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Prompt gamma activation analysis using a chopped neutron beam

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A beam chopper has been developed at the cold neutron PGAA facility of the Budapest Research Reactor. In the open phase of the chopper the usual prompt gamma-spectrum is recorded, while in the decay phase short-lived decay lines can be collected with good counting statistics on an extremely low baseline. A series of elements has been measured with the chopped beam technique to assess the capabilities of the new technique. An archaeological sample was also examined, to demonstrate how spectral interferences can be resolved.

Introduction

Gamma-ray spectra obtained in prompt gamma activation analysis (PGAA) are much more complicated than those in instrumental neutron activation analysis (INAA). While in INAA each observable element produces only a few gamma-peaks with energies typically lower than 2 MeV, in PGAA most of the elements yield several hundreds of peaks up to 8–9 MeV. The summed Compton-plateaus of this large number of peaks may raise the baseline in the prompt gamma-spectra tremendously. Even when a Compton-suppressed detector system is used, the majority of spectrum counts belongs to the background continuum lowering the detection limit of many elements.

The advantages of the prompt gamma measurement can be combined with the simplicity of the decay gamma-spectra obtained in INAA using the technique of chopped beam PGAA. The chopper periodically opens and closes the beam, and the gamma-radiation emitted in the two phases is acquired in two separate spectra. When the beam is open, i.e., in the activation phase, the usual prompt gamma-spectrum is collected. When the beam is shut off, i.e., in the decay phase, only a few gamma-rays are acquired, those emitted by the radioactive nuclides formed during the activation phase.

Decay lines of short-lived reaction products appear in the prompt spectra as well, and some of them are routinely used in prompt gamma-analysis (e.g., the 1633 keV line of ^{20}F or the 1779 keV line of ^{28}Al).[1,2] These lines appear in the decay spectrum with similar intensities. However, due to much lower baseline the detection limits are substantially lowered.

The separation of prompt and decay lines by means of a beam chopper has been suggested in the pioneering work of ISENHOUR and MORRISON,[3] and recently by ZEISLER et al.,[4] with the aim to reduce spectral interferences in the prompt spectrum.

In this paper the first chopped beam measurements at the cold neutron PGAA facility of the Budapest Research Reactor are presented. Contrary to the aforementioned studies, the main goal has been to utilize the decay spectrum for the improvement of peak-to-background ratios, hence, detection limits.

Experimental

A chopper has been installed at the cold neutron PGAA facility at the Budapest Research Reactor. The chopper was particularly designed for the special purpose of the combined PGAA-INAA measurement. The rotation frequency can be adjusted in the region of 3–100 Hz. The circumference of the disk is 255 mm. The openings are machined accurately, so the alternating open and closed quadrants are of equal size within an uncertainty of 0.1%. The absorbing material on the surface of the chopper plates is a ^{6}Li-containing plastic, the attenuation for the cold beam was found better than 4000. ^{6}Li is an ideal shielding material, since it emits no gamma-radiation after capturing the neutron.

The background level at the PGAA system in Budapest is exceptionally low. The count rates measured at different conditions are listed in Table 1. Due to the thoroughly constructed shielding made from ^{6}Li-plastic and lead, the chopper does not increase the background level. The background spectra in the chopper measurements mainly consist of low-energy continuum from scattered gamma-radiation. Short-lived decay background lines are present in both spectra (e.g., 140 keV and 198 keV Ge, and 1779 keV Al), but the prompt gamma-radiation from nitrogen in air is absent from the decay spectrum. The annihilation peak at 511 keV is significantly higher than in the case of normal background spectra, with the chopper removed.

An electronic unit has also been installed to control the chopper and the spectrometer. It provides a gate signal that can be fed into the ADC, thus only one of the spectra, either the prompt, or the decay spectrum can be collected. A newly purchased Polaris type digital spectrometer (made by XIA) will simultaneously collect both spectra, by simply offsetting the addressed channel. The new device is under test.[5]

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Table 1. Background levels as measured at different condition in the PGAA facility at Budapest Research Reactor

