61TRAK - AN ISOTOPE BEHAVIOR ANALYSIS PROGRAM

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ABSTRACT

61TRAK is an isotope behavior analysis computer program designed to comply with Title 10, Part 61 of the Code of Federal Registration. As part of the rule, radioactive waste must be classified based, in part, on concentrations of alpha and beta emitting radionuclides. These radionuclides are not routinely measured at most nuclear plants. 61TRAK was developed to derive scaling factors between gamma emitting (easy-to-measure) isotopes and non-gamma emitting (difficult-to-measure) isotopes. These scaling factors can be used directly with certain easy-to-measure isotopes to arrive at concentrations of the difficult-to-measure isotopes. In addition, 61TRAK monitors changing plant condition indicators to determine when reanalysis of the difficult-to-measure isotopes may be required.

INTRODUCTION

The final rule on Licensing Requirements for Land Disposal of Radioactive Waste was implemented by the NRC on December 27, 1983. The final rule adds a new part 61 to 10CFR. The waste classification method of 10CFR61 is based on concentration limits of a specific list of both short-lived and long-lived radionuclides. Several of the radionuclides in the classification list are alpha and beta emitting radionuclides. These radionuclides are not routinely measured at most nuclear power plants. The NRC is suggesting indirect measurement methods for these difficult-to-measure isotopes to demonstrate compliance. The most common indirect measurement method being proposed is that of determining scaling factors between the easy-to-measure isotopes and the difficult-to-measure isotopes. 61TRAK is a user-friendly interactive computer program developed to derive scaling factors between the easy-to-measure and the difficult-to-measure isotopes.

61TRAK is designed to examine relationships and correlations between easy-to-measure isotopes for various plant conditions. This analysis forms the basis of the justification of scaling factors for the difficult-to-measure isotopes. The relationships are established using statistical methods (linear regression analysis of the natural log of the concentrations of the isotope pairs being evaluated) and analytical expressions representing the dominant behavior processes in the purification and waste treatment systems. Once these scaling factors are established, the program can be used to minimize the cost associated with the radiochemical analysis of difficult-to-measure isotopes.

Changing plant condition indicators are also monitored by 61TRAK. This is accomplished through variances in concentration of specific easy-to-measure isotopes relative to statistical values of correlation and standard deviation. The monitoring technique is used to determine when reanalysis of difficult-to-measure isotopes may be required to account for the changes in plant conditions. The basis for the on-going sampling and radiochemical analysis program are thus provided by the 61TRAK analysis.

61TRAK is a user-friendly program that presents options in a menu format. It provides a step-by-step methodology to utilize all capabilities of the program. The program requests input needed for a given operation and specifies the simplified format required.

The program is also programmer-friendly. All FORTRAN coding, as well as the data files, are commented for easy understanding. Complex coding has not been used. Repetitive operations are carried out by small well-documented subroutines. Plant specific data and input which requires updating are well contained in easily accessible parameter libraries.

61TRAK is written in ANSI standard FORTRAN IV and is compatible with FORTRAN V. It was developed on a DEC VAX-11/780 computer. The effort to implement it on other computers would be minimal since no machine dependent computer language is used and because it is a small program, relatively.

ISOTOPE BEHAVIOR ANALYSIS.

Gamma spectroscopy data from several years back are typically obtained from plant historical data files to be input into the 61TRAK data base. The data obtained is from three basic sources: (a) reactor coolant isotopic analyses, (b) solid waste isotopic analyses used for shipping manifests and (c) plant smear data. These data sets were input into 61TRAK to identify quantitative relationships between easy-to-measure isotopes.

The data can be evaluated by data source, reactor operating condition, waste type, waste stream and dates of interest. These evaluations provide insight into the dominant behavior processes in the plant for all operating conditions which have been experienced over the period the data was evaluated. Once the dominant behavior processes and the behavior relationships are developed for the easy-to-measure isotopes, the model is extended to include the difficult-to-measure 10CFR61 isotopes based on chemical and physical similarities between the two groups of isotopes. The equations developed describing the empirical relationships are then used

to calculate scaling factors between the easy-to-measure and difficult-to-measure isotopes. With the scaling factors, concentrations of the difficult-to-measure isotopes can be calculated based on concentrations of key easy-to-measure isotopes.

