Abstract. JAEA-ISCN has been implementing basic development programs of the advanced NDA technologies for nuclear material (NM) since 2011JFY (Japanese Fiscal Year), which are (1) NRF (Nuclear resonance fluorescence) NDA technology using laser Compton scattered (LCS) gamma-rays (intense mono-energetic gamma-rays), (2) Alternative to $^3$He neutron detection technology using ZnS/B$_2$O$_3$ ceramic scintillator, and (3) NRD (Neutron resonance densitometry) using NRTA (Neutron resonance transmission analysis) and NRCA (Neutron resonance capture analysis). These programs are going to be finished in 2014JFY and have demonstration tests in February – March 2015.

1. Introduction

JAEA has been implementing development programs of basic technologies of the following advanced NDA of NM for nuclear safeguards and security since 2011JFY.

(1) NRF (Nuclear resonance fluorescence) NDA technology using laser Compton scattered (LCS) gamma-rays (intense mono-energetic gamma-rays).
(2) Alternative to $^3$He neutron detection technology using B$_2$O$_3$/ZnS ceramic scintillator.
(3) NRD (Neutron resonance densitometry) using NRTA (Neutron resonance transmission analysis) and NRCA (Neutron resonance capture analysis).

The technology (1) is for an NDA system for precise, quantitative, and selective measurement of any isotopes ($^{239}$Pu and any actinide isotopes) using specific NRF reactions of each isotope with an LCS gamma-ray source of mono-energetic and energy tunable gamma-rays with a few MeV energy range based on a superconducting energy recovery linac (ERL). We are going to conduct demonstration of generation of intense mono-energetic gamma-ray at experimental ERL in KEK Tsukuba in the end of 2014JFY (March 2015).

NRF-NDA would be a powerful tool for the precise measurement of nuclear material if an intense, mono-energetic, energy-tunable, 2-5 MeV class gamma-ray source is achieved. By using 2-5 MeV class mono-energetic gamma-rays with deep penetration capability, NRF-NDA could be applied to non-destructive detection of NM hidden behind very thick shielding material, such as in a cargo container, for example.

The technology (2) is for NDA systems that use alternative neutron detectors to count neutrons from NMs instead of $^3$He detectors for covering serious shortage of $^3$He gas. The present JAEA development program (1) covers development of ZnS/B$_2$O$_3$ ceramic scintillator detector module and demonstration of the ZnS/B$_2$O$_3$ ceramic scintillator detectors and NDA systems using them.

The technology (3) is for a precise NDA system based on NRTA, for determining Pu/U isotopic compositions in target by analyzing the transmitted neutrons according to their energies as determined by the time of flight (TOF) method. NRCA is used to determine neutron-absorbing elements, such as $^{10}$B, in the target, by analyzing gamma-rays specific to the isotopes. The TOF method is also used for NRCA. We started this development program for applying accountancy and control of NM of particle-like debris of melted fuel (MF) in the reactors with severe loss-of-coolant accidents since...
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2012JFY based on a collaboration with JRC-IRMM from its beginning. We are going to demonstrate the NRD method using a beam-line in GELINA of JRC-IRMM at the end of 2014JFY (in February - March 2015).

2. NRF NDA Technology using LCS Gamma-rays (Intense Mono-energetic Gamma-rays)
JAEA has been developing the fundamental parts of NRF NDA technology using LCS gamma-rays [1][2]. There are two major parts for this NDA technology: (1) production of high intensity, mono-energetic gamma-ray beams (LCS gamma-ray sources) and (2) uses of the mono-energetic gamma-ray beams to probe nuclear material.

2-1. Principle of NRF NDA Technology using LCS Gamma-rays
Figure 1 shows a schematic view of NRF with mono-energetic gamma-ray beams. If the energy of incident gamma-ray is identical to the energy of excited state populated directly from the ground state of the nucleus of interest, the incident gamma-ray is effectively absorbed by the nucleus and promptly it emits the same energy gamma-ray (this reaction is called as nuclear resonance fluorescence (NRF)). The energies of the states excited by NRF are inherent to the nucleus of interest, as shown in Figure 1. By measuring the energies of NRF gamma-rays, we are able to detect nuclear species (isotopes) of interest and to know their quantities. The quantity of each isotope can be evaluated by analyzing the NRF peak in the measured energy spectrum.

2-2. New LCS Gamma-ray Source based on Energy Recovery Linac (ERL)
Mono-energetic gamma-rays are produced via inverse Compton scattering of laser photons with relativistic electrons. The advantage of the ERL is that each electron bunch is used only once for the scattering with photons, and thus the ERL is free from degradation of the electron beam emittance. One of the ERL projects in the world is the compact ERL (cERL) project at the high energy accelerator research organization (KEK) in Japan [3][4].

The cERL is a test facility to demonstrate the generation of ultra-low emittance electron beams for a future synchrotron light source based on ERL, which has been proposed at KEK. The cERL facility is used also for basic demonstration of the generation of the LCS gamma-ray beams, under collaboration with JAEA (see Figure 2). The demonstration of the basic technology of LCS gamma-ray beam generation is scheduled at the end of 2014 JFY (in March 2015).

