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Multiple prompt gamma-ray analysis and construction of its beam line

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By combining neutron activation analysis with multiple gamma-ray detection (gamma-gamma coincidence), we have proved better sensitivity and resolution for the trace element analysis than the ordinary single gamma-ray detection method. We now try to apply the multiple gamma-ray detection method to the prompt gamma ray analysis (PGA). We have established a new cold neutron beam line for PGA in Japan Research Reactor, JRR-3M, at Tokai establishment of Japan Atomic Energy Research Institute (JAERI). It consists of a beam shutter, a beam attenuator, a gamma-ray detector array, a sample changer, and a beam stopper. We construct a high-efficiency gamma-ray detector array specially designed for this purpose. Its performance has been evaluated with the Monte Carlo simulation code, GEANT 4.5.0.

Introduction

Prompt gamma-ray analysis (PGA) is a versatile element quantification method with non-destructive, rapid, high-precision and multi-element determination. It is widely used in the fields of environmental science, biology, astrophysics and so on. PGA usually utilizes a single detector to measure gamma-rays depopulating a neutron-captured state. The energy resolution of a high-resolution germanium detector is approximately one thousand, corresponding to less than 2.0 keV full-width at half-maximum for 1 MeV gamma-ray. Since the number of gamma-rays from the sample with multiple elements often exceeds one thousand, the energy resolution is not sufficient and it is impossible to resolve all the gamma-rays. Weak gamma-rays, in particular, are masked by strong ones and their quantification is difficult. Furthermore, the prompt gamma ray strength is seriously fragmented so that one needs better resolution and sensitivity than the case of neutron activation analysis. Because of these reasons, we applied to PGA the multiple gamma-ray detection method, which enables high resolution and high sensitivity measurement.

The principle of multiple gamma-ray detection coupled with PGA

We took advantage of the fact that most of the neutron-capture nuclides emit coincident multiple gamma-rays. These multiple gamma-rays can be detected in coincidence with a gamma-ray detector array and an event data composed of a pair of multiple coincident gamma-ray energies is obtained. From the correlated data we can create a gamma-gamma two-dimensional matrix which incorporates the correlation among the gamma-rays. On this matrix we can have an energy resolution of 1,000,000, because the resolution reduces to the product of those in each axis (Fig. 1). This value is by a factor of one thousand better than the ordinary one-dimensional spectrum. Since the number of gamma-rays produced in PGA for one nuclide often exceeds 100, improvement of the energy resolution is essential to achieve good sensitivity.

Another advantage of the multiple gamma-ray detection is the improvement of signal-to-noise ratio. As shown in Fig. 2, a gamma-ray peak in one-dimensional spectrum is converted typically to one or plural peaks in two dimensional matrix. However, the background counts coming from Compton scattering in the germanium detector mainly localize on a line parallel to the X or Y axes so that in most area on the matrix background counts is reduced to a few counts. Sometimes, when the major nuclide emits only a single gamma-ray like the 2.3 MeV gamma-ray from hydrogen, it will disappear in the two-dimensional matrix originating from gamma-gamma coincidence. In this case the improvement of the signal-to-noise ratio is even better than one thousand. We apply this technique to the prompt gamma ray analysis, where the multiplicity of the gamma-ray is 2–4 in most cases. This multiple prompt gamma ray analysis (MPGA) is successful for the analysis of cadmium in rice and the result is presented also in this Conference.[1]

Experimental

Application of multiple gamma-ray detection method to NAA

To demonstrate the validity of the multiple gamma-ray detection method, we performed an NAA experiment for standard rock samples, JB-1a (basalt) and JP-1 (peridot), issued by Japan Geological Survey.

