Prediction of Changes in Mass for MELCOR Classes and Elements during Fission Product Cooling in Chlorine based Molten Salt Reactor

Eun Hyun Ryu[1,*] and Sung Il Kim[1]

[1]Korea Atomic Energy Research Institute, 1045 Daedeok-daero, Yuseong-Gu, Daejeon, 34057, Korea

**Corresponding author: ryueh@kaeri.re.kr*

1. Introduction

To evaluate radioactivity of the fission product which escaped from the reactor confinements is a crucial point in reactor safety analysis (or severe accident analysis). Considering recent regulation regarding the amount of specific isotope, Cs-137, mentioned evaluation is essential over all nuclear reactor related tasks such as safety analysis report (SAR), code development, safety equipment design and so on.

Before leaking to the environment including human, animal, soil and so on, the fission product will experience transportation through reactor component. They will sometimes be removed by water pool, filters for aerosol, sedimented by the gravitational force. Or they can sometimes be generated in the air again by the fluid flow such as wind and water flow or resuspension by heating of surface which the fission product attached by a specific phenomenon.

While the fission products went through a variety of phenomena of generation and extinction, a different physical process proceeded at the same time, which is the decay (or cooling) mechanism. In fact, the fission product is summation of initial fuel and produced various elements and isotopes appears in the periodic table because all of them can be transported inside of the reactor if circumstance is allowed.

Thus, to identify exact amount of fission product and predicting time dependent changes in fission product have significant meaning in severe accident analysis. For example, the MELCOR classes are categorized by the transportation characteristics of the fission products. This study aims to see the class-wise mass changes during accident progress are significant.

2. Initial Inventory Calculation

Initial invent ory of the fission product will change with many variables such as burnup, operating time, percent power, initial fuel configuration and composition, flux spectrum and so on. Although the pressurized water reactor (PWR) initial inventory is calculated a lot and its initial mass is almost known for reactors of subcategory of the PWR, the initial inventory of the Molten Salt Reactor (MSR) has never been discussed. Thus, the fission product of the MSR core will be dealt with its characteristics in this study. Because the Korean MSRs are based on the chlorine, it is different from other MSRs such as European style which adopts thermal spectrum with the salt based on the fluorine. The target reactor is same as general Korean style MSR. But because of project characteristic, the details regarding design dimension cannot be introduced in this study.

The shape of core is designed as cylinder, rough diameter and height is about 1 meter. The volume ratio between active core and inactive core is about 1:1. The enrichment is set as 20% as commercial limitation. Because the MSR reactor utilizes eutectic phenomenon of the salt, its melting points differs a lot with the composition of the salt. After various iterations from various points of view such as material corrosion, core volume minimization, heat transportation characteristic and so on, the salt is finally determined as KCl-UCl3. The mole fraction between compositions isn't also opened due to the project characteristic.

The OpenMC code is used to produce initial invent tory of the MSR core for selected 6 points of full power years from 0 years to 5 years. No decay calculation is conducted at this study. To reflect the MSR characteristic of flow, periodic mixing is performed at each calculation points. It was verified that sufficient low level of uncertainties are observed for both eigenvalue and flux for the OpenMC calculation.

The number of isotopes in calculation of the OpenMC code is about 1,100 as the McCARD code while the ORIGEN code is famous for its wide range of isotope which includes 1,600 isotopes for precise estimation. It was turned out that the number of isotope in this study is enough for the mass, radioactivity, and decay heat.

In addition to the mass calculation, radioactivity and decay heat are also calculated and should be evaluated. Based on the ANS standard recently issued [1], the in-house program is developed and verified [2]. This program will be utilized to analyze the decay and radioactivity trend after shut down in the future.

The MELCOR code is widely used in the severe accident analysis. To simulated fission product efficiently, the class division as shown in Table I is usually used in the fission product transportation. This division is based on the chemical characteristics of the fission product.

In Table I, the elements inside of parenthesis are minor elements in the aspect of mass, radioactivity and decay heat. Because the decay physics sometimes cause changes in proton and neutron number, the class changes will occur if this physics occurs frequently. In each element will have its own isotopes with various half-lives.

