Comitato Nazionale Energia Nucleare

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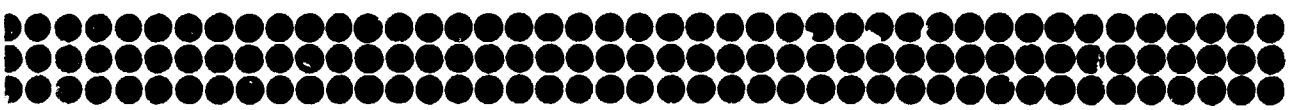

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1. INTRODUCTION

The present paper summarizes the main features of two computer codes, CERPI and CEREL, which perform a fully automatic quantitative analysis of thermal neutron activated samples.

The CERPI program analyses experimental gamma-ray spectra in order to find the photopeak positions and areas. The CEREL program looks for those elements or nuclides which are potential emitters of the gamma-lines. The search may be performed by elements or nuclides, depending on an input parameter. For the sake of simplicity we shall assume from now on that a search by nuclide is to be carried out.

The CERPI and CEREL programs are written in FORTRAN IV language and are linked together working as a single code. A schematic flow diagram is shown in fig. 1.

2. - THE CERPI PROGRAM

CERPI is a code for the fully automatic analysis of complex gamma-ray spectra obtained with Ge (Li) detectors. The significant peaks in the spectrum are detected by observing the behaviour of the second derivative function after random variations in counts per channel have been minimised by the application of a smoothing filter function. Particular care has been taken in the recognition of weak or adjacent peaks, even in the presence of large statistical fluctuations. This was achieved by means of a check procedure included in the fit routine. The detailed description of the CERPI program is given in a previous report[1]. In the following we shall only describe their main features.

2.1 - Peak Identification

In order to locate the peaks in the experimental spectrum, the CERPI program computes the convolution between the spectrum function f (y) (that is the counts per channel versus channel number) and the smoothing filter function given by

$$b\ (y-x) = \frac{d^2}{dx^2}\left[\sqrt{k/\pi}\ \exp\left(-k\ (y-x)^2\right)\right] \qquad (1)$$

where $k = 4\ (\ln 2)\ /\Delta^2$ and Δ is the full width at half maximum (fwhm). If the function f (y) is given by the linear combination

of a Gaussian function (representative of the peak) with a linea- function (representative of the background), the convolution

$$F(x) = \int_{-\infty}^{+\infty} b(y-x)\, f(y)\, dy \qquad (2)$$

is exactly the second derivative of a Gaussian function centered at the peak position and with a fwhm equal to $\Delta\sqrt{2}$. The analysis of the convolution function F (x) makes it possible to locate in the spectrum peaks with a correct shape and a height h above the background greather than

$$h_{min} \geq 2 \sqrt{\frac{f(x_o)}{\Delta}} \qquad (3)$$

where $f(x_o) = \max |\bar{f}(x)|$ in the peak region.

2.2 - Fitting Procedure

Once the peaks have been identified the program determines their mass centers, areas and corresponding errors with a least-squares fit. Each significant peak is fitted with a Gaussian function superimposed on a linear or quadratic background. If several adjacent peaks mutually interfere the whole region surrounding these peaks is examined and up to a maximum of ten Gaussian functions are fitted together, with the same f.w.h.m. The determination of the minimum of the χ^2 function is accomplish

ed by a variant of the gradient method. The behaviour of the fit is checked step by step. New peaks may be introduced when either the f.w.h.m. exceeds its expected value by more than 25%, or when the position of a peak shifts by more than $\Delta/4$ with respect to the initial center of mass. Conversely the smallest peaks in a group may be eliminated any time a satisfactory fit is not reached. In such a case it is safe to assume that one or more spurious peaks prevented a good fit from being obtained.

3. - THE CEREL PROGRAM

The CEREL program was developed for automatic isotope identification on the basis of gamma-ray energy comparison. It contains features such as determination of peak energies and intensities, nuclide identification, and mass computation. For a compiete description of CEREL the reader is referred to a previous report[2].

