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Parameterization of detector efficiency for the standardization of NAA with stable low flux reactors

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With SLOWPOKE and MNS reactors which have reproducible neutron fluxes, the standardization of multielement NAA can be reduced to measuring activation constants once for all elements and then determining relative detection efficiencies for new detectors and counting geometries. In this work, a method has been developed for the parameterization of the efficiency of germanium detectors. The gamma-ray detection efficiency was measured as a function of energy and distance for three detectors. The variation with distance was found to follow a modified EID law, within 1%, for point sources 1 mm to 250 mm from the detector. A model, including coincidence summing corrections, was developed to calculate efficiency for NAA samples; it requires 16 measured parameters. Tests showed that the calculated relative detection efficiencies are accurate to better than 3% for close counting geometries and sample volumes up to a few millilitres. Areas of possible improvement to the accuracy of the method are suggested.

Introduction

Neutron activation analysis permits the determination of many elements with an accuracy of 1% when the relative method is used, with standards irradiated in the same neutron flux and counted in the same geometry as the unknown. However, the considerable effort required to prepare and measure standards of each element with each batch of unknowns has led to the development of other less tedious standardization methods. The best known is the k_0 method[1,2] where the neutron spectrum of the irradiation site is characterized using a few monitor elements which permits the accurate calculation of the induced activities of all radionuclides. The γ-ray counting efficiencies are calculated using the sophisticated Monte Carlo routine SOLANG.[3]

The standardization problem is simplified when stable low flux reactors such as SLOWPOKE and MNSR are used. With these reactors the fuel, and thus the neutron spectrum, remains unchanged for periods approaching 20 years. The reproducibility of the neutron flux depends only on the neutron detector used for reactor control; it typically gives a flux reproducible to within 1%.[4,5] The improved relative method[6] has been developed for use with these reactors: standards of each element are activated once and the sensitivity factors determined are used for all subsequent analyses with the same counting geometry. In the present work this method is extended to other counting geometries by parameterizing the detection efficiency, taking into account γ-ray energy, sample–detector distance, sample size and γ-ray coincidence summing effects. Low flux reactors produce low activities and thus require close counting geometries; great care must be exercised to achieve accuracies of a few percent under these conditions. The aim is to use the sensitivity factors measured for one counting geometry to predict those for any other geometry on any detector. The proposed method is simpler than SOLANG in that it uses measured empirical relations of detection efficiency as a function of counting geometry and it aims at calculating only relative efficiencies.

Experimental

The approach adopted for parameterizing the detector efficiency is essentially that of TIAN et al.[7,8] in their standardization work employing the effective interaction depth (EID) concept. At a point on the axis far from the detector, the absolute efficiency as a function of γ-ray energy is measured. At several energies, the efficiency as a function of distance along the axis is then measured. Coincidence summing is calculated using information from the decay schemes and total efficiencies are determined using measured peak-to-total ratios.

Three intrinsic germanium detectors (p-type) with efficiencies of 14%, 26% and 29% at 1332 keV relative to a 76 mm × 76 mm NaI detector were studied in this work. For each, the efficiency as a function of energy was measured at a distance of 250 mm from the aluminum end cap using calibrated sources of ^{22}Na, ^{54}Mn, ^{60}Co, ^{137}Cs and ^{152}Eu and uncalibrated sources of ^{24}Na, ^{140}La and ^{182}Ta.

To measure the efficiencies as a function of distance along the axis of the three detectors, point sources of ^{52}V, ^{60m}Co, ^{66}Cu, ^{69m}Zn, ^{139}Ba, ^{165}Dy and ^{187}W were activated with the Ecole Polytechnique SLOWPOKE reactor. These radionuclides were chosen because their γ-rays of 1434, 59, 1039, 439, 166, 95 and 686 keV are free from coincidence summing effects. The sources were positioned using a

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mechanical screw device with attached vernier with which the distance from the end cap could be read with an accuracy of 0.1 mm. This accuracy was necessary because, close to the detector, a 1 mm change in distance causes a 6% change in efficiency. For each source a γ-ray spectrum was collected with sufficient counts in the photopeak at 18 distances between 1 mm and 250 mm.

