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R Khelifi

Centre de Developpement des Techniques Nucleaires, Algeria

A Tchantchane

University of Wollongong, tchantch@uow.edu.au

Z Idiri

Centre de Developpement des Techniques Nucleaires, Algeria

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Gamma ray spectra simulation and optimization in neutron activation analysis

R Khelifi, A Tchantchane and Z Idiri, *Centre de Developpement des Techniques Nucleaires, 2 Bd Frantz Fanon, B.P. 399, Alger Gare, Algeria*

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A software package for the simulation of neutron activation spectra has been developed. The simulation of the 14 MeV neutron activation gamma ray spectra has been carried out both for cyclic and conventional modes. The simulation allows the prediction of the optimum parameters for a low level of detection. The parameters are irradiation, counting and decay times. The simulation was based on an accurate calculation of various parameters such as the photoelectric yield, detector efficiency and background estimation. Compton effect, single and double escape are also taken into account.

This software was extensively tested using the IAEA certified material SL1 lake sediment and $\text{Al}(\text{OH})_3$ chemical standard. A high agreement between the theoretical simulated spectra and those measured is obtained.

The second part of the package implements the optimization of the timing parameters. The Monte Carlo method was used to search within a constrained time space for the optimum condition resulting in the lowest detection limit for a given reaction. In order to illustrate the validity of the optimisation routine, sodium is determined in SL1 using the reaction $^{23}\text{Na}(n,\alpha)^{20}\text{F}$. Copyright © 1996 Elsevier Science Ltd

Introduction

The concept of simulation was initiated in Neutron Activation Analysis in order to predict and estimate some fundamental parameters such as detection limits. El Mugarbi and Guinn^{1,2} have investigated this concept for sample analysis in thermal neutron activation.

Several authors have given detection limits for elements with the assumption of zero background noise. Therefore these data cannot be reliable for our needs since the background noise is mostly generated from the activation of elements which are in the sample. Hence the detection limit of a given element may differ from one type of sample to another.

A program for the simulation and optimisation of neutron activation analysis is developed in our laboratory. The detection limit is calculated from nuclear data contained in the library which includes neutron induced nuclear reactions, neutron beam intensity and sample composition. The detector response is also considered.³

First photoelectric peaks are modelled by gaussians of width σ . The gaussian base is spread over $+3\sigma$ and -3σ . The Compton continuum described by Zykowski model,⁴ both the Compton front and the backscatter peak are included in the spectrum. The simple and the double escape peaks are included in the simulated spectrum and treated in the same way as photoelectric peaks.

The optimisation of the experimental time parameters (irradiation, counting and decay times) is based on a Monte Carlo approach:⁵ at each iteration, random values to the time

parameters are generated and new elemental detection limits are calculated using the ratio of noise with respect to the intensity of the photoelectric peak. Time parameters giving a minimum detection limit for a given element are set to be the optimum experimental parameters for the analysis of that element.

In order to test the reliability of the simulation, two samples, SL1 and an aluminium chemical standard $\text{Al}(\text{OH})_3$, are irradiated with a 14 MeV neutron generator. Experimental and simulated spectra are compared and good agreement is obtained. Further, in order to illustrate the validity of the optimisation routine, sodium is determined in the IAEA reference material SL1 where the reaction of interest is $^{23}\text{Na}(n,\alpha)^{20}\text{F}$ (^{20}F half life = 11 s) and the analytical peak energy is 1634 keV.

Software description

The simulation software was developed in Fortran 77 on an IBM personal computer. The program requires as input the sample composition, detector characteristics and the data library named 'LIBRAPID' (for 14 MeV neutrons) containing nuclear tables of elements' isotope nuclear abundance, atomic masses, reaction cross sections, isotopic half life and gamma ray energies with their relative intensities. The time parameters (irradiation, decay and counting times) are also entered into the program. The simulated spectrum is visualised on the screen. Data related to photoelectric peak reactions, energy, area, resolution, detector efficiency, transmission factors, escape peak areas, Compton ratios and background noise are saved with the simulated spectra.

Table 1. SL1 lake sediment composition used as input for the simulation

Element symbol	Concentration (ppm)	Element symbol	Concentration (ppm)	Element symbol	Concentration (ppm)	Element Symbol	Concentration (ppm)
Sb	1.31	Se	2.90	Na	1720	Y	85
Cu	30.0	Th	14.0	P	831	Ga	24
Ag	0.08	Br	6.82	K	15000	Ca	2500
Xe	0.01	Hf	4.16	Pb	37.7	Si	200000
Ge	25.0	Rb	113.0	Zr	241	Cr	104
As	27.5	Au	0.01	Ba	639	Mn	3400
Ti	5170.0	V	170.0	Se	117	B	39
Sc	17.30	Sr	80.0	Al	89000		
Cl	10.0	Mg	29 000.0	Fe	67400		

Based on a Monte Carlo approach, optimisation of time parameters (irradiation, decay and counting) are determined for each element of interest. This method allows us to search for time parameters that give the minimum detection limit for a given element. At each iteration, the three time parameters values are randomly selected within a bounded interval and a new detection limit is calculated using the simulation program. In case the detection limit is improved these new values are kept as being optimum values otherwise they are ignored. The number of iterations may be set by the set user.

