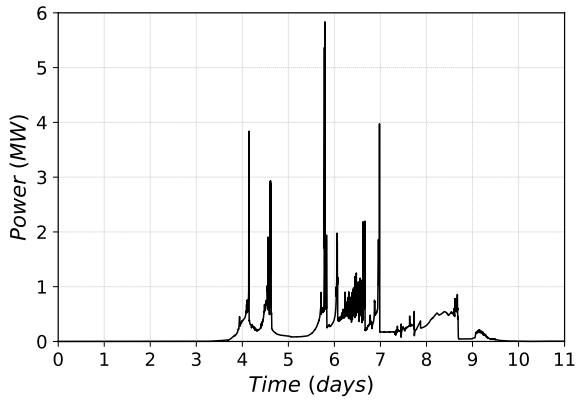

Figure 9. Oxidation heat generation rate

The volume fraction of damaged FAs is shown in Figure 10. The damage reaches 87 % since the third ring, not containing radionuclides and decay power, remains intact. Fuel rod collapse can occur through various means: loss of global support or local temperature exceeding the default temperature-based failure criterion. In the present simulation, rack baseplate failure occurs in radial rings 1 and 2. As shown in Figure 10, the ring 1 rack baseplate fails after around 6.3 days; before, the collapsed FAs are at axial levels 9 to 11. The rack baseplate of ring 2 fails after around 8.6 days, causing the failure of the cold fuel assemblies also if the threshold temperature of 2500 K is not reached in any axial node of ring two.

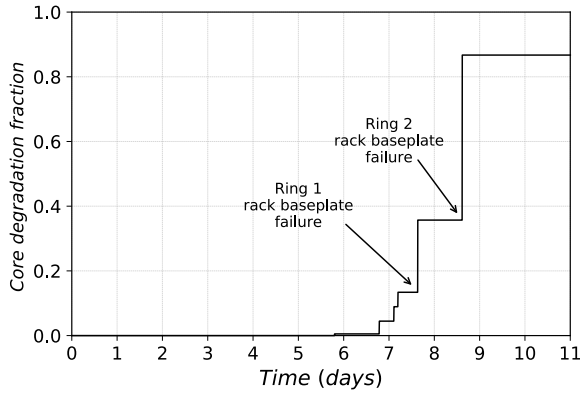

Figure 10. Core degradation fraction predicted by MELCOR

As degradation of core continues, radioactive fission gases contained in fuel assemblies are released and moved toward SPF and reactor building volumes. Source term estimation often focuses on the more volatile radionuclides such as caesium, iodine, and noble gases. Noble gasses are chemically not reactive and can contribute to the radiological doses by external exposure, generally with a small contribution on the total absorbed dose. Iodine and Caesium are volatile and chemically reactive elements and can contribute to internal exposure. Iodine may be present as the very hygroscopic molecules of Caesium Iodide (CsI), dangerous for its high solubility in water. Moreover, the Phebus FP program [8] indicated that Caesium-Molybdate (Cs2MoO4) is the dominant chemical form of the released Caesium, also if it is less volatile than CsOH and CsI. Analyzing MELCOR classes' contributions to released radionuclides, the most significant mass inventory is the chalcogens mass (including radioactive Tellurium), with about 16000 kg (Figure 11). Other relevant radiological MELCOR classes, shown in Figure 12, are: class 1 (grouping noble gases) with 933 kg; class 2 (containing alkali metals, such as CsOH) with 554 kg; class 16 (CsI) and class 17 (Caesium-Molybdate) with 83 kg and 20 kg, respectively.

