Transactions of the Korean Nuclear Society Spring Meeting
Jeju, Korea, May 9-10, 2024

Current Status of Fission Products Behavior Analysis Code Development in Salt Spill Accident Condition

Sung Il Kim*, Hyung Seok Kang, Jae Hyun Ham, Donggun Son, Kwang Soon Ha
Intelligent Accident Mitigation Research Division, Korea Atomic Energy Research Institute, (34057) 111, Daedeok-daero 989 beon-gil, Yuseong-gu, Daejeon, Republic of Korea
**Corresponding author: sikim@kaeri.re.kr*

Keywords : Molten salt reactor (MSR), safety, fission product, source term, salt spill accident

1. Introduction

The molten salt reactor is expected to be a safe nuclear power plant that can fundamentally eliminate severe accident involving nuclear fuel melting, as known for releasing a smaller amount of radioactive material compared to conventional water-cooled reactors in case of accidents [1, 2]. Furthermore, even in the event of a leak of molten salt outside the system, it is anticipated that the heat transfer to the surrounding atmosphere will facilitate the solidification of molten salt due to its low quantity of nuclear fission products(FPs) with FPs removal system(for example, off-gas system), thereby preventing further progression of accidents [3]. Based on this inherent safety feature, research is underway for the miniaturization and modularization of nuclear reactors, with efforts being made to apply them to various fields such as ships and power sources for transportation.

In order to obtain regulatory approval from regulatory agencies is essential for the construction and commercialization of molten salt reactors. To achieve this, it is imperative to demonstrate the safety of molten salt reactors through experimentation or reliable analyses. For instance, it is necessary to evaluate the quantitative time and behavior until molten salt reaches solidification, even though it is expected to be cooled and solidified in the event of a molten salt leak. The temperature and time to solidification of molten salt are directly related to the release of fission products contained within the molten salt, thus requiring consideration in accident prevention and management strategies [4, 5].

The development of reliable fission product behavior analysis codes is essential to assess the quantity of nuclear fission products released into both internal and external environments of marine nuclear power systems in the event of accidents. As shown in Figure 1, through code development, it is anticipated to evaluate the initial inventory of nuclear fission products and the release rate of nuclear fission products according to the temperature of leaked molten salt. Fission products released outside the molten salt move to other containment vessels and exist in the form of gas and aerosols, with aerosols undergoing repetitive growth and removal processes. Interpreting the behavior of fission products based on temperature, pressure conditions within containment vessels, and ultimately evaluating the release rate of major radionuclides into the environment is necessary [6, 7].

To assess the safety of the marine molten salt reactor system, it is necessary to evaluate the leaked molten salt behavior based on the physicochemical properties. Although it is expected that leaked molten salt will readily solidify, entrapping most radioactive materials including nuclear fission products, the technical basis for this assumption is lacking. Moreover, there is minimal information available regarding the physicochemical and thermal properties of leaked molten salt and nuclear fission products, necessitating the acquisition of technical judgment based on fundamental experiments and analyses.

Therefore, in this study, as a fundamental step towards developing a radioactive source term assessment code for accidents in molten salt reactors, we aim to develop a model for the release of fission products from leaked molten salt. To achieve this, we have reviewed existing research and selected the main release mechanisms of nuclear fission products. Additionally, we have organized equations for modeling mechanisms released by concentration gradients and reviewed methodologies for calculating the vapor pressure of nuclear species.

2. Research trend on fission product release

2.1 RN package development in MELCOR

MELCOR is updating its model for accident analysis in molten salt reactors [2, 7]. The current status of overall model development is as follows: The COR package in MELCOR, responsible for fuel damage and behavior, is not directly linked to the RN(RadioNuclide) package, the module for nuclear fission product behavior. Therefore, for current analysis, methods include providing initial values to desired control volumes (CVs) or using mass as a source over time. Movement of fission products employs the same method as before, floating within the primary flow gas between compartments. Precipitation, attachment, and re-suspension mechanisms of fission products use the same methods as in the HS package. The DCH package

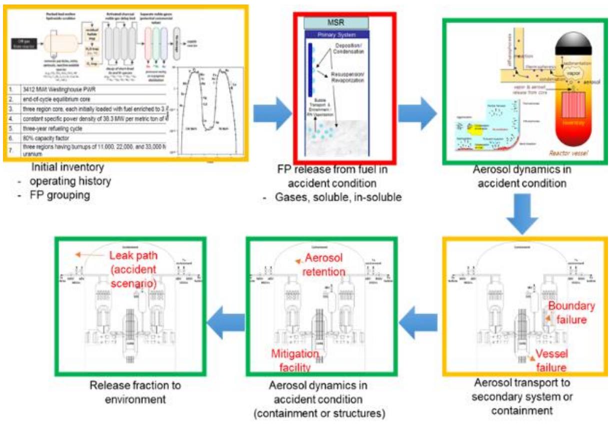

Fig. 1. General procedure for calculating source term.

