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NON-DESTRUCTIVE NUCLEAR DETECTION AND MEASUREMENT TECHNOLOGY DEVELOPMENT PROJECTS OF JAEA FOR NUCLEAR NON-PROLIFIRATION AND SECURITY

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ABSTRACT

The Japan atomic energy agency (JAEA) is implementing technological development projects on non-destructive assay (NDA) for nuclear detection and measurement under the support of the subsidy for "promotion of strengthening nuclear security or the like" of the Japanese government MEXT (Ministry of education, culture, sports, science and technology). In this presentation, three of the projects are briefly overviewed.

The first one is “development of nuclear resonance fluorescence (NRF) techniques” for measurement/detection of nuclear material. This technique utilizes quasi monochromatic gamma-rays. A nuclide of interest resonantly absorbs photons to be excited and then emits gamma rays to be de-excited. Because nuclear resonance transitions are unique to each nuclide, and the penetration ability of gamma-ray beams is high, this technique would be useful for detection or analysis of specific nuclides even inside shields. Developments have been performed on (1) methodological concept, (2) generation of intense and quasi-monochromatic gamma ray, and (3) simulation tools for NRF and elastic scattering of high-energy photons. An experiment was perfumed to demonstrate detection of a material shielded in an iron container at the NewSUBARU facility of Hyogo University.

The second one is “development of active neutron NDA techniques”, in which four techniques utilizing neutron interrogation methods are being developed: i.e., Differential Die Away Analysis (DDA), Delayed Gamma-ray Analysis (DGA), Neutron Resonance Transmission Analysis (NRTA), and Prompt Gamma-ray Analysis (PGA). These techniques are used to achieve complement information of a sample material. They would be useful for nuclear material accountancy of both low- and high-level radioactive nuclear materials (NMs), and for nuclear security purposes such as detection of NM and explosive materials.

The third one is “Development of broad-area covering rapid nuclear and radioactive material detection technologies”. Developments of passive gamma ray and neutron detection systems has been started: e.g. a portable gamma-ray spectrometer combined with GPS and the other sensors, a compact Compton camera mountable on a drone, and fast neutron detectors. They would be useful for preventing nuclear terrorism in a large-scale event.

INTRODUCTION

Non-destructive Assay (NDA) methods are an efficient and quick way for detection and quantification of nuclear materials (NMs). They are commonly used in accountancy and safeguards together with more accurate destructive analysis (DA) methods. Advancements of NDA technologies are required to apply to measurement of highly radioactive samples, such as spent fuel, samples in a reprocessing plant, and next generation fuels. For nuclear security, portable and fixed NDA devices are used for checking objects and monitoring environments to prevent transportation and to find radioactive and nuclear (R&N) materials. They should be improved to be more efficient and effective.

The Japan atomic energy agency (JAEA) is implementing NDA technological development projects for nuclear detection and measurement. In this paper, three of them will be briefly overviewed. The first two are active NDA techniques: i.e., Nuclear Resonance Fluorescence (NRF), and active neutron NDA techniques. The active method induces nuclear reactions with incident particles (such as photons and neutrons), and measures them to extract information of the sample. These methods are potentially applicable to analysis of high-level radioactive NM samples or to detection of NMs hidden in a shield, which conventional passive method could not measure. The third one is development of passive NDA method aiming for improvement of rapid radioactive and nuclear (R&N) material detection covering broad area in a large event to prevent terrorisms.

DEVELOPMENT OF NUCLEAR RESONANCE FLUORESCENCE (NRF) TECHNIQUES

NRF is a phenomenon in which a nucleus resonantly absorbs gamma rays to be excited and then emit gamma-rays in de-excitation as shown in Fig. 2. Since the structure of the excited states of each nuclide are different, a nuclide can be selectively excited by using well-tuned monochromatic gamma rays. The NRF technique enables us to detect and measure NMs with nondestructive manner using few-MeV gamma rays penetrating sample materials without activation of investigating objects [1-4].

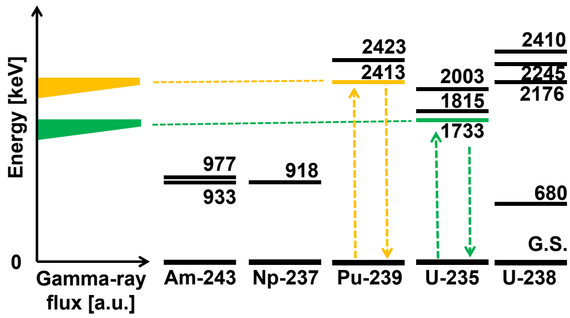

Fig.1: Excited states of nuclear materials and actinides. Using narrowband gamma-rays, a nuclide can selectively be excited, for example the 2.4-MeV gamma ray excites Pu-239.

