The design and implementation of the PGA measurement system in an integrated Active Neutron non-destructive analysis system, "Active-N"

Kazuyoshi Furutaka、Akira Ohzu, and Yosuke Toh
Nuclear Science and Engineering Center,
Japan Atomic Energy Agency

ABSTRACT

In order to facilitate quantification of highly radioactive nuclear materials, we have been developing an integrated active neutron non-destructive analysis system, "Active-N", equipped with an intense D-T neutron source. The system is composed of the following mutually complementing different measurement systems: Differential Die-away Analysis (DDA), Neutron Resonance Transmission Analysis (NRTA), and Prompt Gamma-ray Analysis (PGA). The purpose of the. PGA measurement system is the detection of neutron poisons which disturb DDA measurements, explosives contained in e.g., dirty bombs, and chemical warfare agents. For these, a Ge detector is utilized. In order to do reliable PGA measurements with the integrated system, preliminary measurements were done with a prototype integrated system and the results were examined, and then the combined system of PGA and DDA was designed as a whole. To suppress interfering gamma rays from the DDA system, the use of stainless-steel was reduced as much as possible and lead shields were placed between the two measurement systems.

In order to reduce the number of fast neutrons from the D-T neutron generator which hit the Ge detector and cause radiation damage, an additional neutron shield was designed using series of Monte Carlo radiation transport simulations. With the shield made of high-density polyethylene into which LiF is mixed with 50 wt%, the number of fast neutrons is expected to be reduced about an order of magnitude.

A unified data taking system for the integrated measurement system was developed using waveform digitizers.

Introduction

As nuclear materials can be used for mass-destruction weapons which endanger world peace[1], they must be kept track of. For that, it is essential to have a reliable and accurate method for the nuclear-material accountancy. In order to provide such a means, we have been developing a kind of active neutron interrogation method, namely, Differential Die-away Analysis (DDA) method, and devices based on it. Our method, Fast Neutron Direct Interrogation (FNDI) method[2], is a type of DDA method which can measure the amount of nuclear materials in a large container such

as a drum without large uncertainties caused by localization of the material. In spite of the above-mentioned advantage of the method, it can only determine the total amount of fissile materials and it does not tell how much a fissile isotope is contained. Further, it is not yet clear whether it can be applied to highly radioactive nuclear materials such as those in spent nuclear fuels, fuels used in Accelerator Driven Systems for transmutation of long-lived radioactive isotopes, and fuel debris produced in an accident of nuclear power plants. Though recently the demand increases for a practical accounting method that can be applied to highly radioactive nuclear materials, it is not yet established. In the case of the fuel debris, the possibility that it would be a conglomerated mixture of nuclear materials and neutron poison (such as control rods) makes the accurate measurements with neutron interrogation difficult; it is not clear whether the sample really contains only a small amount of fissile materials or the interrogation/fission neutrons are absorbed by the poisons.

Nowadays, the concern of use of Radioactive Dispersive Devices (RDD),which intentionally disperses radioactive materials with explosives, is also growing, and it is desirable to speedily detect it. The key may be the detection of explosives. A promising candidate is the Prompt Gamma-ray Analysis (PGA) with neutron irradiation. PGA has been used for example to analyze trace amount of elements, and is able to detect and quantify light elements including a typical neutron poison element, boron.

In order to compliment DDA with PGA to enable detection of neutron poison elements and explosive materials, the present authors conceived a combined system of both methods which enables the detection of explosives, toxic materials, chemical warfare agents and neutron poisons in addition to fissile materials. The concept was further expanded to include Neutron Resonance Dosimetry Analysis (NRTA) method which enables the analysis of isotopic ratios of nuclear materials, and the authors have started a project to develop an integrated measurement system named “Active-N”, to proof the concept[3]. In the first phase of the project, a prototype system was built which was optimized for DDA measurements[4], and was then examined to find what was in need to enable coexistence of PGA with DDA, and the DDA+PGA combined system was re-designed with the knowledge obtained in the prototype system. In the present second phase, the re-designed combined system, with NRTA, is under construction.

In this paper, the design and implementation of the PGA part of the system is described. In addition, the data taking of the combined system is briefly presented.

Design and implementation of the PGA facility

Targets of the PGA measurements

The targets of the PGA detection/measurement can be summarized in the following three categories:

- Neutron poisons,
- Elements which signify the existence of explosives, and

· Toxic elements/compounds and chemical warfare agents.

