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Analysis of toxic elements by MPGA

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In order to improve the sensitivity for the quantification of trace elements, we propose the combination of prompt gamma-ray analysis (PGA) and a multiple gamma-ray detection method. A new Ge detector system for multiple prompt gamma-ray analysis (MPGA) was constructed at the neutron guide-hall of the JRR-3M reactor of the Japan Atomic Energy Agency (JAEA). The first demonstration of this system was given with a plastic sample containing traces of cadmium. The quantification limit of cadmium in a plastic sample was found to be about 0.1 ppm.

Introduction

Prompt gamma-ray analysis (PGA) is a highly sensitive, high-precision, multi-element and non-destructive method.[1] The method is used in various fields, such as geology, archaeology, earth science, industry, and environmental science. However, the difficulty of quantification arises when the gamma-ray intensity from the trace element of interest is not strong enough in comparison with the background gamma-rays from large amounts of other elements in the sample. In order to improve the sensitivity of PGA, we propose to introduce a multiple gamma-ray detection method to PGA, and constructed a Ge detector system for multiple prompt gamma-ray analysis (MPGA).

Multiple gamma-ray detection is widely used in the field of nuclear physics, and is known as a coincidence method. In this method, two or more cascade gamma-rays emitted in de-excitation of nuclei are detected simultaneously by several gamma-ray detectors. Events composed of pairs of prompt gamma-rays are collected in the experiment. With the collected data, a histogram of gamma-gamma energy correlation, which is a two-dimensional matrix spectrum, is constructed.[2,5] This enables us to resolve the background and the gamma-gamma cascade events. We have successfully applied this method to instrumental neutron activation analysis (INAA) for trace analysis.[6,7]

In this paper we introduce the detector system for MPGA constructed at the C2 neutron beam port of the JRR-3M reactor of Japan Atomic Energy Agency (JAEA). A preliminary result of quantification of toxic element, Cd, is mentioned as an example of the usage of the newly developed MPGA system.

MPGA detector system

The new Ge detector system for MPGA consists of four Clover Ge detectors, which are placed at 90° with respect to the beam axis. Figure 1 shows a schematic drawing of the MPGA detector system. Each Clover detector consists of four individual crystals sharing a common cryostat. The relative efficiency of each crystal is 25% of a 3"×3" NaI detector. The distance of the Clovers from the beam center is about 5 cm. The Clover Ge detectors are surrounded by Compton shields of bismuth germanium oxide crystals (BGO) scintillator detectors. The total photopeak efficiency of a crystal is about 6% for the 1.3 MeV gamma-ray.

In order to deal with signals from the MPGA detector, a new data acquisition system (DAQ) based on an advanced digital processing technique has been developed. The DAQ consists of three different VME modules, which are for discrimination, for triggering, and for analogue-to-digital conversion. The details of the modules were given in Reference 8. The modules are controlled by computers (FreeBSD or Linux) through the VME bus. All event data are transferred to the computer to be analyzed on-line and to be stored, for example, on magnetic tapes. The data on a storage device can be reanalyzed later.

The MPGA system also contains an automatic liquid nitrogen filling system, an automatic sample changing system, a helium gas delivery system, a neutron collimator, and removable neutron attenuators. An automatic sample changing system can accommodate 160 samples, and changes the samples within 30 seconds. Since a helium gas delivery system replaces nitrogen gas in the neutron beam line with helium gas, we can reduce the background gamma-rays from the neutron capture reaction of nitrogen. Removable neutron attenuators enable us to control the neutron flux.

Data analysis

Because the four crystals in a Clover Ge detector are packed very closely, a Compton scattered gamma-ray from a crystal is often absorbed by one of the other three crystals. Consequently, summing up the all output signals from the four crystals, we can increase the photo efficiency of a Clover detector from about 100% to more than 150% for the 1.3 MeV gamma-ray. This method, which was called as “add-back” mode, improves the peak to total ratio, as well.

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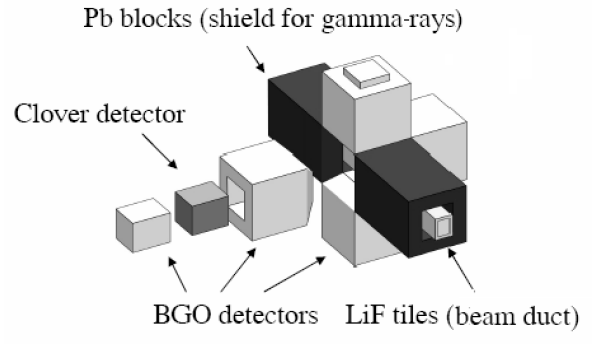

Fig. 1. Schematic drawing of MPGA detector system. Four Clover Ge detectors and BGO Compton suppressors are used and placed at 90° with respect to the beam axis. Neutron beam line is surrounded by enriched ^{6}LiF tiles and lead blocks. The neutron flux is about 10^{6-8} n·cm^{-2}·s^{-1}, and the beam size is about 2 cm by 3 cm

On the other hand, one cannot neglect such events that one Clover detector catches more than two cascade gamma-rays, because the distance of the detector to the target is close. It is impossible to distinguish whether the coincidence event is from cascade gamma-rays or from Compton scattering. The add-back mode, therefore, sometimes makes ghost peaks in a 2D-matrix spectrum as seen in Fig. 2B.

In order to remove such unwanted ghost peaks, we can analyze the same data with a method named "anti-coin mode", in which we discard the events that contain more than two data from a Clover detector. The anti-coin mode, namely, uses the other three Ge crystals as anti-Compton suppressors. The result of the anti-coin mode analysis is shown in Fig. 2C. Although this mode decreases the photo efficiency of a Clover detector, the peak to total ratio is improved significantly. If you have enough events, the data should be analyzed in the anti-coin mode.

