

# A Method of Evaluation of the Decay Constants of Two Radioactive Species Present in a Radioactive Sample from Decay Data

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**ABSTRACT:** Retrieval of decay constants from radioactive decay data is a parameter optimization problem. It may at times, become vexatious if two decay constants of two radioactive species, decaying through common emission, are to be estimated from the count data collected from a sample containing the two species. The tool used is Nelder-Mead Simplex routine. The QBASIC program written by Sivaramakrishnan for use in chemical kinetics study has been appropriately modified and used here. The present method is found to yield accurate results.

**KEYWORDS:** radioactivity, decay constants, parameter optimization.

## I. INTRODUCTION

The activity of a radioactive specimen with two species undergoing radioactive decay through parallel common emission is represented by the equation

$$X_t = A_t + B_t = A_0 e^{-\lambda_A t} + B_0 e^{-\lambda_B t} \quad (1)$$

There is no direct and simple procedure to estimate the decay constants,  $\lambda_A$  and  $\lambda_B$  and the amounts of the two species  $A_0$  and  $B_0$  from this non-linear equation, even if sufficient experimental data are available, as the data always contain random errors. A numerical method can be employed to solve the above equation using experimental data. It yields different sets of solutions with different combinations of the parameters without any hint as to which of them is physically true. Therefore, a statistical method, that seeks to minimize the impact of the errors in the data, is required.

A method consistent with the above requirements, has been designed and tested for its efficiency in this work. The tool used here is Nelder-Mead simplex minimization [1] routine, which requires a large number of data. As real experimental data to the required size are not available, it has been resorted to simulation of data. The present technique is found to yield encouraging results.

## II. INITIATION

Our initiation into this field of research was prompted by the work of the researchers in the field of chemical kinetics study. A two-step, consecutive, first order chemical reaction is governed by the equation [2]

$$X_t = P_t + Q_t = P_0 e^{-k_1 t} + Q_0 e^{-k_2 t} \quad (2)$$

Sivaramakrishnan [3] employed the method of Lanczos [4] for the estimation of the two first order rate co-efficients in a two-step consecutive first order chemical reactions. Though there are many such attempts to optimize rate co-efficients [5, 6, 7, 8, 9], the method of Sivaramakrishnan is found to be elegant and to yield better results. The method

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allows the use of data with moderate errors. The computer program written by Sivaramakrishnan was used by Sukumar [10] and Kanemaney [11] for similar studies. This method, with appropriate modification, has been used in this work for the recovery of  $\lambda_A$ ,  $\lambda_B$ ,  $A_0$  and  $B_0$ .

### III. PRESENT WORK

i) Selection of the form of the system equation:

If  $\lambda_A$ ,  $\lambda_B$ ,  $A_0$  and  $B_0$  are to be recovered, it becomes optimization in four dimensions. However, it could be reduced to three dimensional search by changing  $X_t$  into the fraction of the system yet to disintegrate at time  $t$ .

$$Y_t = \frac{X_t}{A_0 + B_0} = \left( \frac{A_0}{A_0 + B_0} \right) e^{-\lambda_A t} + \left( \frac{B_0}{A_0 + B_0} \right) e^{-\lambda_B t}$$

$$Y_t = L e^{-\lambda_A t} + (1 - L) e^{-\lambda_B t} \quad (3)$$

When 70% of the specimen has decayed  $Y_t = 0.3$ . The parameters to be optimized are  $\lambda_A$ ,  $\lambda_B$  and  $L$ .

ii) Parameter optimization:

The least squares norm due to Gauss is almost universally employed for the parameter optimization. Parameter optimization, in this work, has been carried out under the least squares norm using Nelder-Mead simplex routine. The optimized parameters are those which give the least value for the objective function,

$$F_0 = \sum_{i=1}^N [Y_{t_i} - \hat{Y}_{t_i}]^2 = \sum_{i=1}^N [Y_{t_i} - (L e^{-\lambda_A t_i} + (1 - L) e^{-\lambda_B t_i})]^2 \quad (4)$$

where  $(Y_{t_i}, t_i)$  are the experimental data and  $\hat{Y}_{t_i}$  are the absolute values calculated through eqn.(3) for the guessed values of the parameters at  $t_i$ .

Larger the number of data closer the estimates to the true values and hence the necessity for simulation. It was decided to use as many as 800 datum samples per run, extending upto 70% of the common emission. The program in QBASIC, written by Sivaramakrishnan was used for finding the time for 70 % of the emission process. This time divided by 800 gives the uniform time interval between successive datum samples.

Simulations of the datum samples consists in incorporating errors that obey Gaussian distribution into the error-free data generated through eqn.(3). A sequence of 800 Random Normal Deviates is taken from the Rand Corporation's Tables [12] and incorporated into the set of 800 error-free data at any desired error level  $\sigma$  by subtracting the product of  $\sigma$  and each RND from the corresponding error-free datum. The different error levels used are:  $\sigma = 0.002, 0.005, 0.010, 0.015, \text{ and } 0.020$ . As many as twelve replicates have been obtained by selecting twelve sequences of RND's at random from the Tables.

