DP-MS-75-38
CONF-751009--3

DATA REDUCTION IN THE 100 mg ^{252}Cf ACTIVATION ANALYSIS FACILITY AT THE SAVANNAH RIVER LABORATORY

by

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This paper is proposed for presentation at the
International Nuclear and Atomic Activation Analysis Conference and 19th Annual Meeting on Analytical Chemistry in Nuclear Technology,
October 14-16, 1975
Gatlinburg, Tennessee

This paper was prepared in connection with work under Contract No. AT(07-2)-1 with the U. S. Energy Research and Development Administration.

DISCLAIMER

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ABSTRACT

The automated absolute activation analysis technique developed at the Savannah River Laboratory is based on the ability to predict neutron capture reaction rates from tabulated cross sections and neutron spectra accurately calculated for the 100 mg ^{252}Cf source. A series of computer programs comprises the data reduction system which:

1) reduces the gamma-ray spectra to lists of photopeak energies, areas, and statistical errors for all significant photopeaks,

2) assigns each gamma ray to the appropriate activation product by comparing experimental to tabulated gamma-ray energy,

3) converts each photopeak area into an elemental concentration using the experimental timing data, calculated reaction rates, detector efficiency, and activation product spectroscopic data in the absolute activation master equation.

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* The information contained in this article was developed during the course of work under Contract No AT(07-2)-1 with the U. S. Energy Research and Development Administration.

The data reduction system requires about one second of IBM 360-195 CPU time for the conversion of each 4096 channel spectrum into a qualitative and quantitative list of elemental composition. The accuracy of the algorithm is better than ±15% for most elements.

INTRODUCTION

The data reduction for automated absolute activation analyses of samples in the Savannah River Laboratory (SRL) 100 mg ^{252}Cf facility, described in a companion paper, is accomplished via computer programs RAGS and SIFTER, which were written for the SRL IBM 360-195 computer. The RAGS program reduces a gamma ray spectrum to a list of the energies and areas of all statistically significant photopeaks. The SIFTER program first qualitatively identifies the nuclide which emitted the gamma rays, then quantitatively transforms the photopeak areas into the elemental composition of the sample as ppm. These calculations permit neutron activation analyses of samples without the necessity of simultaneously analyzing comparative standards which must normally be handled under laboratory conditions identical with the samples. The elimination of comparative standards increases the sample throughput of the facility by at least a factor of two.

MATHEMATICAL BASIS

The data reduction for automated absolute activation analysis (AAAA) involves three steps: 1) *analysis* of the gamma-ray spectrum to yield the energies and areas of all the statistically significant photopeaks, 2) *identification* of the nuclides which emitted the gamma-rays, and 3) *conversion* of the photopeak areas into the elemental composition in ppm. The first step is performed by the program RAGS, which has been previously documented [W. W. Bowman, *Nucl. Instr. Meth. 96*, 135 (1971)]. The SIFTER program is responsible for the remaining two steps as discussed below:

The general AAAA expression is:

$$PPM = \left(\frac{P}{E_\gamma\ W}\right)\left(\frac{\lambda}{I_\gamma(TIME)}\right)\left(\frac{A\ 10^6}{B\ N_o}\right)\left(\frac{1}{RATE}\right)$$

The first term is obtained from the experimental parameters, where:

P = photopeak area

E_γ = detector efficiency at the gamma-ray energy

W = mass of the sample

The second term is dependent on the product nuclide, where:

λ = decay constant

I_γ = absolute gamma-ray intensity, photons/decay

TIME = expression containing the experimental timing information = SCDG

where:

$S = 1-\exp(-\lambda T_i)$; T_i = irradiation time

$C = 1-\exp(-\lambda T_c)$; T_c = counting time

$D = \exp(-\lambda T_d)$; T_d = decay time

$G = \frac{M}{(1-Q)} - \frac{Q(1-Q^M)}{(1-Q)^2}$; M = number of irradiation cycles

$Q = \exp(-\lambda T_{cyc})$

T_{cyc} = cycle time

The third term is dependent on the induced reaction, where:

A = atomic weight of the parent nuclide

B = isotopic abundance of the parent nuclide

N_o = Avogadro's number

The last term is dependent on the induced reaction and on the neutron flux and neutron energy distribution, where:

RATE = the specific neutron capture reaction rate

If cross section data in 84 energy groups are available for the reaction, then:

$$RATE = \sum_{i=1}^{84} \sigma_i(E)\ \phi_i(E)$$

If 84-group cross section data are not available, then:

$$RATE = 0.886\ \phi_{th}\ \sigma_{2200} + 0.0221\ [\phi_{epi}\ (R.I.)]$$

where:

ϕ_{th} = thermal flux

σ_{2200} = tabulated 2200 m/s cross section

ϕ_{epi} = epithermal flux

R.I. = tabulated resonance integrals

The constants are used to correct for moderator temperature, lethergy, and non-1/E neutron spectrum above the thermal peak.

DATA REDUCTION PROGRAMS

The block diagram (Figure 1) shows the derivation and organization of the data necessary for evaluating the general expression by SIFTER. Boxes denote names of programs; the capitalized names refer to tape or disc data file names. The gamma-ray spectra are written onto tape by the multichannel analyzer, each spectrum labeled by a tagword. TRANSCRIBE attaches a header record to the translated spectrum and confirms that the header record is attached to the correct spectrum. The header record contains date and time of the analysis, irradiation, decay and counting times, mass of the sample, number of irradiation-counting cycles, and code numbers which describe the source-sample-moderator configuration and the detector-sample geometry. In addition, the header contains space for a 44 alphanumeric-character sample descriptor. ADDER and COPY are required for assembling master and duplicate storage tapes for permanent retention of all spectra collected in the AAAA facility.

Data Bank Libraries

Information to support the data reduction programs is contained in the GAMDAT, FLUXFL, SIGMAF, and EFFCAL libraries.

The GAMDAT library contains the necessary spectroscopic data for all the gamma-rays emitted by each of the possible activation

product nuclides. These data were obtained from the gamma-ray catalogue [*Atomic Data and Nuclear Data Tables 13*, No. 2-3, (1974)] by sorting against a list of all possible product nuclide symbols. The spectroscopic data include the isotopic half-life, energy, and absolute abundance of each gamma ray.

The SIGMAF library contains data specific to a particular reaction, such as: the product nuclide symbol, the reaction symbol, atomic weight and isotopic abundance of the parent nuclide, and the cross sections in the 2- and 84-energy groups, if available. The source of these data are the *Chart of the Nuclides* for 2-group cross sections and an SRL Reactor Physics Division program which calculates the differential cross section from tabulated 2200 m/s cross sections and resonance parameters.

The EFFCAL library contains detector efficiency calibration data which are ordered by sample-detector geometry and by detector number.

These data libraries are stored in library disc space and are called as needed by the SIFTER program.

RAGS Program

RAGS analyzes the spectra and writes the photopeak energies, areas, the 2-sigma statistical uncertainties in the areas, and all the header information into disc file RAGOUT. After RAGS has operated on a particular series of gamma-ray spectra, the RAGOUT file contains all the additional information required for the qualitative and quantitative analysis by SIFTER.

SIFTER Program

The SIFTER program identifies the radionuclides which emitted the gamma rays and converts the photopeak areas to the quantity of each element present in ppm. A simplified flow diagram for SIFTER is shown in Figure 2.

After initialization, the results of the analysis of the first spectrum are read from RAGOUT. Photopeak areas are divided by the detection efficiency as interpolated by energy from the efficiency calibration data sorted in EFFCAL. FLUXFL is next searched for an irradiation geometry code number match which selects the appropriate normalized fluxes. These fluxes are corrected for the ^{252}Cf source decay to the date of the analysis. A "no-match" signals a disintegrations per minute/gram (dpm/g) calculation.

Next, the first isotope (ISO) and its spectroscopic data are read from GAMDAT, which has the gamma rays ordered in decreasing absolute abundance. The first ISO gamma ray energy is searched against the RAGOUT energies within ±EER (usually 0.3 keV).

A "no-match" signals a search with the second ISO gamma ray against the RAGOUT energies. If there is a match, the TIME term is calculated using the nuclide half-life. If a dpm/g calculation has been signaled by a lack of flux parameters, the dpm/g calculation is made and printed, and the search is continued.