Name	Reactor	Beam	Vacuum	Chopper	Phase	Backgound, cps
Room background	Off	–	–	–	–	0.63
Beam-off background	On	Off	–	–	–	1.5
Beam-on background (in vacuum)	On	On	Yes	–	–	4.0
Beam background in air	On	On	No	–	–	5.6
Chopper background in prompt phase	On	On	No	On	Prompt	5.3
Chopper background in decay phase	On	On	No	On	Decay	4.6

The chopper is located 375 mm before the sample position, thus the beam spreads out in time according to its velocity spectrum. To obtain pure decay and prompt spectra, the timing of the phases is of great importance. In the present experiments the proper timing was realized by appropriately positioning the beam collimators and by using electronic delays. The present control unit allows only a few predefined delay time values. The collimator is extended from the geometrical center of the beam to 5 mm away in the direction of the rotation. In the present experiments the prompt phase started 0.48 ms after the chopper plate opened the beam at the center (observed by a pair of diodes in the opposite side of the chopper), and ended 7.25 ms after the beam was closed. The decay phase started exactly when the prompt phase ended, but ended when the chopper opened the beam again. Thus no neutrons reached the sample during the decay phase, significantly lowering the baseline.

The chopper was rotated at 20 Hz. The prompt phases were about 18 ms long, while the decay phases lasted for 6 ms. The chopper passed in front of the 5-mm wide collimator in 1 ms. The actual lengths of the phases were recorded at every measurement, and were used to correct the counting times. Due to the delays and the gradual opening of the beam, it was difficult to relate the observed prompt gamma photons to the number of activating neutrons. The count rate ratio of the 1633 keV singlet peak from ^{20}F, as measured in the decay and in the prompt spectrum, was used to quantify the relative weights of the two phases. This ratio was found to be 0.29±0.01. This value is rather low, but in the ideal case it may reach unity. However, allowing a short counting time in the decay phase provided a decay spectrum free of prompt gamma-peaks, offering better detection limits.

A series of elements with short-lived activation products was examined with this technique. Those elements were selected which have such a short-lived decay line among the major lines in the prompt spectrum that is used for PGAA routinely. The main goal of this investigation was to show that these elements could be analyzed by using the decay spectra with better accuracy and sensitivity. It was verified that a correction could be made for any decay peak disturbed by interfering prompt gamma-lines. It was also checked if in the decay spectra of elemental targets the baseline level was low enough to increase the accuracy of the fit.

Finally, the method was tried in the case of a real sample, in the spectrum of which some of the selected elements appear. The spectra were evaluated using Hypermet-PC.[6,7]

Results and discussion

The following elements have been selected in this series of experiments: F, Al, Sc, V, Cu, Ag, In, Er. All of these have short-lived activation products, whose decay lines appear in the prompt and in the decay spectra as well. The samples were measured in the chopped beam for similar live times, so the accuracy of the decay peaks could be compared directly.

Table 2 shows the results of the measurements with the selected elements. The most intense lines of the prompt and the decay spectra are listed here. If the decay line is the strongest in the prompt spectrum as well, only this line appears in the table. The signal-to-noise (S/N) ratios and the count rates are presented in the right columns. For decay lines the statistical uncertainty also includes the 4% relative uncertainty of the phase lengths calculated from the fluorine rates. The S/N ratios were calculated as the peak area over the background level at the high-energy side of the peak. The count rates were corrected for saturation when needed, as discussed in Reference 1. Masses of the samples and the total counting times for the prompt measurements are also listed in Table 2.

In the case of the selected elements the signal-to-noise ratios were found considerably higher for the short-lived decay peaks measured in the decay phase, than for the highest peaks in the prompt gamma-spectra. The estimated uncertainties for the peak areas in the decay spectra were always better than or equal to the uncertainties of the same peaks in the prompt spectra.

One of the most important practical cases, is the measurement of silver concentrations. Silver has a complicated prompt gamma-spectrum. It contains an extremely large number of peaks and a high continuum. Even the intense peaks typically appear in multiplets, thus the estimated uncertainties of the fits are usually much worse than expected from the peak counts.

