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방사성 물질의 환경영향인자 개발 연구

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Study on Development for Classification Factors of Radioactive Substances

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ABSTRACT

Limitation associated with classification factor of radioactive substance based on the critical volume approach is analysed and the classification factors of radioactive substances are developed using the concept of radiation protection and radiological risk model. To calculate the classification factor of radioactive substance, information on the concentration of radionuclides in medium, the length of exposure period, and average usage rate of medium by population are necessary. The classification factor of radioactive substance developed is age and gender specific and considers both of exposure and effect of radionuclide and different pathway. Effect score of radiological impact can be calculated using the release amount of radionuclide and the classification factor developed in this study.

Keywords : Classification Factor, Radiological Impact Assessment, Risk Assessment, Radiation Protection, Nuclear Fuel Cycle

요 약 문

환경영향인자를 계산하는 기존의 한계량 방법이 방사성 물질의 환경영향인자 개발에 적용 될 때의 문제점을 분석하고 방사선 방호 개념 및 방사선적 위해도 평가 방법을 이용하여 새로운 방사성 물질의 환경영향인자를 개발하였다. 방사성 물질의 환경영향인자를 개발하기 위해서는 매질내의 핵종 농도와 피폭 시간 및 매질 섭취량 등의 정보가 필요하다. 개발된 방사성 물질의 환경영향인자는 피폭자의 연령 및 성별의 특성이 반영되어 있고 다양한 피폭경로 별로 다른 값을 가진다. 개발된 방사성 물질의 환경영향인자와 방사성 물질의 방출량으로부터 LCA 측면에서의 방사선적 환경영향 평가를 할 수 있다.

주제어 : 환경영향인자, 방사선적 영향평가, 위해도 평가, 방사선 방호, 핵주기,

Ⅰ. Introduction

The existing environmental impact assessments of nuclear fuel cycle have focused on two kinds of issues. One is the comparative assessment between other resources for only CO_2 emission, and the other is the radiological risk assessment.

So, it is necessary to consider Life Cycle Assessment (LCA) methodology covering the whole nuclear fuel cycle for the improvement of environmental friendliness of nuclear power generation. However, direct introduction of LCA to the nuclear fuel cycle is difficult more or less due to the absence of the methodology for the radiological impact assessment within the LCA framework and related works to the development for the classification factor of radioactive substance are not carried out.

Therefore, this study shows that the limitation associated with classification factor of radioactive substance based on the critical volume approach is analysed and the classification factors of radioactive substances are developed using the concept of radiation protection and radiological risk model. According to the SETAC nomenclature, impact assessment consists of classification and characterization. But this study doses not distinguish between these two and only talks about classification.

II. Radiation Protection and Risk Assessment

1. Radiation Protection

Several approaches are possible to quantify the environment impact of the radioactive substances using the concept of radiation protection. However, these approaches could be done by dividing the emission quantities of radioactive substances by dose limit or intake standard for that substance. These approaches are based on the critical volume method.[1)]

1) Dose Equivalent Limits

Before the application of the critical volume method to calculate the classification factor of the radiological impact, the dose-response characteristics need to be reviewed preliminarily, which are the fundamental understanding of the radiation protection guide. Observed radiation effects may be broadly classified into two categories, viz. stochastic and non-stochastic effects. Most biological effects fall into the category of non-stochastic effects. The magnitude of the effect increases with the size of the dose. Further there is a clear relationship between exposure and effect. Because of the minimum-dose that must be exceeded before an individual shows the effect, non-stochastic effects are also called threshold effects. And non-stochastic effects are effects such as unacceptable changes in the skin for which a threshold or pseudo-threshold of dose must be exceeded before the effect is induced. Stochastic effects are not unequivocally related to exposure. The main stochastic effects are cancer and genetic effects. Stochastic effects are often called linear, zero-threshold dose-response effects. The International Commission on Radiological Protection (ICRP) recommended the radiation protection guides. ICRPs recommendations are intended to prevent non-stochastic effect and to limit the occurrence of stochastic effect to an acceptable level. So, the conservative assumption is taken in the application of radiation protection that there is no threshold and dose-response function is considered to be linear beginning at the origin.[2)]