Such monochromatic gamma-ray beams can be achieved using laser Compton scattering (LCS), in which a laser photon is scattered by accelerated electron beams [5]. The energy of LCS gamma rays is determined by the incident election energy, the photon energy, and the scattering angle. The energy width of the LCS gamma ray beams is determined by the collimation angle of the scattered photons. In order to obtain high-intensity gamma rays, high flux and high-density electron and laser beams are required. The incident rate of LSC is also an important factor to increase the gamma-ray yield.

An LCS system was constructed under a collaboration of KEK (High Energy Accelerator Research Organization), QST (National Institutes for Quantum and Radiological Science and Technology), and ISCN (Integrated Support Center for Nuclear Nonproliferation and Nuclear Security) of JAEA [6]. An energy recovery LINAC (ERL) was installed in KEK Tsukuba campus. The system specifications are 20 MeV in acceleration energy, 162.5 MHz in repetition rate, 2 ps in bunch width, and 10 mA in maximum beam current. A laser system provided photons of 1064 nm in wavelength, 64 μJ in pulse energy, and 5.6 ps in pulse width. A four-mirror optical cavity was used to accumulate laser power to increase interaction. A demonstration experiment of LCS was performed using 57-μA electron beams. At the interaction point, the electron beam was focused to be 30 μm. Approximately 2.6×10^7 photon/s of 6.9-keV X rays were generated [7], showing a potential of more than 10^9 photon/s generation with the maximum current of 10 mA. Next generation gamma-ray sources will provide 10^{13} photon/s of 2-3 MeV gamma rays, using a system, for example, consisting of an electron LINAC of 350 MeV and 13 mA, and a 700-kW laser storage system [5]. Such a system would be useful for various NRF applications.

Simulation studies are required for designing NRF detection/measurement systems. In an NRF measurement, the NM sample is probably in a container with matrix materials. Gamma-ray elastic scattering background should be taken in account for measurement of NRF. It is known that the photon elastic scattering is caused by three different kinds of processes: Rayleigh scattering, nuclear Thomson scattering, and Delbrück scattering. The contribution of Delbrück scattering becomes significant when the photon energy is more than 1 MeV and the scattering material is heavy elements. However, simulation code incorporating NRF and Delbrück scattering did not exist. Code development was carried out based on Geant4 [8], which is a highly versatile toolkit for simulating the passage of particles in objects. Both NRF and photon elastic scattering are now available using Geant4 simulation tools [1,3,9].

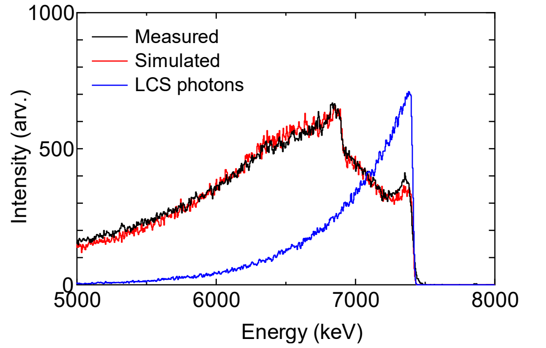

Fig. 2. Incident LCS gamma-ray energy distribution (blue line) was determined by fitting measured response spectrum of NaI detector (black line) and simulated spectrum (read line).

A demonstration experiment of shielded material detection was carried out at the New SUBARU facility of Hyogo University under the collaboration of Hyogo University, QST, and JAEA.

NewSUBARU is an electron storage ring of 0.5-1.5 GeV. LCS gamma-ray beams of 7.4 MeV were generated by the collision of 885-MeV electrons and 2-μm-wavelength laser photons. The gamma-ray beams were guided to an experimental chamber, GACKO, through a 4-mm-diameter collimator. The energy spectrum of the LCS gamma ray was measured by an NaI scintillator (black line of Fig. 2) at GACKO. Analysis of the incident photon energy distribution was carried out so as to reproduce the detector response using the Monte Carlo simulation code EGS-5. Deduced incident gamma ray and simulated response of the detector is given by red and blue

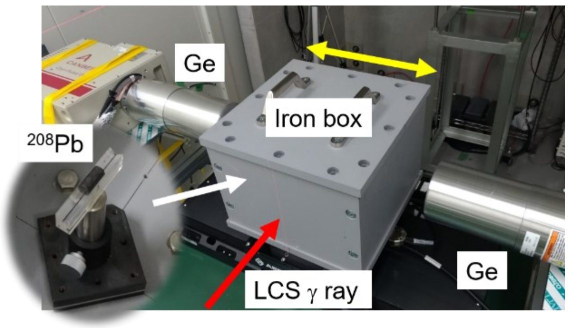

Fig 3. A setup of a demonstration experiment. A cylindrical sample was place in an iron box on a moving stage. The spot size of the LCS gamma ray beams was approximately 4 mm in diameter. NRF gamma rays were measured with HPGe detectors placed on the both side of the box.

lines in Fig. 2, respectively. It was found that the incident gamma-ray beams were 1.5×10^5 photon/s, and the FWHM was about 5%.

