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The new prompt gamma-activation analysis facility at Budapest

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Prompt gamma-activation analysis (PGAA) is an important complementary technique to conventional instrumental activation analysis that can be successfully used in a number of cases when INAA is not applicable. Therefore, a PGAA facility has been constructed at the recently refurbished and upgraded Budapest Research Reactor. It occupies the end position of a new curved thermal guide of 30 m length and 2.5×10 cm^2 cross section which provides a clean beam of low energy neutrons. The sophisticated HPGe–BGO γ-ray spectrometer system can be operated in Compton-suppression and pair-spectrometer modes simultaneously. The octal splitting of the main BGO improves efficient pair mode operation when coincidences between pairs of opposite segments and the HPGe detector are required separately. Gamma–gamma coincidence measurements will also be possible when the new multiparameter data acquisition system is completed. One of the main tasks at the new facility will be the accumulation of new spectroscopic data for detector calibration and standardisation, as well as for the construction of a more accurate prompt γ-ray library for the chemical elements. Various applications are planned, such as the determination of hydrogen in fullerenes and of toxic trace elements in environmental samples.

Introduction

Prompt γ-activation analysis or PGAA, based on the observation of the γ-radiation emitted promptly whenever a nucleus captures a bombarding neutron, is a less known version of neutron activation analysis.[1] (This is the way the neutron binding energy, typically about 8 MeV, is released.) PGAA may be an important complementary method to NAA whenever the end product of the radiative neutron capture reaction does not provide a delayed γ-radiation with convenient half-life or energy, remains stable (non-radioactive), or the isotope with highest neutron capture probability transforms into a stable nuclide. The most important examples are the elements H, B, C, N, P, S, Cd, Pb and some rare earths, especially Sm and Gd.

PGAA is a much more difficult technique, however, the delayed NAA in that the measurement is carried out in situ during the neutron irradiation, not in a separate low-background environment. The real potential of PGAA for multielement (panoramic) and trace element analysis can fully be utilised when guided beams of cold neutrons are used.[2,3] They provide high flux with essentially no fast-neutron and γ-ray background radiation, leading to a substantial increase of analytical sensitivity. The first permanent PGAA facilities on a cold neutron guide have been installed in Germany (1986: KFA Jülich[4]), in the USA (1990: NIST, Gaithersburg[5]) and in Japan (1993: JAERI, Tokai[6]). Progress reports on cold neutron PGAA were given, for instance, at the last MTAA meeting[2,5] and in a recent monograph.[3] Thermal neutron guides, i.e., guides without cold neutron source, are also very useful, although in that case the capture flux is lower due to the shift of the neutron spectrum to lower wavelengths.[7]

The complexity of prompt γ-ray spectra is another important problem which can be cured only by using sophisticated spectrometers and spectrum evaluation procedures. Compton suppression reduces continuous γ-ray background whereas annihilation pair spectrometers eliminate everything but double escape peaks in high energy spectra. Since analytical sensitivities are much more uniform (albeit sometimes lower) with regard to high energy peaks,[8] the pair mode gains importance when a panoramic analysis has to be performed. A modern HPGe–BGO detector system, encompassing both spectrometer modes beside the normal ungated mode has been installed[9] alternatively on a cold and a thermal neutron guide at JAERI, Tokai, leading to substantial improvements in spectral purity. On the other hand, there has been less progress concerning the availability of easy-to-use, yet sufficiently versatile, computer programmes for automatic analysis of complex γ-ray spectra. Also, for many elements the nuclear data libraries available on neutron-capture γ-rays[10,11] do not reach the level of precision, completeness and versatility required for PGAA work, as pointed out recently.[12]

Standardisation also remains an open problem in PGAA, as discussed in recent reviews.[1,3] It has been demonstrated[3,13] that elemental ratios are practically free from analytical biases, hence internal standards can be used safely. According to recent suggestions[3,14,15] the k_0 standardisation method,[16] proving so fruitful in delayed NAA, may be applicable to PGAA as well. This would eliminate the problems associated with uncertain and