3.1 - Peak Energy and Intensities

The energy calibration curve is determined starting from some known energy lines and by means of a fitting procedure with orthogonal polinomials, using the F test (with a 5% signific ance level) for the automatic determination of the polinomial order. An input parameter allows the program to choose among the following options:

- a calibration is available from a previous spectrum,
- the calibration spectrum is superimposed on the spectrum being analysed,
- the spectrum being analysed is a calibration spectrum.

The errors on the interpolated energies include contributions from both peak position uncertainties and errors in the orthogonal polinomial coefficients.

In order to calculate gamma-ray intensities, the photopeak efficiency and the FEP/DEP and SEP/DEP ratios versus energy are approximated by polinomials, whose coefficients are given as input data. The single and double escape peaks are used for a more careful determination of the photopeak intensities and then removed from the observed spectrum.

3.2 - Nuclide Identification

For both nuclide identification and mass calculation the isotope catalogue of Pagden et al.[3] has been used. Some changes were needed in order to make it suitable to our purposes. A new version of the catalogue has therefore been generated with the following main differences: only the isotopes arising from thermal neutron activation are included; for each isotope only those parameters are included that are needed for the mass calculation; the cross sections of the isomeric transitions and the corresponding gamma-ray intensities have been corrected so that the two isomers may be considered as a normal parent-daughter relationship; some parameters have been changed where more recent data were available.

The catalogue is made up of 156 nuclides correponding to 73 elements. Four different types of decay schemes have been identified and the masses of the corresponding activated nuclide have been calculated by four different methods. In

four cases only (Zr^{94}, Pd^{110}, Cd^{116} and Te^{130}) there was an unstable daughter product and in such cases the program lists only the gamma rays belonging to the nephew. The Lu^{177} nuclide was handled separately because of its complicated decay scheme.

The spectrum is examined peak by peak. For each peak a search is made to find those isotopes that emit gamma-rays within the error range of this peak energy. Nuclear data are organised and cross indexed so that the main code can easily find the information it needs, without time consuming searches through the catalogue.

Following the GASPAN code[4] a first test is applied to the list of all possible nuclides that allows the elimination of those of them for which the absence of characteristic lines in the spectrum may by no means be justified in terms of statistical fluctuations. With the remaining nuclides a matrix M is then created in which the element m_{ij} represents the mass of the j-th element calculated on the hypothesis that this element is the only responsible of the i-th line. As far as the experimental error on m_{ij} is concerned, we took into account only the peak area uncertainty.

3.3 - Mass Calculation

The dimensions of matrix M are generally large. It is nevertheless possible[5] to exstract from it independent submatrices, which are solved separately by the program.

For a submatrix with m column and n rows three possibilities may occur, depending on $m \lesseqgtr n$. When $m < n$ a least squares solution is attempted. When $m > n$ a search is made for gamma-rays which are not shared among different nuclides. If such gamma-rays are found the nuclide masses may be independently calculated. If the condition $m > n$ yet holds for the remaining submatrix, upper limits for the masses of the involved nuclides are given.

4 - RESULTS

As an example, we report some results concerning the analysis of a lunar sample of about 10^{-8} g, irradiated for one hour in a thermal flux of $2.6 \cdot 10^{13}$ n $cm^{-2} sec^{-1}$.

Table 1 shows a portion of the CERPI's output for a small region of the gamma spectrum. The χ^2 values and the number of degrees of freedom refer to the peak mass-center and area determination.

Table 2 shows the listing of the same spectrum region after energy calibration and spectrum cleaning have been performed. It can be seen that among the removed peaks, there are the DEP and SEP (peak N. 69 and 75 respectively) of the last gamma-line (2754 KeV Na^{23} line).

