DETERMINATION OF SOME PRINCIPAL RADIONUCLIDES OF POTENTIAL CONCERN FOR DISPOSAL OF LILW

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ABSTRACT

The technical requirements for the land disposal of low- and intermediate- level radioactive wastes, as specified in the Korean law of radioactive waste disposal, necessitate that the classification and characterization of wastes should be performed before their disposal. The waste classification and characterization involve the quantification of radionuclides in the wastes and compliance with their disposal requirements. The determination of radionuclides to quantify the radioactivity of the wastes needs the inventory of radionuclides in the wastes and the evaluation of potential radiation exposure from the disposal of the wastes. The evaluation of potential radiation exposure from waste disposal requires the specification of the location and characteristics of a disposal site. Korea has not yet determined the type and location of disposal site and nevertheless radioactive wastes have been continuously produced from nuclear power plants without classification and characterization. So it is necessary to determine radionuclides to be quantified for disposal of waste as soon as possible. In this study some principal radionuclides of potential concern present in the wastes has been determined for their land disposal and the general exposure scenarios from their land disposal. To do these, radionuclide inventory was obtained from the measured data of reactor coolant, licensed data of final safety analysis report and activated nuclides of plant structure calculated using ORIGEN computer code. The radiation exposure of each radionuclide was estimated and ranked in order from high to low dose radionuclide. From the results of radiation exposure calculation, principal radionuclides occupying most of total radiation exposure from the disposal of LILW were found to be ^{137}Cs, ^{99}Tc, ^{239}Pu, ^{129}I and ^{63}Ni. Dose exposure evaluated by these nuclides would occupy more than 99% of total dose due to the disposal of operation waste or decommissioning waste. But the nuclides used in this study as the reactor coolant inventory were only measurable gamma-ray-radiative nuclides. So there would be some other important nuclides much contributing to the total dose exposure such as ^{3}H, ^{14}C, radioactive inert gas, TRU and beta decay nuclides. The Korea Hydro & Nuclear Power Company temporarily has selected 21 radionuclides as the principal radonuclides for the determination of the total radiation exposure. Finally selected principal radionuclides within the LILW would be determined through the further consecutive study.

INTRODUCTION

Korean law for radiation safety on the disposal of Low- and Intermediate- Level radioactive Waste (LILW) states that specific and total radioactivity of wastes shall not exceed a critical limit [1]. But the radionuclides to be analyzed and their critical limits have not yet determined from related laws. But the specific and total radioactivity of any wastes to be disposed of shall be analyzed before disposal.

Radioactive waste shall meet safety standard for waste disposal in consideration of environmental effect from release of radioactive material out of the waste. For achieving this goal, operator shall 1) classify the wastes with its type and radioactivity, 2) determine the radioactive concentration and total radioactivity of the wastes and 3) estimate the potential radiation exposure from the disposal of the wastes. The radiation exposure depends on the type and activity of the wastes and the location and characteristic of a disposal site. Korea has not yet determined the disposal site, but radioactive wastes have continuously produced from Nuclear Power Plant (NPP) and they do not have sufficient radiation information for their disposal. Hence, it is urgent to determine radionuclides within the LILW to be quantified for disposal and to know the radioactivity of the nuclides in the LILW.

The LILW contains so many radionuclides that the way of analyzing activities of all nuclides present within the LILW may not be a good idea because of the consumption of time and cost. It has been known

that most of radiation exposure from a disposal site is due to a few principal radionuclides [2]. So the determination of a few principal radionuclides giving most of radiation exposure from disposal site and the evaluation of their radiation exposure may be sufficient to meet criteria of LILW disposal.

In this study radionuclides quantifying the radioactivity of the LILW for disposal and giving 99 percent of total radiation exposure were determined by considering the general disposal site and radiation exposure scenarios. The radioactivity inventory was obtained from measured and derived data reported in the Final Safety Analysis Report (FSAR) on the radioactivity of reactor coolant and from the activated radioactivity of plant structure calculated by ORIGEN computer code during operation.

