necessary, printout of answers, and tabulation of collected data will take about ten seconds of machine time.

REFERENCE

[1] R. Von Holdt, *Proceedings of the Western Computer Conference* (1959).

(1-3) CLSQ, THE BROOKHAVEN DECAY CURVE ANALYSIS PROGRAM[1]

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INTRODUCTION

A program for the analysis of multicomponent decay curves by a least-squares procedure has been coded for an IBM 7090 computer. The FORTRAN language has been used for the main calculation and FAP for some of the subroutines. Provision for determining half-lives of the nuclear species is provided by an iterative routine starting from a set of trial values. The general philosophy adopted in coding this problem has been to give the user considerable flexibility in data handling.

MATHEMATICAL METHOD

The data of a radioactive decay curve consist of n measurements of the counting rates, f_i, of the sample at times t_i. If m independent nuclear species are present, then the set of data satisfies n equations of the form

$$f_i = \sum_{j=1}^{m} x_j e^{-\lambda_j t_i} + v_i , \qquad (1)$$

where an individual term in the sum, $x_j e^{-\lambda_j t_i}$, represents the contribution of the jth component to the total activity at time t_i. The residual, v_i, at that point is due to statistical fluctuations and experimental errors. Since the m coefficients x_j enter

[1]Research performed under the auspices of the U.S. Atomic Energy Commission.

these equations linearly, a solution by the least-squares method is possible. The condition for such a solution is that

$$\sum_{i=1}^{n} p_i v_i^2 = \text{minimum} , \tag{2}$$

where p_i is the weight assigned to the square of each residual. In terms of the standard deviation, σ_i, of the ith counting rate

$$p_i = 1/\sigma_i^2 . \tag{3}$$

It is convenient to adopt the matrix notation of Hamilton and Schomaker [1] and used by Harmer [2]. In this notation, Eqs. (1) and (2) become

$$F_{n1} = A_{nm} X_{m1} + V_{n1} , \tag{4}$$

and

$$V'_{n1} P_{nn} V_{n1} = \text{minimum} . \tag{5}$$

In Eqs. (4) and (5) the subscripts indicate the dimensions (rows and columns respectively) of the matrices. The symbol V'_{n1} represents the transpose of matrix V_{n1}. The least-squares solution for the matrix of the unknown coefficients, X_{m1}, is given by

$$A'_{nm} P_{nn} F_{n1} = A'_{nm} P_{nn} A_{nm} X_{m1} . \tag{6}$$

To solve this equation for X_{m1}, we define

$$B_{mm} = A'_{nm} P_{nn} A_{nm} . \tag{7}$$

The B_{mm} matrix is inverted to obtain B^{-1}_{mm} and the solution for the unknown coefficient matrix is given by

$$X_{m1} = B^{-1}_{mm} A'_{nm} P_{nn} F_{n1} . \tag{8}$$

The variance of the ith coefficient is obtained from the corresponding diagonal element of B^{-1}_{mm},

$$\sigma^2_{x_i} = (B^{-1}_{mm})_{ii} . \tag{9}$$

The decay constants, λ, do not enter linearly in Eq. (1); hence, a least-squares solution for their best values is not possible. However, if the terms are expanded in terms of small changes, δx_j and $\delta\lambda_j$, from a set of initial guesses x_j^0 and λ_j^0 as shown below,

$$(x_j^0 + \delta x_j)\, e^{-(\lambda_j^0 + \delta\lambda_j)t_i} \approx (x_j^0 + \delta x_j)\, e^{-\lambda_j^0 t_i} - x_j^0 \delta\lambda_j t_i\, e^{-\lambda_j^0 t_i} \tag{10}$$

a solution for the $\delta\lambda$ terms is now possible. An iterative procedure may then be used until any desired degree of convergence is attained. (A convergent solution will not necessarily be obtained in all cases.) In the matrix notation, one extra column of the form $t_i e^{-\lambda_j t_i}$ is added to A_{nm} for each unknown half-life, and one extra row is added to X_{m1}.