Results and discussion

In order to evaluate the performance of the simulation, an IAEA sediment standard 'SL1 lac' is analysed. The sample (mass of 7.852 g) is irradiated for 600 s using a 14 MeV neutron generator of type SAMES J25. After the irradiation the activity in the sample was measured for 1800 s after letting it decay for 500 s. A hyperpure Germanium detector with a 2 keV resolution and a 20% relative efficiency for 1332 keV gamma energy is used. The detector is coupled to an ORTEC 7150 multichannel analyser. The energy calibration of the detection system is given by:

$$E = 2.73 \times \text{channel} + 87.70(\text{keV})$$

A neutron flux of $4.8 \times 10^7 \text{ cm}^{-2} \text{ s}^{-1}$ is experimentally determined for the $\text{Al}(n,p) \text{ Mg}$ reaction having a 568 s half life and 844 KeV gamma ray (Table 1).

The experimental parameters of the analysis are shown in Table 2.

Figure 1 displays both the experimental gamma ray spectrum and the simulated one for the SL1 sample. We note that the annihilation peaks at 511 KeV due to β^+ emitters are included in the experimental spectrum. This explains the difference in intensity between the two spectra in the region of 511 KeV. Overall, the intensity of the simulated spectrum is slightly less than that of the experimental spectrum. This may be due to the fact that not all the possible reactions are included in the input library since part of the continuum background originates from these reactions. However, this difference does not affect very much the detection limit calculations. The marked peaks on spectra are defined in Table 3.

The simulation program is further tested with a chemical standard, $\text{Al}(\text{OH})$ having a weight of 8.73 g. The neutron flux used was $3.1 \times 10^7 \text{ cm}^{-2} \text{ s}^{-1}$ during 180 s. The activity is measured

Figure 1. Experimental and simulated of the sediment IAEA certifies material SL1.

Table 2. Experimental conditions

Energy = $2.74 \times \text{Channel} + 87.36$
Resolution = $0.002 \text{ Energy} + 0.2$
Solid angle = 1.6467 sr
Sample mass = 7.852 g
Number of cycles = 1

during 600 s after a decay time of 180 s. Both the experimental and the simulated spectra obtained are shown, respectively in Figure 2. Since the sample contains essentially Al (34.5%), only the nuclear reactions induced on this element appear on the spectra. The simulated spectrum shows a perfect similarity with the experimental spectrum. The photoelectric peak areas are the same in the two spectra.

The minimum detection limits and the corresponding time parameters are given in Table 4 for SL1. For each element, the execution of the software which is developed on the VAX 785 had taken up to 3 and 34 min where the number of iterations was set to tens of thousands.

This software is used to predict the timing parameters for the minimum detection limit of sodium using the 1634 keV analytical peak. As shown in Figure 1, the sodium analytical peak is not

observed because of the random choice of the timing parameters. Other random timing parameters and number of cycles simulated spectra are displayed in Figure 3(a) and (b). The detection limits obtained are 2523 ppm and 3696 ppm, respectively. Whereas the detection limit corresponding to the optimal parameters as determined by the optimisation routine is improved to 1017 ppm. The simulated spectra corresponding to the optimal parameters is illustrated in Figure 3(c). This test demonstrates both the validity of the optimisation routine and the utility of the spectra simulation.

Conclusions

Simulated and experimental spectra are shown to be in good agreement and we think that including the Bremsstrahlung contribution and the 511 keV annihilation peaks to the simulation induces an increase in the software execution time without improving significantly the simulated spectra. The spectra simulation and the optimisation routine together are used to investigate detection limits for a given element of interest as a function of the timing parameters and to determine the optimal ones.

This developed software may be extended to radiative capture using instead of 'LIBRAPID' a different library concerning thermal neutron reactions.

Table 3. Peaks' identification as marked on spectra

N°	Energy	Radio	N°	Energy	Radio	N°	Energy	Radio.
1	511	annih.	8	1460	K40	15	2167	K38
2	757	Esc.2	9	1733	Esc.2	16	2235	Esc.1
3	844	Mg27	10	1736	Esc.2	17	2247	Esc.1
4	1010	Mg27	11	1779	Al28	18	2425	Esc.
5	1014	Mg27	12	1810	Mn56	19	2754	Na24
6	1273	Al29 + Esc.1	13	2029	Esc.	20	2758	Na24
7	1369	Na24	14	2125	P34

Figure 2. Experimental and simulated Al(OH)₃ sample.

Table 4. Results of optimisation: minimum detection limits in SLI

Element symbol	Half life (s)	Energy (keV)	Irradiation time (s)	Decay time (s)	Counting time (s)	Number of cycles	Detection limit (ppm)
Al	568	844	895	98	1745	1	497
Fe	9290	1810	1192	505	2357	1	1611
Si	394	1273	617	180	739	1	300
Ba	153	662	559	60	557	1	398
Rb	1220	248	1785	245	2357	1	59
Na	11	1634	33	3	39	15	1017
Ca	1330	1157	1785	491	3514	1	33977
K	458	2168	1796	60	2159	1	2701
Mg	53900	1369	1790	467	7165	1	782
F	27	197	87	3	176	20	288
P	135	1779	299	4	524	7	717
Mn	225	1434	589	9	875	4	3298
N	598	511	1780	98	2300	1	369
Sr	10100	388	2385	494	7028	1	121
Ce	56	757	148	3	176	1	134
Se	17	162	49	3	44	20	214
Sc	18	142	44	3	43	1	479
Ti	18	142	65	3	40	20	4505
Hf	18	214	44	3	43	1	307
Ag	44	93	149	3	132	20	2084
As	18	142	140	3	147	20	2054

Figure 3. Illustration of spectra corresponding to random timing parameter (a) and (b) vs optimal (c) as determined by the optimization code.

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