ESTIMATION OF GASEOUS RADIOACTIVITY RELEASE RATES FROM AN OPERATING BOILING WATER REACTOR

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Abstract

Noble gas activity in BWR stack effluent is commonly estimated by applying a graphic curve-fitting technique to activity measurements on grab samples taken periodically at the steam jet air ejector. It is shown that the graphic solution may conceal significant anomalies in these data, which may be more readily discerned by changing the form of the plot. A better fit to the data is obtained by the method of least squares.

Introduction

Significant quantities of radioactive isotopes of the noble gases, krypton and xenon, are produced in fission. MEEK and RIDER[1] have recently published a tabulation of fission yields for several fissile materials and most of the known fission products. A portion of these products find their way into the primary reactor coolant; the remainder are trapped in the fuel elements. In a boiling water reactor (BWR) most of the gaseous fission products in the primary coolant are removed at the steam jet air ejector (SJAE). After a holdup period to permit decay of much of the short-lived activity and after filtration to remove particulate daughter products, the remaining radioactive gases are vented to the atmosphere via the stack.

Estimation of the radiation dose to the population in the vicinity of a BWR depends on a knowledge of the identity and concentration of the radionuclides present in the stack plume. Twenty-seven radioactive isotopes of krypton and xenon, together with their radioactive decay products are found in the plume. Only about thirteen, plus their decay products, are normally present in sufficient quantity to make them of more than academic interest. As it is impractical to measure the concentration of all important constituents, a combination of measured and calculated values is used to estimate stack activity release rates. Significant overestimation of activity release rates unnecessarily penalizes the public in the form of added costs of power production; underestimation results in a population dose greater than anticipated. For the protection of the public, reactor operating licenses place limits on permissible activity release rates.

Data Acquisition

To demonstrate compliance with radioactivity release rate limits, nuclear power plants are equipped with a continuously operated stack monitoring system - usually consisting of a sodium iodide, NaI(Tl), detector together with associated electronic and recording equipment. It is not used in a spectroscopic mode, but measures gross activity release rates at the stack. In addition periodic grab samples are taken from the SJAE effluent and analyzed to quantify the isotopic activity release rates. The analysis commonly makes use of an additional NaI(Tl) detector or Ge(Li) detector and multichannel analyzer in the plant laboratory. The resulting spectrum is analyzed to estimate SJAE activity release rates for three krypton and three xenon isotopes. Elaborate spectrum stripping techniques are generally beyond the capability of most power plants. It is necessary to plot the data and use a curve fitting technique to interpolate and extrapolate the data to estimate the activity release rates for up to twenty-one additional noble gas isotopes.

Present Data Handling Method

On theoretical grounds and experimental evidence BRUTSCHY[2] has postulated that the isotopic activity release rates (A_i) for each of the noble gas isotopes can be fitted by a second order equation in the one-half power of the radioactive decay constant (λ_i):

$$A_i/y_i = b_o + b_1\lambda_i^{1/2} + b_2\lambda_i \qquad (1)$$

where y_i is the fission yield (atoms/fission) of the ith isotope and the b_i are constants to be determined. An alternative form for eq. (1) is obtained on dividing by λ_i:

$$A_i/y_i\lambda_i = b_2 + b_1\lambda_i^{-1/2} + b_o\lambda_i^{-1} \qquad (2)$$

It is further postulated that each of the terms in eq. (2) represents activity released from the fuel via a different escape mechanism. Then eq. (2) may be separated into three component equations:

$$(A/y\lambda)_{I} = b_2 \qquad (3)$$

$$(A/y\lambda)_{II} = b_1\lambda^{-1/2} \qquad (4)$$

$$(A/y\lambda)_{III} = b_o\lambda^{-1} \qquad (5)$$

where the subscripts on the variables have been dropped for convenience. Taking logarithms of both sides of each of the last three equations it is seen that, on a logarithmic plot of $A/y\lambda$ vs. λ, eq. (3) results in a straight line of slope zero; eq. (4), a straight line of slope -1/2, and eq. (5), a straight line of slope -1. The constants b_1 and b_2 are obtained from the intercepts of these lines with the vertical at $\lambda = 1$. These relationships form the basis of a simple graphic technique for estimating the individual and total activity release rates for the 27 krypton and xenon isotopes.