THE CLSQ PROGRAM

The CLSQ decay curve analysis program has been coded for operation on an IBM 7090 under control of the FORTRAN Monitor System. It is designed to process sequentially an unlimited (subject to time limitations only) number of problems. Each problem has arbitrarily been limited to 200 data points and 10 components. The input

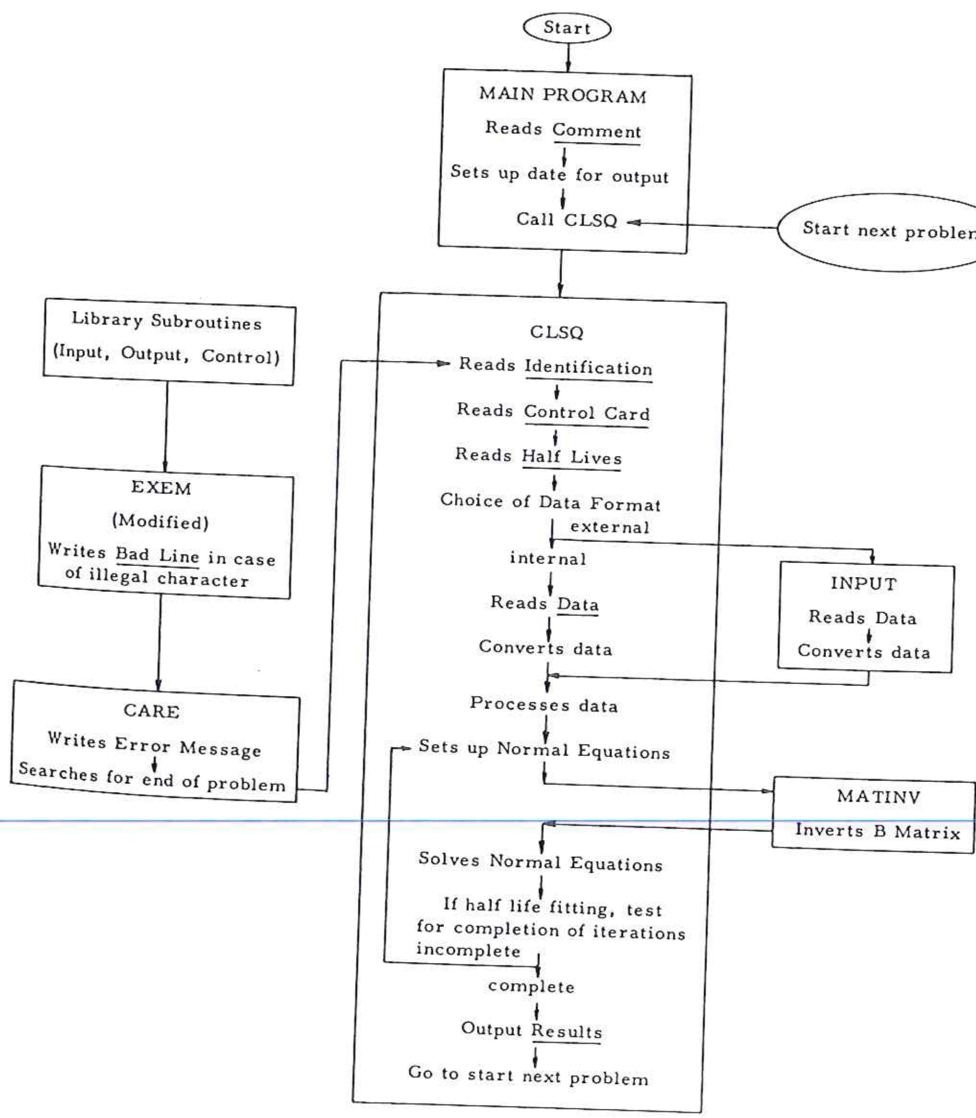

Figure 1.

gram) is shown in Fig. 1. The overall program has been coded as a main program and various subroutines and will now be discussed.

The *Main Program* (MNPRØ) is coded in FAP. It is entered at the start of each run and reads 42 alphanumeric characters (7 six-character words) from one card as a *Comment* (CØMNT) [3]. It then acquires the date from memory and sets it up for output with CØMNT as an output heading on each problem. Control is then transferred to CLSQ.

