DOI: 10.1007/s10967-008-1508-7 *Journal of Radioanalytical and Nuclear Chemistry, Vol. 278, No.3 (2008) 703–706*

Development of a neutron beam line and detector system for multiple prompt gamma-ray analysis

Y. Toh,[1]* M. Oshima,[1] K. Furutaka,[1] A. Kimura,[1] M. Koizumi,[1] Y. Hatsukawa,[1] J. Goto[2]

[1] *Nuclear Science and Engineering Directorate, Japan Atomic Energy Agency, Tokai, Naka, Ibaraki, 319-1195, Japan*
[2] *Radioisotope Center, Niigata University, Niigata, 951-8510, Japan*

(Received July 10, 2008)

A neutron beam line for multiple prompt gamma-ray analysis was constructed at the Japan Atomic Energy Agency. A detector system for the MPGA was constructed at the C2-3-2 beam line in January 2005. It comprised eight (upgraded in March 2007) clover Ge detectors with a BGO Compton suppressor. High efficiency detector system provides an advantage in terms of the detection limit of MPGA when compared to the result of PGA. The supermirror neutron bender was improved and a supermirror neutron guide was installed upstream of the sample position.

Introduction

Prompt gamma-ray analysis (PGA) is a high-sensitive, multielement, and non-destructive analysis method.[1] Therefore, it is employed in a large number of fields such as geology, archaeology, earth science, industry, and environmental science.[2–4] However, a quantification becomes difficult when the gamma-ray intensity from the trace element of interest is not sufficiently strong as compared with the intensities of the background gamma-rays from the elements which are contained in the sample with large amounts (but not of interest). In order to improve the sensitivity of the PGA, a multiple gamma-ray detection method was introduced in the PGA. The multiple gamma-ray detection method, which is known as a coincidence method, is widely used in nuclear spectroscopy. In this method, only those elements that simultaneously emit two or more prompt gamma-rays in a capture reaction are measured. When two or more gamma-rays are detected coincidentally by two or more gamma-ray detectors, they are added to the two-dimensional gamma-ray spectrum, which sets two axes for the energy value for every event. We developed a neutron beam line and a detector system for the combined method (multiple prompt gamma-ray analysis: MPGA), which is the multiple gamma-ray detection method coupled with the PGA.[5,6] The neutron beam line and detector system for the MPGA are constructed at the neutron guide hall of the JRR-3 at the Japan Atomic Energy Agency (JAEA). In the MPGA, the number of gamma-ray events is proportional to the intensity of the neutron beam and to the square of the efficiency of the detector system because the event data is stored when two gamma-rays are detected coincidentally using two detectors. In the MPGA, a high neutron intensity and a high efficiency of the detector system are required. Therefore, the supermirror neutron bender is improved, a supermirror neutron guide is developed, and the detector system is upgraded.

Experimental

Supermirror neutron bender and guide

In 2003, JAEA decided to modify the cold neutron guide tube C2-3 at the guide hall of the JRR-3. The C2-3 line was divided into C2-3-1 (for neutron spin echo spectrometer), C2-3-2 (for MPGA), and C2-3-3 (for neutron radiography) in March 2004.[7] A multilayer neutron bender and supermirrors are now widely used in many neutron sources for neutron control devices. In order to increase the intensity of the cold neutron beam, the supermirror neutron bender was upgraded and a supermirror neutron guide was installed upstream of the C2-3-2 beam line. The neutron bender and guide were made by supermirrors with ion polishing and nickel mirror total reflection. The critical angle of the neutron reflection on this mirror is three times larger than that of natural nickel (3Qc). The reflectivity has been increased, and it has reached 87% at the critical angle of total reflection. The supermirror neutron guide is shown in Fig. 1.

The integral neutron flux of the beam was measured by counting the number of prompt gamma-rays from the standard sample at the end position of the C2-3-2 beam line by using a BaF_2 scintillator. The supermirror neutron guide position was adjusted using a micrometer to obtain the maximum counting rate with the BaF_2 scintillator. In addition, gold-foil (3.2 mg: 5 mm×5 mm) activation was employed to determine the integral neutron fluxes separately. The activity of the gold foil was measured with a simple Geiger-Mueller (GM) counter. The counting rates of the GM tube are shown in Table 1. The integral neutron flux intensity of the beam was approximately 9.3 times greater than the previous.