In the experiment, a cylindrical Pb-208 sample (8 mm in diameter) was placed in an iron box of about 30 cm. The thickness of the entrance and a side was 1 cm, and that of the back and the other side was 3 cm. Figure 3 shows the box placed on a moving stage. The spot size of the LCS gamma-ray beams was approximately 4 mm in diameter. NRF gamma rays were measured with two 140% HPGe detectors placed on the both side of the box. Figure 4 shows gamma-ray spectra obtained in this experiment. The NRF peak appeared when the LCS gamma-ray beam hit the target, while those peaks disappeared when the box was moved. The count rate of the gamma-ray peak of 3.7 MeV was about 0.035 cps.

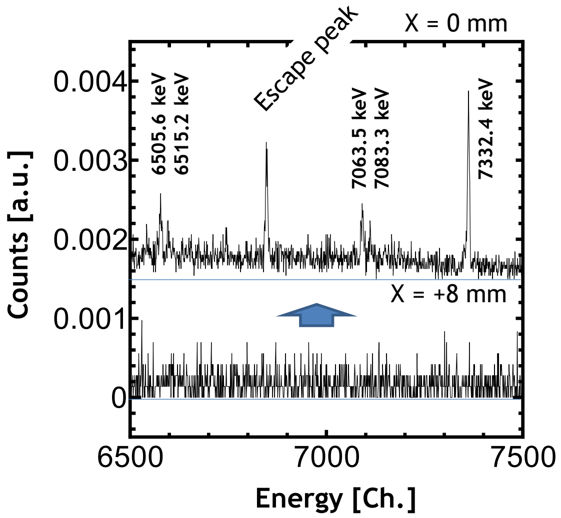

Fig. 4. Spectra measured with LCS gamma beams on the target (top) and off the target (bottom). The blue baselines were moved to show the spectra clearly.

An X-ray imaging system would be useful for finding a suspicious part in a container at a port or harbor. However, distinguishing heavy elements and nuclear materials is difficult without opening to investigate it. NRF technique would be useful to observe inside the cavity penetrating shields. From the result of the demonstration experiment, about 18 cps of NRF signal is expected from a 1-kg U-235 sample with a same detector system placed at 1.5 m distance from the target. An LCS gamma-ray beam of 10^{11} ph/s, and 1% of linewidth is assumed. The difference of reaction probability is also taken into account. The cross section of U-235 is about 3.8×10^{-3} times larger than that of Pb-208. This conclusion agrees with the evaluation given by Negm et al. [10].

DEVELOPMENT OF ACTIVE NEUTRON NDA TECHNIQUES

Development of active neutron NDA techniques started as “Development of neutron resonance densitometry (NRD)” carried out between 2012 to 2014 Japanese Fiscal year (JFY) under the collaboration with NSEC (Nuclear Science and Engineering Centre) and ISCN of JAEA, EC-JRC Geel, and Kyoto university. NRD was proposed as an analyzing method for particle like debris of melted fuel, which has complicated components and shapes, and is highly radioactive. This method is a combination of neutron resonance transmission analysis (NRTA) for NM quantification in a debris sample within few % accuracy, and prompt gamma ray analysis (PGA) or neutron resonance capture analysis (NRCA) for qualification/identification of matrix elements of the sample [6,11-15]. In this research, usage of small electron linac facility for pulsed neutron generation was proposed.

“Development of active neutron NDA techniques” has started since 2015 JFY under the collaboration with NSCE and ISCN of JAEA and EC-JRC Geel and Ispra. This project mainly aims to establishment of a technology of compact active neutron interrogation methods. Four active interrogation methods, differential die away analysis (DDA), delayed gamma ray analysis (DGA), PGA, and NRTA, has been chosen. Basic technological development was conducted in phase I (2015-2017 JPY) using DT and Cf-252 neutron sources. The features of these techniques are given in Table 1. The developments continued as phase II (2018-2021 JPY) aiming mainly for improving the techniques applicable to radioactive sample measurements [16,17].

An integrated interrogation system, named Active-N, is being constructed at NUclear fuel Cycle safety Engineering research Facility (NUCEF) by NSEC of JAEA [16]. The system is equipped with a DT neutron source to provide intense neutrons for three different kinds of measurements: DDA, PGA, and NRTA. The DDA device can be apply for a measurement of fissile material quantification of a sample containing radioactivity in a various object. The system was equipped with a compact NRTA system with a 5-m neutron flight path. The detail of this program will be given by Y. Toh et al. [18,19].