The radiation exposure of each radionuclide from waste disposal was estimated from the DandD computer code [3]. The radiation exposures were ranked in order of from high to low radiation exposure to determine principal radionuclides. From the ranked radiation exposures of the radionuclides the principal radionuclides were selected to give up to 99 percent of total radiation exposure.

RADIATION EXPOSURE FROM LILW DISPOSAL

Inventory of radioactive waste:

The waste to be disposed in a disposal site would be classified into two groups: (a) operating waste produced during power plant operation such as spent resin, spent filter, contaminated clothes and (b) decommissioning waste or building structure having been activated during operation and decommissioned after shutdown such as reactor vessel and containment building.

The operating waste was originated from reactor coolant. The estimation of radiation exposure from waste disposal was carried out using the data obtained from the NPP, Kori unit 1 in Korea. The measured radioactivities of radionuclides within reactor coolant in the NPP, Kori unit 1 are tabulated in Table I The specific radioactivities of radionuclides within reactor coolant are averaged during one fuel cycle.

Table I Radioactivities of principal radionuclides within reactor coolant in the nuclear power plant, Kori unit 1. The radioactivities are averaged during one fuel cycle.

Unit: $\mu Ci/cm^3$

radionuclide	specific radioactivity	radionuclide	specific radioactivity	radionuclide	specific radioactivity
^{54}Mn	1.27×10^{-06}	^{95}Zr	4.64×10^{-06}	^{123m}Te	9.78×10^{-07}
^{57}Co	4.66×10^{-07}	^{99}Mo	1.91×10^{-03}	^{137}Cs	1.38×10^{-05}
^{60}Co	2.44×10^{-05}	^{110m}Ag	4.24×10^{-06}	^{144}Ce	1.27×10^{-06}
^{65}Zn	2.99×10^{-06}	^{113}Sn	1.57×10^{-06}		

The radionuclides in Table I are measurable gamma-ray-radiative nuclides contained within reactor coolant. The radioactivities of some other rest radionuclides than the radionuclides given in Table I within reactor coolant were estimated by using a scaling factor, the ratio of radioactivity of rest radionuclides to that of key nuclide such as ^{137}Cs, ^{60}Co and ^{144}Ce, from FSAR. The estimated radioactivities of the some rest radionuclides by using the scaling factor from FSAR of the NPP, Kori unit 1 are tabulated in Table II.

Table II. Radioactivities of non-principal radionuclides within reactor coolant in the NPP, Kori unit 1. The radioactivities were estimated by using scaling factor or the ratio of radioactivity of rest nuclide to that of key nuclide such as ^{137}Cs, ^{60}Co and ^{144}Ce from FSAR.

Unit: $\mu Ci/cm^3$

radionuclide	specific radioactivity	radionuclide	specific radioactivity	radionuclide	specific radioactivity
^{99m}Tc	7.30×10^{-06}	^{135}I	4.09×10^{-04}	^{134}Cs	1.04×10^{-05}
^{106}Ru	1.22×10^{-04}	^{135}Xe	1.45×10^{-03}	^{187}W	1.23×10^{-04}
^{129}Te	3.98×10^{-05}	^{135m}Xe	2.21×10^{-04}	^{239}Np	7.20×10^{-07}
^{129m}Te	2.89×10^{-07}	^{137}Xe	5.77×10^{-05}		

Inventory of decommissioning wastes

The decommissioning wastes of reactor vessel and containment building are the wastes produced by neutron activation during operation. The radioactivities of decommissioning wastes were calculated by ORIGEN computer code assuming that the NPP would be operated for 30 years. The radioactivities of decommissioning wastes of the NPP, Kori unit 1 after operation completion are shown in Table III and Table IV.

Table III Radioactivities of radionuclides within the reactor vessel of the NPP, Kori unit 1. The radioactivities were estimated by assuming that the NPP would be operated for its design lifetime.