It is further postulated that the iodine isotope concentrations in the bulk coolant should fit an equation identical in form to eq. (2), and that the iodine data and the noble gas data share a common value of b_2, but different values of b_0 and b_1. In practice, the value of b_2 obtained from the iodine data is used arbitrarily and only b_0 and b_1 are determined from the noble gas data.

The measurements and resulting activity release rate estimates are commonly made on a monthly or weekly schedule and form the basis for calibrating the stack monitor after the SJAE activity release rates are corrected for radioactive decay during holdup prior to release at the stack.

Recent Data Handling Experience

The authors have recently made field measurements of airborne alpha, beta and gamma activity on a continuous basis over a period of several weeks in the vicinity of an operating BWR. The monitor is sufficiently sensitive to permit on-line measurements of airborne activity at levels down to the natural background. The monitor has been described elsewhere[3] and some of the results of the survey are the subject of a separate paper[4].

In order to be able to correlate the activity measurements in the field with stack-release data the plant records of the (weekly) measurements of the radioactivity of the SJAE effluent samples were examined, together with the stack monitor record. It was confirmed that the graphic technique had been applied on a consistent basis and that the necessary supporting computations were arithmetically correct. However inspection of the plots of the data and the curves fitted to the data by the graphic technique indicated that the fitted curves did not appear to be good fits to the data. Therefore the data were fitted by the method of least squares, using eq. (2) (the same form of the equation used for the graphic solution). A double-precision program was used on a PDP-10 computer.

With only six data points per plot, one hesitates to ascribe a great deal of significance to the results of various statistical measures of the goodness of fit. However one expects that a consistent improvement (decrease) in the residual sum of squares should be indicative of a better fit to the data. The residual sum of the squares is defined as

$$RSS = \sum_{i=1}^{N} (y_i - Y_i)^2 \qquad (6)$$

where N is the number of data points, y_i is the measured value of the dependent variable, and Y_i is the curve-fit value of the dependent variable. (A least squares solution, by definition minimizes the value of RSS for the form of equation to which the data are fitted.) Twelve sets of weekly data were examined. Not only did the fitting to $A/y\lambda$ fail to improve the average goodness of fit, but the coefficient (b_2) of the second-order term was consistently negative. A negative coefficient is mathematically satisfactory but it conflicts with the theoretical postulate that each term of the equation represents activity released via a different mechanism (how to postulate a negative escape mechanism?). The coefficient was sufficiently negative that it resulted in predicting negative radioactivity release rates for several of the shorter half-life isotopes - in conflict with both theory and experience.

When the data are replotted, on a linear scale, using eq. (1), the reason for the negative coefficient for the quadratic term becomes clear. Figure (1) is a typical example of the data. The abscissa scale is in units of $\lambda^{1/2}$, with λ in sec^{-1}. The ordinate scale is arbitrary. Only by brute force can one fit such data to a quadratic equation with a positive second order term. In practice this is accomplished by using the value of b_2 obtained from the iodine measurements (curves labeled 1 & 2).