Table 2. Signal-to-noise ratios for the most intense peaks as measured in the prompt and decay spectra in different elements

Element	*E*, keV	Sample	Live counting time, s	Sample mass, mg	Prompt spectrum		Decay spectrum		Gain
					Rate	S/N	Rate	S/N	
F	1633	Teflon	6499	690	1.63(2)	1300	1.63(6)	7000	5
Al	1779	Al plate	3400	52	2.62(1)	2000	2.61(1)	200,000	100
Sc	143	$Sc_2(SO_4)_3$	436	18	28(1)	200	29(1)	40,000	20
	147				46.8(7)	400			
V	125	V_2O_5	1438	37	20.0(2)	270			
	1434				9.3(1)	1200	8.8(4)	41,000	34
Cu	159	Cu plate	4578	100	29.6(2)	200			
	1039				0.58(2)	90	0.55(2)	900	10
Ag	198	Ag plate	1293	43	82(1)	62			
	658				7.16(15)	20	7.3(3)	1900	95
In	163	In plate	209	39	157(3)	53	146(6)	3700	70
Er	185	Er_2O_3	183	3.8	32(1)	400			
	208				14.6(8)	180	15.4(6)	11,000	60

S/N = peak area/background level.
The absolute 1σ uncertainty of the last significant digit is given in parentheses.

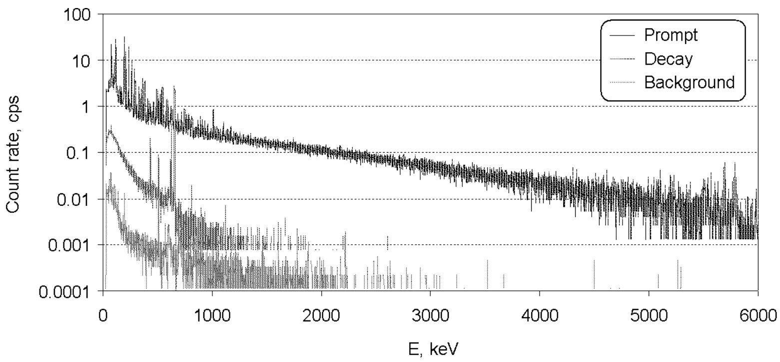

Fig. 1. Prompt and decay spectra of silver and the background measured in a chopped beam

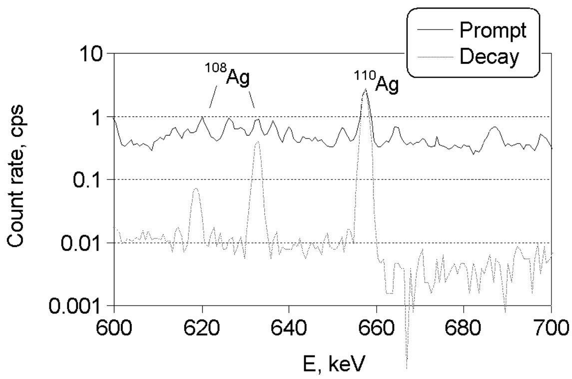

Fig. 2. Prompt and decay spectra of silver measured in a chopped beam in the energy region of 600 to 700 keV

Furthermore, the highest prompt gamma-peak at 198 keV overlaps with a background peak, and several other interferences with prompt gamma-lines from other elements make the identification of silver problematic in a multi-element sample. However, in the decay spectrum only five intense peaks can be found, as the long-lived ^{108m}Ag and ^{110m}Ag isotopes are produced at a much lesser extent. The strongest peak appears at 658 keV, from the decay of ^{110}Ag with a half-life of 24.6 seconds.

As it can be seen in Table 2, the signal-to-noise ratio of the 658 keV peak in the decay spectrum exceeds considerably that of the highest peak in the prompt spectrum. On the other hand, the achieved accuracy was found even better for the 658 keV peak in the decay spectrum than for the 198 keV line in the prompt spectrum, though this latter one's intensity is an order of magnitude higher.

The prompt and decay spectra are shown in Fig. 1. It can be seen that the prompt spectrum was rather complicated from the point of view of analysis, as it contained too few characteristic peaks, while the number of total counts is high. The total count rate in the prompt spectrum was 2200 cps, compared to only 71 cps in the decay spectrum. The baseline level was almost 2 orders of magnitude lower in the latter case, which resulted to a large increase of the signal-to-noise ratio. The background spectrum measured in the decay phase with the total count rate of 4.6 cps is also shown in Fig. 1 for comparison.