The neutron poisons considered in this project are boron, chlorine and the compounds which include either or both of the elements. Most of explosives include nitrogen with high mixing ratios and therefore the prompt gamma rays of nitrogen can be used as an indication of explosives. Elements considered as signs of toxic materials are (in addition to nitrogen and chlorine) phosphorus, sulfur and arsenic. The following table shows examples of the materials which contain these elements.

B	Neutron poison
N	Explosives Toxics: VX gas($C_{11}H_{26}NO_2PS$), Cyanogen chloride (CNCl), N-mustard ($CH_3CH_2N(CH_2CH_2Cl)_2$ etc.)
P	White phosphorus munitions, VX gas ($C_{11}H_{26}NO_2PS$), Sarin ($C_4H_{10}FO_2P$)
S	VX gas ($C_{11}H_{26}NO_2PS$)
Cl	Neutron poison Toxics: Lewisite ($C_2H_2AsCl_3$), sulfur mustard ($C_4H_8Cl_2S$), phosgene (CCl_2O), Cyanogen chloride (CNCl), N-mustard ($CH_3CH_2N(CH_2CH_2Cl)_2$ etc.)
As	Toxics: Lewisite ($C_2H_2AsCl_3$)

Brief description of the FNDI method

For a PGA system to coexist with a DDA system and to be able to do reliable measurements, its design should not end within the PGA part only but be extended to the DDA part. Before describing the overall design of the combined system, the principle of a DDA measurement will be explained first.

The DDA method is a type of active neutron interrogation methods, and in this method, a sample is irradiated by neutrons and the “die-away”, i.e., change in time of the number of neutrons detected, is analyzed, to extract information on the amount of fissile materials. We use a neutron generator which utilizes D-T reactions, and the FNDI method in which the sample is directly irradiated by the 14-MeV neutrons from the generator without moderation. By irradiating fast neutrons with high penetrability and let them moderated in the sample, accurate results can be obtained without uncertainties caused by the materials of matrix contained even if the fissile materials are unevenly distributed in a large-size container. Neutrons arising from the fission reactions are discriminated against thermal ones by surrounding neutron detectors with thermal-neutron absorbing materials such as boron or cadmium. By analyzing die-away of the neutron counts, the total amount of fissile material is deduced.

PGA measurements in Prototype Active-N system and improvements for the new system

A photo of the prototype Active-N is shown in Figure 1. The apparatus is surrounded by neutron

shields made of high-density polyethylene (HDPE) to reduce the number of outgoing neutrons. A neutron generator, a sample rotating table, and a detector bank are placed inside the shields. The detector bank consists of sixteen ^{3}He detectors (9.5atm, 1m in length) each wrapped by a cadmium sheet. Each detector is inserted in a hole of a HDPE block, which is then surrounded by a layer of B_4C resin and placed inside a case made of stainless-steel.

Figure 1: A view of the prototype Active-N. PGA and DDA measurement regions are roughly indicated by dashed lines. The PGA detector is surrounded by a tube made of lead (3 cm in thickness) and then is stored inside the shielding made of boron-mixed HDPE (20%).

Shown in Figure 2 is an energy spectrum of the gamma rays measured by a Ge detector without a sample placed in the prototype system. As can be seen from the figure, many gamma-ray peaks were observed although no sample was placed. Prominent gamma-ray peaks are the following:

- 2223-keV gamma ray emitted in the neutron-capture reaction by hydrogen atoms in HDPE,
- ~478-keV gamma rays due to the ^{10}B(n,α) reaction in the DDA detector bank, and
- Gamma rays from neutron-capture reactions by iron, manganese, nickel and chromium in stainless-steel.

The number of gamma rays emitted without an irradiation sample should be minimized as small as possible, or otherwise weak gamma rays cannot be detected.

Shown in the inset of Figure 2 is an expanded view of the spectrum around the e^{+} annihilation peak. Arsenic, an objective of the PGA measurement, emits a gamma ray with the energy 559 keV and it is the most intense one at thermal neutron energy. A small peak is observed at around 559 keV in the expanded view. This is due to the capture of neutrons by ^{113}Cd (thermal cross

section: 20.17×10^3 barns[5]) in the DDA detector bank. The interfering gamma ray should also be eliminated for the detection of arsenic gamma rays.