For stochastic effects the limiting exposure is based on the principle that the limit on risk should be equal whether the whole body is irradiated uniformly. This condition will be if

$$\sum w_T H_T \le H_{wb} \tag{1}$$

where w_T is a weighting factor representing the ratio of the stochastic risk resulting from tissue (T) to the risk when the whole body is irradiated uniformly.

H_T is the dose equivalent received by tissue (T).

H_{wb} is the stochastic dose-equivalent limit for uniform irradiation of the whole body.

For occupational exposure, the value 50 mSv (5 rem) in any year is recommended. The recommended dose-equivalent limits for stochastic effects are made using the hypothesis that risk of an effect is linearly related to dose equivalent. Commission has reconsidered the time over which this total dose equivalent should be integrated and has concluded that the period of 50 years is appropriate. The total dose equivalent in any tissue over the 50 years after intake of a radionuclide into the body is termed the Committed Dose Equivalent, H_{50} as equation (3). Therefore, exposure limit of workers is as follows;

$$\sum w_T H_{50,T} \le 0.05\ Sv \qquad (2)$$

where $H_{50,T}$ is the committed dose equivalent in tissue T resulting from intakes of radioactive materials from all sources during the year in question.

$$H_{50} = \sum_i \frac{\int^M D_{50,i} Q_i N_i dm}{\int^M dm} \qquad (3)$$

where M is the mass of the specified organ or tissue.

$D_{50,i}$ is the total absorbed dose during a period of 50 years after intake of the radionuclide into the body in the element of mass dm of the specified organ or tissue for each type of radiation i.

Q_i is the quality factor.

N_i is the product of all other modifying factors such as dose rate.[3)]

2) Intake Standards

Intake standards are often stated in terms of maximum permissible concentration (MPC). This is defined for radiation workers as the maximum concentration of a radionuclide in air or water that over a 50-year period from the onset of constant daily intake provides a dose equivalent rate to any organ in excess of the maximum permissible dose rate for that organ.[4)] The MPC values are largely based on calculations performed by ICRP publication 2 during the 1950s. Similar calculations using more modern biological and radiological data were carried out in the 1970s and reported in ICRP publication 30. Although the Commission emphasized in ICRP publication 2 that the rate of intake of a radionuclide could be varied, provided that the intake in any quarter was no greater than that resulting from continuous exposure to the appropriate MPC for 13 weeks, the concept of MPC has been misused to imply a maximum concentration in air or water that should never be exceeded under any circumstances. For the reason described above, the ICRP does not recommend MPCs, and the concept of MPC is not used any more as the intake standard. Instead, the ICRP computes and recommends what it calls the Annual Limit on Intake (ALI) of radionuclides. ICRP also specifies a new quantity the Derived Air Concentration (DAC) either by ingestion and inhalation. ALI of the radionuclide into the body is determined by the value of committed dose equivalent H_{50}.[5)]

2. Limitation of Critical Volume Approach

Modified critical volume approach using the ALI or MPC can be proposed to calculate the classification factor for radiological impact. However, this approach is merely done by dividing the emission quantities of radioactive substances by dose limit or intake standard for that substance. So, there are some problems when dose equivalent limit or intake standard is used as the standard. Firstly dose equivalent limits are not classified by radionuclides. So classification factor considering the characteristics of radionuclides cannot be derived. Also because intake standard such as ALI is used for inhalation and ingestion, external exposure is not considered in classification factor. And also this approach does not consider both of exposure and effect of radionuclides according to the different exposure pathway. Therefore critical volume approach is worst case and differences between substances in terms of half-life and exposure pathway are not considered. So, in this study it is attempted to introduce the concept of radiological impact assessment to calculate the classification factors of radioactive substances. The concept of risk model for the radiological impact assessment is briefly reviewed in following section.