The improvement can be better examined in Fig. 2, where the energy region of the decay peaks is magnified. The intensity of the 658 keV peak is the same in both spectra, but it sits on a much higher background and overlaps with prompt gamma-peaks in the in-beam spectrum.

The precision of these demonstrating measurements was not sufficient to improve the previous partial gamma-ray production cross section data obtained in a continuous beam.[1,2,8] From the ratios of count rates of the decay peaks in both spectra upper limits for the intensities of possibly overlapping prompt gamma-peaks could be derived, however. The 1633 keV peak of ^{20}F was assumed to be interference-free according to ENSDF.[9] As it can be seen from Table 2 most decay peaks produce the same count rate in both spectra within the error limits. Indium and, perhaps, vanadium seem to show significantly lower rates in the decay spectrum. These cases can be regarded as evidence of existing overlapping prompt lines at the energies of the decay lines (overlapping decay lines could be excluded based on the level scheme).[10] The newest compilation of prompt gamma lines[8] and also the earlier versions of ENSDF[9] report prompt gamma-lines at the energies of the short-lived decay lines of our interest. The prompt σ_γ values were calculated from the differences of the count rates in Table 3 and from the published σ_γ values.[1,2,8] Since the obtained uncertainties are too high (more than 50%), the prompt σ_γ values have to be regarded as upper limits. In the other cases the interference with prompt peaks could neither be proved, nor excluded. Upper limits were derived from the uncertainties of the count rates and the σ_γ values.[1,2,8]

The method was finally tested in routine analysis. An archaeological sample, a piece of a wase from a Roman bronze workshop was measured in routine prompt gamma-analysis, and also with the chopped beam technique. The results obtained from the decay spectra are compared to the results from prompt spectra in Table 4. The main constituents of this bronze artifact were Cu, Zn, Sn, and the minor components were Na, Si, Fe and possibly Ag of approximately 400±100 ppm. Since all the silver peaks found appeared in overlaps, the presence of this element in the sample was questionable. Using the chopped beam technique it could be clearly proved, that the bronze piece actually contained silver. The mass ratio of silver to copper was measured, and the silver content proved to be somewhat lower than the value obtained from the prompt spectrum, where the overestimate could be ascribed to peak overlaps. The 472 keV peak from 20.2 ms half-life ^{24m}Na was also observed in the spectra, but the present timing was not suitable for the quantitative analysis of this nuclide.

Table 3. Partial gamma-ray production cross sections for short-lived nuclides important in PGAA and for possible overlapping prompt gamma-lines

Element	Half-life	Energy, keV	Decay line σ_γ, barn	Prompt line σ_γ, barn
F	11.16 s	1633	0.0093(3)	–
Al	2.24 m	1779	0.233(4)	<0.005
Sc	18.75 s	143	4.88(10)	<0.13
V	3.75 m	1434	5.20(10)	<0.3
Cu	5.12 m	1039	0.0600(12)	<0.0023
Ag	24.6 s	658	1.93(4)	<0.08
In	2.18 s	163	15.8(8)	<1.1
Er	2.27 s	208	2.15(9)	<0.18

The absolute 1σ uncertainty of the last significant digit is given in parentheses.

Table 4. The composition of a bronze artifact as determined by the routine analysis in Budapest. The silver content was correctly determined with the chopped beam PGAA technique according to the mass ratio of silver to copper

Element	PGAA concentration, %	Chopped beam PGAA concentration, %
Na	0.56(6)	
Si	1.5(2)	
Fe	0.33(5)	
Cu	79.4(2)	79.4(2)*
Zn	15.2(1)	
Ag	0.040(10)	0.025(3)
Sn	3.0(2)	

* Normalized to the value from the prompt spectrum.
The absolute 1σ uncertainty of the last significant digit is given in parentheses.

Conclusions

Chopped-beam PGAA combines the advantages of the in-beam measurement with the simplicity of the INAA spectra. The spectrum measured in the prompt phase provides the same analytical information as the usual PGAA spectrum, while the decay spectrum has a much lower baseline and the number of peaks is also much lower. Prompt spectra can be used for complete analysis of the sample, while the decay spectra are essentially interference-free, since only a part of the elements appears in it. Using an appropriate beam chopper the measurements can be performed at low radiation background, thus the detection limits for a number of elements can also be lowered significantly.

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