

THE STATISTICAL ANALYSIS FOR THE CHARACTERISTICS OF LOW-LEVEL WASTES GENERATED FROM PWR

Young Min Kim and Kun Jai Lee
Korea Advanced Institute of Science and Technology
P.O. Box 150
Cheongryang, Seoul, Korea

ABSTRACT

The characteristics of the radwastes generated from nuclear power plants cannot be directly determined due to the various problems associated with it. Previously, the scaling factors which correlate the hard-to-measure nuclides to the easy-to-measure nuclides are proposed and used as a tool describing indirectly the characteristics of the radwastes. However, the scaling factors are generic only for selective nuclides and must be calculated repeatedly whenever the plant condition changes substantially. To solve the problems, the characteristics of the radwastes must be correlated to the plant's operational history. Since the existing sample data does not provide the information on the characteristics of the radwastes with the plant's operational history, the characteristics of the radwastes should be correlated to the other characteristics of the radwaste which not only can describe roughly the plant's operational condition, but also can be easily obtained in plant routine basis.

The two variables which are correlated each other in this study are adjusted so that the causal relationship between the variables can be easily identified, and are assumed to have the lognormal distribution. By the graphical analysis, the quantity $Q(\text{Co}^{60}) + Q(\text{Cs}^{137})$ is selected as the independent variable and the quantity $Q(i)/[Q(\text{Co}^{60}) + Q(\text{Cs}^{137})]^2$ as the dependent variable, where Q means the activity concentration and i means the various hard-to-measure nuclides. The correlation of which log mean dispersion is less than 10 is considered as a useful calculational formula. The correlation obtained in this study for C^{14} , Sr^{90} , I^{129} (which are very difficult to characterize in previous study) and Fe^{55} turn out to be very good for all types of waste. Since the formation and removal mechanism of TRU is much more complex than that of corrosion and fission products, the correlations for TRU show mostly relatively high log mean dispersions which are greater than 10. The quantity to which the characteristics of TRU are correlated should be improved in the future study.

INTRODUCTION

Since the total amount of low-level wastes generated from nuclear power plants are increasing rapidly, the safe disposal and management of low-level wastes is very important for the aspects of environmental protection and public health. Generally, the potential hazards of the radwaste are deeply related with the radioactivity level of various radionuclides in the radwaste. In that respect, regulations and guides that were developed to represent the waste classification system require that the waste producers identify and quantify the concentrations of the radionuclides in the radwastes (1). To comply with the rule required by the regulations, the plants must determine completely the compositions of the radwaste. However, the determination of composition of radwaste is involved with a number of problems. Sometimes sampling is very difficult and results in considerable radiation exposure to the sampling personnel. Furthermore, several radionuclides are very difficult to detect or expensive to be measured in the routine analyses. Thus, indirect methods which can correlate the hard-to-measure nuclides to the easy-to-measure nuclides are developed and used. As one of the indirect methods, scaling factor method are developed to carry out the correlation.

Previously the representative sample data obtained from various operating plants are statistically analyzed by EPRI (2,3) and SAIC (4), and a theoretical approach de-

veloped by AIF (5) describes a methodology to quantify the concentrations and the amounts of radionuclides of concern in nuclear plant wastes. These results are generic for selective nuclides, and since the existing data does not provide the information on nuclide concentrations and quantities of the wastes with the power plant's operational history, the scaling factors must be calculated repeatedly whenever the plant condition changes substantially. To solve the problems, the characteristics of the radwastes must be correlated to the plant's operational history. However, since the plant's operational history cannot be easily obtained in plant routine basis, it is useful to correlate the plant's operational history to the other characteristics of the radwastes which can be obtained easily in the routine analyses.

In this study, calculational formula are obtained which correlates the characteristics of the radwastes to the other characteristics of the radwastes which can be obtained easily in the routine analyses. The two variables, independent and dependent, which are correlated each other are adjusted so that the causal relationship between the two variables can be easily identified, and are assumed to have the lognormal distribution. By preliminary graphical analysis, the quantity $Q(\text{Co}^{60}) + Q(\text{Cs}^{137})$ is selected as the independent variable and the quantity $Q(i)/[Q(\text{Co}^{60}) + Q(\text{Cs}^{137})]^2$ as the dependent variable, where Q means the activity concentration and i means the hard-to-measure nuclides.