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The weight was 100 mg each. The samples were irradiated with neutrons from the JRR-4 reactor at JAERI with a flux of $3.3\cdot10^{13}$ $n\cdot cm^{-2}\cdot s^{-1}$. Gamma-rays were measured with a gamma-ray detector array, GEMINI, which comprises twelve BGO Compton suppressed germanium detectors.[2] The detection efficiency of each Ge detector is 40–70% relative to a 3"ϕ×3" NaI scintillator. The peak-to-total ratio is 0.50. The total detection efficiency amounts to 1.5%. Gamma-gamma coincidences were measured for 100 and 24 hours for JB-1a and JP-1, respectively. It turns out that 4 ppb of europium was successfully identified as the minimum quantity in those samples. Broad dynamic range from 10^{-2} percent to 10^{-9} ppb order could be addressed in a single measurement. For those samples totally 27 elements have been quantified and their minimum quantity was 10^{-9} ppb order in europium.[3,4]

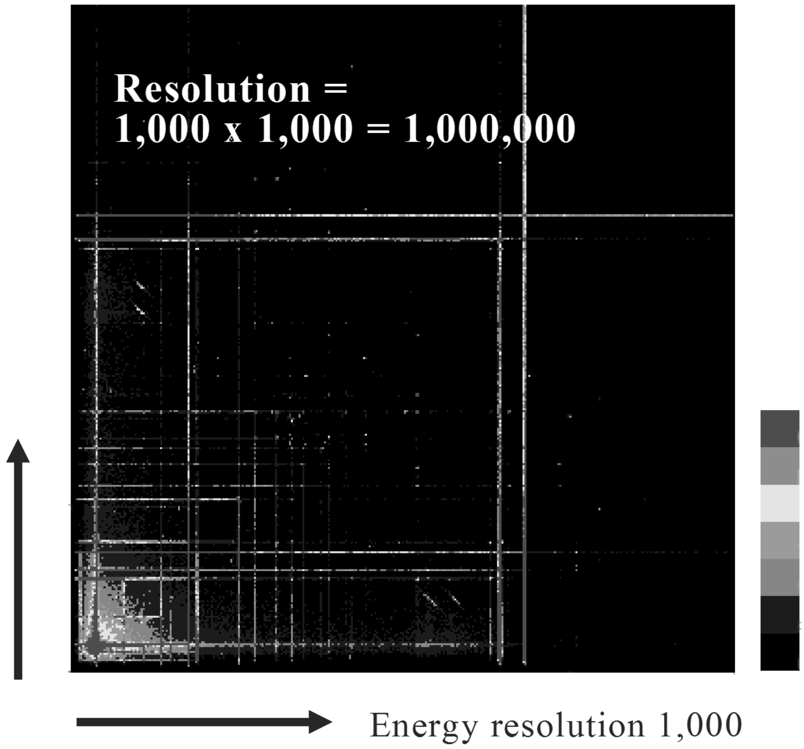

Fig. 1. Improvement of energy resolution. Number of nuclides is less than 2,800 so that one can identify as many nuclides as present in the sample

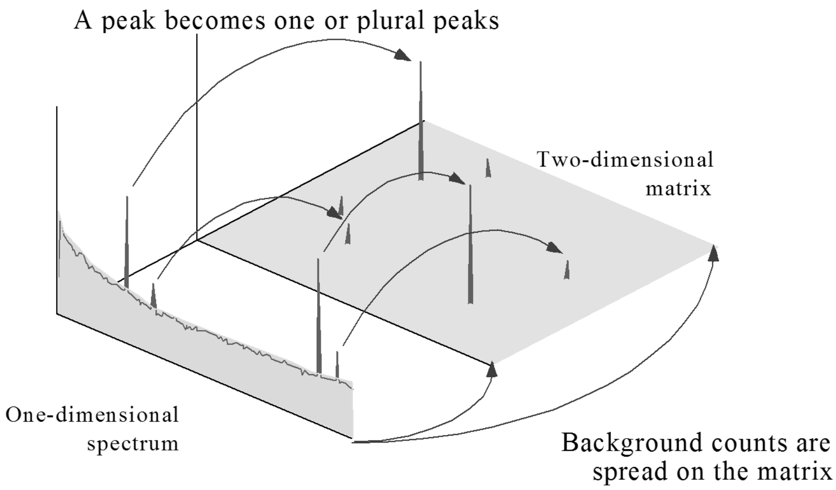

Fig. 2. Improvement of nuclide sensitivity. Two-dimensional peaks appear on the matrix, which can be used for nuclide determination