The total efficiency is needed to correct coincidence effects. Thus, peak-to-total ratios were measured as a function of energy using the single γ-ray emitters ^{60m}Co, ^{109}Pd, ^{134m}Cs, ^{46m}Sc, ^{51}Cr, ^{69m}Zn, ^{52}V and ^{28}Al. Although it is known that peak-to-total ratios vary with distance from the detector,[7] they were measured only at 10 mm because true coincidence losses are important only at small distances.

The above measurements allowed the construction of a model which calculates efficiency as a function of position for point sources on the detector axis. For NAA samples which may have a volume of several ml, the source position was taken as the geometrical centre of the sample. In order to establish a simple relation for the decrease in efficiency due to the sample extending off the axis, point sources were counted at various distances from the axis at 10 mm and at 40 mm from the detector.

The model developed will calculate the efficiency for γ-rays emitted from samples up to a few ml in size at any distance from one of the three detectors. The important thing is the accurate calculation of relative efficiencies because the model will be used to calculate sensitivity factors for a given geometry from those measured on another detector. To test the model, 1.3 ml samples of 25 elements, contained in 12 mm diameter by 25 mm long polyethylene vials, were irradiated and counted 37.9 mm from the 14% detector, as illustrated in Fig. 1, and 7.4 mm from the 29% detector. For 32 intense γ-rays emitted by the 25 radionuclides, the ratio of the measured peak areas, corrected for dead-time and decay time, was compared to the ratio of the calculated efficiencies.

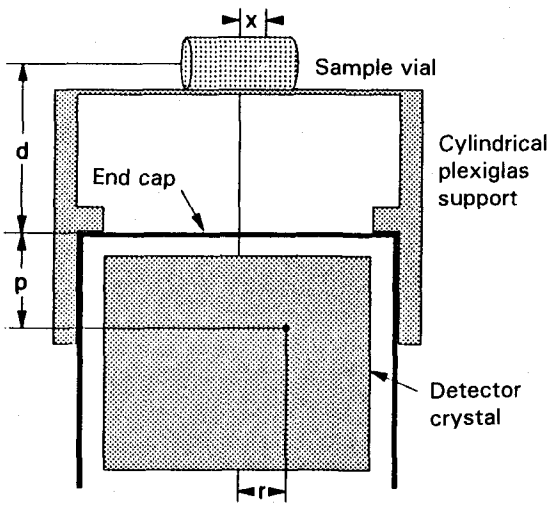

Fig. 1. Counting geometry 1 used to the test the model for a 1.3 ml sample 37.9 mm from the 14% detector

Results and discussion

The efficiencies measured at 250 mm from the 29% detector are shown in Fig. 2 along with the fitted curve. For the three detectors, an excellent fit of logarithm efficiency versus logarithm energy (in MeV) was obtained with a fourth order polynomial:

$$\ln \varepsilon_{250\ mm} = c_1 + c_2 \ln E + c_3(\ln E)^2 + c_4(\ln E)^3 + c_5(\ln E)^4 \quad (1)$$

The effective interaction depth (EID) law[8,9] states that the efficiency for a point source on the detector axis varies inversely as the square of the distance from a point inside the detector and on its axis. All the data accumulated for the three detectors on photopeak count rate versus distance