CLSQ (FORTRAN coded) is the major subroutine in the present program. On entry from MMPRØ it reads 72 alphanumeric characters to serve as *Identification* (NAME) of the particular problem (subheading on output). It then reads a *Control Card* which selects the various calculational and input options and supplies necessary parameters. Figure 2 shows the format of the Control Card. The number of

CLSQ DECAY CURVE ANALYSIS PROGRAM, INPUT DATA

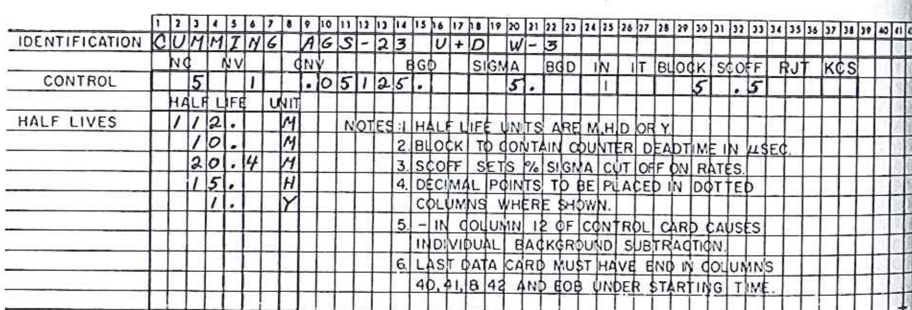

Figure 2.

components, NC, is given in columns 2 and 3. In column 6 the number, NV, of unknown half-lives is specified. The program will treat the first NV of the NC components as variable. The limitation

$$NC + NV \leqq 10 \tag{11}$$

has been imposed arbitrarily. In columns 9, 10, and 11 a number CNV (.05 in the example) is supplied to govern how far the iterations will proceed. Iterations will be continued until the ratio of change in the decay constant to the standard deviation of the decay constant is less than CNV for all NV variable half-lives. If CNV is zero a maximum of nine iterations will be performed.

In columns 12 through 17 the counter background is entered for subtraction from the data. A background standard deviation may be entered in columns 18 through 23 for root-mean-square addition to the standard deviation of each point. An option here (in conjunction with the external input only) allows subtraction of backgrounds which vary from point to point on the curve. To use this, a negative background is entered. This is ignored and background subtraction is performed by the INPUT subroutine.

Columns 24 and 25 (IN) specify the input data format. A zero or blank causes CLSQ to use its own input format while a positive integer transfers to the external subroutine INPUT as shown in Fig. 1.

Columns 26 and 27 (IT) are normally left blank. A number punched in this field causes output of intermediate matrices and a check of the inversion routine by multiplication

$$B_{mm}^{-1} B_{mm} = U_{mm} . \tag{12}$$

Columns 28, 29, and 30 (BLOCK) are the *counter dead time* in microseconds (5 μsec in the example of Fig. 2).

Columns 31, 32, and 33 (SCOFF) set a cutoff (in percent) on the smallest value at which the program will use the standard deviation from statistics alone. In the example the standard deviation of any point will never be less than 0.5% of the rate, regardless of how many counts may have been recorded.

Columns 34, 35, and 36 (RJT) if not blank or zero cause the program to examine its output and reject those points which fall further than RJT times the standard deviation from the curve. The fit is then repeated.

A number in column 38 (KCS) causes the program to enter a *known component subtraction* routine which treats the last KCS of the half-lives as having known intercepts. It reads these intercepts from cards after the data are input, and appropriately subtracts these components from the decay curve before fitting. KCS cannot exceed 5 and the sum NC + NV + KCS cannot exceed 10.

After reading the information on the control card, CLSQ then reads the list of NC half-lives (one per card). The first NV of these are considered first guesses. Half-life units may be minutes (M), hours (H), days (D), or years (Y). If no unit is given it is taken to be M. For internal use, decay constants in $\min^{-1}$ are calculated.

