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# GAMMA-RAY SPECTROMETRIC PRODUCT IDENTIFICATION AND HALFLIFE ANALYSIS FROM PROTON INDUCED SPALLATION AND FISSION REACTIONS OF LEAD AND URANIUM

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Abstract

A computer aided analysis system has been developed for measuring the product yield distribution from proton induced spallation reactions. This system is based on gamma-ray spectrometric identification of product radioactivities in the irradiated target materials lead and uranium. Gamma-ray spectra are collected as a function of time. All peaks and peak areas are identified and measured with the computer code AGAMEMNON.

Decay curves for all gamma-rays observed in the measured spectra are constructed and analyzed by the computer code YELLOW. The essential features of YELLOW are: any number of nuclides may be fitted to the experimental data, parent-daughter decays are included, the complete Erdmann-Soyka library is used, the program prepares a collection of plots containing all neccessary information to decide on the correctness of the program's assignment.

## INTRODUCTION

Development of a high flux neutron source utilizing the spallation reaction necessarily involves extensive study of residual activity produced in the target material. To accomodate this task, gamma-ray spectrometric methods have been developed to deduce mass yield distributions for proton-induced spallation at incident particle energies of 600 MeV and 1100 MeV. This nuclear reaction produces radioactive nuclides, most of them emitting $\gamma$-rays, that cover almost the entire chart of the nuclides. Figure 1 shows a typical $\gamma$-ray spectrum for a lead-target. The complexity of this spectrum is easily seen.

## SCHEME OF THE ANALYSIS

The object of the analysis of these types of spectra is to translate the complicated gamma-ray spectra into a data set consisting of the partial cummulative and independent yield production cross sections. From these data isobaric mass yield distributions can be deduced.
In the development of a computer aided partially interactive analysis system, attention was payed to making all routine data handling and processing completely automatic and simple to use. This includes photo-peak search and analysis, preparation for decay halflife analysis and the halflife analysis itself. At all points sufficient output information is given to the user to be able to recognize readily any systematic errors that the previous automatic part of the analysis may have introduced. The user is able to make all corrections necessary and rerun that part.
The flow diagram of the analysis is given in figure 2. The recorded spectral information on magnetic tape is translated and transferred to easily accessible disc data sets. At this point it is possible to screen for energy calibration drift, incorrectly recorded start and stop times and other necessary information.
In the second step the peak find and peak fit procedure AGAMEMNON /1/ is run. The code outputs all information on a disc for the halflife analysis as well as a printer output of the fit obtained for each peak. The last step is the code YELLOW, which sorts the outputs of AGAMEMNON by energy and time, plots the calculated activity for each peak-energy as a function of time and identifies the reaction products based on their halflives and known gamma-ray transitions. Besides the activity-time plot the present system prints a list of all candidates which fit the experimental points best. The resulting decay curves of the best candidates are drawn in the

graph. If the decay is of parent-daughter nature, this is taken into consiguation. Any number of overlapping nuclides may be taken into account, but at present we only use two for simplicity (for parent-daughter decay, four).
A substantial help in analyzing these plots is the cross reference to the other peak energies of a candidate. The list of candidate isotopes is derived from the compilation of Erdtmann and Soyka /2/.
All programs are written in Pl/1 and are run on an IBM-370/168 TSS computer. This enables the user to work with the conversational or nonconversational mode. All plots may be displayed on a Tectronix- or HP-screen, but in our opinion, it is much more comfortable to prepare a collection of plots. This allows us to observe the computer analysis and make necessary corrections or additional calculations if needed.

## THE ANALYSIS PROGRAMS

Peakfit: All peak area fitting is done with an automatically operating computer code AGAMEMNON /1/. This computer code takes the prepared spectral input from the disc and processes each spectrum individually to determine the energy that corresponds to the centroid as well as the area of all peaks above a controlable significance level. The code outputs this information on disc for the halflife analysis, and prepares a printer output of the fit obtained for each peak.
AGAMEMNON offers a variety of options: internal or external lineshape calibration, only gaussion on a smoothly varying background or gaussian with exponential tail and optional step function under strong peaks.
In this way the problem of high count rate distortions of the peak shape associated with a very radioactive target that is continuously decaying is attacked. Goodness-of-fit criteria are $\chi^2_r$ and RMS for the fitted region and FOM for the peak and the background regions individually.
The resolution of multiplets is performed by the peakfind algorithm which is by the method of convolution with a zero-area gaussian. The peakfind is applied to the original spectrum and (in cases where the goodness-of-fit criteria are not reached) with the updated peakshape parameters to the region under consideration and to the residual spectrum to decide if an additional peak has to be considered.

