

Evaluation on In-vessel Source Term in PGSFR (2017 Results)

Seung Won Lee *, Sang June Ahn, Chi-Woong Choi, Kwi Lim Lee, Taekyeong Jeong, Sunghyuk Im
SFR Reactor Design Division

Korea Atomic Energy Research Institute (KAERI)

*Corresponding author: swonlee@kaeri.re.kr

1. Introduction

The potential for accidents to cause the release of radionuclides into the public environment is the source of safety concern with the use of nuclear reactors for power generation, research and actinide transformation. Safety concerns with nuclear power plants are sufficient that a conservative safety strategy termed “defense in depth” has been adopted essentially universally. This strategy requires nuclear plants to have features that prevent radionuclide release and multiple barriers to the escape from the plants of any radionuclides that are released despite preventive measures. Considerations of the ability to prevent and mitigate release of radionuclides arise at numerous places in the safety regulations of nuclear plants. The effectiveness of mitigative capabilities in nuclear plants is subject to quantitative analysis. The radionuclide input to these quantitative analyses of effectiveness is the Source Term (ST). All features of the composition, magnitude, timing, chemical form and physical form of accidental radionuclide release constitute the ST [1]. Also, ST is defined as the release of radionuclides from the fuel and coolant into the containment, and subsequently to the environment [2].

Since the TMI accident in 1979, extensive experimental and analytical information has been accumulated on the accident ST for LWRs. Such mechanistic models and computer codes as the MELCOR and MAAP have been developed [3]. The results of extensive calculations and experiments have been used to formulate an alternative to the simple TID-14844 ST [4] for regulatory purpose. This Alternative ST (AST), NUREG-1465 ST categorizes radionuclides into eight chemical classes based on chemical and physical similarity [5]. AST specifies the release fractions of each class of radionuclides into the containment during each of the four accident phases: gap release, in-vessel release, ex-vessel release and late in-vessel release. Use of AST is optional for existing LWRs. Future LWRs are required to use AST. AST is certainly not applicable to SFR. AST provides a valuable insights and framework for the development of a mechanistic ST model for SFR to be used in licensing as well as risk analysis.

There are not much experimental data or experience about the source term of metal fuel in SFR. Since one example of the source term about metal fuel comes from ANL-ART-38 report [6], KAERI preliminarily will evaluate the in-vessel ST using ANL methodology in

the Prototype Gen-IV Sodium-cooled Fast Reactor (PGSFR). This paper shows the matters of progress of the preliminary evaluation on the in-vessel ST.

2. Calculation of In-vessel Source Term

2.1 Assumptions of Radiological Consequence Analysis

In-vessel STs are estimated using a nonmechanistic and conservative methodology like that of ANL methodology. The fraction of fuel damaged is assumed 100 % (whole fuel rod failure). The radioactivity inventory is assumed 120 % of the estimated inventory at the end of life that is very conservative but used to cover uncertainties in estimating the Fission Products (FPs) and activated primary sodium inventory. The high primary sodium temperature (1,300 °C) is used for estimating the release fraction in the release into the cover gas region.

2.2 Radionuclide Groups and Inventory

The elements to be evaluated and the radionuclide groups were specified based on NUREG-1465 ST [5] and Regulatory Guide (RG) 1.183 [7]. Because uranium is not defined in RG 1.183, it was included in the cerium group. A separate group was added, because the operation of liquid metal-cooled reactors results in the activation of the sodium coolant. Radionuclides with a half-life of more than 1 minute are considered. The radionuclide groups and the elements are as follows:

1. Nobles Gases: Xe, Kr
2. Halogens: I, Br
3. Alkali Metals: Cs, Rb
4. Tellurium Group: Te, Sb, Se
5. Barium, Strontium: Ba, Sr
6. Noble Metals: Ru, Rh, Pd, Mo, Tc, Co
7. Lanthanides: La, Zr, Nd, Eu, Nb, Pm, Pr, Sm, Y, Cm, Am
8. Cerium Group: Ce, Pu, Np, U
9. Coolant: Na

The radiological inventory is proportional to the thermal power and is gradually accumulated depending on the radionuclide. The inventory of each radionuclide is calculated by ORIGEN-2 code using the peak burnout conditions as shown in the Fig. 1. The radiological inventory may include errors in excess of 20 % in magnitude by taking in account various uncertainties

associated with fuel mass in the core. The nominal value of the radiological inventory is multiplied by a factor of 1.2 as an uncertainty margin to give the radiological inventory.