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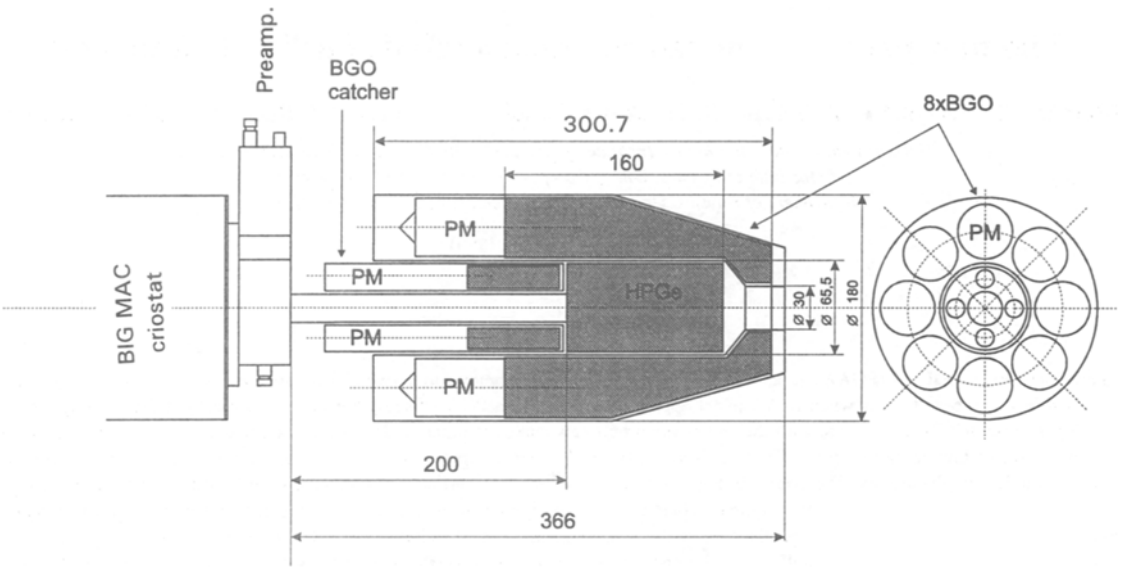

Fig. 1. Schematic view of the HPGe–BGO detector assembly

inconsistent nuclear data for sample and standard, but still mandates an accurate calibration of the detector efficiency for all γ-ray energies involved and the selection of a suitable comparator element.

A new PGAA facility has been installed on one of the thermal neutron guides at the recently upgraded Budapest Research Reactor. In this very first report on the new facility the basic features are outlined. Design principles and actual performance of the Compton-suppression/pair mode γ-ray spectrometer are described in more detail. Finally, current problems with efficiency calibration procedures and standards are mentioned briefly.

The PGAA system

The Budapest Research Reactor has been refurbished and upgraded to 10 MW power in the framework of an eight-year reconstruction and recommissioning project, which ended in 1994. As a result of a four-year effort,[17–19] the new PGAA station has just been completed. One of the curved neutron guides was extended by a 17-m long section made of borosilicate glass with natural Ni coating and a new experimental station was established at the guide end, about 30 m distance from the reactor. The guide has a cross section of 2.5×10 cm^2 which is presently collimated down to a spot of 2×4 cm^2 size at the target position, about 1.7 cm distance from the guide end. An evacuated tube incorporating the final sample holder and an adjustable collimator is still under construction. A sophisticated HPGe–BGO γ-ray spectrometer serves as the basic instrument for PGAA. It is positioned on a turntable moving on a circular rail. A second turntable has also been prepared to enable mounting of an additional detector for low energy γ-ray and γ–γ-coincidence measurements in the future. The system was put in operation in mid-June 1995. It will be complemented by a new multiparameter data acquisition system, which is still under construction, by the end of 1995.