2-3. Probing Nuclear Materials by means of Nuclear Resonance Fluorescence (NRF) using an LCS Gamma-ray Source
There are two main measurement methods to perform NRF NDA. The first one is direct measurement of gamma-rays that are scattered from targets by the NRF process. The second method uses nuclear
A. Direct Measurement of NRF Gamma-rays (Scattering Method)
It is possible to distinguish the NRF-scattered photons by using energy-sensitive gamma-ray detectors that surround measurement targets because these photons have the same energy as the photons in the original gamma-ray beam. Figure 3 (left) shows an idea of this method. The direct measurement of NRF gamma-rays is useful for detection of some specific isotopes relevant to nuclear weapons such as $^{235}$U, $^{239}$Pu. For applications to the precise measurement of nuclear material isotopes in very highly radioactive materials such as spent fuel assemblies and/or melted fuel debris, we need to deal with very high gamma-ray background[5] [6].

B. Measurement of Absorption of Interrogating Gamma-rays (Resonance Absorption Method)
The second method is to measure reduction in the LCS gamma-ray beam (caused by NRF reactions in the target) by measuring the corresponding gamma-rays that are scattered by NRF reactions in a well-characterized piece of material containing nucleus of interest (such as fissile isotopes), that is placed into the beam after it has passed through the target (see Figure 3 (right)). (This isotopically well known piece of nuclear material is sometimes called a “witness plate.”) The number of NRF reactions in the witness plate is especially sensitive to the number of resonance-energy gamma-photons remaining in the beam after it has passed through the target.

![Figure 3](image)

Figure 3 Schematic views of measurement methods.
(left: scattering method, right: resonance absorption method)

Therefore, by measuring the NRF-scattered gamma-rays from the witness plate, we can infer the number of NRF reactions that have occurred in the measurement target. Our calculations [7][8] indicate that this second way to do NRF NDA can be very successful with an LCS gamma-ray beam that has a lower energy of around 2 MeV.

2-4. Possible Applications of NRF NDA Technology using LCS Gamma-rays in Nuclear Security and Safeguards

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<th>Basic technology</th>
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<th>Possible applications</th>
<th>Field</th>
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<tr>
<td>NRF NDA with LCS gamma-rays</td>
<td>necessary improvements</td>
<td>Detection of NM in heavy shield in a cargo container</td>
<td>Security</td>
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<td></td>
<td>necessary improvements</td>
<td>Detection of replacement of fuel rods in a spent fuel assembly</td>
<td>Security/ Safeguards</td>
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<td></td>
<td>necessary improvements</td>
<td>Precise measurement of all Pu/U isotopes in a spent fuel assembly</td>
<td>Safeguards (NM Accountancy/ Verification)</td>
</tr>
<tr>
<td></td>
<td>necessary improvements</td>
<td>Precise measurement of all Pu/U isotopes in melted fuel debris</td>
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After we finish the demonstration of LCS gamma-ray beam generation, the next step is to design and demonstrate the final goal LCS gamma-ray source with energy of 2-5 MeV, for use in nuclear security and safeguards. We could summarize the possible applications of NRF NDA with LCS gamma-rays in Table 1. Figure 4(a) shows a cargo container (hidden nuclear material with heavy shield) inspection system and Figure 4(b) an example of precise measurement system of Pu/U in a canister of melted fuel debris.

3. Alternative to $^3$He Neutron Detection Technology, using ZnS/B$_2$O$_3$ Ceramic Scintillator
The serious shortage of $^3$He gas may cause a deterioration of international safeguards and nuclear security, since both deeply depend on $^3$He gas neutron detectors for the detection and counting of neutrons emitted from nuclear material. JAEA has been developing an alternative neutron detection technology using ZnS/B$_2$O$_3$ ceramic scintillator (hereafter, “ceramic scintillator” also used), since 2011 JFY [9][10][11].

3.1 Alternative Neutron Detector Module (ZnS/B$_2$O$_3$ Ceramic Scintillation Detector Module)
Figure 5 shows a photograph of the exterior view of the unit module built on an experimental basis. The detector module is composed mainly of three components: an aluminum square (32 mm x 32 mm) tube, a rectangular ZnS/B$_2$O$_3$ ceramic scintillator sheet (45 mm x 250 mm) and two PMTs. The scintillator sheet is fit on the diagonal inside the square tube, while the two PMTs are installed at both ends of the tube. Nuclear reactions, between the neutrons that enter the scintillator from outside the tube and the $^{10}$B atoms in the scintillator, induce the emission of scintillation light from the surface of the scintillator. The light photons are divided into two directions, towards both ends of the tube, and are detected individually by the two PMTs as two pulse signals. These two pulse signals are processed by the specially designed electronics circuit that enables to detect neutron signals. The light signals detected with the PMTs are amplified with pre-amplifiers. Then, the amplified signals are discriminated with certain levels.
The neutron signal is finally detected according to the coincidence of the two light signals from the two PMTs, to eliminate the electrical noise signal which arises from various causes.