* E-mail: toh.yosuke@jaea.go.jp

The present intensity of the neutron beam is sufficient for the MPGA. However, the intensity of the neutron beam should be optimized for each sample. Random coincidence degrades the accuracy of the MPGA. The probability of an random coincidence is then simply the probability of occurrence of an event in one detector and an event in another detector within a short interval (approximately 1 μs). The random coincidence rates increase with the square of the intensity of the neutron beam, while true coincidences increase only linearly with the intensity of the neutron beam. Therefore, a neutron attenuator system consisting of acrylic plates with different thickness was installed in front of the C2-3-2 beam line.

The neutron flux can be adjusted by changing the thickness of the acrylic plates. Neutrons are efficiently scattered by hydrogen and other light atoms such as C and O in the acrylic plates. The neutron beam intensity is decreased by approximately 30% for a 1-mm acrylic plate and approximately 50% for a 2-mm acrylic plate.

Gamma-ray detector system for MPGA

Phase-I MPGA detector system was constructed at C2-3-2 in January 2005. This system consisted of three clover Ge detectors with a BGO Compton suppressor, and in March 2007, phase-II detector system was installed, which comprised eight clover detectors. In addition, four coaxial Ge detectors and two twin Ge detectors will be installed by the end of 2007. The geometry of the detectors was optimized by employing GEANT4 calculation[8,9] (Fig. 2). The sample–detector distance is approximately 5 cm. Each clover detector consists of four individual crystals that share a common cryostat. The relative efficiency of each crystal is 25% compared with a 3-inch by 3-inch NaI detector. Because the four crystals in a clover Ge detector are packed very closely, the Compton-scattered gamma-ray from a crystal is often absorbed by one of the other three crystals. Consequently, by summing all the output signals from the four crystals, we can increase the relative photo-peak efficiency of a clover detector from approximately 100% to more than 120% for 1.3 MeV gamma-rays. This method, which is called the "add-back" mode, also improves the peak-to-total ratio. On the other hand, one cannot neglect events such as those in which the clover detector detects more than two cascade gamma-rays because of the proximity between the detector and the target. It is impossible to distinguish whether the coincidence event occurs from the cascade gamma-rays or from Compton scattering. In order to remove such unwanted events, we can analyze the same data with a method called the "anti-coin" mode; in this method, we discard events that contain more than two data from a clover detector. The anti-coin mode, in particular, uses the other three Ge crystals as anti-Compton suppressors. Although this mode decreases the photo efficiencies of the clover detectors, the peak-to-total ratio is improved.

Enhancement of the detection limit mainly depends on the absolute photo-peak efficiency of the detector system and the peak-to-total ratio, because it is determined by the ratio of the peak count to the square root of the background count. Detection limits (3 sigma) of PGA and MPGA are shown in Fig. 3, which is normalized to the PGA detection limit with 1% absolute photo-peak efficiency detector system. In the calculation, background is constructed by Compton scattering of high energy gamma-rays. High efficiency detector system provides an advantage in terms of the detection limit of MPGA when compared to the result of PGA. Therefore, high efficiency detector system is required in MPGA.

A high-speed data acquisition (DAQ) system is required for the detector system of the MPGA because of the proximity between the detectors and the target (approximately 5 cm) and because many large-volume gamma-ray detectors are employed.

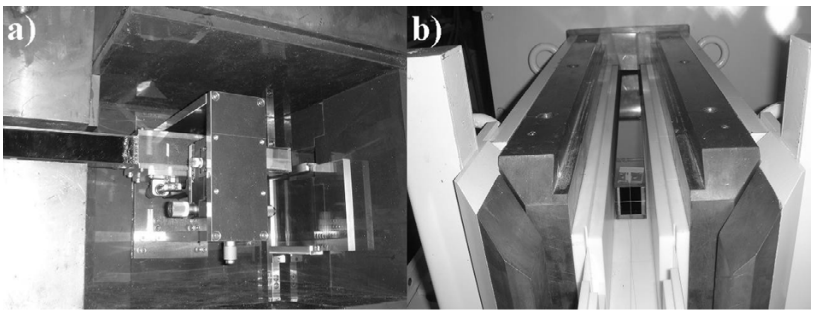

Fig. 1. Neutron guide installed upstream of the C2-3-2 beam line. This guide was made of 3Qc supermirrors; (a) shows the adjuster and (b) depicts the supermirror neutron guide