Compton-suppression/annihilation-pair mode gamma spectrometer

This basic γ-spectroscopic instrument consists of an *n*-type high-purity germanium (HPGe) main detector with closed end coaxial geometry and a bismuth germanate (BGO) scintillator guard detector annulus, as well as of appropriate NIM electronics for coincidence gating and signal processing. The HPGe detector has an energy resolution of 1.8 keV, a relative efficiency of 25% and a peak-to-Compton ratio of 50. The BGO guard detector has been modelled after the one used at JAERI, Tokai.[9] It consists of an octally split nose corn-shaped annulus and a back catcher crystal. The annulus is protected by a lead shielding of 10 cm thickness. The sample-to-detector distance is about 25 cm, which can be reduced by removing some of the three front plates of the lead shielding. A schematic drawing shows the HPGe–BGO detector assembly in Fig. 1.

We have found that the octal BGO geometry can be better exploited in pair spectrometer mode if signals from each optically isolated octant of the BGO guard detector are processed separately. Chance coincidences are reduced due to the increase of spatial resolution for the coincident

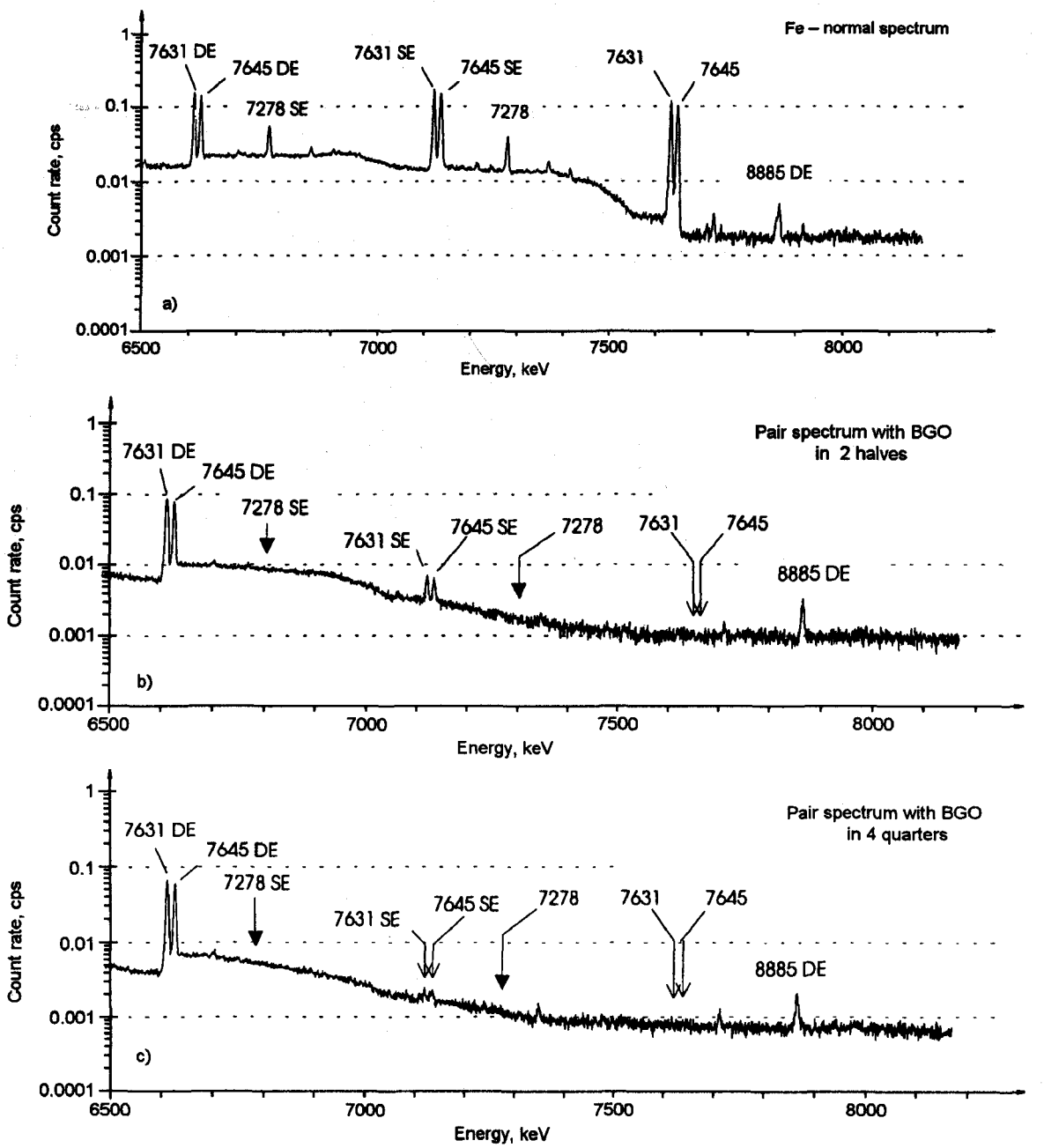

Fig. 2. Comparison of ungated (a) and two differently gated pair-mode spectra (b, c) in the vicinity of the 7631–7645 keV doublet in ^{57}Fe, as measured with a natural Fe target

detection of 511 keV annihilation pair quanta instead of the usual energy gating which requires a slow coincidence circuitry, hence, increases the system dead time. The enormous gain in suppression of unwanted peaks when using opposite pairs of smaller BGO segments is demonstrated in Figs 1 and 2 where partial results from an (n, γ) measurement on a natural Fe sample are presented. It is remarkable that the double-escape/single-escape peak ratio in the pair spectrum increases from about 20 to 100 when using four quadrants instead of two halves, which yields an exceptionally clean pair spectrum with only double escape peak. Note that about 10^5 counts had to be accumulated for the DE peak in order to see the SE peak with reasonable statistics. The price is only a 50% less in double-escape peak efficiency. Even better suppression is expected when eight octants are used.