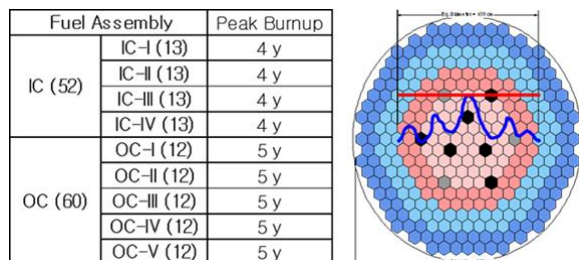


Fig. 1. The peak burnup conditions in ORIGEN-2 code.

2.3 Release Path

Figure 2 shows the radioactive release path used in the PGSFR ST evaluation. The release path includes transport of the fission products from the damaged fuel to the primary coolant, release of fission products and activated sodium from the primary coolant to the cover gas space, leakage from the cover gas space to the containment, and the leakage from the containment to the environment.

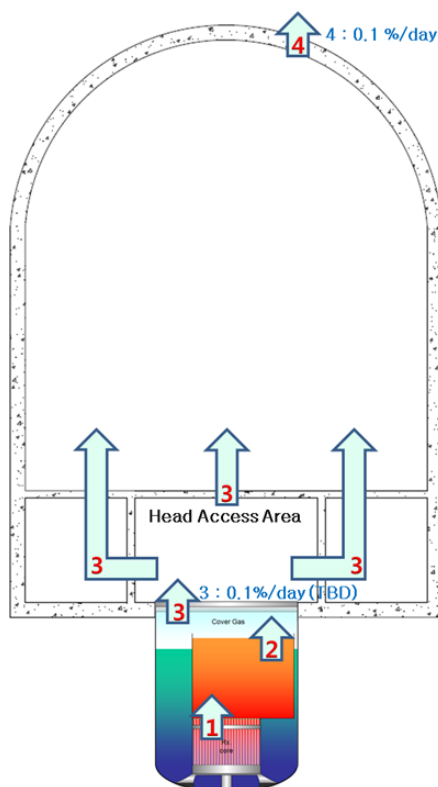


Fig. 2. Release path for ST evaluation in PGSFR.

2.4 Release from the Core to Primary Sodium

Table I shows the release fraction from the core to primary sodium using for this calculation.

Table I: Release Fraction from the Core to Primary Sodium

Radionuclide Groups	Release Fraction (Core to Primary Sodium)
Noble Gases	1
Halogens	1
Alkali Metals	1
Te Group	0.05
Ba, Sr	0.2
Noble Metals	0.05
Ce	Ce 0.15
Group	U&Pu 0.001
Lanthanides	0.3

The main assumptions are as follows [6]:

At very high temperatures (1,700 °C ~ 1,800 °C) for short time periods (<1 min), the ORNL fuel melt experiments saw complete release of the noble gases with fuel at 15% burnup. Additionally, melt tests of uranium metal in sodium conducted by AI saw the complete release of xenon from the fuel samples when melted at extremely high temperatures (> 2,300 °C) over very small time periods (< 20 ms). Both ORNL and AI melt tests indicate that complete (100 %) noble gas release should be expected at very high temperatures, with no dependency on fuel burnup or time at elevated temperature.

Many iodine volatilization tests have been conducted in this temperature range. The EBR-II melt refining tests and FCF (Fuel Cycle Facility) reprocessing experience saw the majority of the iodine volatilized at heating to 1,300 °C for several hours, with essentially complete volatilization with heating to 1,400 °C for several hours. This process formed the basis of iodine removal during reprocessing at FCF, which was successfully utilized for over 35,000 used fuel pins. Similarly, the melt tests conducted at ORNL in air and in steam environments saw on average 26 % ~ 46 % of the iodine volatilized when at 1,700 °C ~ 1,800 °C for less than a minute. This appears to indicate some amount of time dependence, as complete volatilization of iodine did not occur immediately, even with some oxidation occurring, which likely increases volatilization. Lastly, the AI melt tests provide perhaps the best comparison to an SFR accident, as the fuel melting was conducted in sodium. The results find iodine release fractions between 44 % and 72 %. While the time at temperature for these tests was very short (< 20 ms), the complete melting and disintegration of the fuel likely aided in iodine release. At 1,300 °C, a slightly smaller release may occur, but a substantial amount of iodine will still be released. Therefore, the iodine release fraction is set at up to 100 % of the inventory, with a small dependence on time at temperature.