In table 3 the identified nuclides are shown. Nuclides # 1÷4, and 5÷6 result from two independent submatrices, while the remaining ones have no common gamma-rays. In the last case both standard errors and weighted root mean square deviations are given. We point out that no error has been taken into account on either deterctor efficiencies or catalogue gamma-ray intensities. The analysis of a 4096 channel gamma-ray spectrum, from the above mentioned activated sample, requires less than 60 seconds of IBM 360/75 CPU time, and about 500 K bytes of core memory.

5. - CONCLUSIONS

Many complex pulse height spectra taken with a 4096 channel analyser have been studied with CERPI.The high precision and sensitivity make it very suitable to very complex spectrum analyses where many peaks overlap each other. Some attention must be paid to this high sensitivity because often this implies the introduction of some spurious peaks. Particular care must also be taken in the choice of the input parameters because the fitting procedure is affected by this choice.

As far as CEREL is concerned, its limits stem directly from those inherent to the CERPI code. The higher the number of peaks identified, especially the number of spurious ones, the higher is the probability to introduce fictitious elements, and the more complicated is the resolution of the mass matrix.

As a consequence, a full automatic analysis without operator's intervention requires a low sensitivity in the peak identification procedure, even if this implies the loss of some element responsible of low intensity gamma rays. On the other hand it is possible to obtain the maximum possible information by running the CERPI and CEREL programs twice or even more times, varying the most critical input parameters. In order to overcome such a loss of computer time, an interactive version of these two programs should be used.

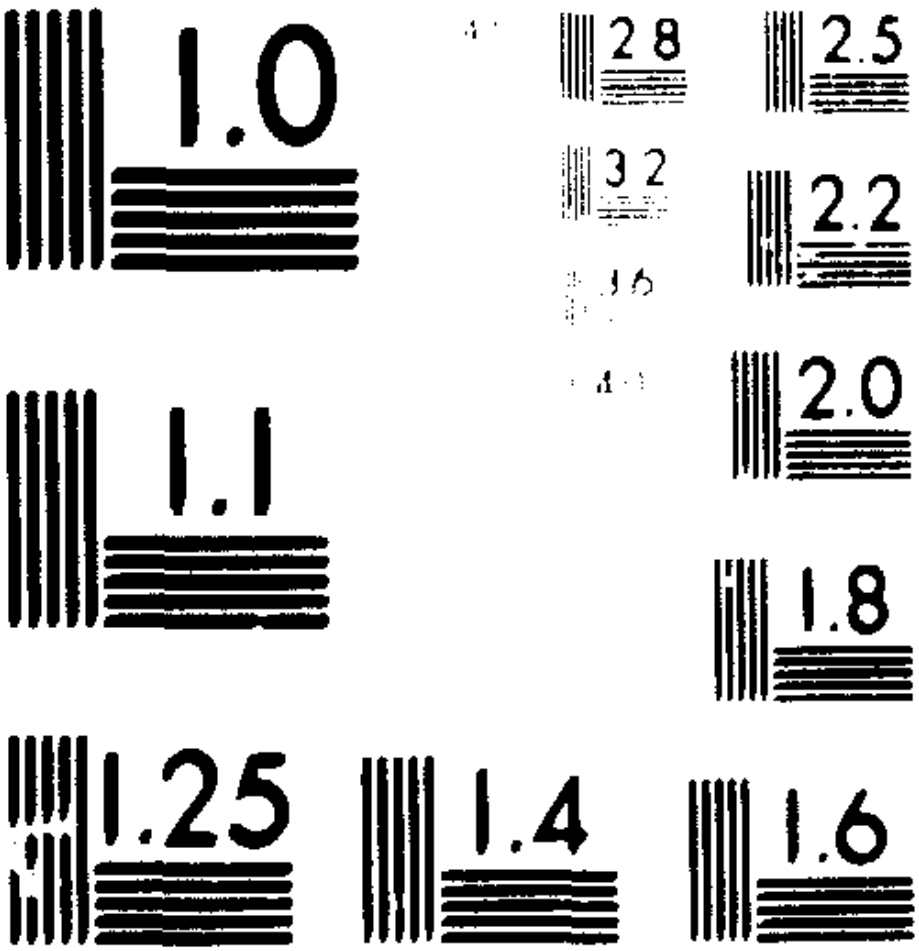
1.0
2.8
2.5
3.2
2.2
3.6
4.0
2.0
1.1
1.8
1.25
1.4
1.6

