ESTIMATION METHOD FOR DETERMINATION OF RADIOACTIVITY WITHIN LILW FOR LAND DISPOSAL

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ABSTRACT

Korea started the first commercial operation of nuclear power plant (NPP) in 1978. Now, there are 19 NPPs in operation in Korea, but it has not yet a disposal site for low- and intermediate- level radioactive waste (LILW). Korea Hydro & Nuclear Power Corporation (KHNP) has studied a methodology to estimate of radionuclide inventory of LILW since 1992 and as a result, the first radioactive waste analysis system was established in 1996. Its application is only limited to parts of wastes produced from the pressurized water reactor (PWR), KORI unit 1 in Korea. So, starting 2002, KHNP has developed a new method of the estimation of radionuclide inventory of LILW and investigated the physical and chemical characteristics of LILW produced from all over the NPP in Korea. To estimate the radionuclide inventory of LILW, KHNP has determined to measure the radioactivity of packed waste directly because it is cost-effective and time-saving. In a previous study in 2003, KHNP has selected 21 radionuclides as principal radionuclides for disposal of LILW and started to seek scaling factors needed for analysis of difficult-to-measure (DTM) radionuclides in LILW site-specifically classified as 13 groups of NPPs in Korea. In this study, the method of estimation of radionuclide inventory of LILW was determined as follows: 55 gallon standard drum containing LILW can be analyzed by tomographic gamma scanner (TGS) or segmented gamma scanner (SGS) spectroscope for the measurement of radioactivity of key gamma emitting nuclides such as ^{60}Co, ^{137}Cs and ^{144}Ce (if it exists) in case the radioactivity on its surface is lower than 10 $R \cdot h^{-1}$ and the weight is less than 1,000 kgf. Non-gamma-emitting nuclides in LILW are estimated directly by using the ratios of radioactivities of non-gamma-emitting nuclides to those of a key nuclide or scaling factor. The wastes, of which radioactivity was difficult to measure by TGS because of not being the standard drum, can be applied by dose-to-curie analysis. Old wastes are difficult to measure the radioactivity of radionuclides within because of lack of their own scaling factor. The radioactivities of those wastes can be obtained by analogy of current scaling factor of the same type of NPP with decay correction. If ^{144}Ce does not exist in LILW, then ^{60}Co and ^{137}Cs will be used for scaling factor instead. In this case, radionuclides such as TRU could be analyzed by ^{60}Co as a key nuclide rather than ^{144}Ce. ^{3}H may need statistical scaling factors made up of ^{60}Co, ^{137}Cs or their average concentration data within the waste because of its volatile characteristics. ^{99}Tc and ^{129}I could be estimated by statistical scaling factors of ^{137}Cs, but also need theoretical approach like radioisotope transportation and removal adjustment factor through the waste stream.

INTRODUCTION

Korea started the first commercial operation of NPP in 1978 and now ranked as the world's 6th largest in terms of nuclear power generation. But Korea has not yet a disposal site for LILW and so the radioactive wastes from Korean NPP have insufficient information on their radioactivity.

In Korea, the regulation and licensing of the disposal of LILW are based on the provisions of

the Atomic Energy Act, its Enforcement Decree, Regulation, and the Notice by the Minister of Science and Technology. The basic concept of them is not only to protect the public health and safety from radiation hazards, but also to protect the environment from any subsequent harmful effect. In the near future, probably before the end of 2005, the Notice about the disposal of LILW will be amended and specified details of the disposal of LILW. According to the regulation, the disposal of the LILW necessitates classification of LILW, physical and chemical characterization of LILW and radiation safety analysis from the disposal of LILW into a disposal site.

KHNP had studied a method to estimate the radionuclide inventory of LILW with assistance of Korea Electric Power Research Institute (KEPRI), Korea Atomic Energy Research Institute (KAERI) and Korea Advanced Institute of Science and Technology (KAIST) from 1992 to 1996 and as a result, the first radioactive waste analysis system was established in 1996 [1]. The system uses a segmented gamma scanner (SGS) for measurement of gamma-ray emitting key nuclides and generic scaling factor for analysis of difficult-to-measure (DTM) nuclides and so applicable to parts of wastes produced from the pressurized water reactor (PWR) NPP, KORI unit 1 in Korea.

So, starting 2002, KHNP has developed a new method of the estimation of radionuclide inventory of LILW and investigated the physical and chemical characteristics of LILW produced from all over the NPP in Korea. The estimation of radionuclide inventory of LILW has been being executed in cooperation with KAERI, KAIST and Korea Power Engineering Company (KOPEC) and the characterization of LILW has been being done with KAERI.