Quantification is often made by the comparison method: a sample of interest and a standard sample including a known amount of any element are irradiated by neutrons at the same condition and their gamma-ray intensities are compared. The quantity of the element can be deduced from their ratio. This procedure does not include any nuclear data (except the half-life for the decay correction) and thus gives an accurate result. However, it has been applied only for single element so far. We have prepared two types of grand standard samples, which include 23 elements altogether with different quantities. The irradiation and measurement for both samples were 10 minutes and 24 hours, respectively. With the multiple gamma-ray detection technique, the reproducibility of the weight from gamma-ray measurement has been tested. In this way, it has been proven that 23 elements can be quantified simultaneously with an accuracy of 3–20% depending on the statistics.[5]

An application test of multiple gamma-ray detection method to PGA

The applicability of the multiple gamma-ray detection method to PGA was examined in a cold neutron beam line c2 for PGA in Japan Research Reactor, JRR-3M, at Tokai establishment of JAERI. We used two coaxial Ge detectors with 24% and 35% efficiencies relative to a 3"$\phi\times$3" NaI scintillator. The 24% detector was surrounded by a BGO Compton suppressor. We put 1g sample of natural iron at the center of the neutron beam line. The flux was $3.4\cdot10^{6}$ $n\cdot cm^{-2}\cdot s^{-1}$. We measured singles and gamma-gamma coincidences for ten hours and obtained the spectra shown in Fig. 3. Figure 3a indicates the singles spectrum obtained by the Ge detector with BGO suppressor and Fig. 3b shows a coincidence spectrum gated on 352.36 keV gamma-ray. The distances of the detectors from the sample were larger than 15 cm in this test experiment so that the geometry was not well suited for the coincidence experiment. Thus, the counts in the gated spectrum is much less than those of the singles spectrum, but the improvement of the signal-to-noise ratio is evident. For example, the ratio of the 1206.60 keV peak is improved by a factor of about 4 in the gated spectrum as compared with the singles spectrum. It should be noted that the statistics can be improved by using a more efficient gamma-ray detector array as described below.

Results

Construction of MPGA beam line

Recently we have established a new cold neutron beam line for PGA. The cold neutron line c2 has been reconstructed and a new beam line c2-3-2 is installed. The beam size is 3 cm×4 cm. The constructed MPGA beam line, shown in Fig. 4, has been completed in March, 2004. The beam line consists of a beam shutter, a beam attenuator, a gamma-ray detector array, a sample changer, and a beam stopper. The neutron flux at the end of the guide tube (entrance of the MPGA beam line) and at the sample position is $1.2\cdot10^{7}$ and $2.1\cdot10^{6}$ $n\cdot cm^{-2}\cdot s^{-1}$, respectively.

The gamma-ray detector array specifically designed for MPGA consists of four clover Ge detectors and four BGO Compton suppressors, which are arranged symmetrically at 90 degree relative to the beam axis (Fig. 5). The clover Ge detector has the energy resolution of 2.3 keV FWHM or better at 1.33 MeV and the relative efficiency of 120% against 3"$\phi\times$3" NaI scintillator. The BGO scintillator has the suppression factor of 8 or larger. Its performance has been evaluated with a Monte Carlo simulation code, GEANT 4.5.0 and the total detection efficiency amounts to 8%. The construction of the new detector system will finish at the end of 2004.

PGA is known to have the following features:[6] (1) non-destructive; (2) high-precision (% order) without chemical separation, and (3) simultaneous determination of multi-elements (up to 70).

Based on the GEANT simulation we expect that the lowest determination limit is 0.05 ppm or better in measuring Cd for ten minutes.[1] Thus, the high sensitivity up to ppb order becomes possible by combining the multiple gamma-ray detection method.