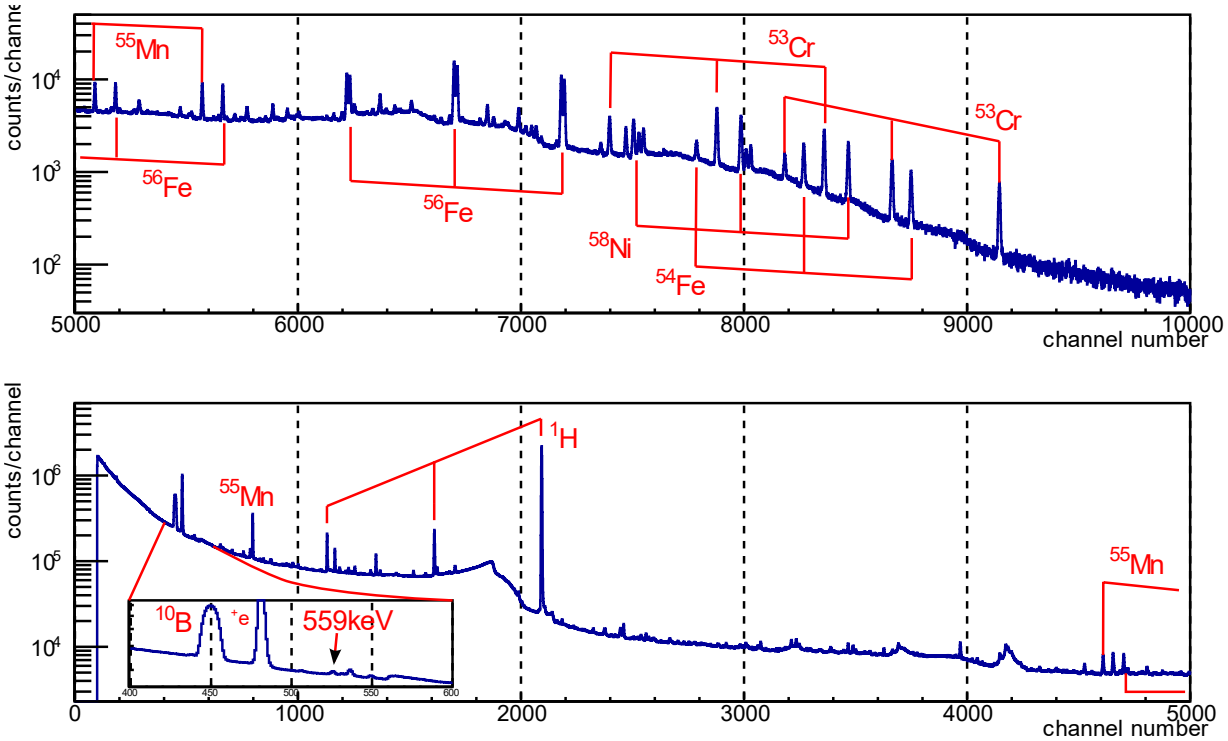

Figure 2: An energy spectrum of gamma rays observed in the prototype Active-N without placing an irradiation sample. Many gamma rays originated in the neutron capture by hydrogen, boron, iron, nickel, and manganese, were observed, most intense one being the one from hydrogen atoms.

Based on the findings obtained with the prototype system, the new Active-N was designed and improved in the following manner: (i)the use of stainless-steel is reduced and aluminum is used instead which has far smaller capture cross sections, (ii)sources of the interfering gamma rays were covered with lead so that the Ge detector does not directly see them.

Figure 3 depicts horizontal/vertical cutaway views of the combined DDA+PGA measurement parts of the new Active-N. The apparatus is of cuboid shape, and the PGA part is placed on top. Three neutron detector banks for DDA measurements are embedded in the side shielding walls, and the neutron generator is set inside a gap in the last side wall. To be used for highly radioactive samples, for DDA neutron detectors, B-10 detectors are employed which has low gamma-ray susceptibility. To compensate its low neutron detection efficiency, a total of 60 B-10 detectors are used. Inside of the cuboid is layered by lead so that the PGA detector does not directly view the DDA detectors which use boron and are covered by cadmium.