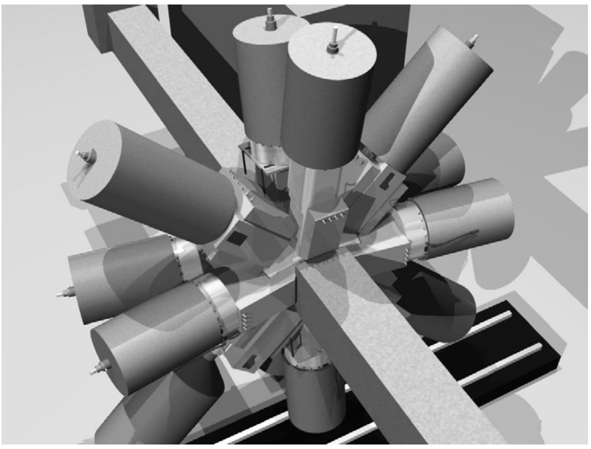

Fig. 2. The computer graphic of the Ge detector arrangement for the MPGA optimized using GEANT4

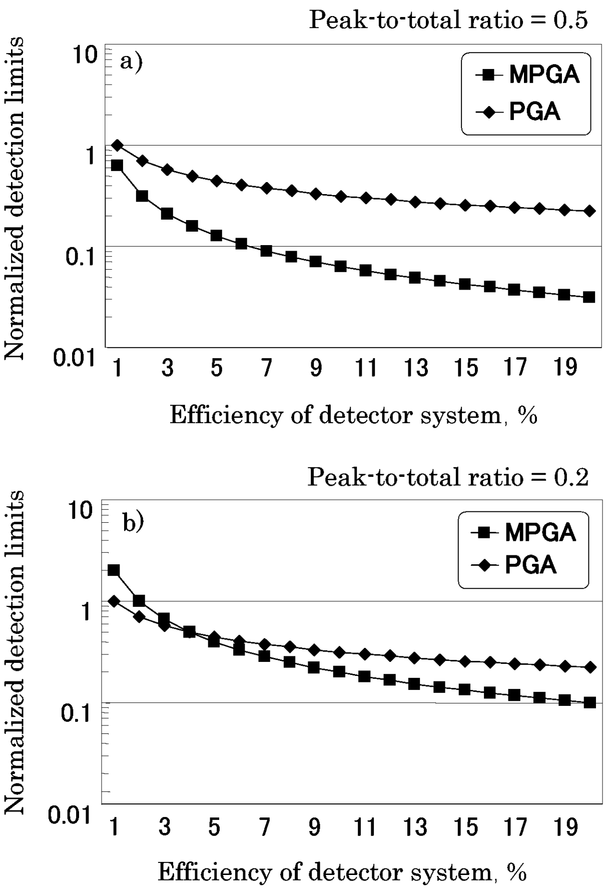

Fig. 3. Normalized detection limits of PGA and MPGA are shown; (a) peak-to-total ratio =0.5; (b) peak-to-total ratio = 0.2

Therefore, we developed a new DAQ system, which consists of main ADC modules, fast timing modules, and a coincidence module.[10–12] These modules comprise a digital signal processor (DSP) and a field-programmable gate array (FPGA)-based data acquisition board. These are 19-inch VME modules and can be mounted on a 19-inch VME rack. Once digitized, the data is ingested by the DSP and FPGA. The system can easily be upgraded to accommodate more detectors (maximum 256 channels). When the Ge detectors gave coincident signals, the data were recorded event by event on magnetic tapes or hard disk devices.