3. Radiological Risk Assessment

Since the mid-1980s United States Environmental Protection Agency (EPA) has issued a series of the Federal guidance documents for providing the technical information to assist their implementation of radiation protection programs. So the EPA provides the risk coefficients for cancer attributable to exposure to any of approximately 100 important radionuclides.

The risk coefficient is expressed as the probability of radiogenic cancer mortality or morbidity per unit activity inhaled or ingested for internal exposure or per unit time-integrated activity concentration in air or soil for external exposure. The risk coefficient is interpreted as average risk per unit exposure for persons exposed throughout life to a constant activity concentration of radionuclide in an environmental medium. And this is interpreted as average risk per unit exposure for persons acutely exposed to the radionuclide through the environmental medium. This interpretation is possible as long as the exposure involved is properly characterized as low acute dose or low dose rate.

One of two basic types of radiogenic cancer risk projection models is used for a given cancer site: absolute risk model or relative risk model. The absolute risk and relative risk at age x, $\varepsilon(x)$, due to a unit absorbed dose received at an earlier age x_e are calculated as equation (4) and equation (5) respectively.

$$\varepsilon_{absolute}(x,x_e) = \alpha(x_e)\varsigma(t) \quad (4)$$

$$\varepsilon_{relative}(x,x_e) = \mu(x)\beta(x_e)\varsigma(t,x_e) \quad (5)$$

where $\alpha(xe)$, $\beta(xe)$ is risk model coefficient defining the potential level of risk.
$\mu(x)$ is the base line force of cancer mortality or morbidity at age x.
$\xi(t)$ and $\xi(t, xe)$ is time period during which the risk is expressed.

So, for an absolute risk model or a relative risk model, age-specific life time risk coefficients, $r(x)$, which is averaged by the survival function, $S(x)$, are calculated as equation (6).

$$r(x) = \frac{\int_x^{\infty} \varepsilon_{relative}(z) \text{ or } \varepsilon_{absolute}(z)S(z)dz}{S(x)} \quad (6)$$

The cancer risk for internal exposure, $r_a(x_i)$, resulting from a unit intake of a radionuclide at age x_i is calculated from the absorbed dose rate $\dot{D}$ as equation (7).

$$r_a(x_i) = \frac{\int_{x_i}^{\infty} \dot{D}(x)\, r(x) S(x)\, dx}{S(x_i)} \quad (7)$$

The intake rate is proportional to a constant environmental concentration. However, usage is also age and gender specific and therefore must be included in the averaging step. Therefore, the averaged lifetime risk for internal intake is calculated from equation (8).

$$\bar{r}_a = \frac{\int_0^\infty u(x) r_a(x) S(x) dx}{\int_0^\infty u(x) S(x) dx} \quad (8)$$

Since the external exposure is not considered to be age dependent and the usage rate, $u(x)$, is unnecessary, lifetime risk for external exposure, $r_e(x)$, is simply reduced to the product of dose per unit exposure coefficient, d_e, and life time risk coefficients as equation (9) and averaged lifetime risk for external exposure is calculated as equation (10).

$$r_e(x) = d_e r(x) \quad (9)$$

$$\bar{r}_e = \frac{\int_0^\infty r_e(x) S(x) dx}{\int_0^\infty S(x) dx} \quad (10)$$

Above two equations for the risk coefficients are applied to a specific cancer site. So the total risk is the sum over all cancer sites.[6)]

4. Advances in Classification Factor Considering Risk Model

This study introduces the risk coefficients to overcome the limit of critical volume approach for the development of classification factors of radioactive substances.