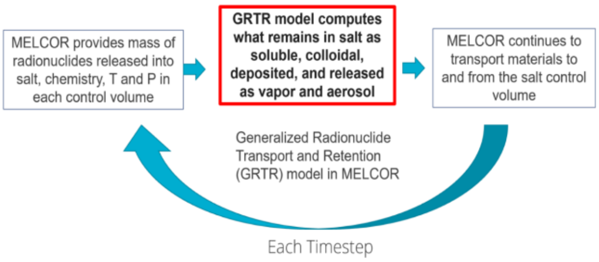

Fig. 2. GRTR development.

reflects the decay heat generated by fission products, distributing heat differently depending on their location (liquid salt, atmosphere, attached to structures). The aerosol behavior model used in the RN package remains consistent in MSR models, with interactions between aerosol and gas phases determined by the temperature and pressure of the respective node. Aerosols in the atmosphere are continuously calculated based on diameter in the RN package, while those present in the liquid salt or gas phase do not undergo diameter calculations. Movements between gas and aerosol phases for each nuclide and their movements based on location are implemented using control functions (CFs) as follows:

· Can set CFs in each CV for any and all RN1 classes

· CFs set mass transfer rates that are multiplied by time-step size in order to ascertain total mass moves by class between forms

· Aerosols in pool transform to Aerosol in atmosphere (user-specified section or section 1 –the largest)

· Vapor in pool transforms to vapor in atmosphere

· Radioactive/total move in like proportion across pool/atmosphere interface

Sandia National Laboratories (SNL) is developing the GRTR (Generalized Radionuclide Transport and Retention) framework and planning to perform calculations as shown in Figure 2. Through MELCOR calculations, the amount of fission product release, chemical reactions, and the temperature and pressure of each node are obtained. Subsequently, the GRTR model evaluates the quantity of nuclear fission products present in the molten salt, including soluble, colloidal, and deposited forms, as well as the amount released in gas and aerosol forms. This information is then received by MELCOR to calculate movement between nodes. Assuming constant temperature and pressure conditions within each timestep, these values are updated at each timestep.

As depicted in Figure 3, fission products in the molten salt reactor can be broadly classified into five types. The first type is the highly soluble form, which is dissolved in the liquid molten salt. The fission products dissolved in the liquid molten salt are evaluated as the total quantity present without considering size distribution. The second type is the insoluble/colloidal form present in the liquid but not dissolved. The quantity of such nuclear fission products, as well as their size distribution, is calculated at each timestep, along with the total quantity present in the liquid molten salt. The third type is the surface insoluble/colloidal form present on the liquid salt surface. Similar to the second type, the size distribution and total quantity present in the liquid molten salt are calculated for these nuclear species. The fourth type is the insoluble/colloidal HS deposition form attached to structures. For these nuclear species, the particle size distribution is calculated, and calculations are performed in conjunction with the HS package.

2.2 GEMS code calculation

At PSI (Paul Scherrer Institut) and ETH Zurich in Switzerland, the release of fission products from leaked molten salt was evaluated using the GEMS(Gibbs Energy Minimization Software) code and HERACLES database, and the results were simulated with MELCOR [8, 9]. This report describes how the release of nuclear fission products was derived in this study.

The initial inventory of nuclear fission products is distributed in the molten salt with depending operating period. By considering these elements, Gibbs Free Energy Minimization calculations are used to select the most probable nuclear species compounds. Using the results of these calculations, the saturated vapor pressure of the potentially released nuclear species compounds can be calculated based on the temperature of the molten salt using HERACLES data and GEMS calculations. When saturated vapor pressure of nuclear species compounds released from the molten salt at the calculated temperature is determined as P_{sat}, the saturation concentration is determined using the equation (1) (where R is the ideal gas constant, M is the molecular weight of the nuclear species, and T is the temperature of the atmosphere where the fission products are released). Additionally, to calculate the release rate, equation (2) is used, where Ci is the concentration of nuclear species (i) in the atmosphere, and A is the surface area of the molten salt where