Table 1. Counting rates of the gold foil (3.2 mg: 5 mm×5 mm) before and after the improvement of the neutron guide and bender at a sample position. The integral neutron flux of the beam was approximately 9.3 times greater than the previous one

Guide and bender	Irradiation time, s	Count, cpm
Before improvement	1800	450
After improvement	1800	4200

Results and discussion

The supermirror neutron bender and supermirror neutron guide were modified and installed simultaneously. Hence, the effect of each of these improvements cannot be measured separately. However, we can estimate this effect by using numerical simulation. The calculation results of the effects of the supermirror neutron bender and neutron guide are approximately 5 and 2.5, respectively. The total effect of the supermirror neutron bender and guide approximately agreed with the experimental value of 9.3.

The MPGA system also contains an automatic sample-changing system, which can accommodate 160 samples and change the samples within 30 seconds. Improvement in the neutron intensity, the upgradation of the detector system, and the automatic sample-changing system allowed the screening analysis using the MPGA. The PGA shows excellent sensitivity for the determination of some toxic elements and heavy metals (Cd, Hg, Gd, Sm, etc.). The MPGA system may be used for the survey analysis for samples of foods, polymers, soil, etc.

*

We would like to express the gratitude to the staff of the JRR-3M for providing and upgrading the neutron beam. The authors would like to thank Dr. K. SOYAMA, Dr. R. MARUYAMA and Dr. I. TAMURA for assistance and useful discussions.

References

1. R. E. CHRIEN, in: Neutron Radiative Capture, B. J. ALLEN, I. BERGQVIST, R. E. CHRIEN, D. GARDNER, W. P. POENITZ (Eds), Pergamon, Oxford, 1984, p. 187.
2. M. KATAOKA, A. KUNO, M. MATSUO, J. Radioanal. Nucl. Chem., 255 (2003) 283.
3. L. SAJO-BOHUS, MA. M. ANTCZAK, ZS. KASZTOVSZKY, E. D. GREAVES, A. ANTCZAK, A. SIMONITS, D. PALACIOS, B. MILLAN, J. Phys., Conf. Series, 41 (2006) 408.

4. S. OHDE, A. A. RAMOS, H. OZAKI, H. SAWAHATA, T. OKAI, J. Radioanal. Nucl. Chem., 258 (2003) 431.
5. Y. TOH, M. OSHIMA, M. KOIZUMI, A. OSA, A. KIMURA, J. GOTO, Y. HATSUKAWA, Appl. Radiation Isotopes, 64 (2006) 751.
6. M. OSHIMA, Y. TOH, A. KIMURA, M. EBIHARA, Y. OURA, Y. ITOH, H. SAWAHATA, M. MATSUO, J. Radioanal. Nucl. Chem., 271 (2007) 317.
7. J. SUZUKI, S. TASAKI, K. SOYAMA, T. EBISAWA, Appl. Phys., A74 (2002) S308.
8. S. AGOSTINELLI, J. ALLISON, K. AMAKO, J. APOSTOLAKIS, H. ARAUJO, P. ARCE, M. ASAI, D. AXEN, S. BANERJEE, G. BARRAND et al., Nucl. Instr. Meth., A506 (2003) 250.
9. J. GOTO, M. SUGAWARA, M. OSHIMA, Y. TOH, A. KIMURA, A. OSA, M. KOIZUMI, M. MIZUMOTO, T. OHSAKI, M. IGASHIRA, H. HARADA, Y. NAGAI, Am. Inst. Phys. Conf., 788 (2005) 792.
10. A. KIMURA, Y. TOH, M. KOIZUMI, A. OSA, M. OSHIMA, J. GOTO, M. IGASHIRA, Am. Inst. Phys. Conf., 769 (2005) 792.
11. A. KIMURA, M. KOIZUMI, Y. TOH, A. OSA, M. OSHIMA, J. GOTO, Y. ARAI, M. SAGARA, S. IRI, IEEE Nuclear Science Symp. Conference Record, Vol. 3, (2004) 1489.
12. A. KIMURA, M. KOIZUMI, Y. TOH, A. OSA, M. OSHIMA, Y. ARAI, M. SAGARA, S. IRI, H. KOBAYASHI, D. MAEDA, T. HIRAYAMA, Y. SUZUKI, IEEE Nuclear Science Symp. Conference Record, Vol. 1, (2005) 570.