Unit: $\mu Ci/cm^3$

radionuclide	specific radioactivity	radionuclide	specific radioactivity	radionuclide	specific radioactivity
^{3}H	1.35×10^{-09}	^{60m}Co	3.65×10^{-03}	^{93m}Nb	1.70×10^{-09}
^{14}C	4.68×10^{-06}	^{59}Ni	4.64×10^{-04}	^{94}Nb	4.21×10^{-07}
^{32}Si	2.34×10^{-13}	^{63}Ni	8.59×10^{-01}	^{93}Mo	6.08×10^{-04}
^{45}Ca	1.33×10^{-17}	^{65}Zn	1.40×10^{-11}	^{99}Mo	$1.26 \times 10^{+00}$
^{54}Mn	$2.62 \times 10^{+01}$	^{90}Sr	8.33×10^{-14}	^{98}Tc	1.42×10^{-14}
^{55}Fe	$5.48 \times 10^{+02}$	^{95}Zr	1.02×10^{-04}	^{99}Tc	1.27×10^{-05}
^{60}Co	9.59×10^{-03}	^{92}Nb	5.94×10^{-03}		

Table IV Radioactivities of radionuclides within the containment building or concrete of the NPP, Kori unit 1. The radioactivities were estimated by assuming that the NPP would be operated for its design lifetime.

Unit: $\mu Ci/cm^3$

radionuclide	specific radioactivity	radionuclide	specific radioactivity	radionuclide	specific radioactivity
^{3}H	5.21×10^{-08}	^{39}Ar	1.57×10^{-04}	^{41}Ca	2.15×10^{-06}
^{14}C	3.29×10^{-08}	^{40}K	4.27×10^{-05}	^{45}Ca	5.28×10^{-04}
^{36}Cl	3.70×10^{-08}				

Dose Exposure from LILW Disposal:

The radiation dose of each radionuclide from waste disposal was estimated from the DandD code, which was developed by the U.S. NRC in 2001. From the general disposal site and general exposure scenarios, dose conversion factor of each radionuclide had been obtained as shown in the Table V. The characteristics of disposal site were set to initial values in the DandD code. All of exposure pathways were included in the estimation such as external, inhalation, secondary ingestion, agricultural, drinking water, irrigation, and surface water exposure.

The radiation dose from the disposal of reactor coolant wastes was calculated from the radioactivities in Table I and II and the dose conversion factor in Table V. The radioactivities of radionuclides were simply calculated by considering their natural decay over time. But the inventory of some nuclides, of which half lives are short and the half lives of their daughter radionuclides are long such as ^{99m}Tc, consisted of the following nuclides:

Inventory consisted of Mother Nuclides where, Elapsed time≤half life of mother nuclides

Inventory consisted of Daughter Nuclides where, Elapsed time>half life of mother nuclides

The activities of daughter nuclides were calculated from the activities of mother nuclides and the decay constant of both nuclides as follows:

$A_m / \lambda_m = A_d / \lambda_d$
where, A_m: activity of mother nuclide
A_d: activity of daughter nuclide
λ_m: decay constant of mother nuclide
λ_d: decay constant of daughter nuclide

Table V Dose conversion factor of each radionuclide calculated from the DandD code and general exposure scenarios.

nuclides	Dose conversion factor (mSv/y)/ (Bq/cm^3)	nuclides	Dose conversion factor (mSv/y)/ (Bq/cm^3)	nuclides	Dose conversion factor (mSv/y)/ (Bq/cm^3)
^{3}H	8.84×10^{-2}	^{63}Ni	1.81×10^{-3}	^{110m}Ag	1.40×10^{0}
^{14}C	1.66×10^{-1}	^{65}Zn	6.43×10^{-1}	^{123m}Te	3.65×10^{-2}
^{36}Cl	2.03×10^{1}	^{90}Sr	5.03×10^{0}	^{129}Te	7.89×10^{-6}
^{40}K	8.89×10^{-1}	^{95}Zr	2.69×10^{-1}	^{129m}Te	6.76×10^{-3}
^{41}Ca	1.09×10^{-1}	^{93m}Nb	1.17×10^{0}	^{135}I	1.53×10^{-3}
^{45}Ca	1.26×10^{-1}	^{94}Nb	2.39×10^{-3}	^{134}Cs	1.20×10^{0}
^{54}Mn	4.73×10^{-1}	^{99}Mo	4.14×10^{-1}	^{137}Cs	6.20×10^{-1}
^{55}Fe	6.43×10^{-4}	^{99}Tc	7.49×10^{-5}	^{144}Ce	3.43×10^{-2}
^{57}Co	4.59×10^{-2}	^{99m}Tc	4.14×10^{-1}	^{187}W	1.35×10^{-3}
^{60}Co	1.80×10^{0}	^{106}Ru	1.36×10^{-1}	^{239}Np	9.44×10^{-4}
^{59}Ni	6.63×10^{-4}				