Analysis of cadmium

Quantification of the toxic element cadmium in rice is one of the important subjects in our study. Located in a volcanic zone, Japanese soil tends to be highly cadmium contaminated. There is a possibility that cadmium contaminated agricultural products may be distributed unevenly. Since rice is the most important staple food in Japan as well as the major crop in Japanese agriculture, for food safety the technical development of a quantification of cadmium in rice is required.

In the quantification of cadmium in rice with PGA, hydrogen is the main source of background, because hydrogen is one of the main components of rice. It has a large neutron capture cross section, and emits single 2223 keV prompt gamma-rays in the neutron capture reaction. Compton scattering of the prompt gamma-rays obscures the gamma-rays from cadmium. Because cadmium emits cascade prompt gamma-rays, we can apply the MPGA system for the quantification of cadmium in rice.

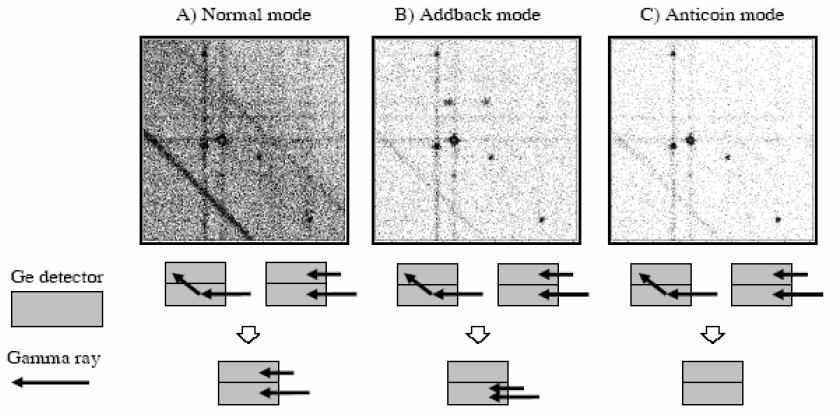

Fig. 2. This is a part of a two dimensional spectrum: (A) normal mode spectrum which is obtained in the NIES rice sample measurement; (B) addback mode sorting spectrum. The peak to total ratio is improved. However, two ghost peaks are observed in the upper part of the spectrum; (C) anticoin mode sorting spectrum. The peak to total ratio is improved significantly

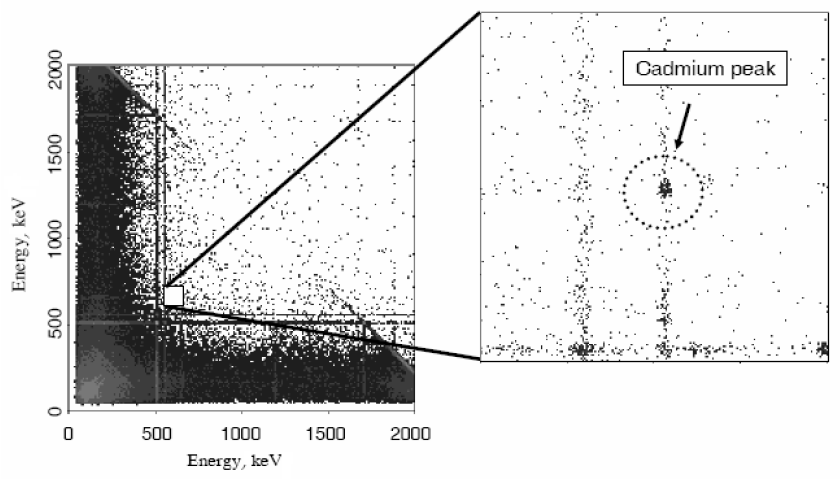

Fig. 3. Two dimensional spectrum of IRMM B6 (54 ppm cadmium) standard plastic sample.
Measuring time was 600 seconds. Cadmium emits a strong gamma-ray pair of 558 and 651 keV and is clearly observed in this spectrum

A cadmium standard sample provided by the Institute for Reference Materials and Measurements (IRMM) was measured with the new MPGA detector system. Counting rate of room background was about 50 count/s. Background neutron capture reaction at the beam duct and the target holder was suppressed by the neutron collimator, which is made of enriched ^{6}LiF tile. Air in the beam line was replaced with helium gas to reduce the background gamma-rays from the neutron capture reaction with nitrogen. Figure 3 shows a part of the two dimensional spectrum of IRMM B6, which is a plastic sample that contains 54 ppm cadmium. The event rate was about 1.7k count/s. Cadmium emits some prompt gamma-rays mainly in the neutron capture reaction of ^{113}Cd(n,γ)^{114}Cd: 558, 576, 651 and 725 keV gamma-rays, for example. The gamma-ray pair of 558 and 651 keV, which is emitted in the $2^{+}\rightarrow 2^{+}\rightarrow 0^{+}$ cascade de-excitation of ^{114}Cd, is the strongest in the two dimensional matrix spectrum as seen in Fig. 3. From the experiment, we estimated the quantification limit of cadmium in a plastic sample to be about 0.1 ppm.

Conclusions

In order to improve the sensitivity for the quantification of trace elements, we proposed the combination of prompt gamma-ray analysis (PGA) and a multiple gamma-ray detection method. A new Ge detector system for multiple prompt gamma-ray analysis (MPGA) was constructed at the neutron guide-hall of JRR-3M reactor in JAEA. The MPGA system consists of four Clover detector and BGO Compton suppressor. Two different methods for data analysis were examined. The first demonstration of this system is given with a plastic sample which contains traces of cadmium. The quantification limit of cadmium in a plastic sample was found to be about 0.1 ppm. The capability of quantification of the MPGA system could be improved by modifications of detector system, shield arrangement, software algorithm and so on.

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