The wastes considered here are ion-exchange resins, evaporator bottoms, filters, DAW and oil, and then overall data for PWR are also considered as a whole. The correlations are obtained by applying the multivariable successive approximation method for ion-exchange resins, evaporator bottoms and filters, while by the method of linear least squares for DAW, oil and all PWR data. The correlation of which log mean dispersion is less than 10 is considered as a useful calculational formula.

GENERATION AND REMOVAL OF RADWASTES IN PWR

The radionuclides in nuclear power plants are generated in fuel, coolant and structural components by fission and neutron activation. Under ordinary operating conditions, the small fraction (< 0.1 percent) of the radionuclides generated in the fuel escape into the reactor coolant. In addition, the radionuclides may be created in the primary coolant due to the neutron activation of corrosion products, chemical additives such as lithium hydroxide or boric acid, and isotopes of hydrogen and oxygen in the primary coolant. Small amounts of radioactivity may also be circulating in the primary coolant by the fissioning of "tramp" uranium which presents as a contaminant on the exterior surface of fuel cladding (1). The formation or the release rate of the radionuclides in the radwaste is related with various plant conditions such as irradiation age of the fuel (burnup), nature of the fuel cladding defect, and chemical and physical perturbations within the reactor coolant system.

The radionuclides generated in the reactor core are free to circulate throughout the entire primary system, including those components of the reactor coolant system designed to remove them. Most of this circulating material is held within the reactor coolant system, and the major means of removal of radioactive material are filters, resins and evaporator bottoms (5,6). The chemical form of the radionuclides will determine, to a large extent, the release rates of the radionuclides from the fuel and activated components and then affect the characteristics of the radwastes. The volatile and nonvolatile (dissolved and undissolved) components of a solution or slurry are separated by evaporation. Suspended, undissolved, particulate solids from a fluid are separated by filtration. The radionuclides which are in ionic form are removed by ion-exchange resins. It is known that relatively significant fractions (about one-fourth) of the corrosion products (especially iron and nickel) tend to be present as suspended solids. Fission products tend to be present dominantly as soluble forms. Transuranic nuclides are very much insoluble in water.

The categories of the waste which input the solid radwaste system are as follows (2,3):

- a. Condensate filter sludges and resins

- b. Clean-up filter sludges and resins
- c. Evaporator concentrates or bottoms
- d. Miscellaneous resins and filters
- e. DAW (Dry Active Waste)
- f. Scrap
- g. Special waste
- h. Oil

WASTE GENERATION MODEL

Generally, since the plant conditions change during normal operation of nuclear power plants, the activity content of the radwaste in a given reactor can be expected to vary. And the specifics of the design of the radwaste system in nuclear power plants have not been standardized and are dependent on each plant characteristics. In order to describe the characteristics of the radwaste generated from a given reactor, the design of the radwaste system for a given nuclear power plant must be well described, and also the characteristics of the radwastes must be correlated with the plant's operational history. So the standard waste generation model which not only can be applied to all radwaste system, but also can represent the plant's operational history is required to describe exactly the characteristics of the radwaste.

In order to formulate the standard waste generation model, it is assumed that at time t , the system reaches equilibrium state throughout the entire system, and material balance approach (5) is applied to each waste process unit. Some factors are transformed to the other type or defined newly to apply the multivariable successive approximation method (7,8). The quantity of isotope i built up on a process unit for process run time T_R , $Q(T_R)$, can be expressed by

$$Q(T_R) = \bar{Q}_{rcs}(t) \bar{F}(t) AF(1 - e^{-\lambda T_R})/\lambda \quad (\text{Eq. 1})$$

where bar means the average for time period T_R , and

$Q_{rcs}(t)$ = total activity inventory of isotope i in the reactor coolant system at time t [Ci],

$F(t)$ = inventory transport factor [sec^{-1}],

$AF = 1 - 1/DF$ = accumulation factor,

T_R = process run time [sec].