The radiation dose of each radionuclide is shown in Fig. 1. When the half life of a radionuclide is short, its daughter nuclide was finally included the inventory and therefore the daughter nuclide instead of the

mother nuclide was represented in the figure as ^{99}Tc instead of ^{99m}Tc and ^{99}Mo, ^{135}Cs instead of ^{135}I, ^{129}I instead of ^{129}Te and ^{129m}Te, ^{239}Pu instead of ^{239}Np.

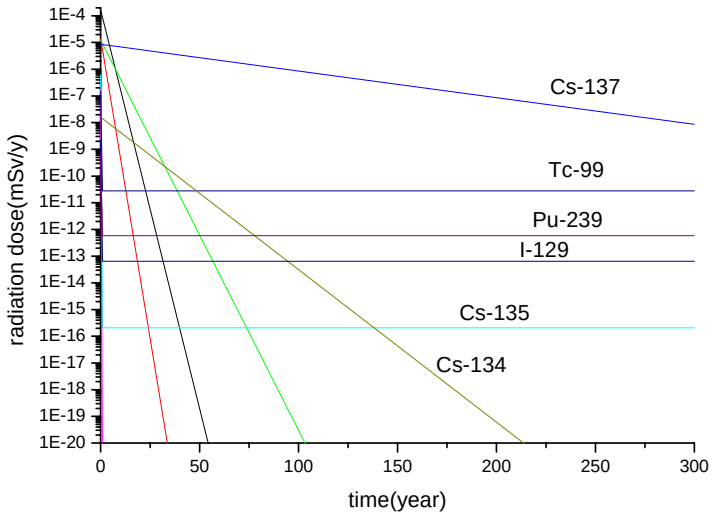

Fig. 1 Radiation Dose from the disposal of reactor coolant waste in a general disposal site.

Even though not shown in Fig. 1, at the early stage of disposal, the principal radionuclides giving 99% of total radiation dose were ^{106}Ru, ^{65}Zn, ^{134}Cs, ^{137}Cs, and ^{95}Zr in order of magnitude of radiation dose. But half lives of ^{106}Ru, ^{65}Zn, ^{134}Cs and ^{95}Zr are short and after several years, they contributed insignificantly to the total radiation dose. After hundreds of years ^{99}Tc, ^{239}Pu, ^{129}I, ^{135}Cs and ^{134}Cs give most of the total radiation dose in the reactor coolant wastes.

The radiation dose from the disposal of decommissioning waste was calculated using the radioactivities in the Table III and Table IV and the dose conversion factor in the Table V. The radiation doses from radionuclides present in the decommissioning waste are shown in Fig. 2.

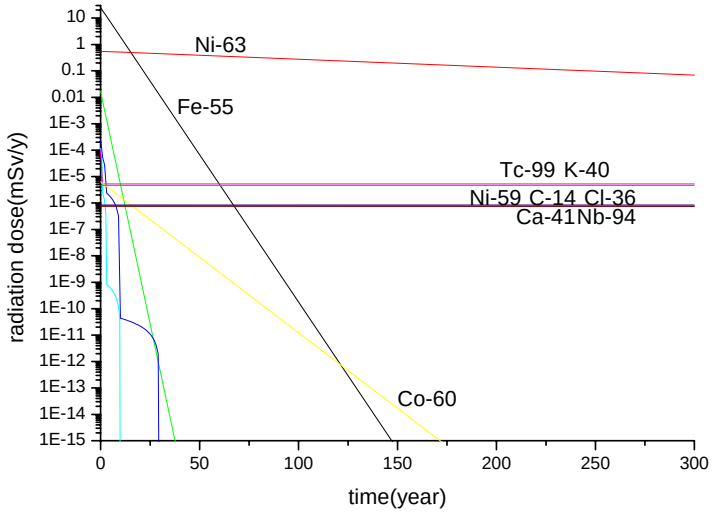

Fig. 2 Radiation dose from the disposal of operation waste and decommissioning wastein a general disposal site.