Obviously, the use of a HPGe + BGO γ-ray spectrometer in Compton-suppressed and pair modes simplifies greatly the spectra for low and high energies, respectively, leading to a net increase of analytical sensitivity and selectivity, thanks to the suppression of unwanted peaks and continuous background. At present, a single-input PC-based multichannel analyser (MCA) allows the collection of only one type of spectrum at a time. This simple MCA is being used for the first calibration and test measurements. A permanent, PC-based, data acquisition system has already been designed and is being built to enable multi-input MCA operation for simultaneous collection of normal, Compton-suppressed and pair spectra with 16k conversion gain, as well as of single and coincidence spectra with additional detectors. It will be connected to the ethernet LAN to facilitate remote on-line control and data analysis on PCs and UNIX workstations. A second, low-energy, HPGe detector is still needed to enable the measurement of X-rays and very low energy γ-rays for increasing the analytical sensitivity for some important elements, such as Pb.

Efficiency calibration

Calibration of the spectrometer also involves the determination of photopeak and escape peak efficiencies up to about 10 MeV γ-energy. Radioactive sources cannot be used at all above about 3.5 MeV, the highest energy from a ^{56}Co source, although the analysts' requirement of at least 2% accuracy sets this limit even lower, at about 2.6 MeV. At energies higher than this the prompt neutron-capture γ-rays themselves have to be used for energy and efficiency calibration. However, the uncertainties of literature data for capture γ-ray intensities are usually too high for this purpose. The only exception is the ^{14}N(n, γ) reaction for which recommended[20] intensity values are available with a precision of about 1% or slightly higher, up to almost 11 MeV. Unfortunately, nitrogen has a very low neutron capture cross section, and there are sample problems as well. Therefore, accurate efficiency standards have to be developed in the first place. The best candidates seem to be Ti, Cr and Cl which provide convenient sequences of γ-rays and have large enough neutron capture cross sections. As to chlorine, the recommended[20] γ-ray emission probabilities vary between 1–4% for the 17 strong lines between about 0.5 and 8.5 MeV energy if one ignores the additional systematic uncertainty of 8% on the absolute intensities, specified by the original authors.[21]