From the point of view of describing the characteristics of the radwaste, it is considered useful that the decontamination factor, DF , is replaced by the accumulation factor, i.e., fraction accumulated on process unit. Usually, since the value of the decontamination factor is greater than or equal to 1, the value of the accumulation factor should be between 0 and 1. $F(t)$ denotes the fraction of which the total activity inventory in the reactor coolant system, which are transported to each process unit per unit time [sec^{-1}] for the given

fluid system. Actually, the value of $F(t)$ is always between 0 and 1. After all, Eq.(1) has been chosen as our waste generation model.

CHARACTERIZATION OF RADWASTES

Various radwastes are generated from nuclear power plants. The activity contents and the distribution of each radionuclides in the radwastes are varying from wastes to wastes. In fact, the sample data obtained from the operating nuclear power plants are scattered in wide ranges. Moreover, since some radionuclides are very difficult to detect or expensive to be measured or are in minute quantities in the routine analyses, scaling factor which can correlate those hard-to-measure nuclides to the nuclides which can be measured easily in the routine analyses are developed and used. The scaling factor can be represented by utilizing Eq.(1) as follows:

$$S_i = Q_i(T_R)/Q_k(T_R) \quad (\text{Eq. 2})$$

where i means the scaled nuclides and k means the scaling nuclides. For a specific isotope i , the scaling nuclides are selected through the theoretical background. Ideally, the hard-to-measure nuclides should be scaled to appropriate nuclides which has essentially identical properties in the entire systems. Then regardless of the treatment processes or the plant chemical environment, the two isotopes would track each other reasonably closely. However, because of uncertainties related with the plant condition, the scaling factors are also expected to be scattered in wide ranges. Furthermore, the extent of the scattering of the scaling factors are known to be much larger if the easy-to-measure nuclides are selected as the scaling nuclides. The scaling factors which are obtained previously (2,3,4) by sampling data are generic only for selective nuclides, and must be calculated repeatedly whenever the plant condition changes substantially. For this reasons, it is quite appropriate to perform the statistical regression analysis on the sample data which correlate the characteristics of the radwastes to any plant operational parameter obtained easily in the plant routine basis.

However, since the currently available sample data dose not provide the information on nuclide concentrations and quantities of the wastes with the reactor's operational history, the characteristics of radwastes should be correlated to the other characteristic of the radwaste which not only can describe the plant condition, but also can be easily obtained in the routine analyses. Since Co^{60} and Cs^{137} are easily measured in the routine analyses, the quantity associated with both nuclides are used the correlating quantity (independent variable). When the characteristics of the radwastes are correlated to the quantity associated with Co^{60} and Cs^{137} , it should be noticed that the two variable (the dependent variable X_1 is $Q(\text{Co}^{60}) + Q(\text{Cs}^{137})$ and the independent variable X_2 is $Q(i)/[Q(\text{Co}^{60}) + Q(\text{Cs}^{137})]^2$) are

both stochastic variables. The relationship between the two stochastic variables is characterized by the corresponding two-dimensional distribution, but this characteristic is often supplemented by a regression analysis, where the mean value of X_2 is determined as a function of X_1 , either because variations of X_1 are the cause of variations in X_2 , or sometimes the regression curve furnishes us with a suitable correlation between the two variables (9).

CALCULATIONAL PROCEDURES

Criteria for the two random variables, X_1 and X_2 , are given as follows:

- a. The independent variable X_1 must be the quantity which not only can represent the plant condition, but also be easily obtained in the routine analyses.
- b. A causal relationship between the two variables must be supplied to obtained the significant correlations. Though the causal relationship between the hard-to-nuclides and any key nuclides can be qualitatively identified through the theoretical background, the measure of the relationship obtained quantitatively cannot exactly describe the extent of the causal relationship between the two nuclides because the data sampled, in fact, show the scattered behavior in wide ranges. In that respect, the two variables are adjusted so that the causal relationship or the stochastic dependence between the two variables can be easily identified.