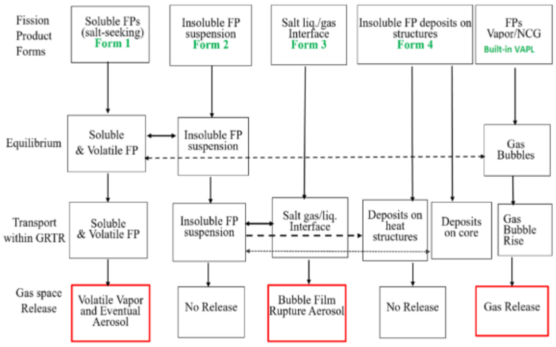

Fig. 3. Classification of nuclides from MSRE.

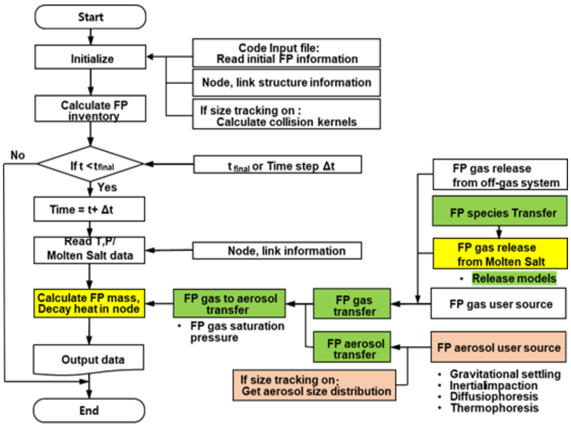

Fig. 4. Concept proposal for code development for evaluating source term.

evaporation occurs. Unknown values can be determined using equations presented in the reference [8].

$$C^s = \frac{P_{sat}(T)M_i}{RT} \quad (1)$$

$$\frac{dm_i}{dt} = \mathrm{A}k_i(C_i^s - C_i^a) \quad (2)$$

In the research using GEMS, the release of fission products and the behavior of aerosols in containment vessels were evaluated under various accident conditions, and the mass of nuclear species compounds was determined [9].

3. Applicability of SIRIUS code

The SIRIUS(Simulation of Radioactive nuclides Interaction Under Severe accidents) code, which is used for analyzing the behavior of fission products in the event of a severe accident in pressurized water reactors, was reviewed for its applicability to molten salt reactors [10, 11]. The domestic integrated accident analysis code, CINEMA, for analyzing severe accidents in pressurized water reactors consists of CSPACE for reactor and reactor coolant system analysis, SACAP for containment building analysis, the SIRIUS module for analyzing radioactive material behavior, and the MASTER module for integrating these components [12]. An evaluation was conducted, and based on this, a preliminary concept draft for a future development of fission product assessment code was derived, as shown in Figure 4.

SIRIUS, used for analyzing the behavior of radioactive material within the reactor coolant system and containment building, interprets various aspects including the initial inventory of nuclear fission products within the fuel rods, release of nuclear fission products from the fuel rods, aerosol generation from released nuclear fission products, transport and removal of nuclear fission product aerosols, and transport of nuclear fission products in the gas phase.

The applicability of this code to the analysis of fission product behavior in MSRs was evaluated as follows:

- Target Elements: Additional considerations may be needed depending on the existing SIRIUS model and the composition of the molten salt.
- Nuclear Fission Product Inventory Calculation Library: Additional references may be needed based on calculations for molten salt.
- Nuclear Fission Product Release Model: Improvements may be necessary as the existing model is based on the release of nuclear fission products from solids. Theoretical and experimental data are needed to predict the release based on the saturated vapor pressure of nuclear fission products within the molten salt.
- Aerosol Removal Mechanisms: The existing SIRIUS model may be applicable. Additional phenomena reflecting the characteristics of molten salt reactors may be incorporated into the model.
- Aerosol Transport: The existing SIRIUS model may be applicable.
- Others: Considerations for additional chemical reaction phenomena among released nuclear fission products may be needed to reflect the characteristics of molten salt reactors.

4. Conclusions

The ultimate goal of this study is to quantitatively assess the amount of fission products released into the environment in the event of an accident in a molten salt reactor. This is a crucial research endeavor as it serves as a significant means to demonstrate the safety of the ongoing MSR research and can be utilized as quantitative data for safety evaluations during the licensing process of MSR in the future.