Statistical Data Analysis

From the initiation of development of 61TRAK, it was recognized that there were uncertainties associated with the historical gamma spectroscopy data which had been collected from the plants. The uncertainties arise from sampling methods, counter efficiencies, counting equipment software limitations, isotope interferences and sample decay times. Additionally, the processes involved in the steps from the point an isotope originates in the fuel or in the neutron flux to the point where it is measured at some concentration in the coolant are very complex and not well understood and are fairly dynamic. These dynamic processes gave rise to uncertainty and variability in the actual data which added to the uncertainty in the interpretation of the data. To account for this, a linear regression analysis was developed for 61TRAK.

A linear regression analysis can be performed for any pair of isotopes sorted singly or by any combination of the following categories: (a) unit number, (b) date, (c) plant system, (d) waste type, (e) plant power level and (f) data source. The program converts all data to the units of μCi/cc and takes the natural log of the concentrations to perform the linear regression analysis. The program performs linear regression analysis using typical least squares fit equations for slope and intercept. 61TRAK also computes the correlation factor and the standard error of the estimate.

Because of the linear regression analysis and the characteristic trend of the data, a straight line association between isotope pairs is typically seen. For the linear regression analysis and statistical evaluations, the slope, intercept, correlation and the standard error of the estimate were determined to characterize the relationship between isotope pairs.

The program calculates 95% confidence limits on the slope and intercept of the derived straight line from the regression analysis. The confidence limits are used as another method of determining the goodness of the fit of the line. This is a measure of the accuracy of the data.

Scattergram plots are generated for each pair of isotopes that linear regression is performed on. The scattergram also includes the calculated statistical parameters associated with the isotope pair.

Isotope Behavior Empirical Models

In the development of the empirical models which describe the relationships between easy-to-measure isotopes, it was recognized that the processes affecting the isotopes were very complex. From their point of origin in the fuel or on the fuel cladding surface to the point they are released to the coolant, the isotopes are not well understood in explicit terms. Also, the processes change with plant conditions further adding to the complexity of the model.

The first step in the model development was to perform regression analysis for isotope pairs on all data in the data file and account for all plant conditions. The isotope data was examined in four basic groupings corresponding to their chemical and physical properties. The first group included the volatile radiohalogens. This is to account for I-129. The second group was the nonvolatile soluble fission products which includes cesium. This group also accounted for strontium and technetium so the models for Sr-90 and Tc-99 could be developed. The third group included the insoluble fission products to account for the transuranics. Finally, the activated corrosion products constituted the fourth group to account for Ni-63.

Volatile Radiohalogen Model

Because the gamma emitting iodines are short-lived, they are not usually seen in any significant concentrations in plant solid waste products. Therefore, there is a necessity to determine the release rate, to reactor coolant, of iodine isotopes relative to a longer-lived isotope such as cesium. The Cs-134/Cs-137 isotope pair was examined to derive the release rates for cesium. In addition, the relationship between two relatively long half-life isotopes was used to derive time constants for processes in the model which are of the same magnitude as the decay constants of the two cesiums. An equation of the following form was postulated:

$$C_i = \frac{K_{cs}\, y\, \lambda_i}{M\lambda_i + f} \prod_{n} (\beta_i)_j = n$$

Where:

C_i	= concentration of ith isotope, μCi/kg
K_{cs}	= radiocesium release rate
y	= fission yield of ith isotope, atoms/fission
λ_i	= decay constant of ith isotope, sec^{-1}
M	= mass of coolant, Kg
f	= purification flow rate, Kg/sec
$\prod(\beta_i)_{j=n}$	= The product of n dominant
n	processes where the ratio of their time-constants to the isotope decay constant is of a magnitude to affect the isotope behavior

A solution of the equation was found for the two cesiums based on plant data to derive numerical values for K_{cs} and the plant-specific constants which describe the dominant model processes.

The equation with the constants for the dominant process derived above was again solved for the I-133/I-131 isotope pair. A second set of constants was determined for the dominant processes with time-constants of a magnitude to affect the relationship of the two isotopes.

The equation with the two sets of constants was solved a final time for the I-132/I-135 isotope pair. A third set of constants was determined for this pair.