PGA measurement system and its shielding against fast neutrons

By absorbing a neutron, Nitrogen emits relatively higher energy gamma rays compared to other elements, the highest being 10.8 MeV. Therefore, it is advantageous to observe the gamma rays to detect the existence of nitrogen, which is contained at high mixing ratios in explosives. To detect the high-energy gamma rays efficiently, a Ge detector (N type) of 70% relative efficiency is utilized. It is cooled with electronic refrigerator. The endcap of the Ge detector is surrounded by a BGO (Bismuth Germanate) detector to suppress Compton events by doing anti-coincidence between the two detectors. The front and side faces of the BGO detector are covered by a lead collimator 5cm in thickness. Placed below the collimator are LiF tiles enriched in ^{6}Li to 95% (1cm in thickness) and LiF-mixed HDPE (50 mm in thickness), in descending order.

In usual neutron PGA measurements, irradiations are done with a beam of neutrons, which is extracted from a neutron source such as a nuclear reactor and transported through guide tubes. This means that only the sample is irradiated by neutrons. On the other hand, in Active-N, neutrons are generated through D-T reaction and are emitted almost isotopically; not only the irradiation sample but also the Ge detector is irradiated by those neutrons. It is well known that fast neutrons ($\gtrsim$ 10 keV) induce radiation damages in germanium crystals[6]. The number of damages increase with the dose and gradually deteriorate energy resolution of the detector and finally become unusable for reliable measurements (although the damage can be repaired by

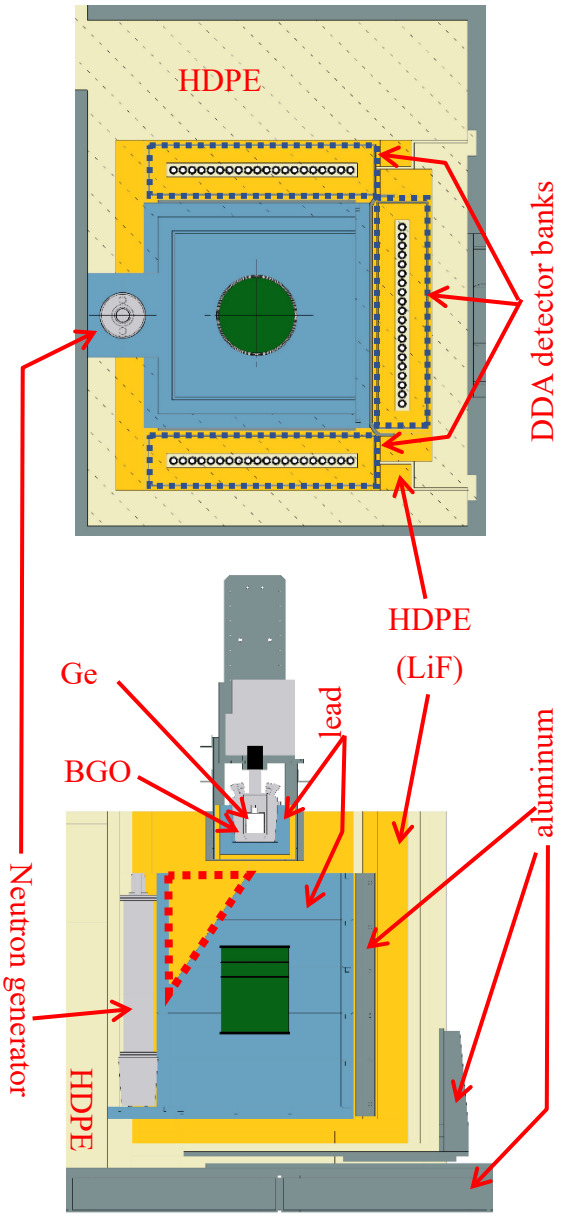

Figure 3: Horizontal (upper) and vertical (lower) cutaway views of the PGA&DDA parts of new Active-N. The position of additional triangular-prism shaped shielding is indicated by red dotted line. The position of a sample stored in 18-littre pale is indicated in green. The region of additional shielding, which will be described in the next subsection, is indicated by a triangle in red dashed line.

annealing). To elongate the operation cycle of the detector, shielding against fast neutrons are essential.

In order to reduce neutron dose at the Ge detector, additional neutron shielding for the detector was designed using a Monte Carlo radiation transport code system[7]. The code system used was PHITS[8]. There is only a limited space left for the additional neutron shielding (at most 60 cm); it is indicated by a red dashed triangular area in Figure 3.