TABLE 1

CERPI'S RESULTS

Peak No.	Mass-center and error		Area and error		Fwhm and error		χ^2 and degree of freedom	
64	1330.09	0.04	2749.	61.	3.88	0.09	42.	12.
65	1532.82	0.70	24.	16.	1.91	1.67	29.	23.
66	1547.81	0.70	55.	16.	4.21	0.19	22.	21.
67	1554.32	0.09	755.	34.	4.21	0.19	22.	21.
68	1685.47	0.31	171.	6.	3.73	0.30	35.	13.
69	1688.97	0.14	488.	32.	3.73	0.30	35.	13.
70	1788.48	0.68	15.	10.	2.07	0.80	22.	41.
71	1793.73	0.69	13.	9.	2.07	0.80	22.	41.
72	1802.02	0.56	16.	9.	2.07	0.80	22.	41.
73	1814.82	0.96	20.	11.	2.07	0.80	22.	41.
74	1821.43	0.73	12.	9.	2.07	0.80	22.	41
75	2192.42	0.24	177.	26.	4.20	0.66	18.	14.
76	2295.73	1.05	19.	13.	2.52	1.06	16.	21.
77	2696.22	0.06	925.	32.	3.93	0.11	28.	26.

TABLE 2

COMPUTED ENERGY AND EFFICIENCY OF OBSERVED GAMMA-RAYS

Peak No.	Mass-center and error		Energy and error		Photopeak efficiency
64	1330.09	0.10	1368.50	0.13	4.549E-03
65	1532.82	0.70	1574.23	0.72	4.049E-03
66	1547.81	0.70	1589.44	0.71	4.019E-03
67	1554.32	0.10	1596.02	0.12	4.006E-03
68	1685.47	0.31	1729.11	0.32	3.768E-03
70	1788.48	0.68	1833.61	0.70	3.605E-03
72	1802.02	0.56	1847.35	0.58	3.585E-03
73	1814.82	0.96	1860.33	0.98	3.566E-03
74	1821.4.	0.73	1867.03	0.75	3.556E-03
76	2295.73	1.05	2349.36	0.59	3.017E-03
77	2696.22	0.10	2754.47	0.14	2.987E-03

TABLE 5

IDENTIFIED NUCLIDES AND CORRESPONDING MASSES AS COMPUTED BY CEREL

N.	Isotope	Mass and error (gm)		Wgh.St.Dev. (gm)	Observed γ-ray energies (KeV)
1	Lu-176	5.27 E-13	5.4 E-13	-	113,208
2	Yb-174	4.85 E-11	9.6 E-12	-	113,283,396
3	Pt-198	5.72 E-11	3.3 E-11	-	159,208
4	Ru-104	9.73 E-10	1.4 E- 9	-	208
5	Eu-151	1.65 E-12	3.1 E-14	-	296,344,122,841, 963,970
6	Er-170	5.36 E-12	2.5 E-12	-	296
7	Pr-141	1.17 E-10	7.6 E-11	0	1574
8	Sm-152	5.87 E-11	4.6 E-13	0	103
9	Nd-150	1.59 E-10	9.9 E-11	5.5 E-11	164,176
10	Ce-142	8.81 E-11	2.6 E-11	0	294
11	La-139	2.23 E-10	5.6 E-12	2.9 E-11	433,487,751,816, 329,1596,2349
12	Sb-123	4.63 E-10	1.7 E-10	0	603
13	Ag-109	2.82 E- 9	1.1 E- 9	0	657
14	Mn- 55	8.51 E-12	7.1 E-13	0	847
15	Sc- 45	1.37 E-10	3.0 E-11	0	889
16	Na- 23	9.35 E-10	1.6 E-11	1.5 E-10	1368,2754