Using the equation above and the three sets of constants derived from the cesium and iodine isotope relationships, iodine release rates (K_h) were derived as a function of cesium release rates using the I-131/Cs-137 relationship. The final form of the radiohalogen equation was then:

$$C_i = \frac{K_h' \, y \, \lambda \quad (\beta_1)\,(\beta_2)\,(\beta_3)}{m\,\lambda_i + f}$$

where K_h' is a function of the cesium release rate, $f\,(K_{cs})$.

The equation for the I-129 to Cs-137 scaling factor is:

$$\frac{\text{I-129}}{\text{Cs-137}} = \frac{Y_{129}\,\lambda_{129}\,(M\lambda_{137} + f)\,K_h'\quad(\beta_1')\,(\beta_2')\,(\beta_3')}{Y_{137}\,\lambda_{137}\,(M\lambda_{129} + f)\,K_{cs}\quad(\beta_1)\,(\beta_2)\,(\beta_3)}$$

Where $\beta_1', \beta_2', \beta_3'$ are iodine-related constants

and $\beta_1, \beta_2, \beta_3$ are cesium-related constants.

Nonvolatile Soluble Fission Product Model

Because cesium is included in the group of soluble fission products, the equation developed for cesium was used to predict the concentration of other soluble fission products. The equation used was:

$$C_i = \frac{K_{cs}\;Y_i\;\lambda_i}{M\lambda_i + f}\quad(\beta_1)_i\,(\beta_2)_i\,(\beta_3)_i$$

Where $(\beta_1, \beta_2, \beta_3)\,i$ are constants related to the ith isotope and K_{cs} is the cesium release rate.

It was assumed that the release rate for cesium was equal to the release rate for the technetium fission product isotope. Based on that assumption, the scaling factor for Tc-99 to Cs-137 was derived as follows:

$$\frac{\text{Tc-99}}{\text{Cs-137}} = \frac{Y_{99}\,\lambda_{99}\quad(M\lambda_{137} + f)\,(\beta_1)_{99}\,(\beta_2)_{99}\quad(\beta_3)_{99}}{Y_{137}\,\lambda_{137}\,(M\lambda_{99} + f)\,(\beta_1)_{137}\,(\beta_2)_{137}\,(\beta_3)_{137}}$$

Sr-90 also belongs to the soluble fission product group. A review of industry data on the ratio of Sr-90 to Cs-137 ranged over several orders of magnitude, though. Therefore, the equation derived, which again assumed that the release rate for cesium was equal to the release rate for strontium, tended to overpredict the strontium concentration. To account for the release rate of Sr-90 relative to Cs-137, an adjustment factor of 1/50 was determined from industry wide sample points from various plants. The resulting scaling factor equation for Sr-90 to Cs-137 was derived as follows:

$$\frac{\text{Sr-90}}{\text{Cs-137}} = \frac{Y_{90}\,\lambda_{90}\quad(M\lambda_{137+f})\,(\beta_1)_{90}\,(\beta_2)_{90}\quad(\beta_3)_{90}}{50\;Y_{137}\,\lambda_{137}\,(M\lambda_{90+f})\,(\beta_1)_{137}\,(\beta_2)_{137}\,(\beta_3)_{137}}$$

Insoluble Fission Product Model

Although the transuranics are not fission products they do originate in the fuel and their chemistries, once released to the coolant, are similar. For that reason, the transuranic model was based on the Ce-144 isotope. The half-life of Ce-144 is 284 days which is reasonably long, however, it is short enough to be affected by accumulaton in filters and resin beds.

Using industry-wide plant data, the concentration relationship between Ce-144 and Pu-238 was determined. Then, relationships between the remainder of the transuranics and Pu-238 were determined. The relationships were derived from linear regression analysis of the industry-wide data. The resulting equations using Ce-144 were derived as follows:

$$C_{Pu\text{-}238} = A_1(C_{Ce\text{-}144})^{a1}$$
$$C_{Pu\text{-}239} = A_2(C_{Ce\text{-}144})^{a2}$$
$$C_{Pu\text{-}241} = A_3(C_{Ce\text{-}144})^{a3}$$
$$C_{Pu\text{-}242} = A_4(C_{Ce\text{-}144})^{a4}$$
$$C_{Am\text{-}241} = A_5(C_{Ce\text{-}144})^{a5}$$
$$C_{Cm\text{-}242} = A_6(C_{Ce\text{-}144})^{a6}$$
$$C_{Cm\text{-}244} = A_7(C_{Ce\text{-}144})^{a7}$$

where A_1 through A_7 and a1 through a7 are constants derived from the linear regression analysis.