To achieve this goal, the study first analyed the codes used for accident analysis in pressurized water reactors to derive a conceptual framework for a fission product behavior analysis code. It distinguished between models that can be adapted from existing ones and those that need to be newly developed to reflect the characteristics of molten salt reactors. One of the major differences between water based nuclear power plants and MSRs is the mechanism of fission product release from leaked molten salt. It is essential to evaluate and identify this mechanism by investigating how specific nuclear fission products dissolved in the molten salt are released outside the salt as a function of temperature in the future.

Initial studies conducted in previous research, such as in MELCOR or SAMOSAFER, have provided preliminary assessments of the amount of fission products released into the environment in the event of an accident in a molten salt reactor [13]. Building upon this, the current study aims to evaluate the release quantity through Gibbs Free Energy minimization calculations and utilize it to develop a comprehensive model that will be incorporated as a module into the final fission product behavior assessment code. Currently, we are collaborating with PSI on research using the GEMS code.

ACKNOWLEDGEMENT

This work was supported by the National Research Foundation of Korea(NRF) grant funded by the Korea government(MSIT) (RS-2023-00261295).

REFERENCES

[1] David E. Holcomb, Alexander J. Huning, Randall J. Belles, George F. Flanagan, Willis P. Poore, Integrating the Safety Evaluation for a Molten Salt Reactor Operation and Fuel Cycle Facility Application, ORNL/TM-2022/2671.

[2] U.S.NRC, SCALE/MELCOR Non-LWR Source Term Demonstration Project –Molten Salt Reactor (MSR), SAND2022-12146 PE, September 13, 2022.

[3] Sara Thomas and Josh Jackson, MSR Salt Spill Accident Testing Using Eutectic NaCl-UCl3, ANL/CFCT-22/32.

[4] Nuclear Energy Institute (NEI), Risk-Informed Performance-Based Technology Inclusive Guidance for Non-Light Water Reactor Licensing Basis Development, Report Revision 1, August 2019, NEI18-04.

[5] KC Wagner, Dave Luxat, and Brad Beeny, Molten Salt Reactor–MSRE Day 1, Option 2, Session 5, MELCOR Workshop, June 12-16, 2023.

[6] Sara Thomas, James Jerden, Mechanistic Source Term Development for Liquid Fueled MSRs -Model Development Update, ANL/CFCT-20/16.

[7] L.L. Humphries, B.A. Beeny, C. Faucett, F. Gelbard, T. Haskin, D.L. Louie, J. Phillips, MELCOR Computer Code Manuals, Vol. 2: Reference Manual Version 2.2.11932, SAND2018-13560 O.

[8] Jarmo Kalilainen, Sergii Nichenko, Jiri Krepel, Evaporation of materials from the molten salt reactor fuel under elevated temperatures, Journal of Nuclear Materials 533 (2020) 152134.

[9] Sergii Nichenko, Jarmo Kalilainen and Terttaliisa Lind, MSR Simulation with cGEMS: Fission Product Release and Aerosol Formation, Journal of Nuclear Engineering, 2022, 3, 105-116.

[10] Kwang Soon Ha, Sung Il Kim, Hyung Seok Kang, and Dong Ha Kim, SIRIUS: A Code on Fission Product Behavior under Severe Accident, Transactions of the Korean Nuclear Society Spring Meeting Jeju, Korea, May 18-19, 2017.

[11] Sung Il Kim, Kwang Soon Ha and Dong Ha Kim, Analysis of Accident of 1400 MWe Nuclear Power Plant with SIRIUS in CINEMA, Transactions of the Korean Nuclear Society Autumn Meeting Gyeongju, Korea, October 26-27, 2017.

[12] JinHo Song, DongGun Son, JaeHyun Ham, JunHo Bae, SungWon Bae, Kwangsoon Ha, ByungJo Kim, Sun Hong Yoon, BubDong Chung, SoonHo Park, ChangHwan Park, JaeHwan Park, YuJung Choi, A comparative simulation of severe accident progressions by CINEMA and MAAP5, Nuclear Engineering and Design 404 (2023) 112181.

[13] Fred Gelbard, Bradley A. Beeny, Larry L. Humphries, Kenneth C. Wagner, Lucas I. Albright, Max Poschmann & Markus H. A. Piro, Application of MELCOR for Simulating Molten Salt Reactor Accident Source Terms, NUCLEAR SCIENCE AND ENGINEERING, VOLUME 197, 2723–2741, OCTOBER 2023.