At first, to select a material or combinations of several materials which can effectively shield fast neutrons, a series of simplified simulations were done in which a pencil beam of 14-MeV neutrons was incident on a circular face of a cylinder, 60 cm-long and 100 cm in diameter, along its axis. The cylinder is divided into 60 equal parts, and each part is filled with a material. The number and energy of neutrons exited through the other circular face were tallied and materials or their combinations were searched for which effectively reduce the number of neutrons with their energies $\gtrsim$ 10 keV. Through the simulations, it is found that tantalum, tungsten, lead and nickel are extremely effective to reduce the number of fast neutrons due to their large elastic cross sections, but they were excluded because they will become the additional sources of gamma rays emitted after inelastic collisions. Further, with finite size shielding the scattered neutrons will then be absorbed somewhere in the experimental room and induce more background gamma rays. Boron-carbide was also excluded for use because it is a compound of an objective material, boron. As the candidate materials for the additional shielding, LiF, LiF in which ^{6}Li was enriched to 95 weight percent (denoted as ^{6}LiF hereafter), HDPE, HDPE (LiF mixed) were chosen[7].

Then, another series of realistic simulations were done with these materials. In the simulations, the triangular prism region for the additional shielding was horizontally sliced to 1 cm-thick layers and each layer was filled with a candidate material, and neutrons as well as gamma rays were tallied at the position of the Ge crystal in the PGA detector. The following configurations were studied: (i)HDPE (^{6}LiF layers are interlaced at 3-cm intervals), (ii)HDPE (^{6}LiF interlaced at 5-cm intervals), (iii) HDPE (^{6}LiF interlaced at 7-cm intervals), (iv)LiF-mixed HDPE (^{6}LiF interlaced at 7-cm intervals), and (v)LiF-mixed HDPE. The gamma-ray fluences obtained by the simulations are more or less the same except for case (iii). As for the neutron fluence, the minimum is with configuration (iv) and is followed by (v) (about 2% larger) and the value increased from (i) to (iii) with the interlace intervals. From these results and economical consideration, LiF-mixed HDPE was chosen for the additional neutron shielding. The result of the simulation shows that by using the material for the additional neutron shielding, the fluence of neutrons with energies larger than 10 keV can be reduced by about an order of magnitude[7].

The data-taking system of Active-N

A highly radioactive sample emits gamma rays and therefore counting rates of the detectors would be high. In order to do reliable measurements and reduce the dead times in such a situation, the time taken to process one detection event should be made as short as possible. In order to enable

data taking at high rate, waveform digitizers have been introduced for the data acquisition. It not only reduces the processing time but also the size of data acquisition system because pulse shaping, pulse-height determination, anti-coincidence and time-of-flight calculation can be done in a few digitizer modules. It is advantageous especially for DDA measurements that use 60 neutron detectors. The following table shows the digitizers used for Active-N:

measurements	detector(s)	# of detectors	digitizers (pulse-processing method)
PGA	Ge	1	CAEN V1724E (DPP-PHA)
	BGO	8	CAEN V1725B (DPP-QDC)
DDA	B-10	60	CAEN V1725B×3 (DPP-PHA)
NRTA	Li-glass	1	CAEN V1725B (DPP-QDC)

With these 6 digitizers housed in only one VME crate, all the signals from the Active-N detectors can be processed. Anti-coincidence between the two PGA detectors is done via a VME programmable logic unit.

Summary

In this paper, the design and implementation of PGA measurement part of an integrated NDA measurement system, Active-N, are described. For PGA to be integrated with DDA and to do reliable measurements, the design of PGA had to be extended to the selection of materials used in DDA apparatus so that it does not become sources of interfering gamma rays. In order to reduce the number of gamma rays originated from hydrogens in HDPE, LiF-mixed HDPE is partly employed for neutron shielding. For DDA, it is inevitable to use cadmium as thermal-neutron absorbers and B-10 detectors as neutron detectors with low gamma susceptibility. In order to prevent gamma rays due to these elements from interfering with those from the objectives of PGA measurements, lead shields are placed between PGA and DDA detectors.

In order to reduce the number of fast neutrons which cause radiation damage to the Ge detector used for PGA measurements and extend usable periods of the detector, additional neutron shield was designed by doing series of Monte Carlo simulation. From the simulation it was found that the number of fast neutrons can be reduced to about an order of magnitude by adding a triangular prism shaped shielding made of LiF-mixed HDPE.

To construct a fast and compact integrated data taking system for Active-N, waveform digitizers were utilized. With six waveform digitizers contained in only one VME crate, signals from more than 60 detectors can be processed.

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