In this study, the isotope mass fraction of stable is calculated and arranged for major element without parenthesis. Also, some elements are added to target element list considering the composition of MSR reactor such as K, Cl, Sm, Cd, In and Pu as shown in Table II (colored as red in Table II). The initial mass information is shown in Table III.

Table I: MELCOR Class Division

Class Number and Name	Member Elements
1. Noble gases	Xe, Kr, (Rn), (He), (Ne), (Ar), (H), (N)
2. Alkali Metals	Cs, Rb, (Li), (Na), (K), (Fr), (Cu)
3. Alkaline Earths	Ba, Sr, (Be), (Mg), (Ca), (Ra), (Es), (Fm)
4. Halogens	I, Br, (F), (Cl), (At)
5. Chalcogens	Te, Se, (S), (O), (Po)
6. Platinoids	Ru, Pd, Rh, (Ni), (Re), (Os), (Ir), (Pt), (Au)
7. Transition Metals	Mo, Tc, Nb, (Fe), (Cr), (Mn), (V), (Co), (Ta), (W)
8. Tetravalents	Ce, Zr, (Th), Np, (Ti), (Hf), (Pa), (Pu), (C)
9. Trivalents	La, Pm, (Sm), Y, Pr, Nd, (Al), (Sc), (Ac), (Eu), (Gd), (Tb), (Dy), (Ho), (Er), (Tm), (Yb), (Lu), (Am), (Cm), (Bk), (Cf)
10. Uranium	U
11. More Volatile Main Group Metals	(Cd), (Hg), (Pb), (Zn), As, Sb, (Tl), (Bi)
12. Less Volatile Main Group Metals	Sn, Ag, (In), (Ga), (Ge)
13. Boron	(B), (Si), (P)
14. Water	(Wt)
15. Concrete	(Cc)

Table II: Class-wise Target Element Information

	# Element	# Target Element	Target Element List
1	8	2	Xe, Kr
2	7	3	Cs, Rb, K
3	8	2	Ba, Sr
4	5	3	I, Br, Cl
5	5	2	Te, Se
6	9	3	Ru, Pd, Rh
7	10	2	Mo, Tc
8	9	3	Ce, Zr, Pu
9	22	6	La, Pm, Sm, Y, Pr, Nd
10	1	1	U
11	8	3	Cd, As, Sb
12	5	3	Sn, Ag, In

Table III: Class-wise and Element-wise Initial Mass Inventory at 5EFPY

	Class Mass (kg)	Class Mass Frac. (%)		Mass (kg)	Frac. (%)
1	1.693	0.042	Xe	1.465	86.5
			Kr	0.158	9.3
2	323.3	8.101	Cs	1.290	0.4
			Rb	0.155	0.05
			K	321.9	99.6
3	0.898	0.022	Ba	0.497	55.4
			Sr	0.398	44.3
4	1376.7	34.489	I	0.050	0.004
			Br	0.008	0.001
			Cl	1376	99.99
5	0.167	0.004	Te	0.145	86.9
			Se	0.021	12.6
6	0.903	0.023	Ru	0.627	69.4
			Pd	0.112	12.4
			Rh	0.164	18.2
7	1.493	0.037	Mo	1.180	79.1
			Tc	0.304	20.4
8	5.713	0.143	Ce	0.944	16.5
			Zr	1.449	25.4
			Pu	3.276	57.3
9	2.817	0.071	La	0.446	15.8
			Pm	0.092	3.25
			Sm	0.222	7.89
			Y	0.210	7.46
			Pr	0.407	14.46
			Nd	1.418	50.34
10	2277.9	57.07	U	2277.9	100.0
11	0.008	0.000	Cd	0.005	60.2
			As	0.000	0.471
			Sb	0.003	39.31
12	0.014	0.000	Sn	0.010	71.32
			Ag	0.003	22.36
			In	0.001	5.4
	3991.69	100.0			

Table IV: Isotope-wise Initial Mass Inventory at 5EFPY from Class 1 to 5

Element	# Isotope	# Target Isotope		Mass (kg)	Frac. (%)
Xe	27	4	131	0.190	12.95
			132	0.294	20.06
			134	0.532	36.31
			136	0.447	30.53
1	Class 1		Summation		99.85
Kr	25	3	83	0.022	13.94
			84	0.041	26.10
			86	0.084	53.25
2	Class 1		Summation		93.29
Cs	25	3	133	0.446	34.59
			135	0.442	34.25
			137	0.400	31.05
3	Class 2		Summation		99.89
Rb	22	2	85	0.043	28.03
			87	0.111	71.96
4	Class 2		Summation		99.99
K	6	2	39	301	93.34
			41	21.4	6.64
5	Class 2		Summation		99.98
Ba	27	3	134	0.001	0.18
			137	0.023	4.71
			138	0.047	94.02
6	Class 3		Summation		98.91
Sr	24	2	88	0.151	37.82
			90	0.239	60.09
7	Class 3		Summation		97.91
I	27	2	127	0.010	20.09
			129	0.038	76.54
8	Class 4		Summation		96.63
Br	24	2	79	0.000	0.01
			81	0.008	99.96
9	Class 4		Summation		99.97
Cl	6	3	35	12.6	0.92
			36	0.027	0.00
			37	1360	99.08
10	Class 4		Summation		100.0
Te	30	6	122	0.000	0.00
			124	0.000	0.00
			125	0.001	0.65
			126	0.000	0.10
			128	0.025	15.56
			130	0.120	82.91
11	Class 5		Summation		99.23
Se	25	5	77	0.000	1.41
			78	0.001	3.82
			79	0.002	8.34
			80	0.005	23.68
			82	0.013	62.74
12	Class 5		Summation		100.0

Table V:Isotope-wise Initial Mass Inventory at 5EFPY from Class 6 to 9

Element	# Isotope	# Target Isotope		Mass (kg)	Frac. (%)
Ru	24	6	99	0.000	0.00
			100	0.002	0.25
			101	0.268	42.80
			102	0.230	36.74
			104	0.112	17.90
			106	0.009	1.45
13	Class 6		Summation		99.14
Pd	28	6	104	0.002	1.90
			105	0.065	58.04
			106	0.022	20.00
			107	0.014	12.72
			108	0.006	5.41
			110	0.002	1.93
14	Class 6		Summation		100.0
Rh	30	1	103	0.164	99.95
15	Class 6		Summation		99.95
Mo	23	5	95	0.282	23.92
			96	0.001	0.05
			97	0.292	24.70
			98	0.286	24.26
			99	0.319	27.01
16	Class 7		Summation		99.94
Tc	23	1	99	0.304	99.98
17	Class 7		Summation		99.98
Ce	22	3	140	0.432	45.82
			142	0.413	43.73
			144	0.000	9.28
18	Class 8		Summation		98.83
Zr	22	6	90	0.015	1.01
			91	0.248	17.10
			92	0.273	18.87
			93	0.292	20.14
			94	0.300	20.68
			96	0.306	21.12
19	Class 8		Summation		98.92
Pu	9	4	238	0.001	0.02
			239	3.22	98.38
			240	0.051	1.55
			241	0.002	0.06
20	Class 8		Summation		100.0
La	21	2	138	0.000	0.00
			139	0.445	99.85
21	Class 9		Summation		99.85
Pm	24	1	35	0.091	99.71
22	Class 9		Summation		99.71

Table VI: Isotope-wise Initial Mass Inventory at 5EFPY from Class 9 to 12

Element	# Isotope	# Target Isotope		Mass (kg)	Frac. (%)
Xe	27	4	131	0.190	12.95
			132	0.294	20.06
			134	0.532	36.31
			136	0.447	30.53
1	Class 1		Summation		99.85
Kr	25	3	83	0.022	13.94
			84	0.041	26.10
			86	0.084	53.25
2	Class 1		Summation		93.29
Cs	25	3	133	0.446	34.59
			135	0.442	34.25
			137	0.400	31.05
3	Class 2		Summation		99.89
Rb	22	2	85	0.043	28.03
			87	0.111	71.96
4	Class 2		Summation		99.99
K	6	2	39	301	93.34
			41	21.4	6.64
5	Class 2		Summation		99.98
Ba	27	3	134	0.001	0.18
			137	0.023	4.71
			138	0.047	94.02
6	Class 3		Summation		98.91
Sr	24	2	88	0.151	37.82
			90	0.239	60.09
7	Class 3		Summation		97.91
I	27	2	127	0.010	20.09
			129	0.038	76.54
8	Class 4		Summation		96.63
Br	24	2	79	0.000	0.01
			81	0.008	99.96
9	Class 4		Summation		99.97
Cl	6	3	35	12.6	0.92
			36	0.027	0.00
			37	1360	99.08
10	Class 4		Summation		100.0
Te	30	6	122	0.000	0.00
			124	0.000	0.00
			125	0.001	0.65
			126	0.000	0.10
			128	0.025	15.56
			130	0.120	82.91
11	Class 5		Summation		99.23
Se	25	5	77	0.000	1.41
			78	0.001	3.82
			79	0.002	8.34
			80	0.005	23.68
			82	0.013	62.74
12	Class 5		Summation		100.0

Table VII: Isotope-wise Initial Mass Inventory at 5EFPY of Class 12

Element	# Isotope	# Target Isotope		Mass (kg)	Frac. (%)
Ag	34	2	147	0.003	99.70
			148	0.000	0.03
32	Class 12		Summation		99.73
In	47	2	113	0.000	0.16
			115	0.001	99.81
33	Class 12		Summation		99.97

As shown in Table III, several elements are added to the target element list considering the MSR composition and fission product mass. Naturally, the K, Cl increases a lot compared with those of PWR while class 9 fraction decreases a lot compared with that of the PWR. For the class 11 and 12, the mass is too low compared with those of the other classes. Thus, small amount of class to class mass transportation to class 11 and 12 will cause a great change in mass of class 11 and 12. Among various isotopes of a certain element, stable (half-life is zero) or isotopes of half-life of more than 107 second are extracted to find out mass fraction of those isotopes as shown in Table IV~Table VII. The reason for the 107 second is from usual sever accident simulation time. In general, 7 days are maximum simulation time. Thus 604,800 seconds, namely, 6.048E5 is the simulation time. Thus isotope of half-life of over 107 second will almost same for mass during severe accident simulation. In Table IV~VII, under line for a certain isotope means that this is meta stable isotope. As shown in Table IV~VII, all classes have at least over 96% mass for stable isotopes including isotope of half-life of over 107 seconds. In this regard, it can be predictable that the mass for each class will not change at all during severe accident simulation even for the MSR type reactor. However, extremely low mass class such as 5, 11 and 12 can change a lot for fraction due to decay chain. This can be verified after decay calculation of the OpenMC code and it will be conducted in the near future.

4. Conclusions

Throughout this study, we found that the mass for each class will not change at all during general severe accident simulation for the MSR reactor by extracting initial fission product mass using the OpenMC code.

In the future, low mass class such as 5, 11 and 12 will be verified as well by the OpenMC decay calculation for each Effective Full Power Year (EFPY).

Not only mass but also radioactivity and decay heat will be investigated for initial amount along with its change depend on cooling time for the MSR reactor.

5. Acknowledgement

This work was supported by Korea Research Institute for defense Technology planning and advancement (KRIT) grant funded by the Korea government (DAPA(Defense Acquisition Program Administration)) (KRIT-CT-22-017, Next Generation Multi-Purpose High Power Generation Technology (Liquid Fuel Heat Generator Transportation and Safety Assessment Technology), 2022)

References

[1] American Nuclear Society, Decay Heat Power in Light Water Reactors, ANSI/ANS-5.1-2014, 2014.

[2] E. H. Ryu and Sung Il Kim, "Decay Heat Calculation Program Development adopting state-of-the-art Standard, ANS-2014", KNS Spring Meeting, Jeju, Korea, May 9-10, 2024.

[3] E. H. Ryu, H. S. Jeong and D. H. Kim "Comparison of the Results of the Whole Core Decay Power using the ORIGEN Code and ANS-1979 for the Uljin Unit 6", KNS Spring Meeting, Jeju, Korea, May 7-8, 2015.

Keywords

Fission Product, Mass, MELCOR Classes, Elements, Cooling