For five years, the FCF at EBR-II reprocessed 35,000 used metal fuel pins by heating them to 1,400 °C for 4 hours and volatilizing the cesium inventory in an inert gas atmosphere. This experience indicates that approximately all the cesium is volatilized from the fuel at this temperature and time range. The only other quantitative data point at this temperature range is the cesium volatilization melt tests conducted by ORNL. Natural uranium held at very high temperatures (1,700 °C ~ 1,800 °C) for less than a minute resulted in the volatilization of 10 % ~ 20 % of the cesium inventory, despite some amount of oxidization occurring. Surprisingly, the AI melt tests did not find cesium in the sodium following the melt tests (and the small quantity of cesium that was found in the cover gas space was likely the result of xenon decay). The FCF experience at EBR-II clearly demonstrates that complete cesium volatilization is possible when at 1,400 °C for multiple hours. The ORNL melt tests show that release fractions may be smaller if the time period is short, even at very high temperatures. Therefore, the cesium release percentage is set at up to 100 %, with a dependence on time at elevated temperature.

The EBR-II melt refining tests examined the volatilization of tellurium at 1,200 °C and 1,400 °C, and found volatilized quantities below 5 %. However, the use of an oxide crucible makes the applicability of these results for metal fuel melting questionable, as the majority of the tellurium reacted with the crucible. The Hanford melt test in helium provides another quantitative indication of tellurium volatilization, with 4.2 % of the tellurium inventory volatilized when at 1,215 °C for 25 minutes. However, like the EBR-II melt refining tests, the use of a zirconia (ZrO₂) boat as the melting vessel may have affected the results through the formation of oxides. The EBR-II and Hanford melt tests provide limited insight, as oxide crucibles affect the applicability of the results. However, the Fermi 1 accident and EBR-II capsule failure appears to reinforce the notion that the tellurium release is limited. The EBR-II experimental capsule used a U-Pu-Zr fuel element, and the presence of zirconium may account for the retention of tellurium. Fermi 1 utilized a U-Mo fuel, but did have a zirconium cladding, although the low burnup may have limited tellurium migration to the cladding. Therefore, the release percentage for tellurium in this temperature range is set at up to 5 %. Although tellurium release was not noted following the Fermi 1 incident and the EBR-II experimental capsule failure, the EBR-II melt refining tests saw some volatilization (below 5 %) even with an oxide crucible.

EBR-II melt refining tests at 1,400 °C for three hours found about 1 % of the barium volatilized, with the rest of the inventory bonding with the oxide crucible. ORNL melt tests found only 0.2 % ~ 0.4 % of barium volatilized in steam and in air at 1,700 °C ~ 1,800 °C for less than a minute, but in an oxide crucible. Perhaps the most relevant test, the AI melt tests in sodium found 2.1 % ~

5.4 % of the barium released at very high temperatures with fuel dispersal (and some vaporization) in sodium. The EBR-II and ORNL melt tests indicate very little barium volatilization, even at temperatures approaching the boiling point of barium (1,897 °C), but with oxide crucibles. Perhaps most notable, the AI melt test in sodium found a relatively small fraction of barium released, despite very high temperatures and complete fuel dispersal in sodium. This would appear to indicate that barium release from the fuel matrix to the sodium is small, even when molten. Therefore, the determined release fraction is 20 % (up to 5 % from the sodium bond, and a conservative 15 % through volatile and non-volatile release from the fuel matrix).

The only data point at this temperature range is the melt tests performed by ORNL in air and in steam. These tests found on average 1 % ~ 2 % of the strontium inventory volatilized when heated to 1,700 °C ~ 1,800 °C for a minute or less. As noted previously, some amount of oxidation likely occurred during these tests. Also, the use of a quartz (SiO₂) crucible may have resulted in the formation of some oxides. However, the relatively short time period of the experiment may have lessened this effect when compared to the Hanford melt test and EBR-II melt refining tests. The ORNL melt tests appear to indicate a strontium release percentage below 5 %. However, due to the conflicting Fermi 1 data from the previous temperature category, the release percentage is set at up to 20 %.

