Development of An Integrated Active Neutron Non-Destructive Analysis System: Active-N

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ABSTRUCT

Since neutrons have exceptional ability to penetrate high-density materials and can induce fission, they are used in non-destructive analysis such as, Differential Die-Away Analysis (DDA), Prompt Gamma-ray Analysis (PGA), Neutron Resonance Capture Analysis (NRCA), Neutron Resonance Transmission Analysis (NRTA) and Delayed Gamma-ray Analysis (DGA). The different analytical methods give us complementary information, which are particularly useful for the quantification of Special Nuclear Materials (SNM) and Minor Actinides (MA) in highly radioactive nuclear materials, including spent fuel and MA transmutation fuel. The Japan Atomic Energy Agency (JAEA) and the Joint Research Centre (JRC) of the European Commission are collaborating to develop an active neutron NDA system for nuclear non-proliferation and nuclear security. In the second phase of the project, an integrated active neutron NDA system, Active-N, which enables the measurements of DDA, PGA and NRTA (and NRCA) has been developed at NUclear fuel Cycle safety Engineering research Facility (NUCEF) in the JAEA. The DDA detects fission neutrons, and it can determine very small amounts of the fissile mass, such as U-235 and Pu-239. PGA is well acknowledged to be especially valuable for the measurement of light elements such as H, B, N, S, and Cl, as well as Cd, Gd, Sm, and Hg which have large neutron capture cross sections. Therefore, PGA is utilized for the quantification of neutron absorber and particularly useful for the detection of explosives because the most typical high explosive materials contain nitrogen. NRTA can determine isotopic composition by relating neutron resonance absorption and/or scattering. Therefore, Active-N can be used to quantify almost all medium and high-Z elements and considered as one of the most accurate NDA to quantify the amount of SNM and MA. In this presentation, we will provide an overview of Active-N and report the recent experimental results at NUCEF.

Keywords: active neutron NDA techniques, differential die-away analysis, neutron resonance transmission analysis, prompt gamma-ray analysis, neutron resonance transmission analysis, D-T pulsed neutron source

INTRODUCTION

Neutron is a powerful tool in the non-destructive analysis (NDA), since it has exceptional ability to penetrate high-density materials, and can induce fission and radiative capture. Nuclear material accountancy (NMA) is important for nuclear safeguards and security. Nevertheless, to the best of our knowledge, NDA that allows us to accurately quantify the amount of Special Nuclear Materials (SNM) and Minor Actinides (MA) in highly radioactive nuclear materials has not been established so far.

The Japan Atomic Energy Agency (JAEA) and U.S. Department of Energy (DOE) provided recommendations for measurement systems of fuel debris in Fukushima Daiichi nuclear power plant [1-3]. Neutron Resonance Densitometry (NRD) was developed in collaboration Action Sheet-1 (2012-2015) of the Joint Research Centre of the European Commission (EC-JRC) and JAEA. NRD is promising and enable us to quantify the amounts of nuclear materials in small debris [4-7]. However, it is only applied for a thin sample, and hard to measure a thick sample.

Action Sheet-7 (AS-7) between EC-JRC and JAEA for joint collaboration on development of NDA techniques started in 2015 [8-10]. AS-7 aims to study the active neutron NDA techniques and develop new active neutron NDA system using a D-T pulsed neutron source for nuclear non-proliferation and nuclear security (Fig.1). Differential Die-Away Analysis (DDA) can quantify a very small amount of the fissile materials [10]. Neutron Resonance Transmission Analysis (NRTA) is known as one of the most accurate NDA to quantify the SNM and MA [11,12]. It measures the energies of neutron resonances to identify nuclides by the time-of-flight technique, and can quantify almost all medium and high-Z elements. Prompt Gamma-ray Analysis (PGA) is one of the most efficient and useful NDA. It measures neutron capture gamma rays, which are characteristic of each individual nuclide [13,14]. These provide the means to identify and quantify the elemental composition of a sample, especially boron (as a neutron poison) and nitrogen (as an explosive). Nuclear fission reaction releases several neutrons and fission products. Delayed Gamma-ray Analysis (DGA) measures delayed gamma-rays emitted from the fission products. The intensities of individual gamma-ray peaks in the DGA spectra give us information about the ^{235}U/^{239}Pu and/or ^{241}Pu/^{239}Pu ratios [15]. Thus, these techniques give complementary results which is indispensable for quantification of SNM and MA in highly radioactive nuclear materials. In 2017, we developed an integrated NDA system at NUclear fuel Cycle safety Engineering research Facility (NUCEF) in JAEA, which can measure DDA and PGA. Since firstly we have to ensure the performance of DDA, the system is optimized for the DDA measurement.

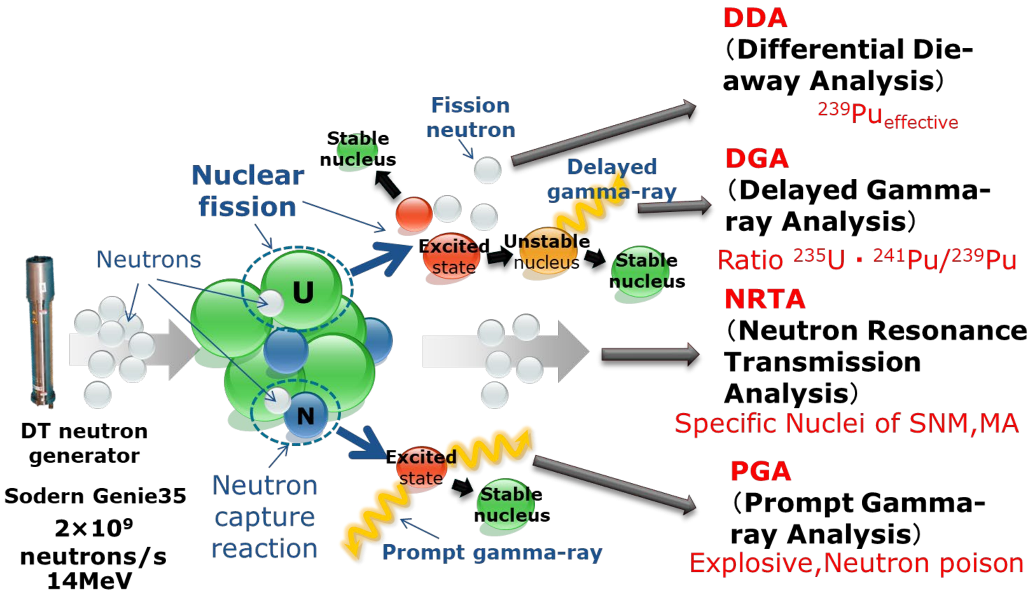

Figure 1. Active neutron NDA techniques. Differential Die-Away Analysis (DDA), Prompt Gamma-ray Analysis (PGA), Neutron Resonance Capture Analysis (NRCA), Neutron Resonance Transmission Analysis (NRTA) and Delayed Gamma-ray Analysis (DGA)

DEVELOPMENT OF NEW Active-N (DDA, PGA and NRTA systems)

In 2018, the second phase has started and aims to develop the active neutron NDA system for highly radioactive materials, such as spent nuclear fuel and highly radioactive nuclear waste [16-19] (See Fig.2). In this phase, further research is required to improve the methodologies and develop a new integrated NDA system which consists of NRTA as well as DDA and PGA. As a first step toward the development of new Active-N, we designed the new Active-N system with Monte Carlo simulations codes such as PHITS [20], MVP [21] and MCNP [22]. In 2019, the DDA and PGA system was deployed at NUCEF in the JAEA Tokai-site. After that, the NRTA system was installed in 2020. Upper right of figure 2 also shows appearance of the new Active-N, which consists of three individual NDA measurement systems (DDA, PGA and NRTA systems). A D-T neutron generator Sodern Genie35 was adopted as the interrogation pulsed neutron source of new Active-N, which can produce 14 MeV neutrons of 1.0×10^9 n/sec. The width of the pulsed neutron and the repetition rate are 10 μs and 100 Hz, respectively. The D-T neutron generator is mounted in the side wall of the DDA and PGA systems. High-density polyethylene including boron can usually be used as an ideal neutron shield since it can effectively suppress cold, thermal and epithermal neutrons. However, since boron emits 478 keV gamma rays in the ^{10}B (n,α) ^{7}Li reactions, the PGA measurement is interfered with by the gamma rays. Although the abundance of Li-6 is only 7.5%, Li-6 has large cross section (941 barns) of the ^{6}Li(n,α)^{3}H reaction for thermal neutrons. Moreover, Li-6 emits no gamma rays in the reaction. We, therefore, adopted the high-density polyethylene including Li instead of boron as the neutron shields of new Active-N.

NRTA system of new Active-N requires rather low energy neutrons (< 1 keV) since they are suitable for applying the time-of-flight method with short flight path length. A neutron moderator is, therefore, important for NRTA measurements and has been investigated by simulation, especially moderator shape and thickness dependences of the neutron beam profile.

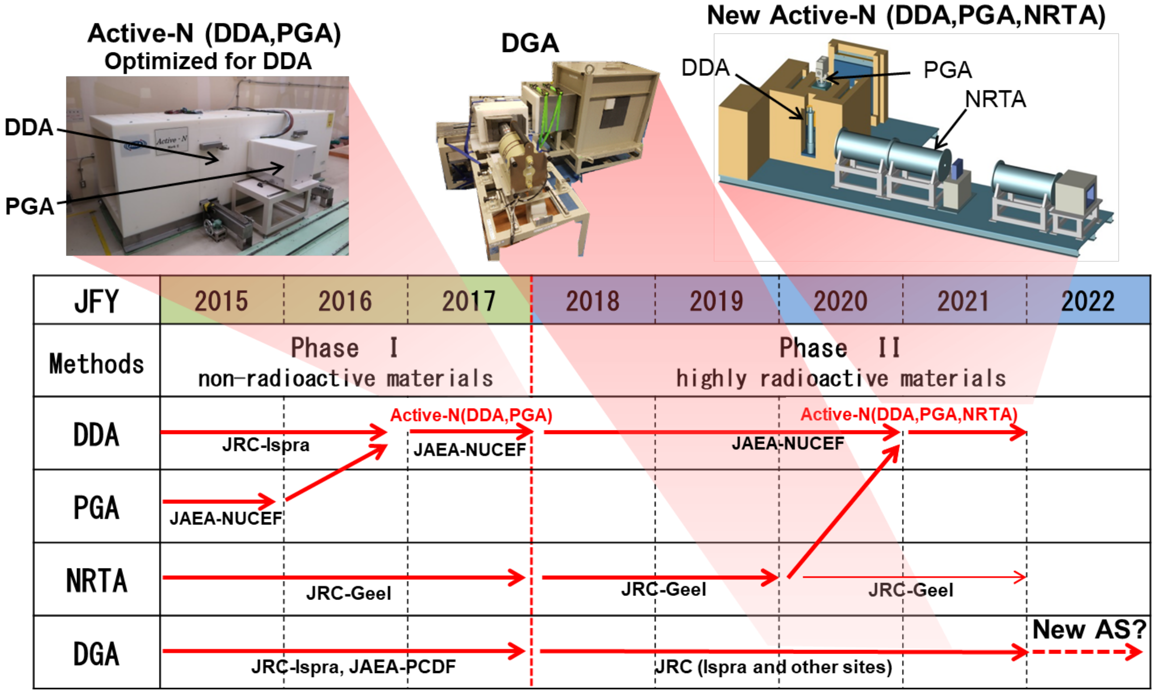

Figure 2. Research timeline of EC-JRC – JAEA collaboration.

Neutron absorbers of Cd and B are used to improve the time response of the neutron detector bank in DDA system of the present Active-N [23]. To make DDA and PGA compatible in new Active-N, Cd and B are not preferred to be adopted in the detector bank and neutron shields, since they emit so many gamma rays in their neutron capture reactions. Therefore, we have developed a new detector bank of DDA system for the new Active-N, which consists of the high-density polyethylene including Li. Lead is also mounted in the sample cavity of DDA and PGA to reduce background gamma-rays from the highly radioactive nuclear materials and the neutron capture reactions, and also behave as the neutron reflector to increase the neutron flux. The DDA system of the old Active-N utilizes only one detector bank composed of He-3 detectors. We have adopted B-10 detectors in new Active-N, since although the B-10 detector has lower neutron sensitivity than the He-3 detector, it can endure much higher intensity of the gamma radiation. New Active-N is equipped with three B-10 detector banks on three sides in order to achieve a large solid angle and high neutron detection efficiency. The PGA system consists of the Coaxial Ge detector and BGO Compton suppressor which is used to suppress the background gamma rays. We have developed a new integrated data acquisition system which can handle all signals obtained from DDA, PGA and NRTA systems.

Figure 3 shows the layout and photographs of the new Active-N, which is installed next to the old Active-N in NUCEF. The length, width and height of DDA and PGA systems of the new Active-N are approximately 1.3m, 1.7m and 2m. The size of sample cavity is a length of 700mm, a width of 700mm and a height of 950mm. The flight path of the NRTA system has two options: the long flight path of 5 m (low statistics high resolution) and the short flight path of 3 m (high statistics low resolution). These options can be selected by relocation of the flight path tubes.

It was experimentally confirmed that the dose rate on the outside wall of the experimental room was low enough for radiation safety. After that, we have begun experimental studies with the new Active-N. First of all, primary tests of the detector bank of the DDA system

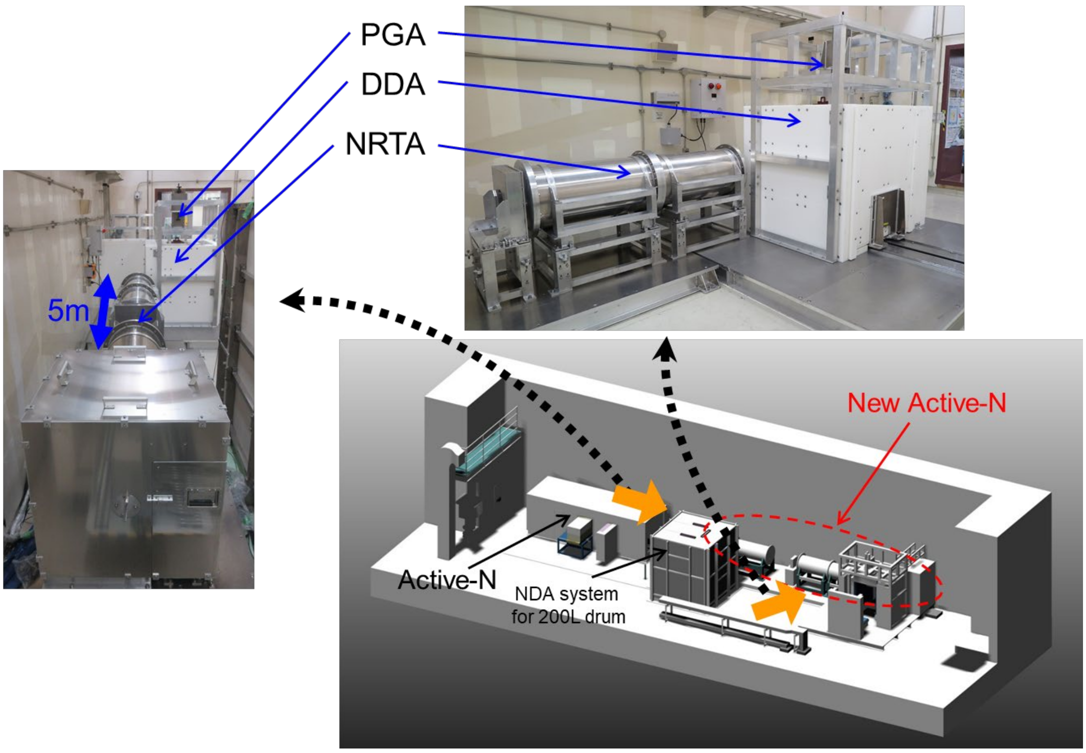

Figure 3. The location and photographs of the new Active-N.

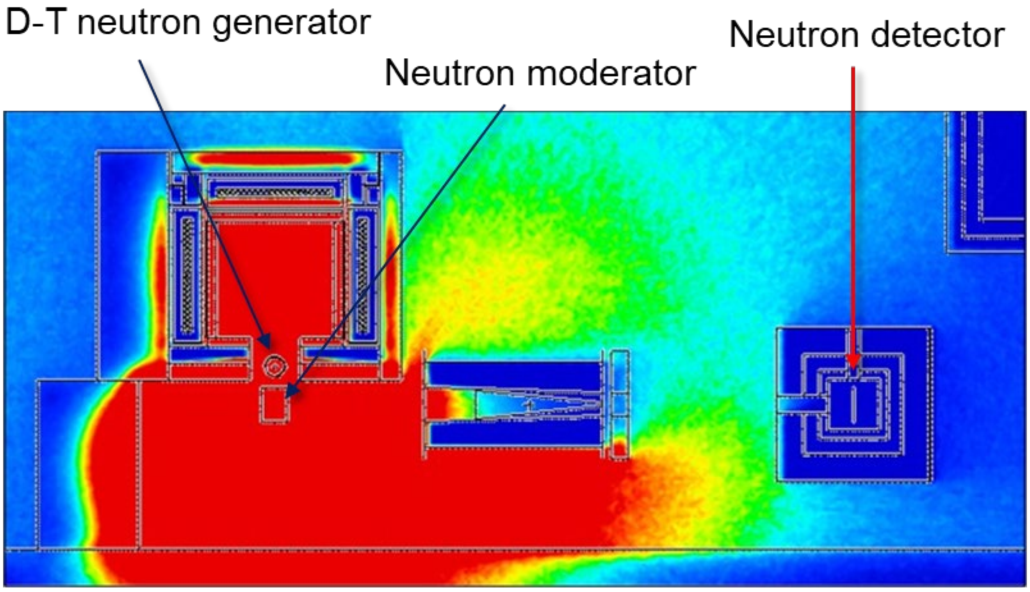

Figure 4. The simulation results of the neutron detector test measurements of DDA by the new Active-N: Blank (black line) and Pu 1g (red line).

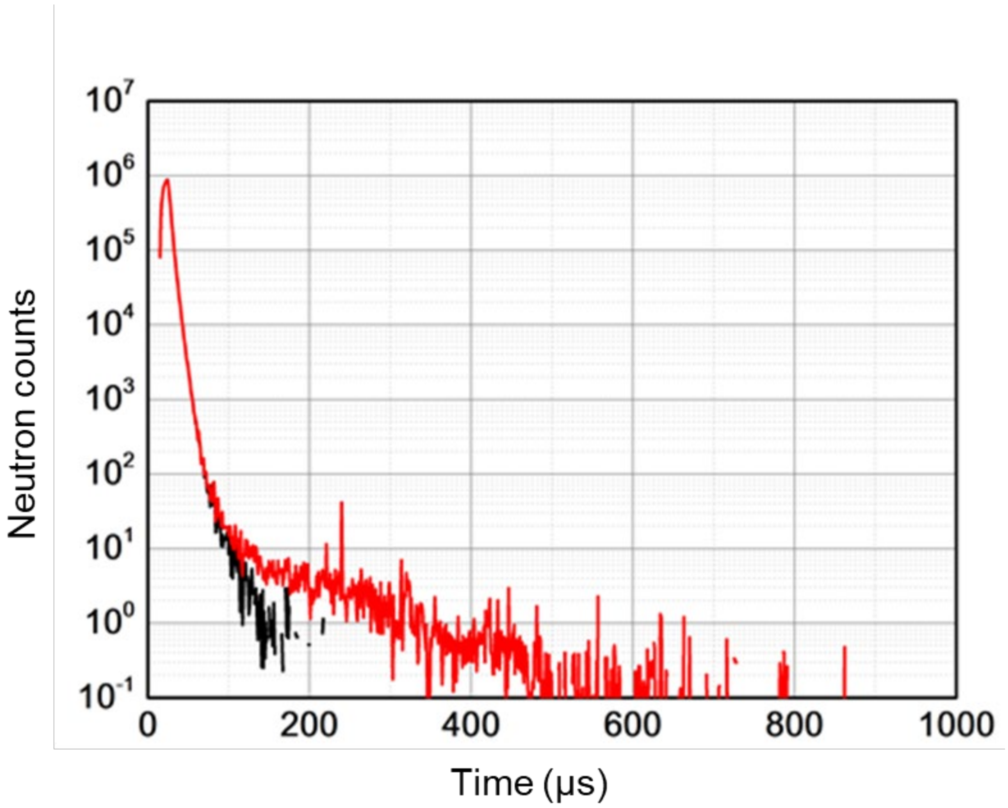

Figure 5. The simulation results of the scattered neutron distribution of around 1eV between 200-300 μs after the trigger pulse of the D-T neutron source.

were carried out. As we mentioned above, the DDA system of the new Active-N has three neutron detector banks. Each bank is mounted twenty B-10 detectors. