FLUSHING WATER CONTAMINATION IN DIFFERENT CONCRETE MODELS FOR PSA LEVEL 3 SEVERE ACCIDENT SCENARIO

Oyeon Kum[1] and Deokjung Lee[2]

[1] *University of Southwest America: 2954 W 8th ST STE 101, Los Angeles, CA 90005, and okum@uswa.ac*
[2] *Ulsan National Institute of Science and Technology: 50 UNIST-gil, Ulsan, 44919, and deokjung@unist.ac.kr*

Concrete is the most common material for radiation shielding in nuclear reactors and radiation facilities, and for the prevention of radiation leakage from radioactive sources because it is widely used for structures, floor slabs, and cladding systems. Compositions of commercial concrete consisting of cement, sand, aggregate of stones and gravels etc., and water, vary from batch to batch, resulting in large variations in physical and chemical properties even within the same facility. Many different concrete models are known and published for Monte Carlo simulations. It is worthwhile to evaluate the differences in their properties in terms of nuclear safety, specifically, for PSA-level 3 accidents. In this study, we chose three different concretes, "ORNL", "Samseong", and "ANSI" concretes, to calculate contamination when a tsunami/flood flushes out of an activated part of a concrete used in heavy-ion therapy accelerator facility after 50 years of operation and one year cool-down time. For conservative estimation, isotopic trace elements such as Eu-151, Eu-153, Co-59, and Cs-133 are included in the chemical composition of the concrete, in addition to the main ingredients. The combination of MCNPX2.7.0 and CINDER'90, one of the best activation simulation tools in the world, is used. To ensure a conservative estimate of concrete activation, one hour of accidental maximum beam loss (1×10^9 pps) after 50 years of normal operation (7.2×10^{15} particles/year) was assumed as the beam parameters. After a decay time of one year, the sums of values of all isotopes, Σ(specific activity/permitted limit), are 0.1, 0.3, and 0.1 for "ANSI", "ORNL", and "Samseong", respectively, which ensures that the concrete waste can be recycled with no radiological precautions because the sums are all below 1.0. However, when tsunami/flood flushes out the activated part of the concrete and resolves the radioactive isotopes into water in full, the waste water is contaminated severely. Many radioactive isotopes with long half-lives such as K-40, Co-60, Cs-134, Eu-152, and Eu-154 exceed the unrestricted release limit values. Therefore, the contaminated water by tsunami/flood must not be discharged immediately onto the environment under this severe accident scenario (PSA-level 3) even after one year of decay time.

I. INTRODUCTION

The recent Fukushima nuclear power plant accident gives us a clear lesson that preparation against catastrophic accidents (natural disasters) such as a tsunami, earthquake, fire, plane crash, bomb attack, and flood is pivotal, although their occurrences may be rare. Thus, it is desirable to build safety management systems to prepare for PSA-level 3 incidents by simulating the effect of radiation exposure of the surrounding population and environment.

Concrete is the most common material for radiation shielding in nuclear reactors and radiation facilities [1] and for the prevention of radiation leakage from radioactive sources because it is widely used for structures, floor slabs, and cladding systems. Compositions of commercial concrete, consisting of cement, sand, aggregate of stones and gravels *etc*., and water, vary from batch to batch, resulting in large variations in physical and chemical properties even within the same facility. Thus, concrete material data for shielding and activation simulations in general uses a typical model developed for explaining various sites material properties. Many different concrete models are known and published for Monte Carlo simulations. It is interesting to evaluate the differences in their properties in perspective of nuclear safety, specifically, for PSA-level 3 accidents.

In this study, we chose three different concretes, "ORNL", "ANSI", [2] and "Samseong" [3], to calculate flushing water contamination when a tsunami/flood flushes out of the most activated part of the concrete used in heavy-ion therapy accelerator facility, with an operation time of 50 years and cool-down time of one year. For a conservative estimation, isotopic trace elements such as Eu-151, Eu-153, Co-59, and Cs-133 are included in the composition of the concrete, in addition to the main ingredients. The reason why we chose three model concretes in this study is because the real expected

activity of the concrete is highly dependent on the actual chemical composition of the concrete used, which is not known at the moment, and therefore this estimate can only be seen as a first guideline for the expected activation of the concrete.

The combination of MCNPX2.7.0 [4] and CINDER'90 [5], one of the best activation simulation tools in the world, is used. The Activation Script reads most of the problem information such as cell properties, material compositions, and neutron fluxes, which are printed in the MCNPX output file. With an input file, CINDER'90 code produces nuclear inventory for a requested list of MCNPX cells for a requested time history. A second script extracts the decay photon sources from the CINDER'90 output for a requested list of cells and for a requested irradiation or decay time step, which then builds the source deck for subsequent MCNPX calculation.

Simulations of generic concrete radioactivity were performed by using simplified geometry models with an air-filled cylindrical chamber which mimics the most activated part of the facility. To ensure a conservative estimate of the concrete activation, one hour of accidental maximum beam loss (1 x 10^9 pps) after 50 years of normal operation (7.2 x 10^{15} particles/year) was assumed as the beam parameters. The inclusion of the one hour maximum intensity beam loss can explain the significant contributions of the radioactive isotopes with short half-lives.

II. METHODS AND MATERIALS

II.A. Geometry Model of Concrete and Beam Irradiation Scenarios

For the beam operation of the accelerator facility, radioactive isotopes are generated in the vicinity of beam loss points or intentional beam landing points. Actually, the high energy accelerator facility can keep unintentional radioactive isotopes generated from the concretes and other materials used in the accelerator even when the power is off. Thus, the estimation of the radioactivity production in the concrete and other materials is important. For a conservative estimation of the radioactivity of the concrete walls, simplified symmetric cylindrical (see Figure 1) geometry is used. To obtain sufficient statistics, isotope production in a reasonable time was carried out with the Monte Carlo MCNPX code for particle transport and CINDER'90 code for activation.

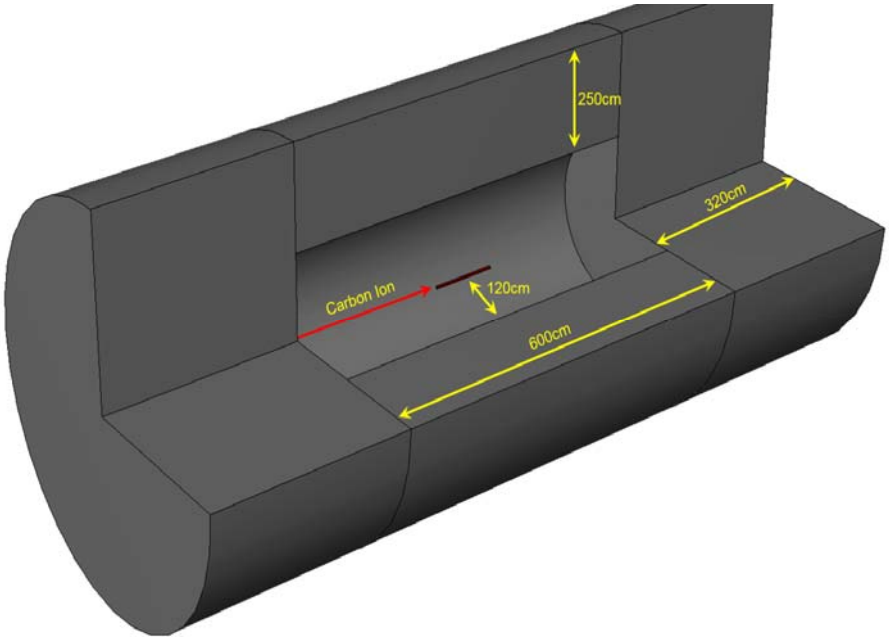

Fig. 1. A simplified cylindrically symmetric concrete geometry. The simulation was used to simulate the activation of the concrete in the middle part. In the center of the geometry, cylindrical iron and copper targets (diameter 5 cm, length 100 cm), which are the major components consisting of the accelerator, are bombarded with high energy carbon ions.

The arrangement consists of an iron/copper target having a length of 100 cm and a diameter of 5 cm, whose top surface is irradiated by a high energy carbon ion beam with energy of 430MeV/u. The use of these target materials ensure that they are the most popular materials used in the design of high-energy accelerator components. Substitutive components and high lateral secondary production is guaranteed, which makes a conservative assumption true for real beam losses. This target is laterally surrounded of 1.2 m of air, which is the minimum distance between the beam line and the building foundation.

Behind it, a concrete cylinder with a lateral thickness of 2.5 m represents the foundation or the closest walls of the facility. The secondary radiation must pass through the thickness of the foundation before it reaches the ground or any existing underground water layers. In this study, we did not simulate underground earth or the water layer, which was studied separately. The region is longitudinally divided into three areas as shown in Figure 1. For a conservative estimation, the concrete isotope production in the middle part (total length of 6.0 m) is calculated only, for the given irradiation scenarios.

As mentioned previously, to ensure a conservative estimate of the activation in the concrete, an operating period of 50 years was used. In a year, the average beam irradiation intensity is 1.142×10^8 particles per second (pps), which translates to a total of 7.2×10^{15} carbon ions with an energy of 430 MeV/u bombarding the top of the target. In addition to long-term activation, short-term activations may also be relevant during the mentioned operation of 50 years, which can be expanded to an operational cycle of one hour at the maximum achievable intensity (1×10^9 pps). The inclusion of this maximum attainable intensity is mainly considered for the concrete simulation to cover a relatively short activation time. It should again be noted that these simulations are to be regarded as extremely conservative, so as to prove the safety of this plant for both the environment and people. Several scenarios of decay times were simulated between one minute and one year after the irradiation.

Compositions of commercial concrete, consisting of cement, sand, aggregate of stones and gravels *etc.*, and water, vary from batch to batch, resulting in large variations in physical and chemical properties even at the same site. A precise specification of these three types of concrete was developed by the RADIATION PORTAL MONITOR PROJECT, "Compendium of Material Composition Data for Radiation Transport Modeling", PIET-43741-TM-963 PNNL-15870 Rev. 1. The chemical composition for each concrete model is given in Table I. The fractional units are shown as the same as those of simulation with MCNPX code which automatically converts the same units.

Table I. Chemical compositions of three different concretes published in PIET-43741-TM-963 PNNL-15870 Rev. 1.

Element	ANS/ANSI 6.6.1	ANS/ANSI + Trace	ORNL + Trace	Samseung + Trace
	Atomic fraction	Atomic fraction	Weight fraction	Weight fraction
	Density: 2.3 g/cm3	Density: 2.3 g/cm3	Density: 2.3 g/cm3	Density: 2.3 g/cm3
H	7.86	7.86	0.006187	0.76433
Li	-	-	-	0.2
C	-	-	0.175193	-
O	43.8	43.8	0.400184	49.224
Na	1.05	1.05	0.000271	1.6985
Mg	0.14	0.14	0.032649	0.25478
Al	2.39	2.39	0.010830	4.5435
Si	15.8	15.8	0.034479	31.3075
S	-	-	-	0.12739
K	0.69	0.69	0.001138	1.9108
Ca	2.92	2.92	0.320287	8.3287
Fe	0.31	0.31	0.007784	1.2314
Co	-	0.1	0.01	0.1
Ni	-	-	-	0.2
Cs	-	0.1	0.01	0.1
Eu-153	-	0.055	0.000055	0.0055
Eu-151	-	0.045	0.000045	0.0045

The concrete with trace elements including the main components (used for the simulation) can provide a significant contribution to concrete activation. The relevant isotopes, which can be either converted by neutron capture in a radioactive isotope or simply present in a sufficient quantity, are Co-59, Cs-133, Eu-151 and Eu-153. In Figure 2, the neutron captures of these four isotopes are shown as a function of neutron energy. The influence of these isotopes on the specific activation was examined by means of a Monte Carlo calculation for the carbon ion with energy of 430MeV/u. In this calculation, the concretes were added to 1% cobalt, 1% cesium and 0.01% natural europium. Because the exact data for the ratio and type of each trace element in a concrete were not available, the real trace elements must be resorted to literature values and assumptions. The ratio of these admixtures to one another corresponds approximately to the ratio of the low-energy neutron cross sections of these elements, as shown in Figure 2. This procedure was chosen in order to obtain all isotopes with similar statistical uncertainty in a single simulation.

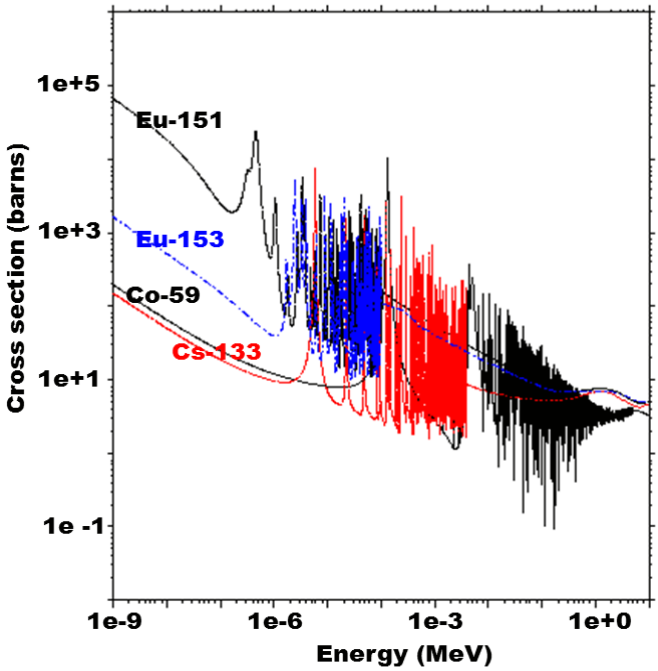

Fig. 2. Neutron capture isotopes: Co-59, Cs-134, Eu-151 and Eu-153.

II.B. Governing Equation for Nuclear Transportation Model

Nuclear transmutation (a conversion of one chemical element or isotope into another) is caused by a process that occurs either through nuclear reactions (in which an outside particle reacts with a nucleus), or through radioactive decay (where no outside particle is needed), although not all nuclear reactions or radioactive decay cause transmutation. Specifically, artificial nuclear transmutation may occur in machineries (i.e. accelerators or fission/fusion reactors) that have enough energy to cause changes in the nuclear structure of the elements. Although the terminology transmutation is rooted back to alchemy, which transforms the base metals into gold in the middle age, Rutherford and Soddy were the first observers by discovering the natural transmutation as a part of radioactive decay of the alpha decay type in the early 20th century. Along with the development of computing technology and analysis software, the CINDER code was developed for rigorous atomic transmutation study.

Brief summary of the transmutation equation is described here. The governing equation describing the rate of change in the atom density, $N_m(t)$ of a nuclide *m* is the sum of the loss and gain rates in the nuclide density. The loss rate is due to two mechanisms: (1) the radioactive-decay of nuclide *m* producing daughter nuclides and (2) particle-absorption reactions of nuclide *m* producing residual nuclides different from *m*. The gain rate is the rate of other nuclides in the system that becomes nuclide *m* as a daughter/residual and particle production/depletion by spallation due to high energy reaction.

$$dN_m(t)/dt = -N_m(t)\beta_m + \bar{Y}_m + \sum_{k \neq m} N_k(t)\gamma_{k \to m}, \qquad (1)$$

where $\gamma_{k \to m}$ is the probability of nuclide *k* decaying or absorbing to nuclide *m* and β_m is the total transmutation probability of nuclide *m* defined by,

$$\beta_m = \lambda^m + \phi\sigma_a^m. \qquad (2)$$

Here ϕ is the energy integrated neutron flux, σ_a^m is the flux weighted average cross section for neutron absorption by nuclide *m*, and λ^m is the total decay constant of nuclide *m*. Absorption reactions of σ_a^m are all of those with residual nuclides other than nuclide *m* and thus include only inelastic scattering to states other than that of nuclide *m*. $\bar{Y}_m$ is an

additional constant corresponding to significant particle production (or depletion) due to neutrons with high energies above thresholds given to the CINDER library (>25 MeV). The underlying assumption of this governing equation is that the transmutation probabilities β_m and $\gamma_{k\to m}$, and thus the flux ϕ are constant for the time interval for which the solution is desired. In usual activation simulation code, any temporal history can be approximated using a histogram of constant-flux intervals. A few different solution methods can be applicable: direct integration, matrix diagonalization, and reduction to a set of independent, linear differential equations using the Markov method resulting in an analytical solution *etc.*.

There are many known activation simulation tools such as FLUKA2011, MCNPX+CINDER, PHITS+DECHAIN, MCNPX+FISPACT, *etc.*. In the aspect of availability, FLUKA2011 and MCNPX+CINDER Monte Carlo codes are used in this study. For the FLUKA2011 code, results for production of residuals, their time evolution and residual doses due to their decays, can all be obtained in the same run for an arbitrary number of decay times with a given irradiation profile. Although FLUKA2011 is freely available, it is an inconvenient code for general use since the source code of the core algorithm is not publically released. It is so called an "all-in-one" code. On the other hand, the combination of MCNPX and CINDER can be used with confidence because all source codes and data libraries are available.

In order to make use of the CINDER particle transmutation code, accurate knowledge of the average flux in the material that is to be transmuted is necessary. Thus, a transport code must be plugged into the front end of the calculation to estimate the neutron distributions in the medium. As the transport wrapper, MCNPX code is used with CINDER. At present, the connection uses ancillary scripts that provide an interface into the output files of the multi-particle transport code MCNPX. The inter-relation of the scripts and the CINDER and MCNPX codes is outlined in Fig. 3. One script, the ACTIVATION script, drives activation calculations of multi-region problems preparing for CINDER input files and executing CINDER with minimal user input. The other script, the GAMMASOURCE script, prepares decay gamma source decks that can be directly fed back into MCNPX calculation of gamma-ray radiation arising from activation products. Because of the separate modular structures and the use of additional script codes, CINDER is hard to use and may provide inaccurate answers due to the lack of sufficient significant digits by the human interrogation of the process.

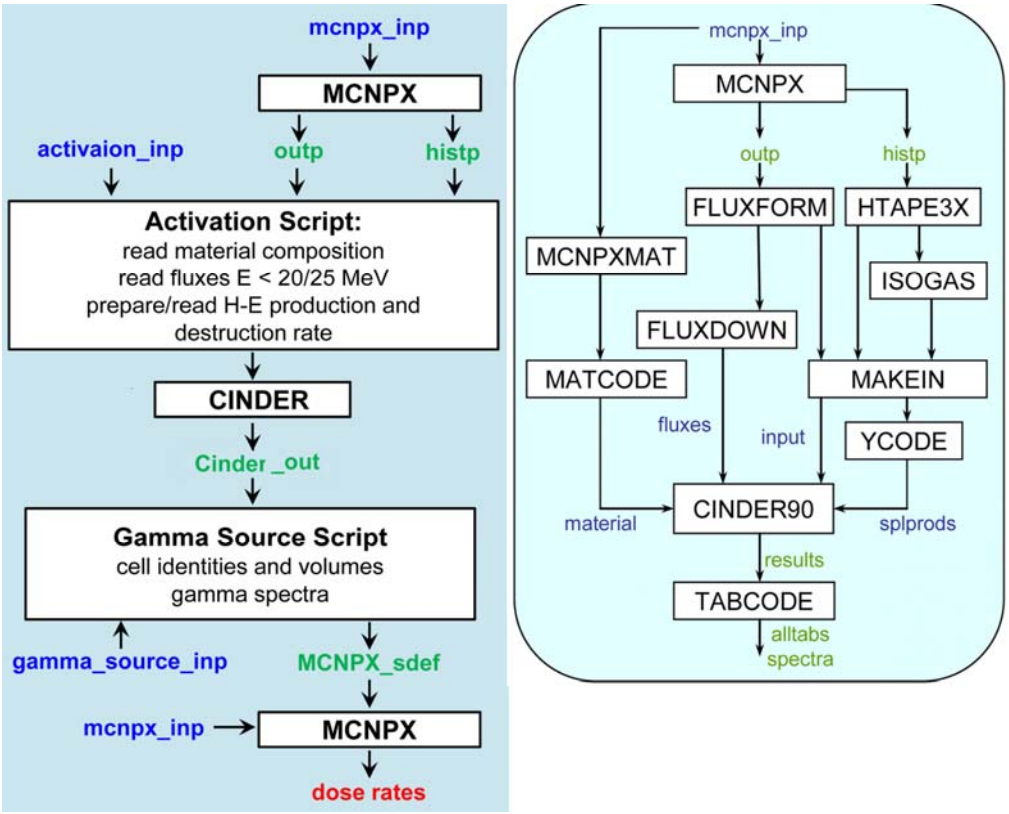

Fig. 3. Transmutation computation flow with two scripts. Right side shows the details of output and "histp" files. CINDER also uses the results of the two scripts for CINDER.

III. RESULTS AND DISCUSSION

The simulations were carried out for the concrete activation, whose location is 1.2 meters below the beam loss spot. As for the irradiation scenario mentioned earlier, an annual beam loss of 7.2 $x10^{15}$ carbon ions was used on the iron/copper rod. This irradiation extends over 50 years and is terminated with a subsequent beam loss of 1 x 10^9 pps at 430MeV/u over an hour. Only the high-energy carbon ions operating at 430MeV/u was used for a conservative estimation (i.e., the additional low energy operating modes were neglected for the estimation). The six evaluated decay times after the irradiation scenario extends from one minute to one year.

The specific activities are also compared in these tables with limits that are acceptable for unrestricted release according to the General Radiation Protection Ordinance. Table II shows the sum of the values of all isotopes Σ (specific activities / permitted limit) obtained as a function of the decay times in the middle part of a concrete cylinder, which extends over a range of ± 3.0 m at the beam spot (see Figure 1).

Table II. The sum of values of all isotopes Σ (specific activities / permitted limit) obtained as a function of the decay times in the middle part of a concrete cylinder, which extends over a range of ± 3.0 m at the beam spot (see Figure 1).

		ANSI (No)		ORNL(with)		Samseung(with)		ANSI(with)	
up time	decay	Cu	Fe	Cu	Fe	Cu	Fe	Cu	Fe
50 year +1 hour irradiation	Just after up time	1.18E-02	1.20E-02	3.76E-01	3.35E-01	1.49E-01	1.35E-01	1.54.E-01	1.38.E-01
	1 minute	1.11E-02	1.13E-02	3.73E-01	3.32E-01	1.46E-01	1.32E-01	1.53.E-01	1.38.E-01
	1 hour	7.41E-03	7.46E-03	3.46E-01	3.04E-01	1.33E-01	1.19E-01	1.49.E-01	1.33.E-01
	1 day	6.44E-03	6.45E-03	3.39E-01	2.97E-01	1.27E-01	1.12E-01	1.45.E-01	1.29.E-01
	1 week	6.40E-03	6.42E-03	3.37E-01	2.96E-01	1.26E-01	1.11E-01	1.44.E-01	1.28.E-01
	1 month	6.39E-03	6.40E-03	3.32E-01	2.91E-01	1.25E-01	1.11E-01	1.43.E-01	1.27.E-01
	1 year	6.29E-03	6.30E-03	2.77E-01	2.43E-01	1.13E-01	9.99E-02	1.31.E-01	1.17.E-01

It is noted that the value for a cool-down time of one minute is below 1.0, which means that our unrestricted release limits for all isotopes are generally very high according to our General Radiation Protection Ordinance compared with other countries such as Europe and America. When an Austrian regulation was applied, the values for a decay time of one week and one month are just over the limit of 1.0. The authors suggest a re-evaluation of our limiting values in the General Radiation Protection Ordinance, specifically, a limit value of H-3(1.0 x 10^6 vs 6.0 x 10^4). Actually, the values in Table II were averaged over a 6.0 m long concrete cylinder jacket which lies in the immediate vicinity of the maximum loss point that may have a significantly higher radioactivity than the average value of the entire concrete. The activation of the concrete also depends, among other things, of the self-shielding of the irradiated components (local shielding). This self-shielding was not considered in these calculations, as only the exposed part of the copper/iron conductor and not the entire assembled component was simulated. For a first order approximation of the long-standing concrete activation for other loss points, the linear extrapolation can be used with the result above for the rate of beam loss.

Because the sum of the values of all isotopes Σ (specific activities / permitted limit) obtained as a function of the decay times for the contaminated materials (radioactive waste) are in the safe range, we evaluate the differences in their properties in terms of nuclear safety by modeling a PSA-level 3 accident of a tsunami/flood flushing out the most radioactive part of the concrete used in this study.

Table III shows the ratio of specific activity of the concrete at 1.2 meters below the iron cylinder target. The table compares the specific activities for the cool-down time of one year to the values for unrestricted release (General Radiation Protection Ordinance). The comparison with the shared values are only performed for isotopes with a ratio more than 0.0001. Empty space has a value of 0.0000. The result suggests that we should be careful to discharge the contaminated water into the environment if a tsunami/flood flushes out of the most activated part of the concrete even after one year of cool-down time. The major contribution of the expected radioactivity can be attributed to K-40 ($T_{1/2}$ = 1.251 x 10^9 years), Co-60 ($T_{1/2}$ = 5.2714 years), Cs-134 ($T_{1/2}$ = 2.0652 years), Eu-152 ($T_{1/2}$ = 13.516 years), and Eu-154 ($T_{1/2}$ = 8.593 years).

The actual radioactive activity of the concrete highly depends on the chemical composition of the concrete used. For the ANSI concrete without trace elements, K-40 is the main element, but Cs-134 is the main element if trace elements are included. Thus, the chemical composition of the concrete seems to be sensitive to the radioactivity. For the ORNL concrete, the main element is Cs-134, but K-40 for the Samseong concrete. Letting the isotopes decay for several more years is recommended for unrestricted discharge of the waste water in contact with the concrete. However, the exact composition of the concrete is not known at the moment of construction and, therefore, this estimation can only be seen as a first guideline for the expected activation of the concrete. The radiological classification of the concrete must be carried out by measurement. The waste water with resolved radioactive materials from waste concrete must be discharged in accordance with the Radiation Protection Act.

Table III. Ratio of specific activity of the concrete at 1.2 meter below the iron cylinder target. The table compares the specific activities for the cool-down of one year to the values for unrestricted release (General Radiation Protection Ordinance). The comparison with the share value is only performed for isotopes with a ratio each more than 0.0001. Empty space has a value of 0.0000.

Nuclide	Specific Activity (Bq/m^3)				Release Limit (Bq/m^3)	Specific Activity/Release Limit			
	Ansi (Wo)	Ansi(W)	ORNL(W)	Sams(W)		Ansi(Wo)	Ansi(W)	ORNL(W)	Sams(W)
H 3	74296	73926	213786	251711	2.0E+07	0.0037	0.0037	0.0107	0.0126
C 14	24.938	25.0601	64.417	80.031	1.0E+06			0.0001	0.0001
Na 22	7818.1	7729.3	12827.9	26451.3	2.0E+05	0.0319	0.0386	0.0641	0.1323
K 40	1369740	80401	1391940	1391940	1.0E+05	13.6974	0.8040	13.9194	13.9194
Ti 44	5.7868	6.6563	15.1145	20.3278	1.0E+05	0.0001	0.0001	0.0002	0.0002
Mn 54	108.262	211.936	2497.5	2876.75	1.0E+06	0.0001	0.0002	0.0025	0.0029
Fe 55	312.539	520.59	7144.7	9568.2	2.0E+06	0.0002	0.0003	0.0036	0.0048
Co 56		4.1958	36.7965	39.257	3.0E+05			0.0001	0.0001
Co 57		287.453	2275.5	1416.73	3.0E+06		0.0001	0.0008	0.0005
Co 58		55.87	452.51	131.979	9.0E+05		0.0001	0.0005	0.0001
Co 60		82880	2890810	494320	2.0E+05		0.4144	14.4541	2.4716
Ni 63				4151.4	5.0E+06				0.0008
I 125		4.5991	17.3345	1.57139	5.0E+04		0.0001	0.0003	
Ba 133		31.3242	56.388	5.0505	7.0E+05			0.0001	
Cs 134		271506	1905500	264106	4.0E+04		6.7877	47.6375	6.6027
Pm 144		45.251	1.01565	1.52366	7.0E+05		0.0001		
Pm 145		509.12	19.7099	21.4341	6.0E+06		0.0001		
Sm 145		232.878	9.028	9.9937	3.0E+06		0.0001		
Pm 146		79.994	2.6899	2.24183	8.0E+05		0.0001		
Gd 148		10.8743			1.0E+04		0.0011		
Eu 152		1501830	694490	1268360	5.0E+05		3.0037	1.3890	2.5367
Eu 154		690420	65416	102601	3.0E+05		2.3014	0.2181	0.3420

IV. CONCLUSIONS

The total values of all isotopes Σ (specific activities/permitted limit) obtained as a function of the decay times for the contaminated concrete (radioactive waste) after 50 years operation were in the safe range just after the end of operation, which suggests re-evaluation of unrestricted release limit values. In addition, we evaluated the differences in their properties in terms of nuclear safety by modeling a PSA-level 3 tsunami/flood accident when the flood flushes out from the most radioactive part of a cylindrical concrete. We chose three different concretes, “ORNL”, “Samseong”, and “ANSI”, to calculate the contamination of a concrete used as a heavy-ion therapy accelerator for 50 years operation and one year cool-down time. Isotopic trace elements such as Eu-151, Eu-153, Co-59, and Cs-133 are included in the composition of the concrete for a conservative estimation. The combination of MCNPX2.7.0 and CINDER’90 is used to simulate concrete activation with an assumption of one hour of accidental maximum beam loss (1×10^9 pps) after 50 years of normal operation (7.2×10^{15} particles/year) as the beam parameters.

When a tsunami/flood flushes out the activated part of the concrete and resolves the radioactive isotopes into the water in full, the waste water is contaminated severely. Many radioactive isotopes with long half-lives such as K-40, Co-60, Cs-134, Eu-152, and Eu-154 exceed the unrestricted release limit values. Therefore, the contaminated water must not be discharged immediately into the environment even after one year of decay time. The actual radioactive activity of the concrete highly depends on the chemical composition of the concrete used. For the ANSI concrete without trace elements, the K-40 is the

main radioactive element, but the Cs-134 becomes the main element if trace elements are considered. Specifically, with trace elements, the K-40 contribution reduced below 1.0 for the ANSI concrete. Thus, the chemical composition of the concrete seems to be sensitive to the radioactivity. The main radioactive element for the ORNL and Samseong concrete is found to be Cs-134 and K-40, respectively. Waiting for several more years allowing further decay can therefore decrease the radioactivity of the concrete below the limit provided for unrestricted discharge of the contaminated water. However, the exact composition of concrete is not known at the moment of construction and, therefore, this estimation can only be seen as a first guideline for the expected activation of the concrete. The radiological classification of the concrete must be carried out by measurement. The waste water with resolved radioactive materials from the radioactive concrete must be released in accordance with the Radiation Protection Act.

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