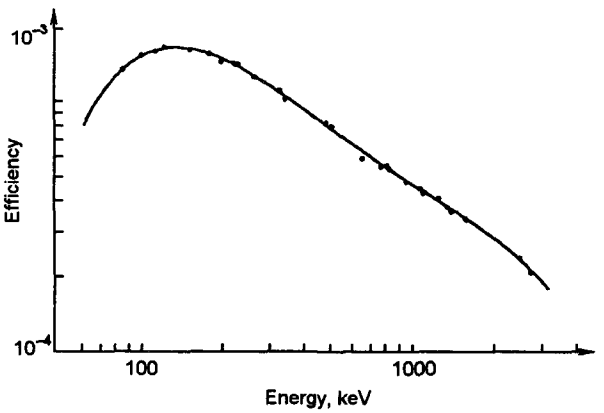

Fig. 2. Absolute efficiency of the 29% detector for a point source at 250 mm

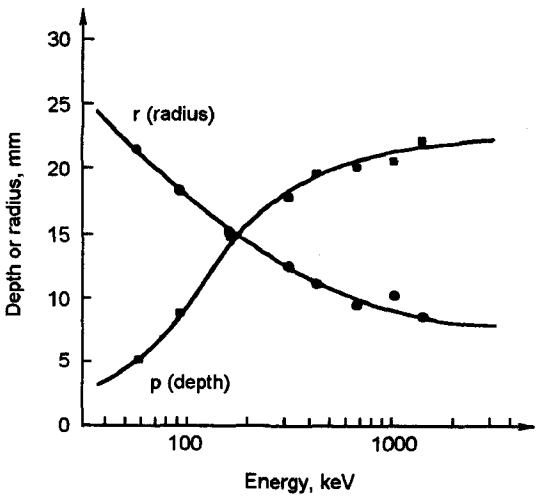

Fig. 3. The effective interaction depth and radius measured for the 14% detector as a function of γ-ray energy, along with the fittd curves

from the detector (1 mm to 250 mm) showed that the EID law does not hold, especially at low γ-ray energies. As was proposed by GUNNINK and NIDAY,[10] the interaction should be imagined to take place not only at an effective interaction depth (or penetration depth) p inside the detector but also at an effective interaction radius r, which leads to the following relation:

$$\varepsilon = \frac{\text{constant}}{(d+p)^2 + r^2} \tag{2}$$

where d is the distance from the point source to the end cap. For convenience, p is taken as the penetration depth from the end cap because the distance from the end cap to the crystal is not known. Equation (2) gave good fits to all the data measured with the three detectors and was used to determine p and r at the energies of γ-rays of the 7 nuclides measured. The variation of p and r with energy is shown in Fig. 3 for the 14% detector. The measured variations of p and r with energy for the three detectors were fit with the following relations:

$$p = c_6 + c_7 \frac{|\ln E - c_8|}{\ln E - c_8} [1 - \exp(-c_9 |\ln E - c_8|)] \tag{3}$$

$$r = c_{10} + c_{11} \ln E + c_{12}(\ln E)^2 + c_{13}(\ln E)^3 \tag{4}$$

The curves of p versus $\ln E$ were found to be symmetrical about an inflection point (parameter c_8). The inflection point is at 110 keV for the 14% detector and at 140 keV for the 26% and 29% detectors.

The measured peak-to-total ratios, illustrated in Fig. 4 for the 29% detector, were fitted with the polynomial of

$$\ln P/T = c_{14} + c_{15} \ln E + c_{16}(\ln E)^6 \tag{5}$$

The efficiency calculations were programmed with a spreadsheet, Lotus 123. A detector is characterized by the parameters c_1 to c_{16}. The parameters determined for the 14% and 29% detectors are listed in Table 1. The efficiency at distance d is calculated by the relation:

$$\varepsilon_d = \varepsilon_{250\text{ mm}} \frac{(250+p)^2 + r^2}{(d+p)^2 + r^2} \tag{6}$$

where $\varepsilon_{250\text{ mm}}$, p and r are calculated by Eqs (1), (3) and (4). The efficiency ε_s for a sample which extends a median radius x off the detector axis is calculated by the empirical relation:

$$\varepsilon_s = \varepsilon_d \left[1 - 1.6 \frac{x^2}{(d+p+r)^2}\right] \tag{7}$$

Table 1. The parameters used to calculate peak and total efficiency as a function of source position and gamma-ray energy for two detectors