It is a common practice to estimate the cancer risk from internal or external exposure to a radionuclide as the simple product of a probability coefficient and an estimated effective dose. A nominal cancer fatality probability coefficient of 0.05Sv-1 is given in ICRP Publication 60 for all cancer types combined. This value contains the uncertainties in radiation risk estimates and is based on the population receiving the uniform dose over the whole body. These assumptions such as nominal cancer fatality probability coefficient and uniform dose may cause the less accurate estimation when the distribution of the dose is non-uniform. However, risk coefficients consider the age and gender dependence, which have not been taken into account in the general risk estimate based on the simple product of a nominal probability coefficient and effective dose.

Classification factors using the risk coefficients need the information on average usage rate of media and the length of exposure period. Because media usage rate and survival function of members of population are different according to the gender and age, classification factor should consider the difference of gender and age. Risk coefficients are dependent on the gender and age and useful for the case of non-uniform as you can see in equation (8) and (10). So gender and age specific classification factors can be calculated by risk coefficients. Also the calculation of classification factor using risk coefficients makes it possible to consider the various exposure pathway; inhalation, ingestion, and external exposure. Therefore classification factors of radioactive substances using risk coefficients consider characteristics both of radionuclides and health.

Ⅲ. Calculation of Classification Factor

1. How to Calculate

For a given exposure scenario, the computation of lifetime cancer risk, R, associated with intake I, or external exposure X involves $R = r \cdot I$ for intake by inhalation or ingestion and $R = r \cdot X$ for external exposure. However, to calculate the classification factor of radioactive substance from the radiological impact assessment method directly, some problems should be solved. Firstly, some kinds of manipulation steps are added to calculate the classification factor from the source term. Generally, only amounts with the unit of Bq^{-1} of radioactive substances released from the nuclear fuel cycle are used in the life cycle inventory. Table 1 shows radionuclides released from the nuclear fuel cycle. It must be possible to calculate the effect score of radiological impact by multiplying the source term by classification factor. Therefore following information is considered. For the external exposure, estimation of the time-integrated activity concentration X requires information on the concentration of the radionuclide in the medium and the length of the exposure period. For the internal exposure scenario, estimation of the per capita activity intake I of the radionuclide requires the same information, plus an estimate of the average usage rate of the medium by members of the population during the exposure period. This study assumes that lifetime of a person is 75 years and average usage rate of environmental media is equal to data prepared by the National Center for Health Statistics for the U.S. Decennial life table in 1997. The scheme of calculation of classification factor is suggested in Fig. 2.

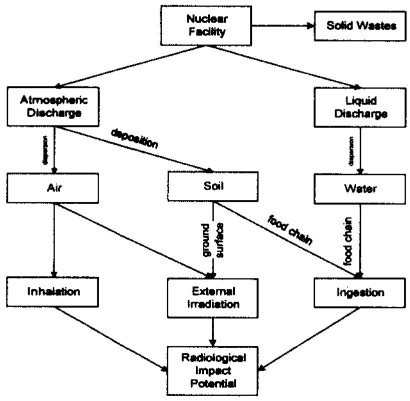

Fig. 1. Exposure pathway.

Table. 1. Radionuclides released from Nuclear Fuel Cycle.[7]

6Mining and Milling	
Gas	Ra-222, U-234, -235, -238
Liq.	U-234, -235, -238
Conversion, Enrichment, Fuel fabrication	
Gas	U-234, -235, -238
Liq.	U-234, -235, -238
Reactor Normal Operation	
Gas	H-3, C-14, Co-58, -60, Kr-85, I-131, -133, Xe-133, Cs-134, -137
Liq.	H-3, Mn-54, Co-58, Co-60, Ag-110m, Sb-124, I-131, Cs-134, -137
Reprocessing	
Gas	H-3, C-14, Kr-85, I-129, -131, -133, Pu-238, -239
Liq.	H-3, C-14, Co-60, Sr-90, Ru-106, I-129, Am-241, Sb-125, Cs-134, -137, U-238238, Pu-238, -239,
Low Level Waste Disposala	
Sol.	H-3, C-14, Co-60, Ni-59, -63, Sr-90, Zr-93, Nb-94, Mo-93, Tv-99, Pd-107, I-129, Cs-135, -137, U-234, -238, Pu-239, -241, Am-241, Np-237
High Level Waste Disposal	
Sol.	Se-79, Zr-93, Tc-99, Pd-107, Cs-135, Th-229, U-233, Np-237, Am-241

Secondly, it is necessary to take into account the exposure pathway divided as followings and details are shown in Fig. 1.