If flux parameters are available, the SIGMAF file is searched for a match between ISO and the reaction product symbol, JSO. A match signals that RATE may be calculated based on 2- or 84-energy-group expressions if cross sections are available for that particular reaction; if unavailable, RATE is set to unity, and comparative standards must be run for that element. At this point, all the terms have been collected for the calculation of ppm (and its error), which is printed along with the nuclide symbol, gamma ray energy, the reaction symbol, and a comment denoting the type of RATE expression used in the calculation.

Several nuclides may be produced by more than one neutron-induced reaction, such as, (n,γ), (n,p), and/or (n,α) on different parent nuclides. To account for these reactions, the entire SIGMAF file is searched for all product symbol matches. The end of SIGMAF signals that the next most abundant gamma ray in the product ISO is to be sorted against the RAGOUT energies. This process is repeated until the end of the list of gamma rays for ISO is reached, then the next possible product nuclide (a new ISO) is read for GAMDAT. This searching loop continues until the end of the GAMDAT file is reached, which signals the listing of

each RAGOUT gamma ray, photopeak area, and error, along with a list of all nuclide symbols having gamma rays that matched in energy. This provides a quick check for demonstrating if all significant gamma rays were accounted for in the search procedure. Following this listing, the next spectral analysis data block is read from RAGOUT, and the entire process is repeated until all spectra have been scanned.

APPLICATION

The elemental composition reported for a liquid unknown was calculated using SIFTER. An example of the output obtained from the analysis of one spectrum is shown in Tables 1 and 2. The columns in Table 1 are: product nuclide symbol, the matched gamma-ray energy, reaction symbol, ppm and its statistical error, and a column for comments to denote the method of calculating the specific neutron reaction rate.

Two reactions are possible for producing ^{24}Na: ^{23}Na (n,γ) ^{24}Na and ^{27}Al (n,α) ^{24}Na. Because tabulated cross sections are not available for the (n,α) reaction, the rate was set to unity. Note also that the calculations were performed for both ^{24}Na gamma rays. A major problem in interpreting the output is illustrated by the 10^{11} ppm result calculated for ^{60m}Co. The 1332.48 keV gamma ray is due to ^{60}Co; however, since the energy is identical in ^{60m}Co decay, the peak was matched, eventhough the long decay time permitted complete decay of ^{60m}Co. Similarly, both ^{152}Eu and ^{152m}Eu are responsible for the gamma ray match.

However, in the calculation the photopeak areas have not been proportioned between the two nuclides, leading to one high and one low value for the concentration of europium. A much earlier or much later spectrum would yield the correct result for one isomer and an "impossible" result for the other. The ^{104m}Rh, ^{110}Ag, ^{134}I, and ^{197}Pt entries result from random chance energy matched to photopeaks belonging to other nuclides, or to statistical fluctuations in the spectrum which were analyzed as "photopeaks".

Table 2 lists the data for each photopeak in the spectrum in columns corresponding to energy, peak area (efficiency corrected), percent statistical uncertainty, and the number of product nuclides (and their symbols) which had energy matches to that photopeak. Large photopeaks not matched and the reason for lack of a match are listed below:

336.27 keV	room background, not in GAMDAT library
510.99 keV	annihilation of pair-produced positrons, not in library
841.96 and 963.67 keV	fell outside the ±0.30 keV search window, so these peaks were not matched to ^{152m}Eu peaks at 841.53 and 963.36 keV
1731.45 and 2241.92 keV	double and single escape peaks resulting from the 2753.92 keV radiation from ^{22}Na
39.19 and 3772.85 keV	spectrum artifacts from the initial shoulder and amplifier saturation

FUTURE DEVELOPMENT

Future development of the AAAA data reduction programs will include elimination (or at least tagging) of results which are obtained under experimental conditions (such as decay time) which are not conducive to accurate analyses for a particular nuclide. Expansion of the 84-group cross section library to include (n,p) and (n,α) reactions will significantly increase the accuracy of results based on these reactions.

TABLE 1. Analysis Printout I

TAG-6 6/14/74 15:55: 0 CT- 3.000E 03 DT- 1.740F 04 TIR- 2.540E 05

WT- 1.000E 04 IGEO- 4 ITUB-13 ICYC- 1

LIQUID UNKNOWN #1 CROP TUBE SECOND COUNT 6 NPK- 36

RNG 1 PCS 1-5 H2C MCD CASE III 84-CASE

Product Nuclide Symbol	*Matched γ-ray Energy, keV*	*Reaction Symbol*		*Elemental Conc., ppm*	*Statistical Error, ppm*	*Comments*
^{24}Na	1368.53	^{23}Na	GO	6.312E 02	7.385E 00	PPM 2 GP
^{24}Na	1368.53	^{27}Al	AO	8.019E-14	9.382E-16	RATE = 1.0[a]
^{24}Na	2753.92	^{23}Na	GO	6.024E 02	9.879E 00	PPM 2 GP
^{24}Na	2753.92	^{27}Al	AO	7.653E-14	1.255E-15	RATE = 1.0[a]
^{60}Co	1332.48	^{59}Co	GO	1.430E 02	3.044E 01	PPM 2 GP
^{60}Co	1173.21	^{59}Co	GO	1.134E 02	4.142E 01	PPM 2 GP
^{60m}Co	1332.48	^{59}Co	G1	9.977E 10	2.124E 10	PPM 2 GP
^{69m}Zn	438.70	^{68}Zn	G1	6.746E 02	1.128E 02	PPM 2 GP
^{76}As	559.47	^{75}As	GO	7.310E 01	1.733E 00	PPM 2 GP
^{76}As	657.20	^{75}As	GO	6.442E 01	8.561E 00	PPM 2 GP
^{76}As	1216.19	^{75}As	GO	4.851E 01	1.673E 01	PPM 2 GP
^{99}Mo	181.06	^{98}Mo	GO	5.037E 03	8.075E 02	PPM 2 GP
^{99}Mo	140.51	^{98}Mo	GO	3.706E 03	6.560E 01	PPM 2 GP
^{99}Mo	739.58	^{98}Mo	GO	3.320E 03	5.209E 02	PPM 2 GP
^{104m}Rh	77.50	^{103}Rh	G1	4.553E 23	4.781E 21	PPM 2 GP
^{110}Ag	657.50	^{109}Ag	GO	2.349E 06	3.122E 05	PPM 2 GP
^{115}Cd	527.90	^{114}Cd	GO	2.048E 03	7.945E 01	PPM 2 GP
^{115}Cd	492.30	^{114}Cd	GO	2.003E 03	2.257E 02	PPM 2 GP
^{117}Cd	344.51	^{116}Cd	GO	6.346E 04	1.313E 04	PPM 2 GP
^{134}I	847.08[b]	1.352E 05 ± 1.345E 04 DPM/GM AT TO				
^{152}Eu	121.78	^{151}Eu	GO	1.447E 02	6.483E 00	PPM 2 GP
^{152}Eu	344.31	^{151}Eu	GO	3.276E 01	6.777E 00	PPM 2 GP
^{152ml}Eu	121.78	^{151}Eu	G1	2.989E-01	1.339E-02	PPM 2 GP
^{152ml}Eu	344.31	^{151}Eu	G1	6.837E-02	1.415E-02	PPM 2 GP
^{197}Pt	77.35	^{196}Pt	GO	5.173E 02	5.432E 00	PPM 2 GP
^{197}Hg	77.35	^{196}Hg	GO	2.349E 04	2.466E 02	PPM 2 GP
^{203}Hg	279.21	^{202}Hg	GO	9.031E 02	1.023E 02	PPM 2 GP

a. Tabulated cross sections for the (n,α) reaction were not available, therefore the rate was set at unity.

b. Random chance energy match (see text).

TABLE 2. Analysis Printout II

LIQUID UNKNOWN #1 DROP TUBE SECOND COUNT 6

γ-ray Energy, keV	Peak Area, Corr.	Statistical Uncertainty, %	No. of Matches	Nuclides with Matching Energy Peaks
39.190	9.586E 06	3.93	0	
45.440	5.173E 05	35.63	0	
77.360	1.060E 07	1.05	3	^{104m}Rh ^{197}Pt ^{197}Hg
121.690	1.632E 06	4.48	2	^{152}Eu $^{152ml}Eu$
140.550	4.260E 06	1.77	1	^{99}Mo
158.670	3.519E 05	19.64	0	
181.000	4.339E 05	16.03	1	^{99}Mo
279.160	8.571E 05	11.33	1	^{203}Hg
336.270	4.406E 06	2.33	0	
344.370	3.563E 05	20.69	3	^{117}Cd ^{152}Eu $^{152ml}Eu$
438.570	3.661E 05	16.72	1	^{69m}Zn
492.390	7.157E 05	11.27	1	^{115}Cd
510.990	6.133E 06	2.33	0	
527.920	2.484E 06	3.88	1	^{115}Cd
559.410	6.285E 06	2.37	1	^{76}As
657.280	7.686E 05	13.29	2	^{76}As ^{110}Ag
739.780	5.874E 05	15.69	1	^{99}Mo
841.960	2.789E 06	4.58	0	
847.280	8.494E 05	9.95	1	^{134}I
963.670	2.390E 06	6.40	0	
970.120	2.148E 05	36.34	0	
1145.710	2.690E 05	48.31	0	
1173.310	2.877E 05	36.51	1	^{60}Co
1216.170	4.028E 05	34.48	1	^{76}As
1314.940	2.025E 05	42.52	0	
1332.610	3.630E 05	21.29	2	^{60}Co ^{60m}Co
1358.280	2.331E 05	37.99	0	
1368.570	4.053E 07	1.17	1	^{24}Na
1460.380	2.890E 05	29.99	0	
1731.450	5.087E 06	4.70	0	
1809.840	3.766E 05	41.67	0	
2241.920	5.664E 06	4.66	0	
2613.740	5.132E 05	32.59	0	
2753.920	3.864E 07	1.64	1	^{24}Na
3754.410	1.959E 05	41.88	0	
3772.850	3.469E 06	6.51	0	

