Correlation Properties of H-3 and C-14 with Co-60 in Concentration Bottom

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1. Introduction

10CFR61.55 (a) and (b) regulation allows indirectly radionuclide assay for disposal of low-level radwaste due to difficulty with measuring essential α- or β-emitted nuclides. This method means using scaling factor calculated through correlation of α- or β-emitted nuclides with γ- nuclide which has systematically similar properties with α- or β- nuclides. In order to obtain scaling factor, every radwaste has to be classified in detail as sample type and plant type owing to difference in operating process of each NPP. The scaling factor related to disposal of radioactive waste in foreign NPPs has been calculated and updated at least every year for Class B and C wastes and biannual assayed for Class A since the beginning of 1980s. Class A, B, and C are divided as a half-life and radioactivity of radionuclides[1]. Meanwhile, the statistic method for the scaling factor of α- or β-emitted radionuclide which was generated from a PWR type of domestic NPPs was expected to different from that of foreign NPPs, so that as such, correlation properties of H-3 and C-14 with Co-60 as key nuclide were investigated.

In this study, concentration bottom(CB) as radioactive waste, which was generated from a PWR type of NPPs were partially obtained and analyzed for H-3 and C-14 using distillation and wet oxidation method, and their radioactivity concentrations were drawn up to the database. The correlations of each nuclide were performed by means of linear regression with Arithmetic mean or geometry mean. These methods for obtaining scaling factor have been generally introduced by various equations as a function of γ-emitted nuclide by using statistic program. Thus, H-3 and C-14 which were separated and determined from concentration bottom solution, were evaluated whether to have possibility to the most suitable correlation with key nuclide using statistic method.

2. Methods and Results

2.1 Chemical assay of H-3 and C-14 from CB solution

For chemically quantitative evaluating H-3 and C-14, they were separated and determined from CB solution of PWR type of NPPs by wet oxidation and distillation method at the same time. A certain measure of CB solution with 5 g of $K_2S_2O_8$ and 0.5 g of $AgNO_3$ used as oxidizing agent was taken carefully into 200 ㎖ round flask and 3 N H_2SO_4 solution was dropped to that slowly during heating up to 90~96℃ until completely reacting with the material to generate $^{14}CO_2$. The generated $^{14}CO_2$ gas was adsorbed to Carbo-sorb trap (Packard) for C-14 counting. After that, the determination of H-3 was carried out through distillation of residue CB solution. A portion of purified H-3 and C-14 were mixed with Permafluor E+ and Ultima Gold XR cocktail (Pakcard) respectively, and allowed to be stabilized in a cool and dark place for 24 hour to prevent chemiluminescence. The samples were counted using liquid scintillation conuter.

Preliminary experiment results of HTO and $Na_2{}^{14}CO_3$ as radioactivity standard samples showed each 104.9±4.3 and 92.2±3.3 %. Through radioactivity standards samples, the verification tests of recovery rates were performed. The result appeared with being excellent and it was possible to be applicable to radioactive waste assay. With the identical method, the concentration of radionuclides in radwastes was determined to 246.3 ~14,041 Bq/g for H-3 and 8.350~1,381 Bq/g for C-14. Their radioactivity showed to distribute widely, and the concentration of H-3 was about 10 times higher than that of C-14.

2.2 Correlation of H-3 and C-14 with Co-60

Table 1. Determination of correlation coefficient using box-cox prediction method

Correlation Coefficient	Key nuclide	
	Co-60	Log Co-60
H-3	0.669	-
Log H-3	0.858	0.695
C-14	0.753	-
Log C-14	0.837	0.603

To obtain the most suitable correlation of H-3 and C-14 with Co-60 using the regression analysis methods with arithmetic mean or geometry mean, box-cox prediction method was applied. Box-cox prediction method deduces ramda, λ and help converting the data distributed abnormally into normally distributed one. The concentrations of H-3 and C-14 correlated with that of Co-60 were able to be converted to log scale as the ramda, λ was at near 1/2value. The correlation coefficient of H-3 and C-14 of logarithm which was each 0.669 and 0.753 before not being applied log scale, was elevated to each 0.858 and 0.837 as shown in Table 1. These correlation properties using the Box-cox prediction method are shown in Figure 1. In Figure 1, only H-3 and C-14 were applied to log scale, and regression line was expressed to two different methods. That is, a solid line in the regression line was expressed as parametric method which generally becomes known as a fitting method and a dash dot line was known as Theil's method which is the non-parametric method. Theil's method determines the slope of a regression line as the median of the slopes calculated from selected pairs of points, and the intercept of the line is the median of the intercept values calculated from the slope and the co-ordinates of the individual points. Figure 1(a) and (b) shows the interval of two different methods is slightly wide, but two kind of linear equations derived from parametric and non-parametric method have to be all applied when the number of data point is under 30.