		14% detector	29% detector
$\ln\varepsilon_{250\text{ mm}}$	c_1	−8.436	−7.657
	c_2	−0.964	−0.696
	c_3	0.061	0.036
	c_4	−0.070	−0.073
	c_5	−0.053	−0.053
p	c_6	11.000	16.900
	c_7	10.800	15.900
	c_8	−2.207	−1.966
	c_9	1.000	0.800
r	c_{10}	9.500	4.600
	c_{11}	−1.900	−4.500
	c_{12}	0.780	1.600
	c_{13}	0.010	0.250
$\ln P/T$	c_{14}	−1.930	−1.720
	c_{15}	−0.800	−0.680
	c_{16}	−0.0026	−0.0025

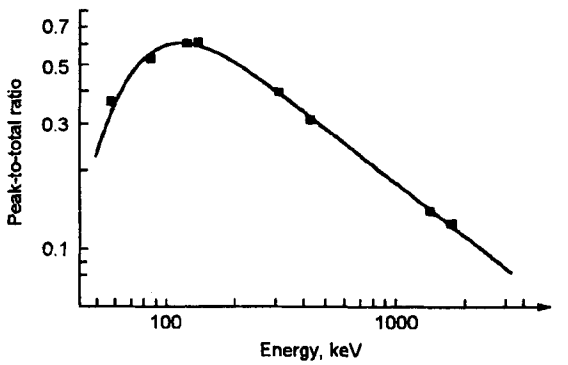

Fig. 4. The peak-to-total ratio measured for the 29% detector as a function of γ-ray energy, along with the fitted curve

which was found to fit the measured data, where x is the radius which encloses one-half of the activity of the sample. For a disk of diameter 40 mm, for example, x is 14 mm and ε_s is $0.8\varepsilon_d$ when the sample is 10 mm from the end cap. ε_s is corrected by a simple exponential calculation for γ-ray attenuation by the support and the sample. The attenuation in the sample is calculated using one-half the average thickness of the sample.

Finally, ε_s is corrected for coincidence summing using a method similar to that of TIAN et al.[7,8]

$$\varepsilon = \varepsilon_s(1 - CP_1\,\varepsilon_{T1})(1 - CP\,\varepsilon_{T2})\ldots \tag{8}$$

Table 2. Comparison of measured and calculated efficiency ratios for 1.3 ml samples counted 37.9 mm from a 14% detector (geometry 1) and 7.4 mm from a 29% detector (geometry 2). The fifth column contains the coincidence summing losses calculated for geometry 2

Nuclide	Energy, keV	Calculated efficiency				Measured ratio	Calculated measured
		Geometry 1	Geometry 2	Coincidence, %	Ratio		
^{165}Dy	95	0.03556	0.1166	–	3.28	3.36	0.98
^{75}Se	136	0.03150	0.1022	19	3.24	3.33	0.97
^{99m}Tc	141	0.03249	0.1248	–	3.84	3.94	0.97
^{139}Ba	166	0.02905	0.1162	–	4.00	4.12	0.97
^{75}Se	265	0.01781	0.0661	20	3.71	3.78	0.98
^{203}Hg	279	0.01778	0.0793	–	4.46	4.63	0.96
^{51}Ti	320	0.01530	0.0703	–	4.59	4.72	0.97
^{51}Cr	320	0.01530	0.0703	–	4.59	4.76	0.97
^{115m}In	336	0.01449	0.0672	–	4.64	4.91	0.95
^{69m}Zn	439	0.01074	0.0527	–	4.90	5.02	0.98
^{128}I	443	0.01063	0.0522	–	4.91	4.92	1.00
^{82}Br	554	0.00759	0.0279	35	3.68	3.98	0.93
^{122}Sb	564	0.00815	0.0421	–	5.16	5.43	0.95
^{124}Sb	602	0.00749	0.0371	7	4.95	4.98	0.99
^{187}W	686	0.00662	0.0356	–	5.37	5.52	0.97
^{82}Br	776	0.00536	0.0215	33	4.02	4.21	0.95
^{58}Co	811	0.00552	0.0295	5	5.34	5.23	1.02
^{27}Mg	843	0.00537	0.0301	–	5.60	5.62	1.00
^{56}Mn	847	0.00528	0.0283	6	5.35	5.45	0.98
^{46}Sc	889	0.00494	0.0246	15	4.99	5.17	0.96
^{27}Mg	1014	0.00448	0.0260	–	5.81	5.89	0.99
^{86}Rb	1077	0.00423	0.0249	–	5.88	5.91	0.99
^{59}Fe	1099	0.00414	0.0242	1	5.86	5.90	0.99
^{46}Sc	1120	0.00394	0.0205	15	5.21	5.28	0.99
^{59}Fe	1292	0.00356	0.0216	–	6.09	6.14	0.99
^{24}Na	1368	0.00328	0.0181	12	5.52	5.51	1.00
^{152}Eu	1408	0.00313	0.0175	14	5.58	5.62	0.99
^{52}V	1434	0.00322	0.0200	–	6.21	6.14	1.01
^{124}Sb	1691	0.00265	0.0148	16	5.59	5.61	1.00
^{28}Al	1779	0.00261	0.0168	–	6.46	6.15	1.05
^{56}Mn	1811	0.00248	0.0141	15	5.69	5.62	1.01
^{49}Ca	3084	0.00138	0.0098	–	7.11	6.83	1.04