The calculational procedures of the present study are summarized as follows:

First, the added quantity of each activity for Co^{60} and Cs^{137} , $Q(\text{Co}^{60}) + Q(\text{Cs}^{137})$, is selected as the independent variable, X_1 , because it not only can roughly describe the plant condition, but also can reduce the uncertainties associated with the plant condition. The quantity of $Q(\text{Co}^{60}) + Q(\text{Cs}^{137})$ increases approximately linearly for the each quantities of $Q(\text{Co}^{60})$ and $Q(\text{Cs}^{137})$, respectively, judging from the graphical analysis on the sample data. For a given X_1 , X_2 is chosen by graphical analysis such that the selected X_2 can represent the most narrow contour ellipse. The X_2 is expressed by

$$X_2 = Q(i)/[Q(\text{Co}^{60}) + Q(\text{Cs}^{137})]^2 \quad (\text{Eq. 3})$$

where i means the hard-to-measure nuclide. Throughout the analysis, it is assumed that both X_1 and X_2 have a lognormal distribution because the lognormal distribution has been proven to represent the sample data most accurately.

Second, the function which describes the relationship between $\log X_1$ and $\log X_2$ is determined by the following linear function from the graphical analysis.

$$\log X_2 = a + b (\log X_1 - c) \quad (\text{Eq. 4})$$

The assumptions used for the regression analysis are as follows:

- a. $\log X_2$ is normally distributed for every value of $\log X_1$.
- b. The variance of $\log X_2$ corresponding to a given value of $\log X_1$ is constant.

The waste considered here are ion-exchange resins, evaporator bottoms, filters, DAW and oil. And overall data for PWR are also considered as a whole. Since the waste generation model cannot directly applied to DAW and oil wastes, the empirical regression lines for DAW, oil waste and overall PWR data are obtained by the method of linear least squares. The empirical regression lines for resins, evaporator bottoms and filters are obtained by the multi-variable successive approximation method. The waste generation model, Eq.(1), is used to estimate the unknowns (a, b and c) in Eq.(4) and then the factors (terms in the right side of Eq.(1)) in the waste generation model become the corresponding unknown population parameters. In the multivariable successive approximation method, the values of the unknown population parameters are obtained by minimizing the sum of squares of the differences between the observed data and the calculated value, then the empirical regression lines are obtained. The unknown population parameters and their ranges are listed in Table I. The process run time, T_R , is assumed to be between 0 and 1 year because all the radwastes are supposed to be treated at least within one year.

TABLE I

Unknown Population Parameters and their Ranges

Unknown Population Parameters	Column No. of Orthogonal Array	Range
$Q_{rcs}(i)$	1	*
$Q_{rcs}(Co^{60})$	2	*
$Q_{rcs}(Cs^{137})$	3	*
F(i)	4	0-1
$F(Co^{60})$	5	0-1
$F(Cs^{137})$	6	0-1
AF(i)	7	0-1
$AF(Co^{60})$	8	0-1
$AF(Cs^{137})$	9	0-1
T_R	10	0-1 year
Angle of slope	11	0-1 degree

* The ranges are determined by analysis on the sample data.

RESULTS

The factor 10 as log mean dispersion is frequently referred to as the allowable variance in a certain scaling factor from its average value (3). The variation of the random variable X_2 to a given value of X_1 is completely originated from the variations of the activity concentrations of the hard-to-measure nuclides analyzed or the corresponding isotopic ratio because X_2 is, in fact, the isotopic ratio divided by $X_1 [Q(Co^{60}) + Q(Cs^{137})]$. So it can be said that if the standard deviation is less than one in common logarithmic scales, the correlations are considered reasonably achieved. Log mean dispersions determined through the current study are listed in Table II. It is noticed that the calculated log mean dispersion would have significant meanings only when correlated with the quantity of $Q(Co^{60}) + Q(Cs^{137})$. Figures 1 to 8 show the correlations of C^{14} , Fe^{55} , Sr^{90} and I^{129} for ion-exchange resin and all PWR data.