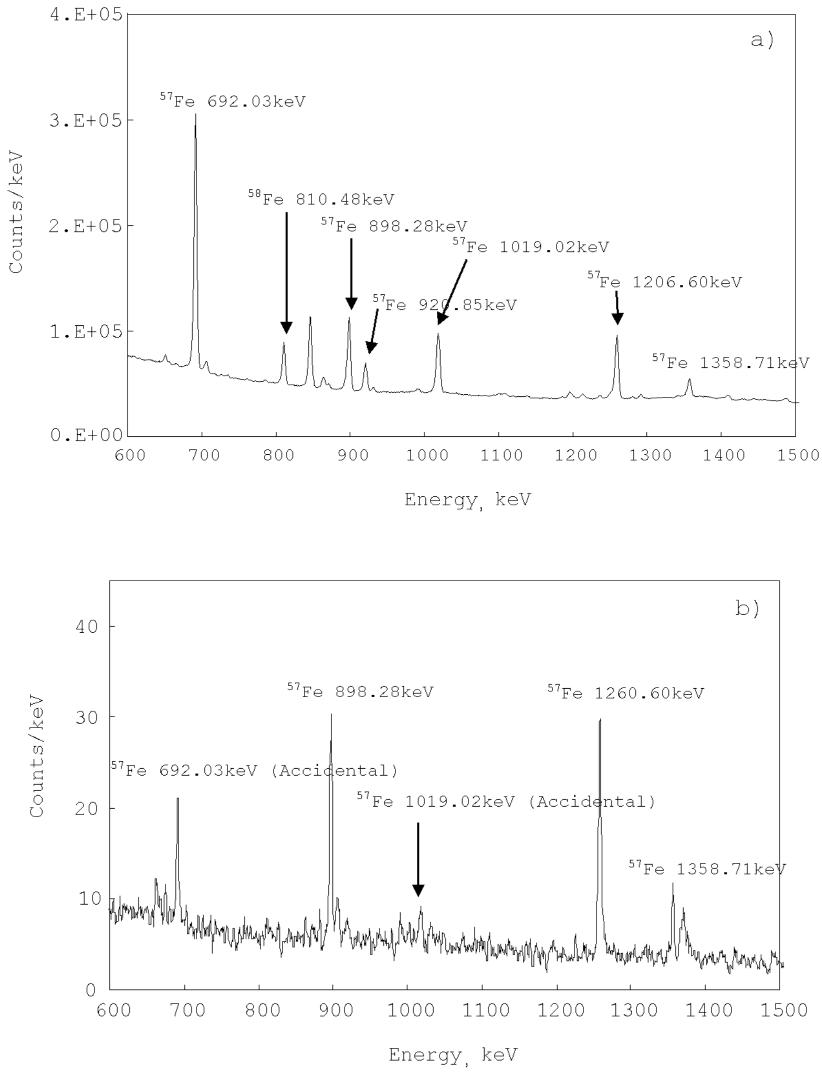

Fig. 3. Prompt gamma-ray spectra for a ^{nat}Fe sample; (a) is a singles spectrum and (b) a gated spectrum obtained by putting a gate on 352.36 keV transition in ^{57}Fe. Gamma-rays of $^{57,58}Fe$ are observed which originate from neutron capture of $^{56,57}Fe$