The Nelder-Mead routine starts with an initial simplex. The simplex in 3-dimensional optimization is a tetrahedron with four vertices. If one of the four is chosen as origin, the other three points define the vectors of the three parameters to be optimized. These vectors span the 3-dimensional vector space.

The objective function is calculated at each vertex of the simplex and the worst point giving the highest value of  $F_0$  is shifted to a better position identified through a series of operations as per the prescribed scheme. A new simplex is constructed by substituting this position for one of the vertices of the original simplex and the process is repeated.

In the absence of any anomalous step, the QBASIC program written by Sivaramakrishnan, makes the simplex to ooze down the valley and pass through the best point by contracting itself in all directions finally pulling itself around the minimum in the error hypersurface. The values of the parameters corresponding to the minimum are the optimized estimates of the parameters.

Judicious specifications of the decay constants are required to be made confining to the range 1 to 100 for  $\lambda_A / \lambda_B$ .  $\lambda_A$  is assigned the value  $0.1(\text{time}^{-1})$  and  $\lambda_B$  the values 0.08, 0.05, 0.03, 0.02, 0.01, 0.005, 0.002 and 0.001. The following values of  $L$  have been considered to be appropriate as they cover the entire range from 0 to 1:  $L = 0.1, 0.3, 0.5, 0.7 \text{ and } 0.9$ . In all cases,  $8 \times 5 = 40$  combinations of  $\lambda_A$ ,  $\lambda_B$  and  $L$  have been used.

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The error levels considered are 0.002, i.e., 0.2%, 0.005, 0.010, 0.015 and 0.020. As there are twelve replicates, it works out to 60 sequences of RND's with 800 RND's in each sequence.

With 40 parameter combinations and 60 RND sequence, it works out to 2400 simulation exercises. In addition, the program was run with 45 initial simplexes (45 starting points). There are, therefore, 45 outputs for each exercise corresponding to the 45 starting points, i.e., in each of the 2400 simulation runs of the program 45 outputs are obtained. Each of the 45 outputs of a set contains the estimates of the parameters in that set. The one output with the least value for  $F_0$  gives the optimized values of the parameters. The ultimate mean of such optimized values of each parameter is the best fit value of that parameter.

## IV. GENERAL FEATURES OF THE OUTPUTS

The proposed method leads to a single minimum for the objective function. However, one simulation is yielding a somewhat flat minimum with many points in the error hypersurface having equal or nearly equal values for  $F_0$ .

In some of the outputs in each set,  $\lambda_A$  and  $\lambda_B$  are found to be interchanged along with  $L$  and  $(1-L)$ . Therefore, only those sets of outputs with higher value for  $\lambda_A$  have been recorded in the results. The errors of 1.7% in  $\lambda_A$ , 0.5% in  $\lambda_B$  and 3.7% in  $L$ , in a typical set, indicate the level of accuracy in the estimates upholding the efficiency of the present method.

It is of no guarantee that there exists only one minimum on the error hypersurface. As the error is symmetric about the  $\lambda_A$ ,  $\lambda_B$  and  $L$  axes, one would quite naturally, expect there to be two minima with the same value for  $F_0$ . In that case, two sets of parameters with  $\lambda_A$  and  $\lambda_B$  interchanged are obtained. The situation is not either that simple always. There are many instances [3, 10, 11, 13] where the outputs contain estimates of the parameters, which differ from one another moderately or sometimes even widely in the case of chemical kinetics study. In addition to the global minimum, which corresponds to the lowest value for  $F_0$ , there may even be more than a dozen other minima in the error surface.

The existence of multiple minima complicates the evaluation of the parameters unless the local minima are significantly removed from the global minimum. Otherwise, the assignment of the decay constants  $\lambda_A$  and  $\lambda_B$  and the system property  $L$  requires a careful analysis.

There may also arise a few more complications, two of which are mentioned here. The decay constant  $\lambda_A$  and  $\lambda_B$  may approach each other and become identical at some error level  $\sigma$ . This kind of "coalescence" may lead to wrong conclusion that two decay processes have identical decay constants even when it is not so. The other is the unfortunate situation of there occurring negative value for one of the optimized decay constants. It is, therefore, essential to evolve a criterion for successful resolution of radioactive kinetics.

## V. CONCLUSION

Many workers have determined single decay constants with less than 1% error. Such level of accuracy cannot be expected in the case of resolution of two or three parameters. Zhang et al [14], using 1800-6000 photometric absorbance readings, estimated the four rate co-efficients of a three step, consecutive chemical reaction with errors as high as 60%. While such errors are commonly expected and also accepted, the present optimization technique, yielding the decay constants with less than 2% error, is believed to be accurate.

A detailed account of such complications as the ones mentioned in the last section and the essential criterion for successful resolution of the radioactive kinetics in spite of the complications have been completed and will appear elsewhere along with results of our attempt on four dimensional optimization of the radioactive parameters.

It is aimed at applying the present method to appropriate radioactive samples. When the experimental data are supplemented with simulated data, the search could be made on a much wider range and the optimized parameters are expected to be much closer to the true values.

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