TABLE II

Log Mean Dispersion of Calculational Formula Obtained in the Present Study

	Evaporator					All PWR Data
	Resins	Bottoms	Filter	DAW	Oil	
C^{14}	7	10	10	6	8	10
Fe^{55}	6	3	7	4	9	10
Ni^{63}	19	25	10	12	2	20
Sr^{90}	6	4	5	4	6	7
Tc^{99}	14	11	8	6	8	11
I^{129}	7	6	9	5	5	10
Ce^{144}	5	11	9	14	5	14
Pu^{238}	11	19	10	13	6	18
Pu^{239}	14	12	10	16	4	15
Pu^{241}	10	20	9	9	2	16
Am^{241}	19	21	11	8	2	15
Cm^{242}	8	13	9	8	3	14
Cm^{244}	10	18	11	8	3	16

In this study, the sample data from the survey of all U.S. nuclear plants (2,3) are covered and all the sample data points are treated such that the two random variables become much more strongly correlated as the population size increases. The calculational formula obtained here depends on the population size and so must be supplemented by the other new sample data. In that respect, calculational formula for C^{14} , Fe^{55} , Sr^{90} and I^{129} , which are usually known to be very difficult to characterize from the previous study,

shows quite reasonable correlation because log mean dispersions for all waste type as well as all PWR data are less than 10. The correlation results for C^{14} , Fe^{55} , Sr^{90} and I^{129}

are listed in Table III. The correlations for TRU show mostly relatively high log mean dispersions which are greater than 10, and it is a expected results because the

TABLE III

Correlation Results for C^{14} , Fe^{55} , Sr^{90} and I^{129}

		C^{14}	Fe^{55}	Sr^{90}	I^{129}
Resins	a	-0.3890	0.5107	0.5423	-2.5115
	b	1.4980	1.2886	1.2131	1.7546
	c	-1.4043	-1.0482	-1.0643	-1.1674
Evaporator Bottoms	a	-0.8524	0.6274	0.4238	-2.3038
	b	1.4826	1.4826	1.1009	1.5622
	c	-1.4774	-1.2264	-1.0854	-1.5494
Filter	a	-1.1397	1.0775	0.4909	-3.0058
	b	1.2131	1.0651	1.1009	1.4826
	c	-1.2339	-1.2940	-1.0593	-0.1984
DAW	a	-1.0488	1.2432	0.3877	-2.2926
	b	-1.4795	-0.9998	-1.1297	-1.7151
	c	-1.0039	-1.1280	-1.0381	-1.0518
Oil	a	3.3171	4.5000	4.7490	3.6990
	b	-1.3269	-1.5622	-2.3579	-2.0449
	c	-4.8645	-4.8645	-4.8654	-4.8654
All PWR Data	a	-1.0410	0.7839	0.4344	-2.5568
	b	-1.3600	-1.1273	-1.0708	-1.5426
	c	-1.1203	-1.2004	-1.2003	-1.1571

Note. The correlations are expressed as follows:

$\log X_2 = a - b (\log X_1 - c)$ for resins, evaporator bottoms and resins, and

$\log X_2 = a + b (\log X_1 - c)$ for DAW, oil and all PWR data.

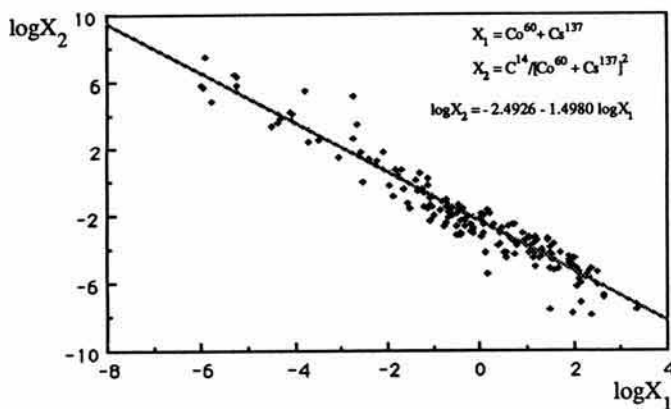


Fig. 1. Correlation of C^{14} for resin data.