- inhalation of a radionuclide in air
- ingestion of a radionuclide in water and food
- external exposure to radiation from a radionuclide in air, on the ground surface and in soil

Risk coefficients for inhalation of radionuclide in air mean the probability of cancer mortality or morbidity per unit intake (Bq^{-1}). The intake rate for a radionuclide in air is assumed to depend on age and gender, so intake is averaged over all ages and both genders. Risk coefficient for ingestion is expressed as that for the inhalation. For the assessment of the intake of a radionuclide in food, its activity concentration in food and an average usage rate are necessary. The risk coefficient for external exposure is expressed as the probability of cancer mortality or morbidity per unit time integrated activity concentration in air, on the ground surface, or in soil.

2. Results of Classification Factor

The result of classification factor developed according to the calculation method mentioned above is in Table 2. The scheme to calculate classification factors in Table 2 is given in Fig. 2. Classification factor considers both of exposure and effect of radionuclides. Exposure is related with the concentration of radionuclide in medium, which is determined by the emission quantity and the length of exposure period. And risk coefficients represent the effect of radionuclides. So if only the emission released from the nuclear fuel cycle is

Table 2. Results of Classification Factor Developed[1/Bq].

Pathway	Inhalation of Air	External Exposure from Air	External Exposure from Ground	Ingestion of Soil	Ingestion of Water
U234	-	-	-	5.61E-15	8.09E-14
U235	-	-	-	5.48E-15	7.84E-14
U238	-	-	-	5.10E-15	7.21E-14
Ar41	-	3.28E-24	3.73E-22	-	-
Kr85	-	6.78E-24	-	-	-
Kr85m	-	3.50E-25	4.56E-23	-	-
Kr87	-	2.09E-24	2.31E-22	-	-
Kr88	-	5.22E-24	5.40E-22	-	-
Xe131m	-	1.46E-26	4.04E-24	-	-
Xe133	-	6.47E-26	1.13E-23	-	-
Xe133m	-	6.15E-26	1.01E-23	-	-
Xe135	-	5.71E-25	7.36E-23	-	-
Xe135m	-	1.00E-24	1.28E-22	-	-
Xe128	-	2.92E-24	3.20E-22	-	-
I131	4.55E-14	8.91E-25	1.13E-22	6.25E-16	-
I132	8.27E-15	5.61E-24	6.80E-22	1.07E-16	-
I133	1.58E-14	1.46E-24	1.83E-22	2.20E-16	-
I134	4.43E-15	6.49E-24	7.77E-22	1.68E-17	-
I135	4.56E-15	4.03E-24	4.55E-22	6.42E-17	-
Ag110m	4.48E-13	6.83E-24	8.12E-22	6.49E-16	-
Fe59	1.76E-13	3.00E-24	3.46E-22	6.42E-16	-
Nb95	4.54E-14	1.86E-24	2.28E-22	-	-
Mo99	1.76E-14	3.60E-25	4.62E-23	1.93E-16	-
Zr95	1.45E-13	1.79E-24	2.20E-22	3.41E-16	-
Co58	3.85E-13	2.36E-24	2.90E-22	1.84E-16	2.02E-15
Co60	3.78E-13	6.36E-24	7.27E-22	2.04E-16	1.77E-14
Ru106	7.71E-13	5.2E-25	-	3.16E-15	2.69E-14
Sr90	8.85E-13	1.04E-27	1.34E-25	1.34E-15	5.59E-14
Ra226	-	-	-	-	3.23E-13
Sb124	-	-	-	-	8.34E-15
Sr89	-	-	-	-	8.76E-16
T-3	1.24E-15				5.17E-17