All these features of the program make it possible to detect reliably and automatically over 100 photopeaks in a single 4K-spectrum, and to calculate

their associated decay rates in the target.
Halflife analysis: It is a well-known fact that difficulties occur when one tries to fit experimental data with a sum of exponentials $f(t) = \sum_{i=1}^{n} A_i e^{-\lambda_i t}$, especially if all the parameters are unknown. As shown by Lanczos /3/, the classical methods (least-squares,...) generally fail to provide a meaningful solution. Even the Golub-Pereyra /4/ algorithm is inadequate.
A method in which the analytical behaviour of the exponential functions is taken into account has been proposed by Gardner /5/. The method makes use of Fourier transform and decomposes so to say the experimental data in Fourier space into the leading components of the decay constants $\lambda_i$.
The success of Gardner analysis is strongly related to the success of the numerical Fourier transformation (the Fast Fourier Transformation) which calls for sufficient data points to integrate the fast oscillating functions with enough precision. From a practical point of view at least some hundred data points are necessary to fit the problem. Our experiments had some ten data points in time only, hower with additional spectral information. This is why we decided to use a more hand-taylored method consisting of sorting for peak-energy first and then computing decay-curves with the known halflifes of the candidates, thereby reducing the number of free parameters by a factor of two.

The complete analysis is as follows:
All outputs of the peakfit program AGAMEMNON for one foil are sorted into energy-groups with a maximum allowable width of 2 keV. To each group of spectral peaks possible candidates are assigned by comparing the peak energies with energies stated in the Erdtman-Soyka Compilation of gamma lines. At this point some criteria enter to reject assignments. These are: at least one out of the three most intense gamma-lines of the nuclide must be present in the spectra, the cut, what is meant by most intense, is set to 1% of absolute intensity, the halflife of the nuclide must be longer than, say, one half hour. In this way about half of the chart of nuclides is rejected. The information gained is stored in memory nuclide by nuclide with pointers serving as connectors between spectral-peaks and the nuclide entry in memory (just as a catalogue entry points to a special drawer). The advantage of this procedure is that all gamma lines of a nuclide are accessible to the program and the associated spectral peaks can be used for further analysis.

We are thinking about rejecting nuclides if the activities calculated from the individual peaks differ by a given value. But because of the strong overlapping of peaks this criterion is very delicate.

For the final halflife analysis the candidates for a given energy group are combined and the decay curve is fitted to the experimental lines with the now known halflives. At present we either use only one nuclide to fit the data points or a combination of any two. It is up to the user to decide of the number of nuclides to be combined. For simplicity and mainly by experience we decided to use two at maximum, which is augmented to four if the decay of both nuclides is of parent daughter nature.
The fitting procedure is a simple analytical ohne. For a decay curve of the type

$$f(t) = \sum_{i=1}^{n} A_i^o \, e^{-\lambda_i t}$$

the minimization of $\chi^2$ is performed

$$\chi^2 = \sum_i \frac{1}{\sigma_i^2} \left\{ R_i - \sum_{j=1}^{n} A_j^o \, e^{-\lambda_j t_i} \right\}^2$$

where

$\sigma_i^2$ : experimental variance of the measured activity

$R_i$ : measured activity at time $t_i$

$t_i$ : time

$A_j^o$ : initial activity of the $j^{th}$ isotope

$\lambda_j$ : decay constant of the $j^{th}$ isotope

The minimization with respect to the parameters $A_j^o$ gives a set of n independent linear equations which are easily solved by a Gauss-Pivot algorithm /6/.