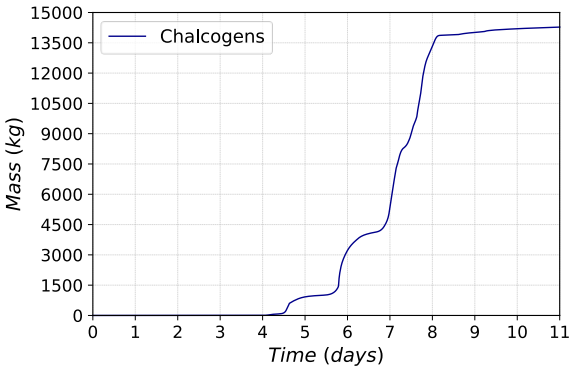

Figure 11. Mass of Chalcogens (Te) released from FAs predicted by MELCOR

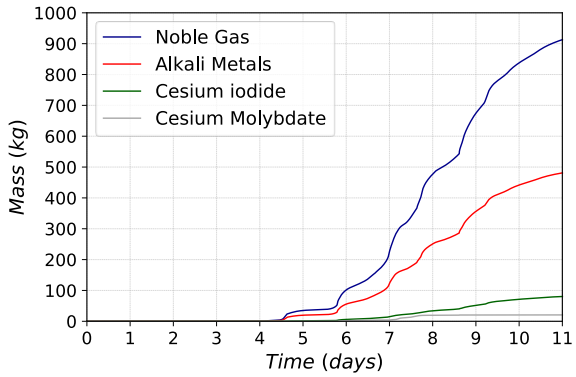

Figure 12. Mass of radionuclides released from FAs predicted by MELCOR

In Table 1, the comparison between initial and released mass is reported for the most relevant classes. Other classes have not been reported since radionuclides masses are very low (<5 kg) with respect to the ones shown in Table 1. As a final investigation, the release in different volumes was analyzed. Almost the totality of released radionuclides from the fuel assemblies has been deposited on the metal structures inside the pool and on the concrete structures of the pool. The released fraction outside the SFP is reported in Table 1.

Table 1. Radionuclides masses

Class	Initial masses	Mass released from FAs	Release fraction from FAs	Mass released outside the pool	Release fraction outside the pool
Class 1	1493.9 kg	933.15 kg	62.5%	871.27 kg	58.32%
Class 2	822.326 kg	554.47 kg	66.2%	66.24 kg	8.05%
Class 5	35018.0 kg	16094.3 kg	46%	1796. kg	5.12%

5. Conclusions

In the present work, the spent fuel pool of Fukushima Daiichi unit 4 has been modelled with MELCOR 2.2.18019 to evaluate the consequences of a loss of cooling accident in terms of fuel degradation and radionuclides release. Main results of the severe accident progression include fuel degradation, clad and racks structures heat-up and failure, hydrogen generation and radionuclide release. In particular, the large amount of water leads to the formation and production of large amounts of oxides and hydrogen. The studied accident, even starting from very conservative conditions such as the water in saturated conditions, let however a long time for mitigative actions to prevent fuel uncover and consequent overheating of the fuel rods since the progression of accident events is quite slow.

Release of radionuclides from cladding begins about 4 days after the initiating event and it strongly follows the progression of core damage and the ongoing oxidation. It was also shown that, in the long-term scenario, almost the whole core fails except for the ring containing fresh fuel that, having no decay power, does not heat up as fast as the other two rings. The failing of the core leads to the production of debris, but there is no damage to the pool walls, so the only concern for radionuclide release is the atmospheric one that is anyway a low part of radionuclides initial mass.

The analyses highlighted as the MELCOR could provide a valuable means for characterizing the consequences of severe accidents in spent fuel pools. However, as there are no corresponding experimental results, the validity of the simulation results cannot be assessed in detail, and uncertainty analyses are needed. As future work, this MELCOR model, developed in the framework of the MUSA project [5], will be used to perform sensitivity and uncertainty analyses to better explore the variability in the potential responses of SFPs systems undergoing severe accident conditions**.**

References

[1] OECD- Organisation for Economic Co-operation and Development, Phenomena Identification and Ranking Table, R&D Priorities for Loss-of-Cooling and Loss-of-Coolant Accidents in Spent Nuclear Fuel Pools, NEA/CSNI/R(2017)18

[2] Condreau O, Jäckel B, Rocchi F, Alcaro F, Angelova D, Bandini G, Barnak M, Behlerg M, Da Cruz DF, Dagan R, Drai P, Ederli S, Herranz LE, Hollands T, Horvath G, Kaliatka A, Kljenak I, Kotsuba O, Lind T, López C., Mancheva K, Matejovic P, Matkovič M, Steinbrückh M, Stempniewicz M, Thomas R, Vileiniskis V, Visser DC, Vokáč P, Vorobyov Y and Zhabin O 2018 *Severe accident code-to-code comparison for two accident scenarios in a spent fuel pool*, Annals of Nuclear Energy, 120, pp 880–887, https://doi.org/10.1016/j.anucene.2018.06.043