The data from the decay curve are now read in. The external subroutine INPUT (coded in FORTRAN and called by IN=1 on the control card) has been used most frequently. Its input format is shown in Fig. 3. For each point the subroutine calculates the counting rate, its variance, and the time (in minutes) at the midpoint of the count. Point by point background subtraction is also performed if called for by a negative background on the control card. Control is then returned to CLSQ. The internal input of CLSQ reads midpoint time, counting rate, and variance for each point in a 3E13.6 format. In either input, the last data card is either blank or contains a time which will be interpreted as the time of the end of bombardment.

After rates, variances, and midtimes are calculated, CLSQ converts times to times relative to the first count, corrects for deadtime, subtracts background, applies the SCOFF criterion to the variances, and performs the KCS option if called for. The intercepts of the known components and their standard deviations are obtained from cards which follow the end of bombardment card. The format is 2E13.5 with one intercept and its standard deviation on each card.

The program now proceeds to set up the necessary equations for the least-squares analysis as outlined above. Inversion of the B matrix is accomplished by the subroutine MATINV [4] and the normal equations are solved. If half-lives were to be determined, this first pass considered them fixed at the initial guesses. This pass then supplies the initial guesses for the intercepts which are used in the next iterative analysis. Results of the first pass are also output for comparison. A typical

CLSQ DECAY CURVE ANALYSIS PROGRAM, INPUT DATA

	STARTING TIME DAYS	HRS	MINUTES	COUNTS	LENGTH OF COUNT (MIN)	BGD (C/M)	SIGMA BGD (C/M)	
DATA	28	17	3.5	37.696	2			
			12.5	303852				
			21.	258464				
			31.	218757				
			43.	183000				
		↓↓	54.	158024				
		18	10.	134323				
			33.	538863	10			
		↓↓	45.	1393000	30			
		19	15.	1177				
		↓↓	45.	1010				
		20	15.	880				
		↓↓	45.	774				
		21	15.	685				
		↓↓	45.	609				
		22	15.	545				
		↓↓	45.	495				
		23	15.	450				
	↓↓	↓↓	45.	410				
	29	0	15.	378				
		↓	45.	351				
		1	15.	329				
		↓	45.	307				
		2	15.	291				
		↓	45.	274				
		3	15.	262				
		↓	45.	248				
		4	15.	237				
		↓	45.	228				
		5	15.	219				
		↓	45.	213				
		6	15.	206				
		↓	45.	199				
		7	15.	192				
		↓	45.	187				
		8	15.	182				
		↓	45.	177↓↓↓	↓↓			
		13	20.	23628	5			
	↓↓	14	39.	31272	7			
	39	15	0.	9535	60			
	28	16	23.1					END

Figure 3.

output is shown in Fig. 4. The quantity FIT is given by

$$\mathrm{FIT} = \sqrt{\frac{V'_{n1} P_{nn} V_{n1}}{n-m}} \, . \tag{13}$$

It should be pointed out that $V'_{n1} P_{nn} V_{n1}$ is essentially χ^2 for the number of degrees of freedom $(n-m)$. Its expectation value is $(n-m)$ and its variance is $2(n-m)$.

If half-lives are to be fitted, the program now proceeds to perform the necessary operations. The changes in a given decay constant at each iteration are damped so that they can never exceed one-half the value of the decay constant. Each change in a decay constant is also tested to ensure convergence. There is some evidence that the test for convergence in the present program is too strong and should be relaxed to allow a slightly larger change to follow a smaller one. When all changes in

CLSC DECAY CURVE ANALYSIS PRØGRAM 10/03/62

CUMMING AGS-23 U+C H-3

NP= 4C NC= 5 NV=0 CNV=0.05 BGD=125.00 SBGD= 5.00 BLØCK= 5.0 SCØFF=0.5 RJT=-0. KCS=0

	HALF LIFE	SIGMA	H	CPM AT EØB	SIGMA	DECAY FACTØR
CØMP(1)	112.000M	0.	M	0.88832E 05	0.25032E 03	0.12920E 01
CØMP(2)	10.000M	0.	M	0.11027E 06	0.33324E 05	0.17631E 02
CØMP(3)	20.400M	0.	M	0.41368E 06	0.59996E 04	0.40824E 01
CØMP(4)	15.000H	0.	H	0.11904E 05	0.20326E 02	0.10324E 01
CØMP(5)	1.000Y	0.	Y	0.36485E 02	0.53433E 01	0.10001E 01