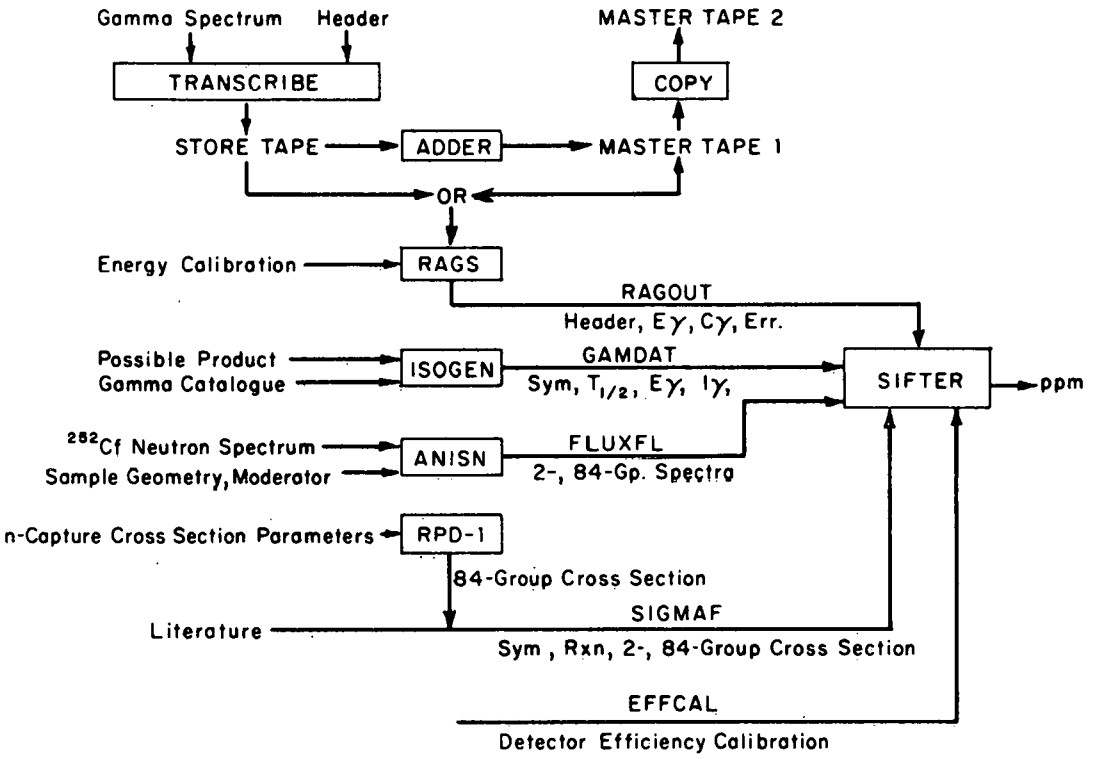

FIGURE 1. Data Reduction Programs

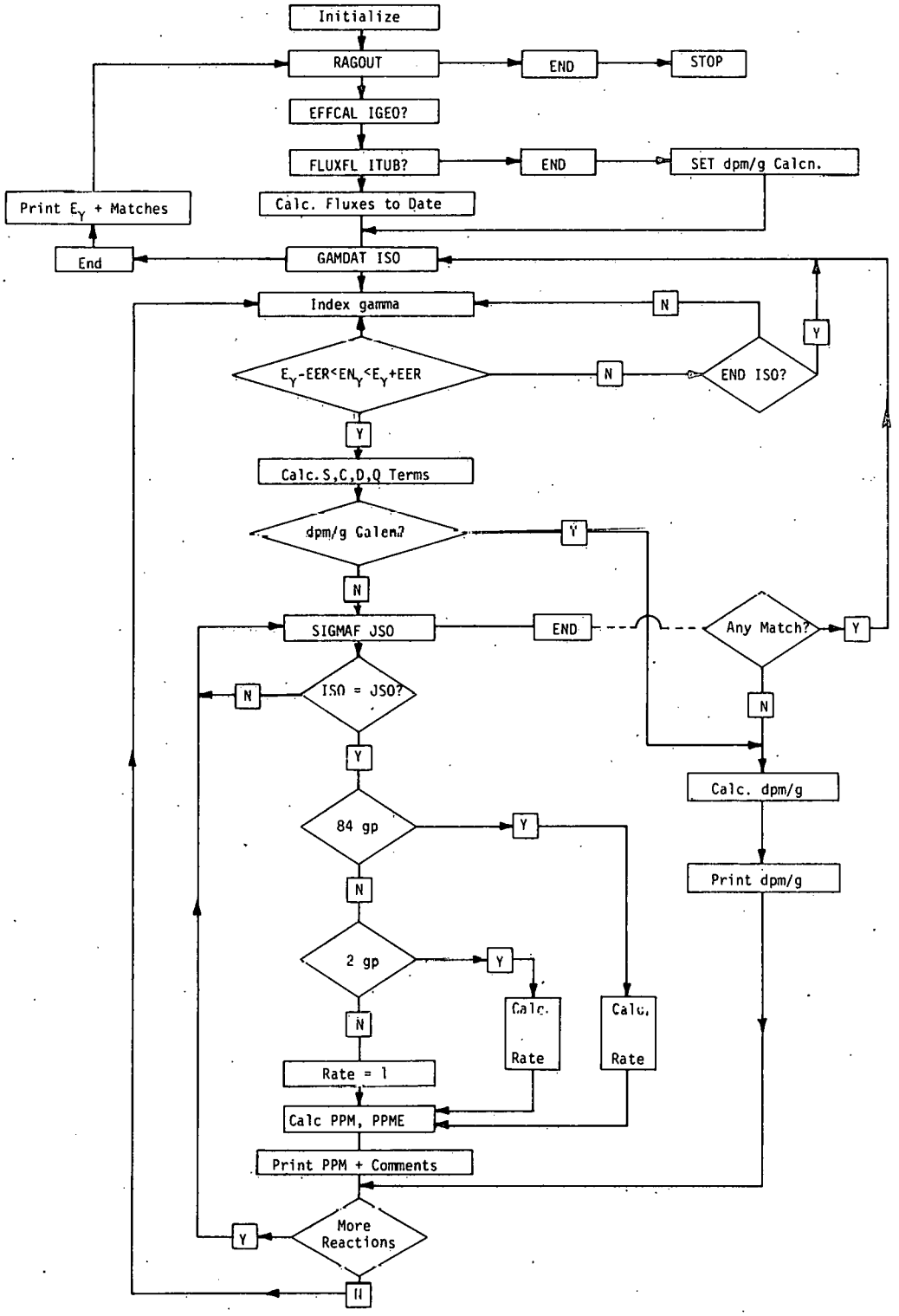

FIGURE 2: SIFTER Program