3-2. Alternative NDA Systems using the ZnS/B₂O₃ Ceramic Scintillation Detector Modules
On the basis of the current specifications and performance characteristics of the ceramic scintillator detector, two novel types of safeguards NDA systems equipped with the ceramic scintillator detectors, have been designed. One of them is a small size NDA system with references of INVS. Figure 6 shows schematic views of the basic design of the Alternative-INVS (ASAS).

3-3. Performance (Demonstration) Tests
In the beginning of March 2015, we are going to conduct performance tests of (1) the ZnS/B₂O₃ ceramic scintillation detector module (detection efficiency of neutrons / gamma-ray sensitivity) and (2) the ASAS by comparisons with the INVS (He-3) using the same MOX objects at TRP-PCDF.

4. NRD using NRTA and NRCA
(For precise quantification of Pu/U in particle-like melted fuel debris)
JAEA has proposed a system of neutron resonance densitometry (NRD) for the quantification of NMs in particle samples in thin disk-type containers. The measurement samples can be composed of any unknown materials such as particle-like debris of MF in reactors that had been produced by severe accidents. (Hereafter, “measurement samples” means “MF debris”.) NRD is the combination of neutron resonance transmission analysis (NRTA) and neutron resonance capture analysis (NRCA) for the quantification of NM in samples (measurement targets) (see Figure 7) [12] [13] [14] [15].

4-1. Particle-like MF Debris in thin Disk-type Containers as Measurement Objects of NRD
It is said that particle-like debris of MF is formed not only at rapid cooling of very high temperature
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MF by water but also at breaking rock of MF into small size chunks which is shown in Figure 8. The particle-like MF debris could be collected with pumping up with water and could be classified into sizes by sieves. The same size particle-like debris is mixed into large sizes, which are shown in Figure 8 and 9. After taking samples for NRCA, the particle-like MF debris is put into thin disk-type containers for measurement by NRTA.

4-2. NRTA for Quantification of NM in Debris

In NRTA, neutron transmission is measured as a function of neutron energy with a time-of-flight (TOF) technique. Uranium and plutonium isotopes resonantly absorb neutrons in the energy range of 0-30 eV, as shown in Figure 10. It is possible to analyze the quantities of Pu and U isotopes using characteristic neutron absorption peaks in Figure 10. The measurements are not disturbed by the strong gamma-ray radiation from MF.

4-3. NRCA for Quantification of Impurities in Measurement Samples

Particle-like debris is a mixture of the fuel, rod cladding, concrete and other construction materials. Consequently, unknown quantities of isotopes would be included in it. Among these isotopes, $^{10}$B is considered to have the strongest influence on NRTA measurements. In the nuclear reactor, boron was used in control rods, and boric acid was injected to prevent nuclear reaction at the time of the accident. In order to evaluate the quantities of such isotopes, we have introduced the technique of neutron resonance capture analysis (NRCA) by means of high energy-resolution gamma-ray detectors. However, the measurement of the 478-keV gamma rays of $^{10}$B is strongly affected by the Compton edge of the 661.5-keV gamma-rays of $^{137}$Cs, which appears at 477.2 keV. To reduce the Compton edge, we are developing a well-type LaBr3 detector (see Figure 11) [16][17].

4-4. Estimated Precision of NRD based on Experiments at GELINA of JRC-IRMM

JAEA and JRC-IRMM have been studying on precision of NRD based on simulated experiments at GELINA of JRC-IRMM resulting that the total precision of NRD is about 3% for quantification of amount of Pu/U in particle like debris, removed from accident reactors, in a thin (~2 cm) disk-type container. Statistical uncertainty less than 1% is achieved for all important U and Pu isotopes with 20 min measurement using $10^{12}$/sec pulsed beam source as shown in Figure 12. The other systematic uncertainty is about 2%.

4-4. Demonstration of NRD Detectors at GELINA of JRC-IRMM

JAEA and JRC-IRMM are going to demonstrate performance of the NRD method at GELINA in March 2015 (see Figure 13).

4-5. Possible Applications of the NRD in Nuclear Safeguards and Security

We started the basic technology development of NRD for application to accountancy of NM of particle-like MF debris (The expected actual system for this purpose is shown in Figure 14).
This technique could be applied to detect and measure nuclear material in other objects, such as safeguards samples taken at nuclear fuel cycle facilities, for verification of the isotopic composition (and also quantity). Possible applications of NRD in the fields of nuclear safeguards and security are shown in Table 2.

Table 2 Possible applications of the NRD in nuclear safeguards and security

<table>
<thead>
<tr>
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</tr>
</thead>
<tbody>
<tr>
<td>Basic technology of NRD</td>
<td>necessary improvements</td>
<td>Precise quantity and Pu/U isotopic compositions measurement (NDA) of samples in nuclear fuel cycle facilities</td>
<td>Safeguards</td>
</tr>
<tr>
<td></td>
<td>necessary improvements</td>
<td>Precise measurement (NDA) of all Pu/U isotopes in particle-like MF debris for material accountancy</td>
<td></td>
</tr>
<tr>
<td></td>
<td>necessary improvements</td>
<td>NDA of all Pu/U isotopes in unknown mixtures with other material</td>
<td>Security</td>
</tr>
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References