FIT= 0.956

T(I)	F(I)	FCALC(I)	V(I)	SIGMAF(I)	RATIØ(I)
0.	0.18865E 06	0.18791E 06	0.74564E 03	0.94326E 03	0.79
0.90000E C1	0.15376E 06	0.15450E 06	-0.74583E 03	0.76878E 03	-0.97
0.17500E 02	0.13052E 06	0.13088E 06	-0.36405E 03	0.65258E 03	-0.56
0.27500E C2	0.11026E 06	0.11005E 06	0.20730E 03	0.55131E 03	0.38
0.39500E C2	0.92079E 05	0.91946E 05	0.13366E 03	0.46040E 03	0.29
0.50500E C2	0.79412E 05	0.79835E 05	-0.42331E 03	0.39706E 03	-1.07
0.66500E C2	0.67415E 05	0.67190E 05	0.22559E 03	0.33708E 03	0.67
0.93500E C2	0.54005E 05	0.53549E 05	0.45598E 03	0.27002E 03	1.69
0.11550E 03	0.46489E 05	0.46229E 05	0.25994E 03	0.23245E 03	1.12
0.14550E 03	0.39237E 05	0.39007E 05	0.23054E 03	0.19619E 03	1.18
0.17550E 03	0.33637E 05	0.33575E 05	0.61483E 02	0.16818E 03	0.37
0.20550E 03	0.29280E 05	0.29246E 05	0.34048E 02	0.14640E 03	0.23
0.23550E C3	0.25731E 05	0.25696E 05	0.35137E 02	0.12865E 03	0.27
0.26550E 03	0.22752E 05	0.22742E 05	0.10188E 02	0.11376E 03	0.09
0.29550E 03	0.20209E 05	0.20266E 05	-0.56991E 02	0.10105E 03	-0.56
0.32550E 03	0.18069E 05	0.18183E 05	-0.11353E 03	0.90346E 02	-1.26
0.35550E 03	0.16398E 05	0.16423E 05	-0.25018E 02	0.81989E 02	-0.31
0.38550E 03	0.14894E 05	0.14931E 05	-0.37617E 02	0.74469E 02	-0.51
0.41550E 03	0.13557E 05	0.13664E 05	-0.10631E 03	0.67786E 02	-1.57
0.44550E C3	0.12488E 05	0.12582E 05	-0.93687E 02	0.62441E 02	-1.50
0.47550E 03	0.11586E 05	0.11656E 05	-0.69148E 02	0.57932E 02	-1.19
0.50550E 03	0.10852E 05	0.10859E 05	-0.70879E 01	0.54259E 02	-0.13
0.53550E C3	0.10117E 05	0.10170E 05	-0.53183E 02	0.50585E 02	-1.05
0.56550E 03	0.95829E 04	0.95722E 04	0.10619E 02	0.47914E 02	0.22
0.59550E 03	0.90153E 04	0.90500E 04	-0.34689E 02	0.45077E 02	-0.77
0.62550E 03	0.86147E 04	0.85913E 04	0.23448E 02	0.43074E 02	0.54
0.65550E C3	0.81474E 04	0.81858E 04	-0.38472E 02	0.40737E 02	-0.94
0.68550E 03	0.77802E 04	0.78253E 04	-0.45071E 02	0.38901E 02	-1.16
0.71550E 03	0.74798E 04	0.75025E 04	-0.22689E 02	0.37399E 02	-0.61
0.74550E C3	0.71795E 04	0.72117E 04	-0.32211E 02	0.35897E 02	-0.90
0.77550E 03	0.69792E 04	0.69478E 04	0.31367E 02	0.34896E 02	0.90
0.80550E 03	0.67456E 04	0.67070E 04	0.38621E 02	0.33728E 02	1.15
0.83550E 03	0.65120E 04	0.64857E 04	0.26319E 02	0.32560E 02	0.81
0.86550E 03	0.62784E 04	0.62811E 04	-0.27102E 01	0.31392E 02	-0.09
0.89550E 03	0.61116E 04	0.60909E 04	0.20632E 02	0.30558E 02	0.68
0.92550E 03	0.59447E 04	0.59132E 04	0.31577E 02	0.29724E 02	1.06
0.95550E C3	0.57779E 04	0.57461E 04	0.31773E 02	0.28890E 02	1.10
0.12180E 04	0.46025E 04	0.45858E 04	0.16670E 02	0.31147E 02	0.54
0.12980E C4	0.43441E 04	0.43018E 04	0.42253E 02	0.25753E 02	1.64
0.15745E C5	0.33919E 02	0.35796E 02	-0.18774E 01	0.52582E 01	-0.36