where CP_i is the probability that γ-ray i is also emitted simultaneously when the γ-ray of interest is emitted, and its total efficiency, ε_{T_i}, is ε_s divided by P/T (both calculated at the energy of γ-ray i). The CP_i were taken from the table of Ref. 7 and from the decay schemes of the Table of Isotopes.[11] At 10 mm distance from the detector, coincidence losses may be as high as 22% for one γ-ray in coincidence.

For point sources 10 mm from the detector emitting β-rays with energies as high as 2.9 MeV (^{28}Al) in coincidence with the γ-ray, the coincidence losses were found to be about 4%. However, in normal NAA work where the betas must go through a sample support and sample vial of total thickness 2 mm or more, the coincidence losses due to betas are less than 1%; they are neglected in this work. Very few nuclides used in NAA emit betas with energies higher than 2.9 MeV. An exception is ^{20}F (end-point energy 5.4 MeV) for which β–γ coincidence losses may be important.

Efficiencies were calculated as described above for γ-rays emitted from 1.3 ml samples 37.9 mm from the 14% detector (geometry 1) and 7.4 mm from the 29% detector (geometry 2). The ratios of the calculated efficiencies are compared in Table 2 with the ratios of the peak areas of 32 γ-rays measured for these two geometries.

The measured ratios are believed to be accurate to about 1% and the average difference between calculation and measurement is 2.4%. The agreement is poorest for the 554 and 776 keV γ-rays emitted by ^{82}Br which is the nuclide with the most complicated decay scheme and the greatest coincidence losses. This indicates that the proposed coincidence correction procedure is inaccurate in the most severe cases. For ^{28}Al the calculated ratio is 5% higher than the measured ratio; the reason may be that the calculation does not take into account coincidence losses caused by betas and bremsstrahlung.

The method developed in this work permits the prediction of relative detection efficiencies for most

nuclides with an accuracy of 2% even for the close geometries needed in neutron activation analysis with low flux reactors. It should be possible to attain similar accuracy for all nuclides used in NAA with improvements to the coincidence summing calculations and consideration of coincidences with betas and bremsstrahlung. Standardization of NAA with reactors like SLOWPOKE and MNSR with reproducible neutron fluxes then becomes a question of measuring accurate sensitivity factors (counts/μg) once for each nuclide. These values are divided by the calculated detector efficiencies to give activation constants. Since all SLOWPOKE's, and possibly all MNSR's, have the same neutron spectrum, the set of activation constants should be universal. Standardizing for any geometry is achieved by multiplying the activation constants by the calculated detector efficiencies. The task of standardization is then reduced to calibrating each new detector by determining the constants c_1 to c_{16}.

Our work on the development of this method will continue with improvements to the coincidence calculations and testing the model for the calculation of detection efficiencies for samples larger than a few millilitres. The activation constants for 85 nuclides will be measured.

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