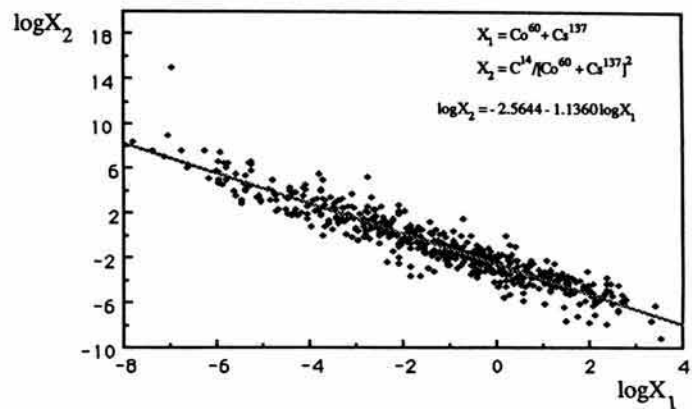


Fig. 2. Correlation of C^{14} for all PWR data.

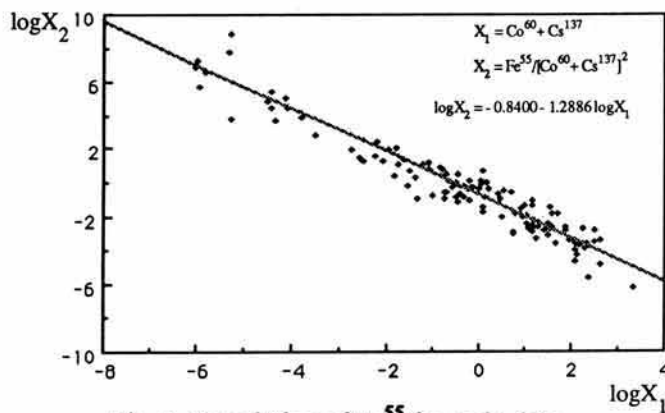


Fig. 3. Correlation of Fe⁵⁵ for resin data.

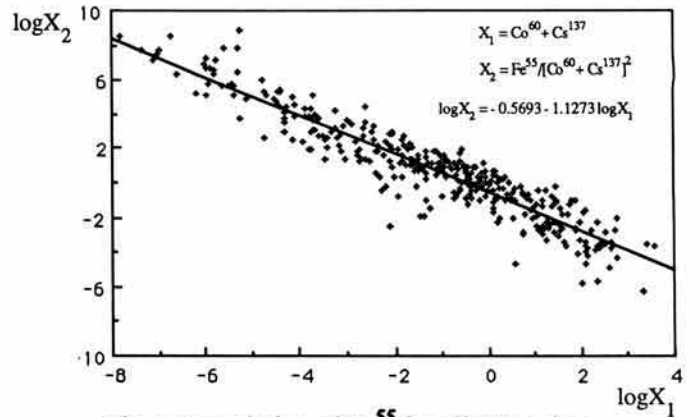


Fig. 4. Correlation of Fe⁵⁵ for all PWR data.

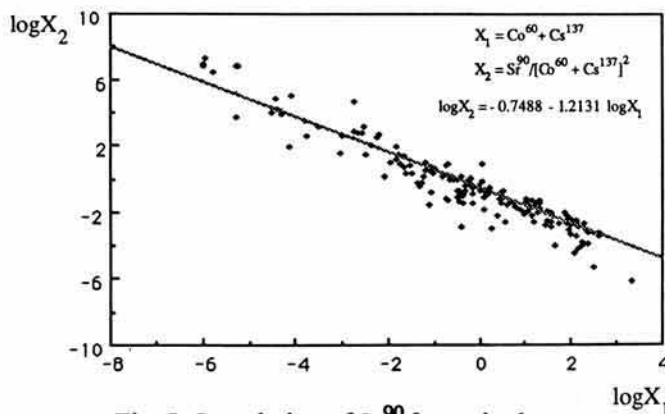


Fig. 5. Correlation of Sr⁹⁰ for resin data.

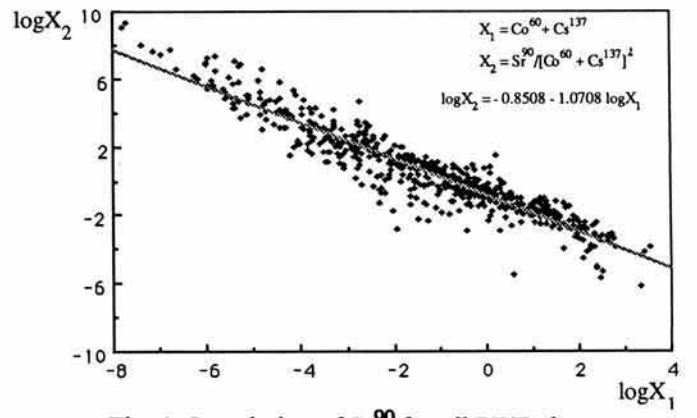


Fig. 6. Correlation of Sr⁹⁰ for all PWR data.

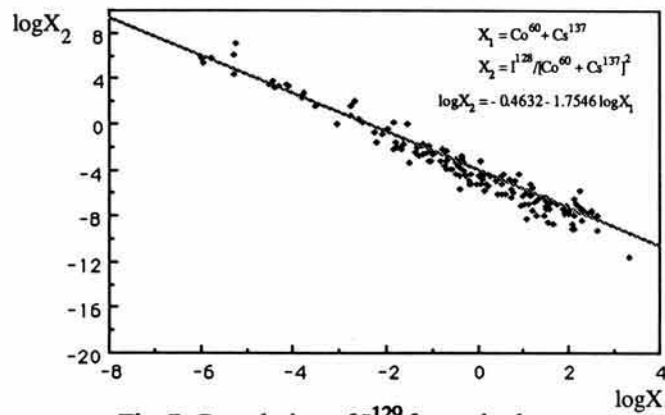


Fig. 7. Correlation of I¹²⁹ for resin data.

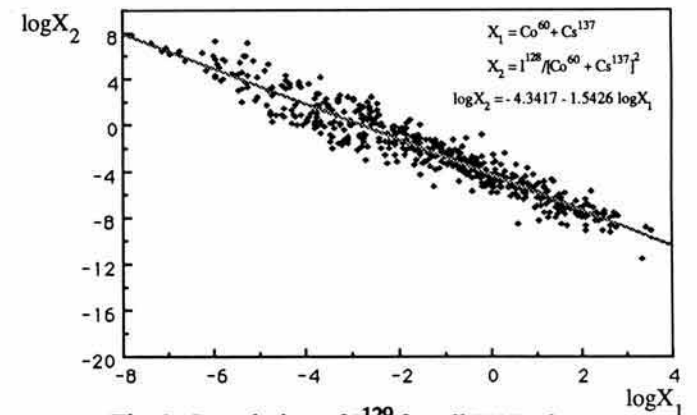


Fig. 8. Correlation of I¹²⁹ for all PWR data.

formation and disappearance mechanism of TRU is more complex than that of activation and fission products. For example, TRU are produced by the decay of other TRU even in the radwaste. The quantity which correlates the characteristics of TRU will be improved as long as the quantity which can describe the formation and removal mechanism of TRU much accurately in physical background is obtained.

CONCLUSIONS

To overcome the difficulty associated with how to correlate the hard-to-measure nuclides to the easy-to-measure nuclides, calculational formula are obtained where some characteristics of the radionuclides are correlated to the other characteristic of the radwastes that can be easily obtained in the plant routine analysis and also can describe the effect of the plant's operating condition. From this study, the following conclusions are drawn;

1. Generally the variations of the characteristics of the hard-to-measure nuclides are greatly reduced when those are correlated to the total activities of Co^{60} and Cs^{137} rather than the independent each activities of Co^{60} and Cs^{137} , respectively, or the isotopic ratio $\text{Co}^{60}/\text{Cs}^{137}$.
2. The characteristics of the activation products except for Ni^{63} in the radwastes can be determined by using the currently developed calculational formula. The previously well-known results are used for the characteristic of Ni^{63} since the results are proven to be generic.
3. The characteristics of Sr^{90} and I^{129} , which are known to be difficult to characterize, can be easily determined by using the currently developed calculational formula.
4. The determination of the characteristics of TRU turned out to be still very difficult even by the our calculational formula as before.

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