There are several ways to determine radionuclide inventory of LILW. Among them direct measurement of packed waste seems to be the most cost-effective and time-saving method because it can be accomplished only by scaling factor of LILW and gamma scanning of waste drum or measurement of dose-rate on drum surface. There are some improvements to the system of estimation of radionuclide inventory of LILW, that is to say, the principal radionuclides of which scaling factors have to be obtained, gamma ray spectroscope for measurement of gamma-ray-emitting key nuclides, scaling factor for estimation of DTM nuclides and the kinds of waste to be analyzed.

Improvement of Estimation Method of Radionuclide Inventory

In the first study between 1992 and 1996, KHNP selected ^{3}H, ^{14}C, ^{55}Fe, ^{60}Co, ^{63}Ni, ^{90}Sr, ^{94}Nb, ^{99}Tc, ^{129}I, ^{137}Cs and total alpha as important radionuclides for land disposal of LILW and sought generic scaling factors for those radionuclides which are applicable only to parts of wastes produced from the PWR NPP, KORI unit 1 in Korea.

However, in this study, KHNP has selected 21 radionuclides as the principal radionuclides for disposal of LILW, that is, ^{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, ^{244}Cm and total alpha. In our calculation, the absorbed dose due to the disposal of LILW to general disposal site shows that the radionuclides occupying 99 percent of total radiation dose from the disposal of LILW are merely ^{3}H, ^{14}C, ^{59}Ni, ^{63}Ni, ^{90}Sr, ^{94}Nb, ^{129}I, ^{99}Tc, ^{137}Cs, and some transuranic nuclides (TRU) [2].

The TGS is used to measure the radioactivity of gamma-emitting key nuclides within a waste drum instead of the previous SGS. TGS requires more time to analyze the radioactivity within the waste drum than SGS, but its accuracy and uncertainty is better than that of the SGS because the TGS analyzes three-dimensional image instead of two-dimensional or segmented image of SGS. The TGS can also be operated in SGS mode for homogeneous waste to save the time for measurement.

In the first study, the scaling factors needed for analysis of radioactivity of DTM nuclide in LILW was obtained generically and applied to PWR NPP. But now the scaling factors have been being sought site-specifically over all NPP in Korea from 2003 classified as 13 groups,

that is, 9 PWR and 4 PHWR NPPs [3]. The site-specific scaling factors are affected by the operation condition of the NPP. But the estimated results of radionuclide inventory of LILW by them report better quality than results by the generic scaling factor.

If there are insignificant differences in site-specific scaling factors of NPPs of the same nuclear power station, then all data needed to obtain the site-specific scaling factors will be averaged over the nuclear power station. It becomes the generic scaling factor of the nuclear power station and may be improved in accuracy and uncertainty than site-specific generic factor. The scaling factors which have been being developed in cooperation of KAERI and KAIST will be obtained by the end of 2005.

Radioactive wastes produced before 2004 lack the information on their radioactivity, waste stream of which waste is produced, operation condition of NPP or the scaling factors. In order to estimate the radionuclide inventory of those radioactive wastes, it is necessary to obtain the scaling factor resulting from the analysis of radioactivity between the principal DTM nuclides and gamma-ray emitting key nuclides at the time the waste produced.

KHNP will estimate the scaling factor of those radioactive wastes which have little information on the scaling factor by using the current scaling factor of the same NPP or nuclear power station and radioactivity correction with regard to radioactive decay. If it is difficult to have the scaling factors obtained from the method be within a certain uncertainty such as 0.1 to 10 times of measured results, then a conservative means like adapting generic scaling factor or safety margin eligible to meet the criteria of Korean regulation should be introduced.

Measurement of Radioactivity in LILW

Standard Drum

The radioactivity of gamma-ray emitting key nuclides such as ^{60}Co, ^{137}Cs and ^{144}Ce (if it exists) in a 55 gallon standard LILW can be analyzed by gamma ray spectroscope using TGS or SGS in case the radioactivity on the surface of waste drum is lower than 10 $R \cdot h^{-1}$ and total weight of waste is less than 1,000 kgf.

The TGS (or SGS) consists of a Canberra broad energy germanium (BEGe) detector of which relative efficiency is 50% compared to a 3 inch by 3 inch NaI(Tl) detector. The TGS analyzes three-dimensional material and/or radioactivity distribution of waste drum containing even inhomogeneous radioactive waste with transmission source and the BEGe detector.
Uncertainty due to inhomogeneous distribution of waste within a drum is as like table 1 [4]. As shown Table I, TGS has less uncertainty than SGS. The uncertainty of measured results by the SGS increases dramatically in case the density of waste is greater than 2 $g \cdot cm^{-3}$.