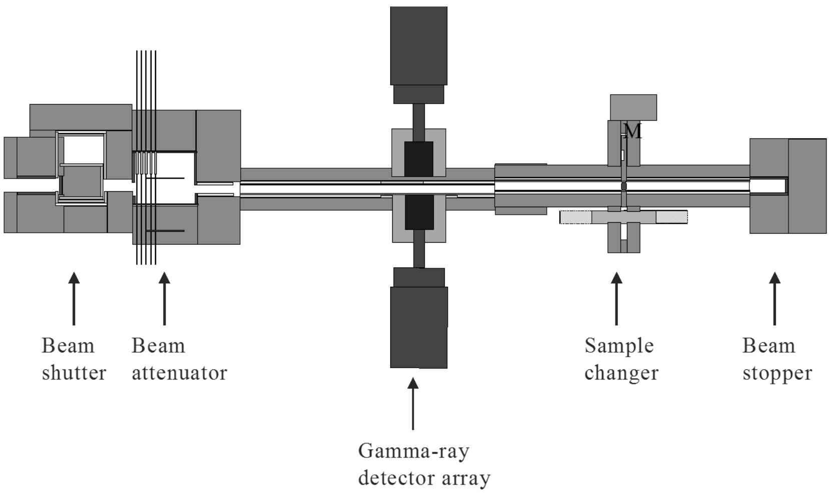

Fig. 4. Arrangement of MPGA beam line. It consists of a beam shutter, a beam attenuator, a gamma-ray detector array, a sample changer, and a beam stopper

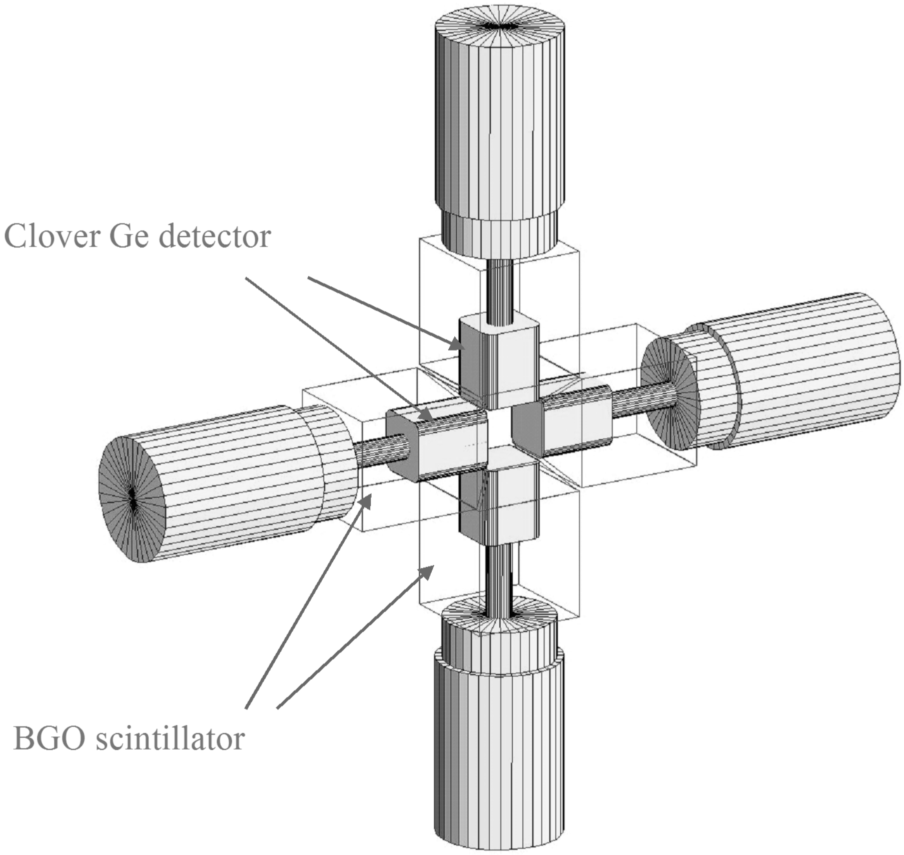

Fig. 5. A gamma-ray detector array designed for MPGA experiment

Summary and future perspectives

By combining the multiple gamma-ray detection method with prompt gamma ray analysis, we can develop a rapid non-destructive trace element analysis technique, which enables simultaneous quantification of 70 elements with high sensitivity (ppb order at maximum) and high precision (% order except statistical uncertainty). As a price for these merits, the other 12 elements are not suited for this method, because the neutron capture cross section is too small or the neutron-capture nuclides do not emit multiple coincident gamma-rays. Ordinary one-dimensional PGA analysis needs to be applied in the latter case. The new MPGA beam line has been already completed. The new detector system will be finished at the end of 2004. Then the applications based on collaboration among more than ten universities and institutes is expected in the fields of astrophysics, geology, archaeology, environmental and medical sciences and so on.

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