The AI melt tests in sodium found a ruthenium release of 0.66 % ~ 1.9 % with complete fuel melting and dispersal (and some fuel vaporization). As with the previous temperature category, the AI melt tests appear to provide an upper bound on the possible release amount of ruthenium in sodium. Therefore the release percentage of ruthenium is again estimated to be up to 5 % of the ruthenium inventory.

The ORNL melt tests in sodium found a cerium release of 0.004 % ~ 0.3 % when at 1,700 °C ~ 1,800 °C for less than a minute in air and in steam, but in an oxide crucible. Although the ORNL melt tests were performed in an oxide crucible, the short timeframe may have limited cerium oxide formation, meaning some insights may still be valid. It appears that cerium volatilization, even at this temperature range, is small. This is consistent with its very high elemental boiling point (3,443 °C). This likely implies that the release of cerium is dominated by the mechanisms discussed in the preceding temperature ranges: migration to the bond sodium, and liberation from melted cladding. Determining an appropriate cerium release percentage in this temperature range continues to be difficult. Volatilization is likely small, but that does not guarantee that the overall cerium release will be small. Therefore, a release percentage of up to 15 % is estimated.

The AI melt tests in sodium saw the fuel material reduced to very small fragments (on the order of 100

microns) with some fuel vaporization at temperatures up to 3,600 °C. Fuel temperatures in this range are highly unlikely for most SFR accidents. The AI tests demonstrate that even with very high fuel temperatures and some fuel vaporization, uranium and plutonium dissolution in sodium is small, as fuel remains in fragments. Therefore the 0.1 % release percentage is continued, with a dependence on fuel burnup level at the start of the accident.

The AI melt tests in sodium found a Zr/Nb release of 10 % ~ 24 % with complete fuel melting and dispersal (and some fuel vaporization). The AI melt tests offer valuable quantitative insight into lanthanide release in this temperature range. A release percentage of up to 30 % is estimated based on these findings. This release percentage is likely very conservative for many of the lanthanides, but without additional data, it is difficult to justify lower release fractions.

2.5 Release from the Primary Sodium to Cover Gas Space

ST in the release from the primary sodium to cover gas space is calculated. Table II shows the release fraction from the primary sodium to cover gas space using for this calculation.

Table II: Release Fraction from the Primary Sodium to Cover Gas Space

Radionuclide Groups	Release Fraction (Core to Primary Sodium)
Noble Gases	1
Halogens	$1.4 \cdot 10^{-3}$
Alkali Metals	$1.2 \cdot 10^{-2}$
Te Group	$9.0 \cdot 10^{-4}$
Ba, Sr	$9.0 \cdot 10^{-4}$
Noble Metals	$9.0 \cdot 10^{-4}$
Ce	$9.0 \cdot 10^{-4}$
Group U&Pu	$9.0 \cdot 10^{-4}$
Lanthanides	$9.0 \cdot 10^{-4}$

Figure 3 shows the temperature dependency of the sodium mass fraction in the cover gas space using equations and experimental materials [8].

3. Conclusions

The in-vessel STs of PGSFR are estimated using ANL methodology. The in-vessel STs are calculated through several phases: The inventory of each radionuclide is calculated by ORIGEN-2 code using the peak burnup conditions. The nominal value of the radiological inventory is multiplied by a factor of 1.2 as an uncertainty margin to give the radiological inventory.

ST in the release from the core to primary sodium is calculated by using the assumption of ANL methodology. Lastly, ST in the release from the primary sodium to cover gas space is calculated by using equation and experimental materials.

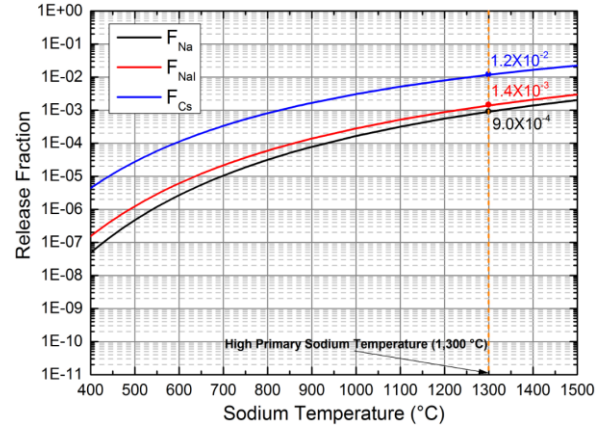


Fig. 3. Release fraction of sodium, NaI and Cs from coolant to cover gas.

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