[3] Humphries LL, Beeny BA, Gelbard F, Louie DL and Phillips J 2015 *MELCOR Computer Code Manuals, Vol. 1: Primer and Users' Guide, Version 2.1.6840*, SANDIA REPORT SAND-2015-6691R, Sandia National Laboratories, Albuquerque, New Mexico. https://www.nrc.gov/docs/ML1704/ML17040A429.pdf

[4] Humphries LL, Beeny BA, Gelbard F, Louie DL and Phillips J 2017 *MELCOR Computer Code Manuals, Vol. 2: Reference Manual,* Version 2.1.6840, SANDIA REPORT SAND2017-0876O, Sandia National Laboratories, Albuquerque, New Mexico. https://www.nrc.gov/docs/ML1704/ML17040A420.pdf

[5] Luis E. Herranz, Sara Beck, Victor Hugo Sánchez-Espinoza, Fulvio Mascari, Stephan Brumm, Olivia Coindreau and Sandro Paci, The EC MUSA Project on Management and Uncertainty

of Severe Accidents: Main Pillars and Status, Energies 2021, 14(15), 4473, https://doi.org/10.3390/en14154473

[6] Gauntt RO, Kalinich DA, Cardoni JN, Phillips J, Goldmann AS, Pickering SY, Francis M, Robb K, Ott LJ, Wang D, Smith C, Germain S, Schwieder D and Phelan C 2012 *Fukushima Daiichi Accident Study (Status as of April 2012),* SAND2012-6173; Sandia National Laboratories, USA, https://doi.org/10.2172/1055601

[7] AIR-SFP Benchmark, Microsoft Excel file, Inventories_for_HF_and_CF_Revised_25_05_2020_V1, not publicly available

[8] Bernard C and Roland Z 2013 *The objectives of the Phébus FP experimental programme and main findings*, Annals of Nuclear Energy, 61, pp 4-10, https://doi.org/10.1016/j.anucene.2013.03.037

[9] Wang D, Gauld IC, Yoder GL, Ott LJ, Flanagan GF, Francis MW, Popov EL, Carbajo JJ, Jain PK, Wagner JC and Gehin JC 2012 *Study of Fukushima Daiichi nuclear power station unit 4 spent-fuel pool*, Nuclear Technology, 180, pp 205-215, https://doi.org/10.13182/NT12-A14634

[10] Carbajo JJ 2012 *MELCOR Model of the Spent Fuel Pool of Fukushima Dai-ichi Unit 4*, American Nuclear Society Meeting, Chicago, Illinois, USA

[11] Wu X, Li W, Zhang Y, Tian W, Su G and Qiu S 2014 *Analysis of the loss of pool cooling accident in a PWR spent fuel pool with MAAP5,* Annals of Nuclear Energy, 72, pp 198-213, https://doi.org/10.1016/j.anucene.2014.05.030

[12] Urbanic VF and Heidrick TR 1978. *High-temperature oxidation of zircaloy-2 and zircaloy-4 in steam*, Journal of Nuclear Materials, 75, pp 251–261, https://doi.org/10.1016/0022-3115(78)90006-5

[13] Coindreau O, Duriez C and Ederli S 2010 *Air oxidation of Zircaloy-4 in the 600-1000 °C temperature range: Modeling for ASTEC code application,* Journal of Nuclear Materials, 405, pp 207-2015, https://doi.org/10.1016/j.jnucmat.2010.07.038

[14] Martina Adorni, Luis E. Herranz, Thorsten Hollands, Kwang-II Ahn, Christine Bals, Francesco D'Auria, Gabor L. Horvath, Bernd S. Jaeckel, Han-Chul Kim, Jung-Jae Lee, Masao Ogino, Zsolt Techy, Alexander Velazquez-Lozad, Abdelghani Zigh, Radomir Rehacek, 2016, *OECD/NEA Sandia Fuel Project phase I: Benchmark of the ignition testing,* Nuclear Engineering and Design, 307, pp 418-430, https://doi.org/10.1016/j.nucengdes.2016.07.016