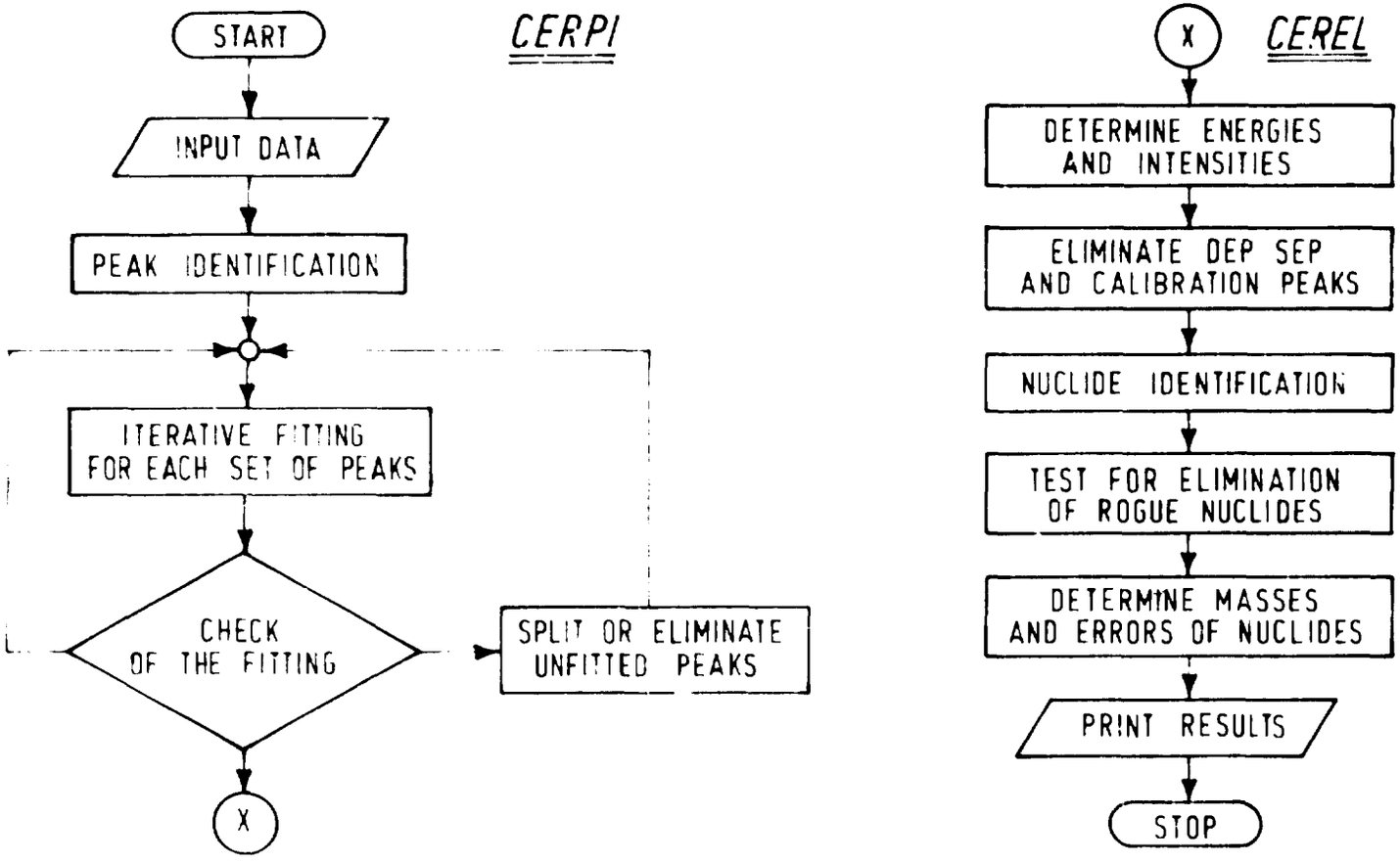

Fig. 1 - Schematic flow diagram of CERPI and CEREL programs

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