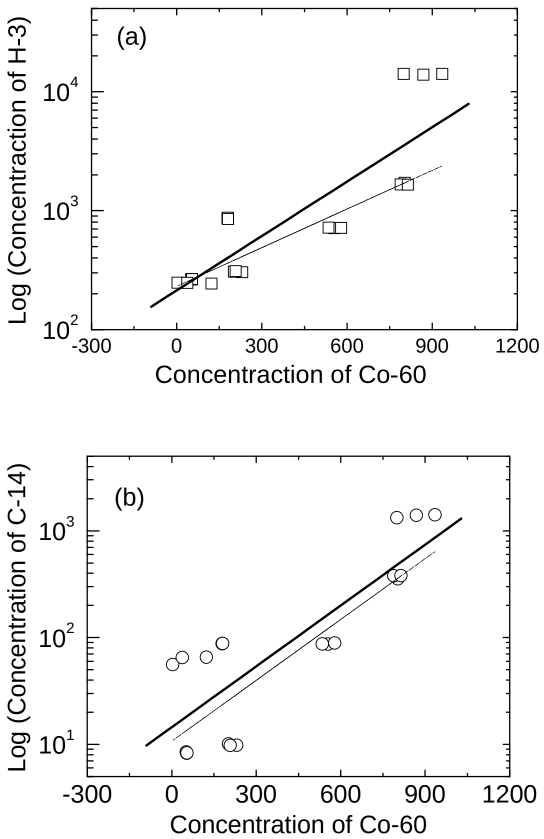

Figure 1. Correlation of H-3 and C-14 with Co-60 by using parametric method(a solid line) and nonparametric method(a dash dot line)

Table 2 shows the correlation coefficients with nuclides were compared between obtained methods which is Pearson product-moment correlation coefficient as parametric method and Spearman rank correlation coefficient and Kendall's tau as non-parametric method. Pearson product-moment correlation coefficient, r which is generally known method, measures the strength of the relationship among observations drawn from bivariate populations. On the other hand, as non-parametric method, Spearman rank correlation, r_s is strictly speaking a measure of the degree of correspondence between the ranks of the sample observations rather than between the observations themselves (x_i, y_i). Like the spearman rank correlation coefficient, Kendall's tau, $\hat{\tau}$ is based on the ranks of observation, and it can assume values between -1 and +1. One of the important differences between $\hat{\tau}$ and r_s is that $\hat{\tau}$ provides an unbiased an unbiased estimator of a population parameter, while the sample statistic r_s does not provide an estimator of a population coefficient of rank correlation[3]. Table 2 represents the correlation coefficient calculated by Pearson and Spearman method was shown in difference slightly but was similar measure to 0.818 ~ 0.857 for H-3 and C-14 correlated with Co-60. Kendall's $\hat{\tau}$ was 0.612 and 0.642 for H-3 and C-14, respectively, which was 27 ~38 % lower than above two sort of methods. But the correlation coefficients for each methods were not comparable with each other because the parameter estimated by $\hat{\tau}$ may be defined as the probability of concordance minus the probability of discordance.

Table 2. Correlation coefficients with radionuclide by using parametric and non-parametric method

Correlation Coefficient		Correlation Coefficient	
		Co-60 & Log H-3	Co-60 & Log C-14
Parametric method	r	0.858	0.837
Non-Parametric method	r_s	0.825	0.818
	$\hat{\tau}$	0.621	0.642

3. Conclusion

H-3 and C-14 determination in radwaste were performed with wet oxidation and distillation method. Their radioactivities were in range of 246.3 ~14,041 Bq/g for H-3 and 8.350~1,381 Bq/g for C-14. Correlations were achieved under plotting H-3 and C-14 of logarithm with Co-60 as the ramda, λ was at near 1/2 value by using parametric and non-parametric method. The correlation coefficient of H-3 and C-14 of logarithm which was each 0.669 and 0.753 before not being applicable to logarithm, was elevated to 0.858 and 0.837 respectively.

REFERENCES

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