The full-energy peak efficiency has been measured for the new system by using ^{56}Co and ^{152}Eu sources, capture γ-rays from ^{36}Cl and ^{49}Ti, as well as recommended γ-ray emission probabilities from Reference 20 and the on-line ENSDF database.[22] A typical relative efficiency curve is shown in Fig. 3 where a sixth-order polynomial has been fitted to the data points on a log–log scale. Due to the complexity of the chlorine spectrum several of the originally recommended peaks proved unsuitable for calibration purposes. Additional information from a precise energy measurement[23] with crystal spectrometers has been used when deconvoluting multiplet peaks with our new Hypermet-PC computer programme.[24] Consequently, several of the recommended peaks had to be eliminated when constructing the efficiency curve. The experience with titanium has been better, although even in the latest ENSDF compilation[22] discrepancies with our data have been found. Hence, work is needed to improve the

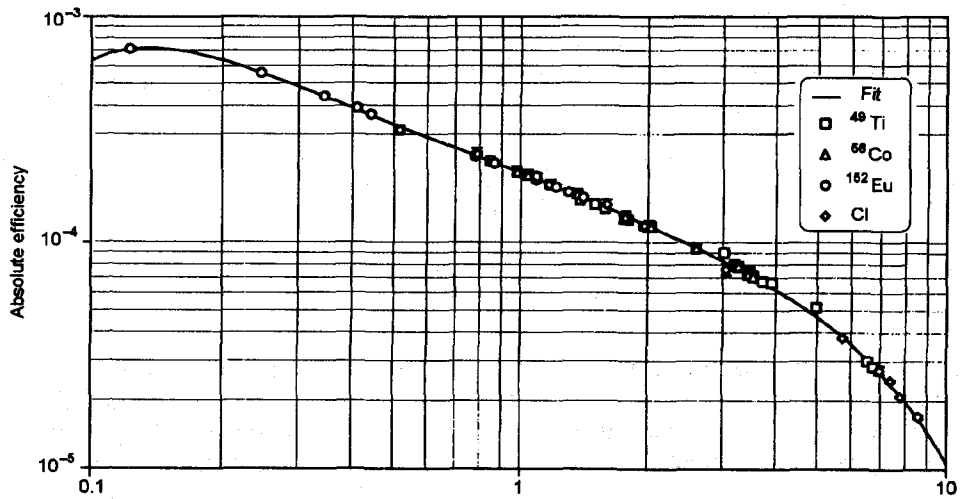

Fig. 3. Full energy-peak efficiency of the HPGe detector

accuracy of the present Cl and Ti data to make them suitable for more precise detector efficiency calibration. This will be done by measurements related to the nitrogen standard. If the required accuracy is achieved, both Cl and Ti might serve as an efficiency calibration standard as well as a comparator standard in routine PGAA. Chlorine is most attractive in that if forms compounds with many elements (an important feature for internal standards) and has intense peaks throughout a wide energy range. On the other hand, Ti has the advantage of a simple γ-ray spectrum and its 1382-keV γ-ray may serve as a convenient external flux monitor.[13]

Conclusions

Prompt γ-activation analysis with guided beams is an important new technique which may find widespread use and unique applications, especially if suitable routine calibration and standardisation procedures are developed. In the present paper we describe briefly the new PGAA facility, installed at the end position of a curved thermal neutron guide at the 10-MW Budapest Research Reactor. The unique design of the Compton-suppression/annihilation-pair mode γ-spectrometer yields good performance, especially in pair spectrometer mode where even single escape peaks are practically eliminated from the γ-ray spectra. Problems with detector efficiency calibration have been attacked. Construction of an efficiency curve using easily measurable standards such as Cl and Ti has been attempted.

Future work is planned in this direction. The high selectivity of the system will be explored in the development of standards for PGAA. For the near future applications of PGAA to high-tech materials, such as fullerenes, and to environmental samples are planned. For the latter, toxic element determinations with more-or-less uniform sensitivity might be possible due to the superb performance of the pair spectrometer.

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