Because Ce-144 is not always found in the "in-plant" gamma spectroscopy analysis of plant wastes, the industry-wide data for transuranics were evaluated against Co-60. Again, the relationships were derived from 61TRAK's linear regression analysis of industry-wide data. A linear curve was found with reasonably high correlation. Although there is no transport model available which supports the high correlation, a statistical relationship was found. In the absence of a Ce-144 measurement, Co-60 could be used to scale the transuranics. The resulting equations using Co-60 were derived as follows:

$$C_{Pu\text{-}238} = B_1(C_{Co\text{-}60})^{b1}$$
$$C_{Pu\text{-}239} = B_2(C_{Co\text{-}60})^{b2}$$
$$C_{Pu\text{-}241} = B_3(C_{Co\text{-}60})^{b3}$$
$$C_{Pu\text{-}242} = B_4(C_{Co\text{-}60})^{b4}$$
$$C_{Am\text{-}241} = B_5(C_{Co\text{-}60})^{b5}$$
$$C_{Cm\text{-}242} = B_6(C_{Co\text{-}60})^{b6}$$
$$C_{Cm\text{-}244} = B_7(C_{Co\text{-}60})^{b7}$$

where B_1 through B_7 and b1 through b7 are constants derived from the linear regression analysis.

Activation Product Model

Because of the absence of suitable basic model parameters, an acceptable generic model for the activation products could not be developed. Primarily, the quantity of the corrosion product parent species residing in the core neutron flux could not be accurately determined. Subsequently, the relationship between Ni-63 and Co-60 was derived based on industry-wide data. A 61TRAK linear

regression analysis of the Ni-63/Co-60 was computed for both PWR's and BWR's. Both had consistently high correlation factors from plant to plant, typically above .90. Based on these linear regression analyses, the following equation was derived:

$$C_{Ni-63} = A(C_{Co-60})^a$$

where A and a are constants derived from the linear regression analysis.

C-14 Model

C-14 is another difficult-to-measure nuclide included in Part 61 and required to be reported on manifests. A very small percentage finds its way into solid waste streams. Typically, it is released with various gaseous waste streams. Because of its means of production, C-14 does not lend itself to the development of a generic model. Though, through 61TRAK's linear regression analysis, C-14 was found to correlate well with Co-60. There are no logical chemical or nuclear characteristics that predicts this relationship even though correlations were usually above .83. For this reason, an equation was developed to predict its concentration instead of using a single concentration for all waste streams. Based on these linear regression analyses, the following equation was derived:

$$C_{C-14} = K(C_{Co-60})^k$$

where K and k are constants derived from the linear regression analysis.

Model Validation

The four isotope behavior models and the C-14 Model discussed above were checked against actual measurements of the 10CFR61 isotope concentrations made for several plants, by radiochemistry laboratories. For the most part, the comparison showed good agreement between calculated and actual concentrations. Some of the differences that occurred were attributed to plant specific parameters which affect the equation coefficients for some of the model's equations.

PLANT CONDITION INDICATORS

To account for some of the plant specific parameters, each model's basic equation was modified to reflect specific plant condition indicators. An evaluation was made to determine which linear regression analysis of gamma emitting isotopes would most closely track each basic model during typical plant transients. Once the trend was established for a model, the model's basic equation was modified to incorporate the trending. By doing this, changes in the plant's operation could successfully be monitored to allow for variances in the difficult-to-measure isotopes.

It was decided that the best gamma data to use would be from reactor coolant since that was the initial source of all the waste streams. Theoretically, that meant scaling factors could change slightly every day if coolant isotopics were input daily into 61TRAK. Obviously, it was not realistic to change the scaling factors daily, so a scaling factors review was incorporated into the procedures, for operating 61TRAK, at times of shipment. The change that occurs is only a small change and designed to account for typical small shifts that occur in plant parameters.