Table I. Uncertainties of measured results of radioactivity within the various types of waste drums due to inhomogeneous distribution of content

Density ($g \cdot cm^{-3}$)	0.25	0.50	0.75	1.00	1.25	2.0	3.0
SGS	4%	9%	14%	21%	28%	-	-
TGS	2%	4%	6%	9%	11%	17%	26%

Radionuclides other than gamma-ray emitting nuclides or DTM nuclides in LILW are difficult to measure their radioactivity by direct measurement of the packed waste without radiochemical analysis with destruction. A good way to know the radioactivity of DTM nuclides is measuring the ratio those nuclides to any key nuclide or scaling factor.

The scaling factor has been being obtained by radiochemical analysis and radioactivity measurement in the waste such as letdown filters, primary mixed bed resins, radioactive waste evaporator concentrates, dry active wastes and reactor coolants over 13 groups of NPPs from 2003 until 2005 by KAERI. In case that there are little differences in scaling factors of

NPPs in the same nuclear power station, the site specific scaling factor can be added and be generic scaling factor.

Non-standard Drum

There are wastes in which radioactivity of gamma-ray emitting nuclide was difficult to measure by TGS because the container is not the standard drum. In that case, the radioactivity can be estimated by measuring the surface dose of the container and using dose-to-curie conversion factor as follows:

1) To obtain the ratio between radioactivities of nuclides emitting gamma-ray in waste by radiochemical analysis when the waste produced.
2) To simulate the interaction between gamma ray emitted from radioactive waste and waste material within the container such as scattering, absorption etc using a computer code as like Monte Carlo N-Particle (MCNP) simulation code
3) To calculate the dose rate on the surface of the drum due to gamma ray emitted from the waste per unit radioactivity (Ci or Bq) of nuclide of the waste
4) To calculate the radioactivity of the waste by multiplying reverse value of conversion factor of the process 3) by the measured dose rate on the surface of waste drum

The waste container other than standard drum of which radioactivity could not analyzed by gamma ray spectroscope with TGS or SGS can be examined to measure the radioactivity of gamma-ray emitting key nuclide by in-situ spectroscopy system or portable gamma ray spectroscope with a portable HPGe detector. But the system has poor accuracy and uncertainty compared to SGS because it analyzes the radioactivity of a waste drum by assuming that it is homogeneous and measures once without any movement of waste drum such as rotation or up/down.

Waste Drum having no Sufficient Information on Scaling Factor

Waste drum produced before 2004 has no sufficient information on scaling factor and difficult to estimate the radioactivity of nuclide in it. The radioactivity of the waste can be obtained by analogy of current scaling factor of the same type of NPP with decay correction. The decay correction has to be executed to both the ratio between radioactivity of gamma-ray emitting nuclides and scaling factor.

It is assumed that the ratio between radioactivities of gamma-ray emitting nuclides of former year waste at the time the waste produced is the same as the ratio between radioactivities of gamma-ray emitting nuclides of current waste. And the scaling factor of former year waste at the time the waste produced is the same as the scaling factor of current waste, too. So, the decay correction has to be made as follows:

1) The radioactivity of gamma-ray emitting nuclide at the time the waste produced can be sought by dose-to-curie method with the measured dose rate on the surface of the drum at the time the waste produced and the ratio between radioactivities of gamma-ray emitting nuclides of current waste of the same type of NPP
2) To estimate the radioactivity of DTM nuclide at the time the waste produced using the scaling factor of current waste of the same type of NPP
3) To modify the radionuclide inventory of the waste at the time through 1) to 2) to current radionuclide inventory using decay correction.

Some Consideration

Gamma-ray emitting key nuclides used to estimate the DTM nuclides are ^{60}Co, ^{137}Cs and ^{144}Ce. If ^{144}Ce does not exist in LILW, then ^{60}Co and ^{137}Cs will have to be used instead. In this case, radionuclides such as TRU could be analyzed by ^{60}Co rather than ^{144}Ce.

^{3}H may need statistical scaling factors made up of ^{60}Co, ^{137}Cs or their average concentration

data sought from the waste because of its volatile characteristics. ^{99}Tc and ^{129}I could be estimated by statistical scaling factors of ^{137}Cs, but they need theoretical approach like radioisotope transportation and removal adjustment factor through the waste stream.

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