Figure 4.

decay constants satisfy the CNV requirement the program proceeds to output the results as shown in Fig. 5. By comparison with Fig. 4 it is seen that FIT is significantly improved by inclusion of the variable half-life and that 110 min is a better value than the first guessed 112-min value.

Since the present program runs under control of the FORTRAN Monitor, a change has been made in the library routine EXEM. Rather than skipping the entire days' run in case of an illegal character in the input, EXEM now writes out the bad line and transfers to subroutine CARE as shown in Fig. 1. CARE then searches for a

CLSQ CECAY CURVE ANALYSIS PRØGRAM 10/03/62

CUMMING AGS-23 U+D K-3

NP= 4C NC= 5 NV=1 CNV=0.05 BGD=125.00 SBGD= 5.00 BLØCK= 5.0 SCØFF=0.5 RJT=-0. KCS=0

ITERATIØNS PERFØRMED= 3 CØNVERGENT

	1ST CØMP	2ND CØMP	3RD CØMP	4TH CØMP	5TH CØMP
D	0.630442E-02				
CELTA(2)	0.115122E-03				
CELTA(3)	0.48C803E-06				
SIGMA	C.265903E-04				

	HALF LIFE	SIGMA H		CPM AT EØB	SIGMA	DECAY FACTØR
CØMP(1)	109.946M	0.464M		0.91337E 05	0.54585E 03	0.12982E 01
CØMP(2)	10.000H	0.	M	0.18830E 06	0.37895E 05	0.17631E 02
CØMP(3)	20.4C0M	0.	M	0.39086E 06	0.79976E 04	0.40824E 01
CØMP(4)	15.000H	0.	H	0.11993E 05	0.28498E 02	0.10324E 01
CØMP(5)	1.000Y	0.	Y	0.34733E 02	0.53580E 01	0.10001E 01