The situation is not as simple for a parent-daughter decay because constraints are imposed on the fit.
For a parent-daughter decay of the type

$$N_1 \overset{\lambda_1}{\rightarrow} N_2 \overset{\lambda_2}{\rightarrow} N_3$$

one finds for the activity of the nuclide $N_2$

$$A_2(t) = \frac{\lambda_2}{\lambda_2-\lambda_1} A_1^o \left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right) + A_2^o e^{-\lambda_2 t}$$

where $A_1^o$, $A_2^o$ are the activities at time t = 0 for the nuclides $N_1$, $N_2$ respectively. This may be written as

$$A_2(t) = C_1 e^{-\lambda_1 t} + C_2 e^{-\lambda_2 t}$$

Now $C_1$ and $C_2$ are not necessarily positive but only the time-zero activities $A_1^o$ and $A_2^o$ are. Thus we have inequality constraints imposed on the parameters of the type

$$A_1^o = \frac{1}{\lambda_2} C_1(\lambda_2-\lambda_1) \equiv h_1(C_1,C_2) \geq 0$$

$$A_2^o = C_1 + C_2 \qquad \equiv h_2(C_1,C_2) \geq 0$$

We now minimize $\chi^2$ with respect to the parameters $C_1$ and $C_2$. If the minimum occurs at a point $(C_1^+, C_2^+)$ which is in the interior of the feasible region, that is $A_{1,2}^o > 0$, then the constraints are irrelevant and the fit is like an unconstrained one. If the minimum found is within the infeasible region or on the boundary, the procedure has to be repeated in a modified version. The modification is to change $\chi^2$ in such a way that it remains almost unchanged in the interior of the feasible region, but increases drastically as one approches the constraints. To accomplish this, we assign a function to each inequality constraint, which is nearly zero when the constraint function is strongly positive, but increases sharply as the constraint function aproaches zero from above. To the constraints above we assign the functions

$$B_i(C_1,C_2) \equiv \alpha_i/h_i(C_1,C_2), \qquad i = 1,2$$

Where $\alpha_i$ are small positive constants. We now modify $\chi^2$ by adding to it the functions assigned to the constraints:

$$\tilde{\chi}^2(C_1,C_2) \equiv \chi^2(C_1,C_2) + \sum_i \alpha_i/h_i(C_1,C_2)$$

The numerical procedure to find the minimum in the feasible region is now as follows /6/:
select the $\alpha_i$ and a feasible initial guess of $C_i$, find $(C_1^*, C_2^*)$ using the Marquard algorithm for minimzing the now unconstrained problem $\tilde{\chi}^2$, reduce the values of the $\alpha_i$ and repeat the minimization using the values $C_1^*$ , $C_2^*$ as initial guesses.

The process is continued until $C_1^*$, $C_2^*$ do not change significantly upon reducing the $\alpha_i$. Then we accept $C_1^*$, $C_2^*$ as our best estimates of the parameters. It has been proven by Fiacco and Mc Cormick /7/ that under suitable conditions this type of procedure is convergent.

Having performed all fits of the candidate nuclides to the experimental points, we are in the situation to select the best fits by their values of the reduced $\chi_r^2$ . Fits with reduced $\chi_r^2$ less than 2.5 are presented besides the plot of experimental data. At least three of single-nuclide type and three of double-nuclide type are printed even if the reduced $\chi_r^2$ is greater than 2.5. The best fits are drawn in the plot for visual aid.

## OUTPUT INFORMATION OF YELLOW

The ourput of YELLOW contains all information necessary to identify the nuclides. Figure 3 presents an example of such an output.
Besides the plot of the calculated peak count rate versus time for the peak energy indicated in the right corner a list of data is printed in the left part. This list includes the identification of the sample ("51 Pb 2"), the tagward (5214), the time since end of bombardment for the midpoint of the counting interval in hours, the counting live time, the FOM value computed in AGAMEMNON, the error of the peak count rate and the peak count rate itself, the energy of the peak centroid, the full width at half maximum of the peak, the counting efficiency and the dead time correction. All of this information serves as control output.
The next lines present estimated halflives and time-zero-activities assuming one or two nuclides fitting the experimental points. These numbers are not used by the program later. They only help the user orienting in the order of magnitude of halflives encountered.
The next lines represent the essential output of YELLOW. This list of candidates is ordered by the candidates atomic numbers and masses. A hint is given to the user showing which candidates fit the experimental data

best. These are the entries E1, E2, for single-nuclide fits and Z1, Z2,.... for double nuclide fits.
Double nuclide fits and parent-daughter decays are indicated below the plot of experimental data. The name of the respective nuclide, its halflife, the time-zero-activities and the reduced $\chi^2$ are given. The complete description of the nuclide is presented in the left part. This includes: the halflife, the time-zero-activity assuming one nuclide fitting the data, the mean difference between spectral peak energy and listed peak energy for all $\gamma$-transmissions of the nuclide, the factor $e^{-\lambda t}$, the ratio of the total line intensities of the nuclide found by the peak search to the one listed in the Erdtmann-Soyka-Compilation.

The next entries give the spectral peak energies which are assigned to the lines of the nuclide listed. A zero in this column means that this line of the nuclide is missing in the spectra. The number is the number of the plot (the page-number) where the respective peak energy and the type of analysis described here can be found. The last column is the nuclides gamma-line intensity. All time-zero-activities have to be divided by this number to get the activity of the nuclide identified.
In this way the user gets a complete set of information helping him to decide on the proper assignments of nuclides to each peak.

## CONCLUSION

The analyzing scheme described here has been developed to translate the extremely complicated gamma-ray spectra resulting from gamma-ray spectrometry of irradiated foils into a data set of spallation and fission products present in these irradiated foils. The complexity of this task makes it impossible to the experimenter to do so by hand. Only with the help of the programs described it was possible to extract information from the gamma spectra resulting from the many nuclides produced from spallation in lead.

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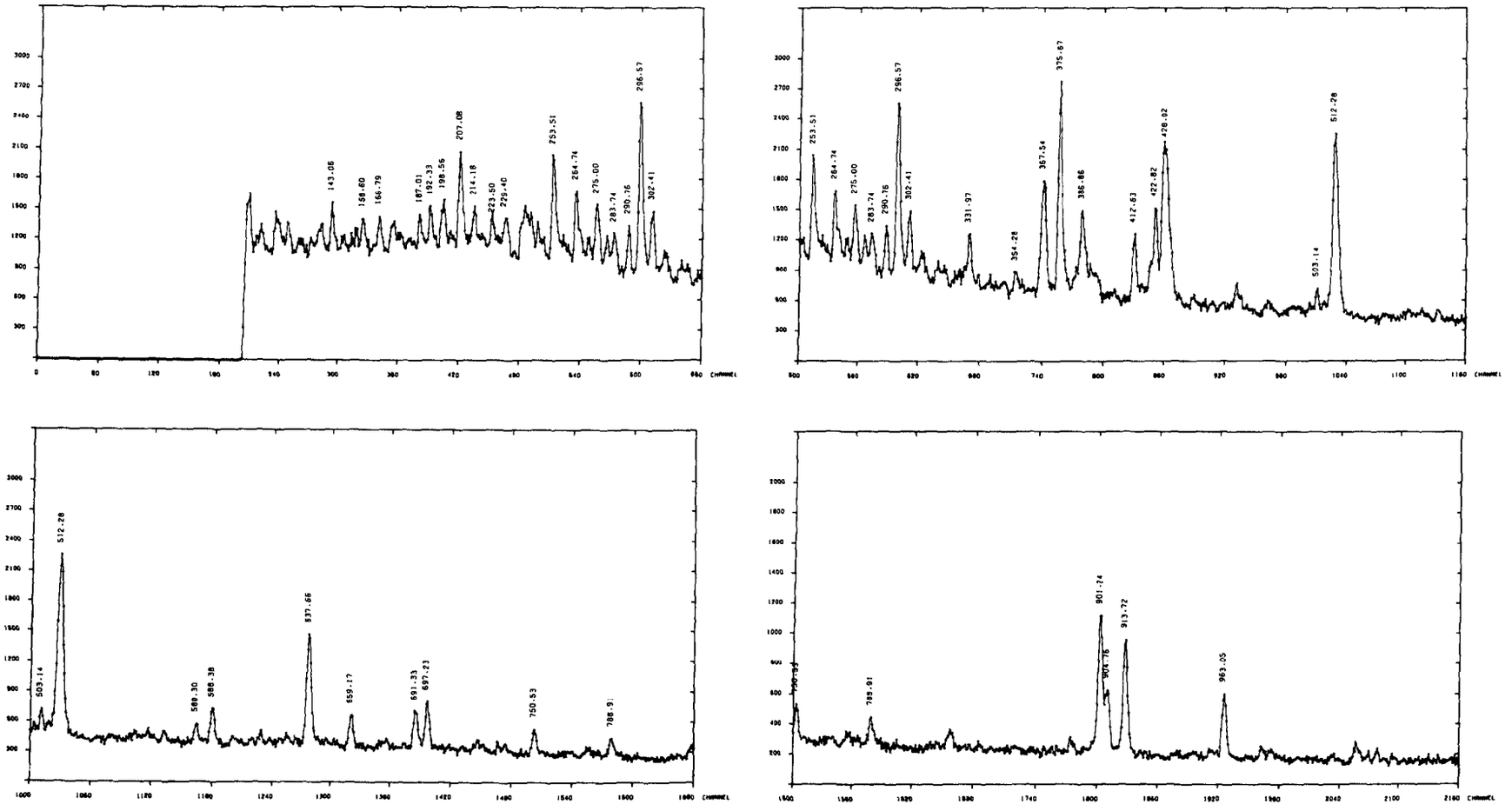


Fig. 1: Typical spectrum from Pb target 5 cm deep in the over-all target taken one houer after the proton irradiation (numbers on peaks are the energies)

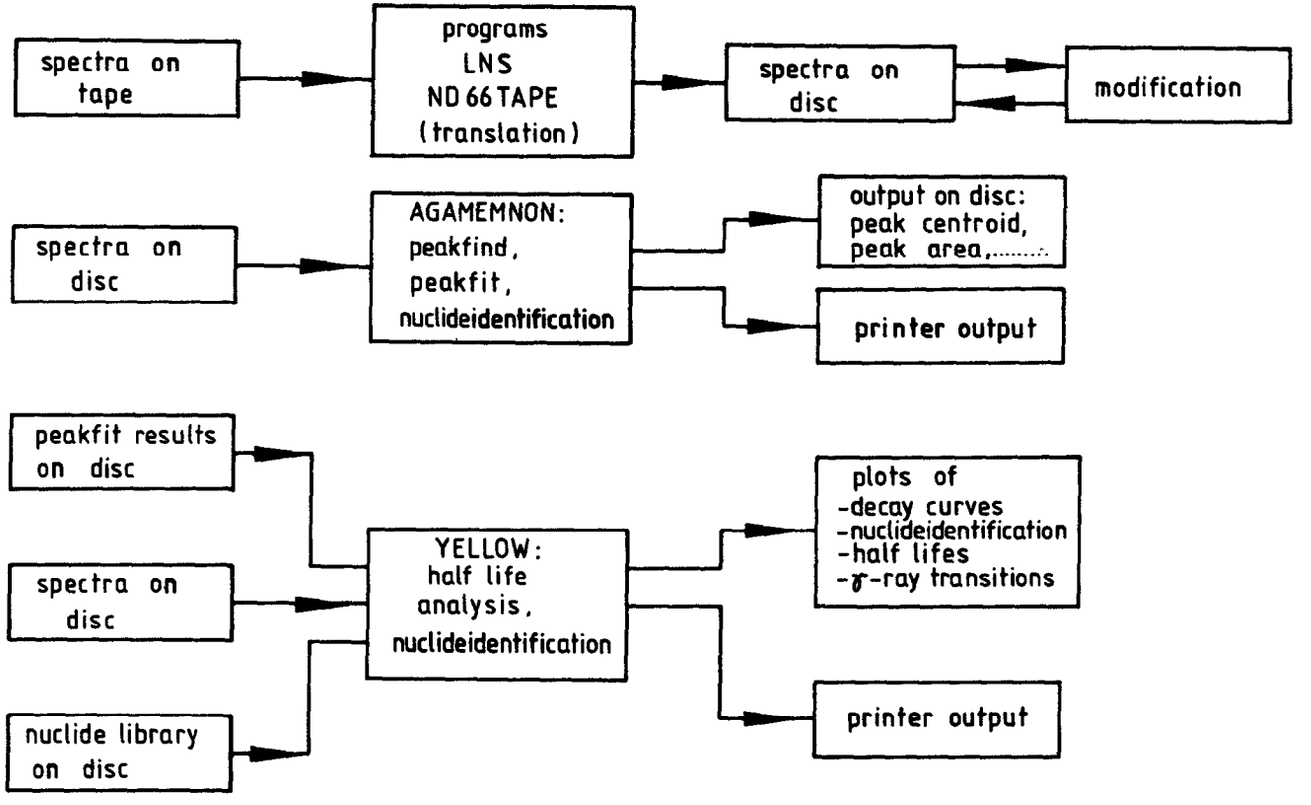


Fig. 2: Flow diagram of the analysis. Refer to text for further details

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 ENERGY       ERROR     CORR
  621.118      0.583   1.060E+00
   ID          S-EOB      LIVETIME     % RESIDUE  CPS ERROR     CTS/S      ENERGY     FWHM     EFFICIENCY    DTCORR
S1PB2         6.002E+00  3.370E+02  3.181E+00  1.611E+04  1.289E+05  621.12  2.862E+00  1.022E-04  1.105E+00
S1PB2         1.274E+01  6.270E+02  6.773E-01  9.026E+03  7.851E+04  621.76  2.706E+00  3.149E-04  1.151E+00
S1PB2         2.067E+01  1.186E+03  1.383E+00  1.201E+03  6.133E+04  621.23  2.328E+00  3.149E-04  1.085E+00
S1PB2         3.085E+01  1.329E+03  1.544E+00  8.348E-02  4.841E+04  621.20  2.290E+00  3.149E-04  1.046E+00
S1PB2         5.486E+01  3.295E+03  2.280E+00  4.562E+02  1.703E+04  620.95  2.297E+00  3.149E-04  1.020E+00
S1PB2         1.614E+02  1.030E+04  1.414E+00  5.892E+01  1.278E+03  620.38  2.351E+00  9.941E-04  1.009E+00
S1PB2         1.735E+02  1.035E+04  1.540E+00  5.347E+01  1.125E+03  620.43  2.403E+00  9.941E-04  1.009E+00
S1PB2         2.126E+02  2.094E+04  2.789E+00  3.355E+01  7.948E+02  619.93  2.507E+00  9.941E-04  1.006E+00
S1PB2         2.742E+02  2.114E+04  5.869E+00  2.446E+01  4.344E+02  620.21  2.281E+00  9.941E-04  1.004E+00
S1PB2         3.355E+02  2.126E+04  3.867E+00  2.085E+01  3.322E+02  620.09  2.305E+00  9.941E-04  1.002E+00
S1PB2         3.596E+02  2.129E+04  3.914E+00  1.957E+01  2.595E+02  620.03  2.232E+00  9.941E-04  1.002E+00
S1PB2 S403    6.451E+02  4.289E+04  2.144E+00  8.129E+00  9.293E+01  620.42  2.209E+00  9.931E-04  1.000E+00
S1PB2 S407    7.042E+02  4.222E+04  1.725E+00  8.361E+00  4.834E+01  620.37  2.178E+00  9.931E-04  1.000E+00
HLF=  4.55E+01 HRS    AO =  5.81E+04 1/SEC    LOG10(AO*SEC) =  4.76E+00
AO-LONG-    =( 2.08E+03 +- 4.16E+01) 1/SEC HLF-LONG-  =( 1.33E+02 +- 1.68E+00) HRS
AO-SHORT-   =( 1.36E+05 +- 1.45E+03) 1/SEC HLF-SHORT- =( 1.85E+01 +- 1.45E-01) HRS
CHISQR      = 8.90E-00
 621.12  119          HALFLIFE      HLF/H        AO   (E-EO)/N     DECAY      1/IO    PEAK  NR    LINE     INTENS
    Z3  65 TB 152   1.75E+01 H  1.75E+01  4.73E+05  9.97E-01  7.70E-13  8.35E-01  344.43  61  344.28  8.60E+01A
                                                                              511.38  94  511.00  1.74E+01A
                                                                              585.36 110  586.29  1.25E+01A
                                                                              621.12 119  622.80  1.25E+00A
+ E1    67 HO 160M  5.10E+00 H  2.86E+01  1.26E+05  1.04E+00  3.78E-08  4.91E-01 1272.46 234 1271.50  2.90E+00A
  CHISQR= 9.32E+01                                                            621.12 119  620.80  0.00E+00
◆ E4    71 LU 170   2.00E+00 D  4.80E+01  5.43E+04  9.87E-01  3.83E-05  7.79E-01 1280.96 236 1280.25  7.93E+00A
  CHISQR= 4.45E+02                                                            985.52 188  985.10  5.38E+00A
                                                                              621.12 119  621.40  4.35E-02A
                                                                              621.12 119  622.75  2.46E-02A
+ E4    71 LU 170   2.00E+00 D  4.80E+01  5.43E+04  9.87E-01  3.83E-05  7.79E-01 1280.96 236 1280.25  7.93E+00A
  CHISQR= 4.45E+02                                                            985.52 188  985.10  5.38E+00A
                                                                              621.12 119  621.40  4.35E-02A
                                                                              621.12 119  622.75  2.46E-02A
    Z2  72 HF 170   1.60E+01 H  1.60E+01  6.51E+05  9.41E-01  5.64E-14  9.89E-01  165.19  12  164.71  3.35E+01A
                                                                              621.12 119  620.70  2.29E+01A
                                                                              573.60 108  572.90  1.85E+01A
    Z1  77 IR 186   1.58E+01 H  1.58E+01  6.82E+05  1.04E+00  3.83E-14  8.76E-01  296.18  48  295.75  4.62E+01A
                                                                              137.28   4  137.15  2.80E+01A
                                                                              434.96  82  434.78  2.38E+01A
                                                                              621.12 119  622.20  2.31E+00A
Y E3Z1  77 IR 188   1.73E+00 D  2.45E+02  2.00E+04  1.03E+00  1.36E-01  8.73E-01  154.94   9  155.03  3.34E+01A
  CHISQR= 2.75E+02                                                            633.80 123  633.10  2.16E+01A
                                                                              479.44  89  478.10  1.60E+01A
                                                                              621.12 119  620.60  1.00E+00A
* E2    77 IR 194   1.91E+01 H  5.26E+04  1.57E+04  1.04E+00  9.91E-01  8.19E-01  326.81  56  328.54  1.30E+01A
  CHISQR= 1.01E+02                                                            645.80 127  645.32  1.16E+00A
                                                                              621.12 119  622.30  3.10E-01A
I E6    79 AU 194   1.65E+00 D  3.95E+01  7.09E+04  8.63E-01  4.30E-06  7.38E-01  326.81  58  328.50  6.09E+01A
  CHISQR= 4.98E+02                                                              0.00   0  293.60  1.07E+01A
                                                                              621.12 119  622.00  1.70E+00A
                                                                              621.12 119  621.20  6.09E-01A
* E6    79 AU 194   1.65E+00 D  3.95E+01  7.09E+04  8.63E-01  4.30E-06  7.38E-01  326.81  56  328.50  6.09E+01A
  CHISQR= 4.98E+02                                                              0.00   0  293.60  1.07E+01A
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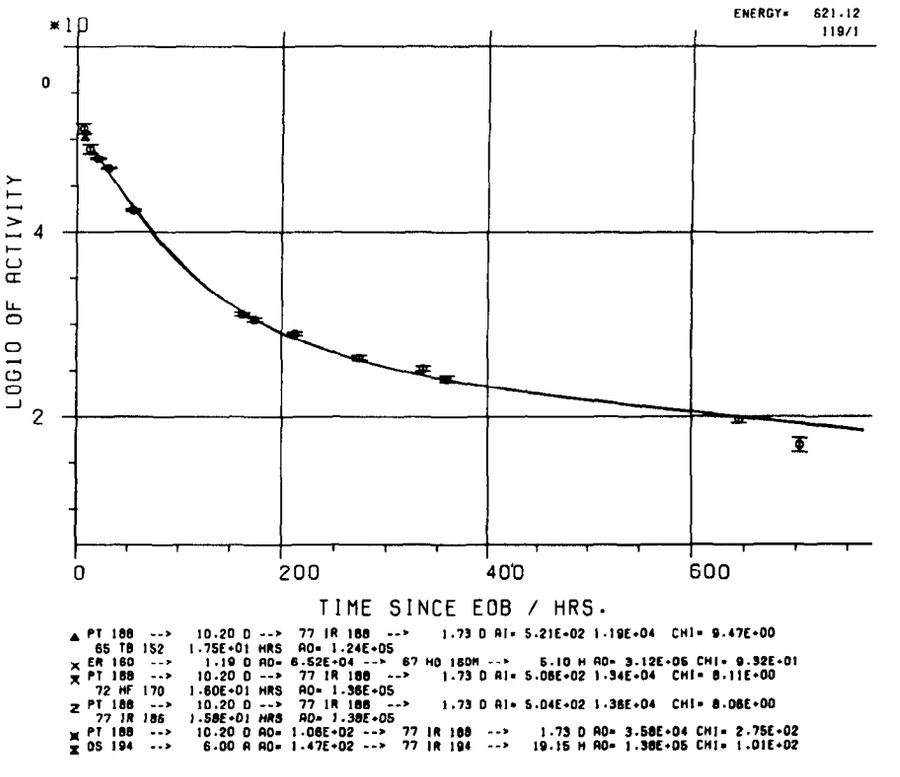


Fig. 3: Typical output information of YELLOW. Refer to text for further details