It should be noted that the noble gas data spans values of $\lambda^{1/2}$ from 0.001 to 0.030 $sec^{-1/2}$. The abscissa scale must be extended to 0.85 $sec^{-1/2}$ to include all of them. It is asking too much of any curve-fitting technique to provide reliable extrapolation over such a range. However extending the abscissa to 0.060 $sec^{-1/2}$ will include all isotopes which remain in significant quantity after a fifty minute holdup prior to release from the stack. (From the standpoint of a potential accidental release within the plant after only a brief holdup it would be useful to have a technique which could be used with confidence to estimate SJAE activity flow rates for the shorter-lived isotopes.) The critical unmeasured long-lived isotope is Kr-85. Its value of $\lambda^{1/2}$ is 0.45 x 10^{-4} $sec^{-1/2}$, essentially zero on our abscissa scale. Because of the shape of the data and the resulting poor fit of both curves #1 and #2, there is probably a factor of two uncertainty in the estimated emission rate of Kr-85 for these data.

A negative second order term which results in estimations of negative activity release rates is unacceptable, a priori. It is also inconsistent with the three-mechanism concept. So the brute force approach may be the best that can be done with data which is inconsistent with the model. We have used the method of least squares to accomplish this type of force-fitting, as well as to examine the behavior of RSS (eq. 6) when the noble gas data are fitted to equations (1) and (2) under varying assumptions. Figure (1) shows the resulting curves.

The lowest value of RSS (best fit in the sense of least squares) was consistently obtained when the data were fitted using (A/y) (curve #3) as the dependent variable (eq. 1) rather than (A/yλ) (curve #4) (eq. 2). It is not understood why this should be so. (As expected, the second order terms remained negative) The reduction in RSS in going from A/yλ to A/y was commonly by a factor of five and occasionally more than an order of magnitude.

Since the least squares method, applied directly to the noble gas data, resulted consistently in negative second order terms in both equations (1) and (2), it is clear that arbitrarily changing the value of this coefficient to some positive value must result in worsening the goodness of fit (increasing

RSS). When a least squares fit is performed first on the iodine data (curve #2) to determine the value of b_2 to be used in fitting the noble gas data, eq. (1) is reduced to a first order equation in $\lambda^{1/2}$ whose remaining constants (b_0 & b_1) are also found by the method of least squares. The resulting value of RSS lies between that for fitting to A/y and that due to the graphic solution. It is consistently closer to RSS(A/y) when the plotting variable (A/y) is multiplied by the fission yield (y) to give isotopic activity release rates, the discrepancy between the graphic and the least squares results is on the order of five to fifty percent for the data examined. There appears to be a consistent bias in the graphic results yielding lower release rates at longer half-life and higher release rates at shorter half life than the release rates estimated by the method of least squares (cf. curves #1 & #2). It is unclear whether this observation is due to inherent bias in the graphic technique or to the peculiar shape of the data examined. Neither the graphic nor the least squares method yields any information about the reason for the unexpected shape of the data. The study is continuing and it is expected that there will be an opportunity to examine data from other time periods for the same BWR to determine whether the "misshapen" data were peculiar to the particular time period of the study. Ad interim, it is impossible to know whether either technique results in over- or under-estimating absolute levels of radioactivity release for the period studied.

Conclusions

We conclude that the use of the method of least squares on the data for the iodine isotopes in the bulk coolant and on the data for the noble gas isotopes in the steam jet air ejector effluent is preferable to the graphic solution, given the availability of a programmable calculator or computer. We also note that the graphic solution itself can be reduced to a simple routine suitable for a programmable calculator. This should be done to eliminate errors of human plotting judgment in accomplishing the solution, whenever a programmable calculator or computer is available - if it is determined that there is a valid argument for not replacing the graphic method with the method of least squares.

It is suggested that BWR operators more closely examine the data they are acquiring by spectroscopic analysis of bulk coolant iodine levels and SJAE off-gas noble gas levels. The discrepancies discovered in our data are not readily apparent from the usual log-log plots of $A/y\lambda$ vs. λ and an uninquisitive mechanical application of the graphic method; they are apparent by inspection of a linear plot of A/y vs. $\lambda^{1/2}$.