At the early stage of disposal as shown in the Fig. 2, principal radionuclides giving 99% of total radiation dose were ^{55}Fe and^{63}Ni in order of magnitude of radiation dose. But ^{55}Fe has short half life nuclide and therefore after tens of years, it contributes insignificantly to total radiation dose and ^{63}Ni becomes a principal nuclide.

DETERMINATION OF RADIONUCLIDES TO QUANTIFY

From the results of the radiation dose determination, principal nuclides to occupy most of total radiation dose from the LILW disposal were appeared to be ^{137}Cs, ^{99}Tc, ^{239}Pu, ^{129}I and ^{63}Ni. Radiation dose evaluated using these nuclides would occupy more than 99% of total radiation dose from the LILW disposal and therefore the five principal radionuclides might be sufficient for estimation of radiation dose from LILW disposal. Those selected five nuclides are important nuclides only in the inventory which consists of measurable gamma radionuclides in reactor coolant waste, in FSAR, and in decommissioning waste calculated from ORIGEN computer code. There could be some other important nuclides contributing to total radiation dose such as ^{3}H, ^{14}C, radioactive inert gas, TRU and beta decay nuclides. The KHNP temporarily has selected 21 radionuclides as principal nuclides such as ^{3}H, ^{14}C, ^{55}Fe, ^{58}Co, ^{59}Ni, ^{60}Co, ^{63}Ni, ^{90}Sr, ^{94}Nb, ^{99}Tc, ^{129}I, ^{134}Cs, ^{137}Cs, ^{144}Ce, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{241}Am, ^{242}Cm and ^{244}Cm. Final determination of principal radionuclides would be completed from the temporarily selected 21 radionuclides through the consecutive future study.

CONCLUSION

The evaluation of potential radiation dose from waste disposal requires specification of the location and characteristic of a disposal site. Korea has not yet determined the type and location of the disposal site and nevertheless radioactive wastes have been continuously produced from nuclear power plants without classification and characterization. Therefore it is necessary to determine radionuclides to be quantified for disposal of LILW wastes as soon as possible.

In this study the radionuclides of potential radiation dose for disposal of LILW waste has been determined assuming a general disposal site and a general exposure scenario. Radionuclide inventory was obtained from measured data of reactor coolant wastes, licensed data of final safety analysis report and calculation from ORIGEN computer code for neutron activated plant structure. The radiation dose of each radionuclide was estimated and ranked in order of from high to low dose radionuclide. From the results of radiation dose determination, principal nuclides to occupy most of total radiation dose from the disposal of LILW were appeared to be ^{137}Cs, ^{99}Tc, ^{239}Pu, ^{129}I and ^{63}Ni. Radiation dose evaluated using these radionuclides would occupy more than 99% of total radiation dose from the LILW disposal. The five selected principal radionuclides might be sufficient for estimation of radiation dose from LILW disposal and are important radionuclides only in the inventory which consist of measurable gamma radionuclides in the reactor coolant and decommissioning wastes calculated from ORIGEN computer code. There could be some other important nuclides contributing to total radiation dose such as ^{3}H, ^{14}C, radioactive inert gas, TRU and beta decay nuclides. The KHNP temporarily has selected 21 radionuclides as principal radionuclides. The final determination of principal radionuclides present within the LILW would be completed from the temporarily selected 21 radionuclides through the future consecutive study.

REFERENCE

1 Ministry of Science & Technology's notification 2001-32 - regulation on the transfer of low- and intermediate- level radioactive waste (2001).

2 KINS/HR-494," Research on the Assessment Technology of the Radionuclide Inventory for the Radioactive Waste Disposal (II)" (2003)

3 NUREG/CR-5512 Vol. 2 - Residual Radioactive Contamination from Decommissioning (2001).