DGA development is being carried out under the collaboration between EC-JRC Ispra and ISCN JAEA [20]. Experiments were performed at EC-JRC Ispra using PUINITA (pulsed neutron interrogation test assembly) equipped with a DT neutron source. A devise developed by JAEA, which consists of a Cf-252 neutron irradiator and a gamma-ray measurement system, was tested at PERLA (performance laboratory) of EC-JRC Ispra. Spectra of neutron induced fission products of U and Pu samples were observed. The detail of the progress will be given by D.C. Rodrigues et al. [20].

Table 1: four neutron interrogation techniques

Techniques	Description	Quantification
DDA	Pulsed neutron interrogation method that measures a time-dependent neutron die-away curve depending on the neutron emission of induced fission reactions.	^{239}Pu-effective mass
PGA	Measurement of prompt gamma rays induced by (n, gamma) and the other reactions. Characteristic gamma rays are used to identify nuclides within the sample.	Existence, qualification, or quantification of specific nuclide
NRTA	Neutron time-of-flight (TOF) measurement, in which nuclide characteristic dips are observed in an energy-dependent transmission spectrum at the nuclear reaction resonance energy.	Quantity of each of U/Pu nuclides
DGA	Measurement of gamma rays in decay of fission products resulting from neutron induced fission reactions	Ratio of ^{235}U/^{239}Pu/^{241}Pu

DDA: differential die away analysis
PGA: prompt gamma ray analysis
NRTA: neutron resonance transmission analysis
DGA: delayed gamma ray analysis

Feasibility study of laser driven neutron source (LDNS) for NRTA application is being carried out under a collaboration between Osaka university, Kyoto university, and ISCN JAEA [20].

LDNS is newly rising neutron generation technology, which can provide intense and short pulse neutron beam. Such a system can be installed in a facility where installation of accelerator is difficult. An evident resonance absorption of a sample was observed in neutron time of flight (TOF) spectrum [21].

DEVELOPMENT OF BROAD-AREA COVERING RAPID NUCLEAR AND RADIOACTIVE MATERIAL DETECTION TECHNOLOGIES

Developments of passive gamma-ray and neutron detection systems are being implemented to prevent nuclear terrorism in a large-scale event as a four-year project (2020-2023 JFY) under the collaborations of ISCN and the Collaborative Laboratories for Advanced Decommissioning Science (CLADS) of JAEA, and Kindai university. The procedure of broad area covering rapid N&R martial detection is as follows: (1) find an increase of radiation field, (2) figure out the source position, and (3) identify the radioisotopes.

An aerial dose rate mapping using Global Positioning System (GPS) would be useful for finding a trace of N&R materials outside. This technique could not be applied to inspection inside of stadiums and buildings, where GPS signals could not be received. Therefore, requirement is incorporating a system with Simultaneously Localization and Mapping (SLAM) system using, for example, Light Detection And Ranging (LiDAR).

When an increase of counting rate is found in an area survey or mapping, the location of radioactivity has to be quickly figured out. In such a case, dose rate mapping procedure is not effective any more. Rapidly the source object has to be figured out. Compton cameras and the other detector techniques would be required for finding the location. A program that predicts the location from the trace of measured count would be useful. Video devices should be combined to record the view of source object or person.

In addition, identification of radioactive isotopes is required to decide the next action. Naturally occurring radioactive material (NORM), medical, and industrial radioisotopes are poetically the source of the alarm. Development of AI software for rapid diagnose of the alarming spectrum would be helpful.

Figure 5 shows relevant technologies to be applied to a broad area monitoring system. The detail of our development project will be given by T. Takahashi et al. [22].

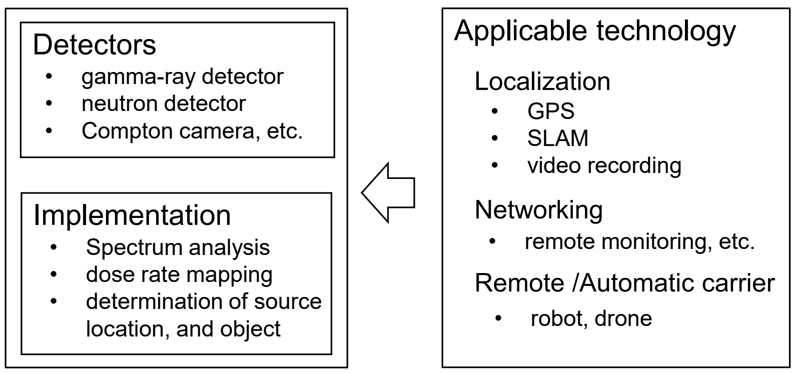

Fig. 5. Image of technological developments of a system incorporating various technologies.

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