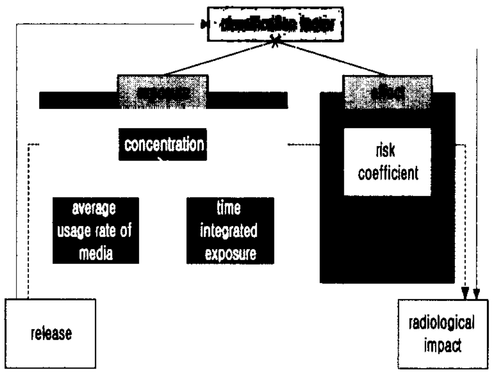

Fig. 2. Calculation scheme of classification factor.

given in the life cycle inventory table, effect score of radiological impact can be calculated by multiplying the emission by classification factor developed.

In Table 2, classification factors of 32 radionuclides developed in this study are shown according to 5 types of exposure pathways. Classification factors of internal exposure have the order of about 10E-15 and those of external exposure have the order of about 10E-22 or 10E-24. And some radionuclides hyphenated are not available for calculation of the classification factor because risk coefficients of those substances are not supplied or those radionuclides are not emitted through the relevant exposure pathway. So classification factors of remaining radionuclides need to be continuously supplemented.

Therefore classification factors in Table 2 overcome the limit of critical volume approach, and introduce the risk coefficients that are gender and age specific and able to cover the whole body non-uniform dose according to the exposure pathway. So these factors developed in this study make it possible to calculate the effect score of radiological impact by multiplying the source terms (Bq^{-1}). But this study needs the supplements. For the classification factor for external exposure does not consider the effect of half-life of radioactive nuclide when the time-integrated concentration of radioactive substance is calculated. Therefore, it is recommended that computational code for radiation dose simulation of nuclear facility be introduced for more accurate calculation of radiological risk from the external exposure.

Ⅳ. Conclusion

Classification factors of radioactive substances are important to apply LCA to nuclear fuel cycle, however, LCA framework does not supply these factors and related works are not carried out. Therefore, in this study, the limitation associated with classification factor of radioactive substance based on the critical volume approach is analysed and classification factors of radioactive substances are developed.

Critical volume approach using dose limit or intake standard is possible to quantify the environment impact of the radioactive substances. This approach is to divide the emission quantity by the standard such as ALI for that substance. But this approach does not consider both of exposure and effect of radionuclide according to the different exposure pathway and half-life.

So, this study introduces risk coefficients to overcome the limit of critical volume approach. The calculation of classification factor of radioactive substance using risk coefficients makes it possible to consider the gender and age dependent effect, various exposure pathways and non-uniform dose. To develop the classification factor of radioactive substance, information on the concentration of radionuclides in medium, the length of exposure period, and average usage rate of medium by population are necessary. Classification factors of radioactive substances developed have the order of about 10E-15 for internal exposure and about 10E-22 for external exposure.

Therefore, from the result of this study, provided that only the release information (Bq^{-1}) is known, the effect scores of radiological impact can be calculated. And this study could help the promotion of LCA methodology and the advances of environmental management of nuclear fuel cycle.

Terminology

Dose Equivalent	The weighted absorbed dose by the type and energy of radiation
Committed Dose Equivalent	The time integral of the equivalent dose rate
Absorbed Dose	The energy absorbed per unit mass of the irradiated material
Quality Factor	A factor used in radiation protection to consider the different biological effects
rem	The conventional unit of equivalent dose 1 rem=0.01Sv
Sv	The SI unit of dose equivalent
Bq	Becquerel The basic unit of radioactivity 1Bq = 1 disintegration per second

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