FIT= 0.614

T(I)	F(I)	FCALC(I)	V(I)	SIGMAF(I)	RATIØ(I)
0.	0.18865E 06	0.18843E 06	0.22546E 03	0.94326E 03	0.24
0.90000E C1	0.15376E 06	0.15429E 06	-0.52894E 03	0.76878E 03	-0.69
0.17500E C2	0.13052E 06	0.13050E 06	0.11999E 02	0.65258E 03	0.02
0.27500E 02	0.11026E 06	0.10976E 06	0.49989E 03	0.55131E 03	0.91
0.39500E C2	0.92079E 05	0.91857E 05	0.22217E 03	0.46040E 03	0.48
0.50500E C2	0.79412E 05	0.79918E 05	-0.50599E 03	0.39706E 03	-1.27
0.66500E C2	0.67415E 05	0.67436E 05	-0.20541E 02	0.33708E 03	-0.06
0.93500E 02	0.54005E 05	0.53876E 05	0.12840E 03	0.27002E 03	0.48
0.11550E 03	0.46489E 05	0.46526E 05	-0.37102E 02	0.23245E 03	-0.16
0.14550E 03	0.39237E 05	0.39218E 05	0.18995E 02	0.19619E 03	0.10
0.17550E C3	0.33637E 05	0.33700E 05	-0.63301E 02	0.16818E 03	-0.38
0.20550E 03	0.29280E 05	0.29301E 05	-0.20442E 02	0.14640E 03	-0.14
C.23550E 03	0.25731E 05	0.25699E 05	0.32104E 02	0.12865E 03	0.25
0.26550E 03	0.22752E 05	0.22710E 05	0.42190E 02	0.11376E 03	0.37
0.29550E 03	0.20209E 05	0.20212E 05	-0.29536E 01	0.10105E 03	-0.03
0.32550E 03	0.18069E 05	0.18117E 05	-0.47264E 02	0.90346E 02	-0.52
0.35550E 03	0.16398E 05	0.16351E 05	0.46320E 02	0.81989E 02	0.56
0.38550E C3	0.14994E 05	0.14860E 05	0.33724E 02	0.74469E 02	0.45
0.41550E 03	0.13557E 05	0.13596E 05	-0.38431E 02	0.67786E 02	-0.57
0.44550E 03	0.12488E 05	0.12520E 05	-0.31518E 02	0.62441E 02	-0.50
0.47550E C3	0.11586E 05	0.11600E 05	-0.14042E 02	0.57932E 02	-0.24
0.50550E 03	0.10852E 05	0.10811E 05	0.40266E 02	0.54259E 02	0.74
C.53550E 03	0.10117E 05	0.10131E 05	-0.13802E 02	0.50585E 02	-0.27
0.56550E 03	0.95829E 04	0.95407E 04	0.42141E 02	0.47914E 02	0.88
0.59550E 03	0.90153E 04	0.90260E 04	-0.10689E 02	0.45077E 02	-0.24
0.62550E 03	0.86147E 04	0.85743E 04	0.40403E 02	0.43074E 02	0.94
0.65550E 03	0.81474E 04	0.81754E 04	-0.27998E 02	0.40737E 02	-0.69
0.68550E 03	0.77802E 04	0.78207E 04	-0.40474E 02	0.38901E 02	-1.04
0.71550E 03	0.74798E 04	0.75032E 04	-0.23356E 02	0.37399E 02	-0.62
0.74550E 03	0.71795E 04	0.72170E 04	-0.37542E 02	0.35897E 02	-1.05

Figure 5.

card with the characters END in columns 40, 41, and 42; hence, this word should appear in this location on the last card of each problem. On finding an END it then transfers to CLSQ and starts the next problem.

Further details on the program may be obtained from the author.

CONCLUSIONS

Experience has shown that when such a program is available it will be used for analyses of most decay data with a considerable saving in time over graphical procedures. Furthermore its results are not subjective and its error estimates are considerably more meaningful than those guessed from the graphical analyses. Small effects, such as presence of impurities or improper half-lives which would not have been seen in a graphical analysis, become apparent in the least-squares procedure.

The author is indebted to Mrs. R. Larsen and Mr. K. Fuchel for their assistance during the coding of this problem.

REFERENCES

[1] W. Hamilton and V. Schomaker (unpublished).

[2] D. S. Harmer, BNL 544(T-141) (1959) (unpublished). Note: The present paper has interchanged the notation for the X and A matrices from that used by Harmer.

[3] During current operation, CØMNT reads "CLSQ Decay Curve Analysis Program." However, CØMNT offers a convenient way to wish users of the program a "Merry Christmas," etc.

[4] This program, share distribution No. 664 AN F402, was coded by B. S. Garbow, Argonne National Laboratory (1959).

(1-4) ANALYSIS OF MULTICOMPONENT DECAY CURVES BY USE OF FOURIER TRANSFORMS

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INTRODUCTION

Frequently it happens that experimental data may best be represented by a sum of exponentials of the form

$$f(t) = \sum_{i=1}^{n} N_i^0 \exp(-\lambda_i t) . \qquad (1)$$

The problem is not one of mere curve fitting because the parameters have physical significance. Therefore it is necessary that the true parameters be accurately estimated. This implies that the number of components n must also be determined, for if