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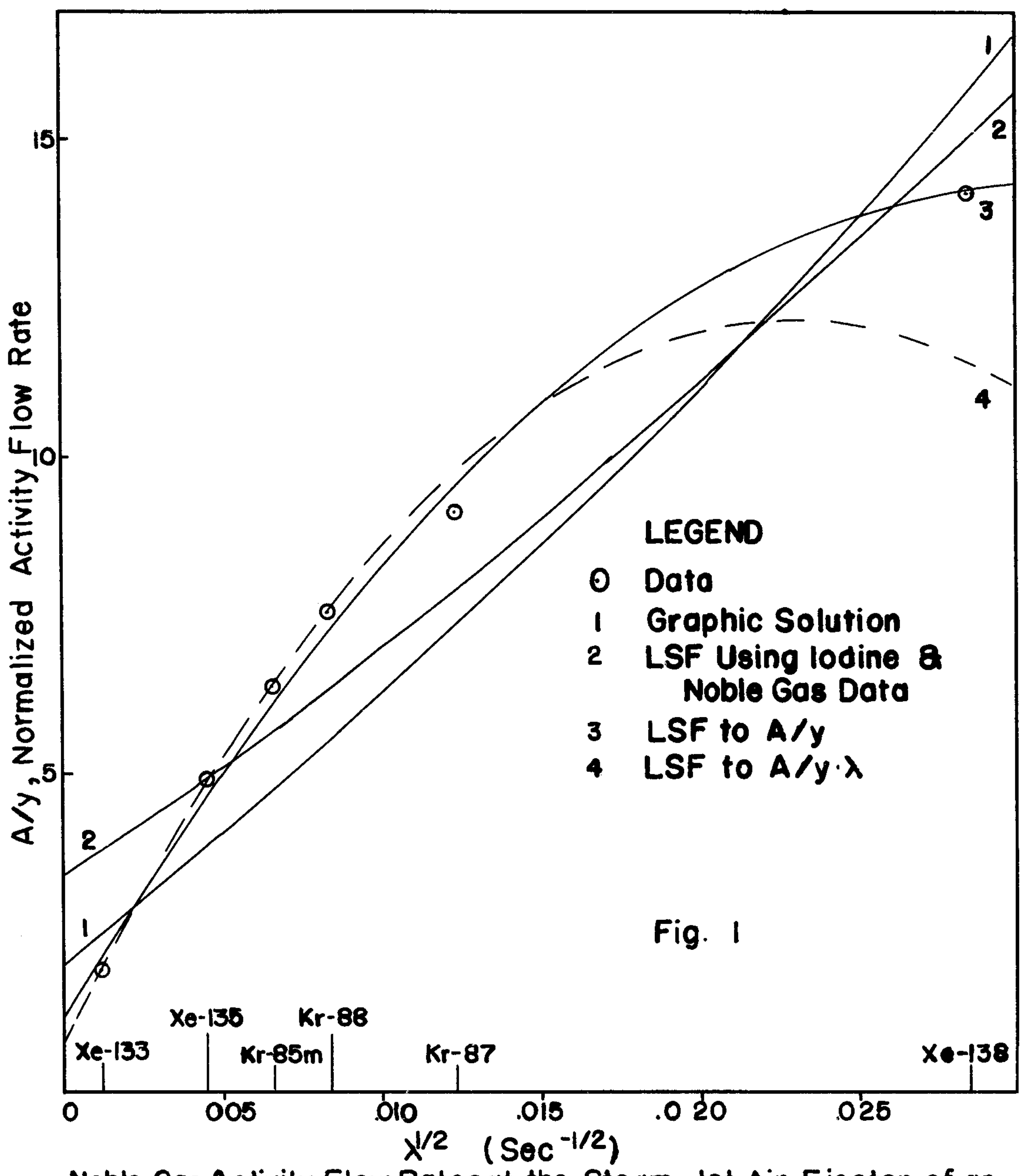

Noble Gas Activity Flow Rates at the Steam Jet Air Ejector of an Operating BWR

Results of Selected Curve Fitting Attempts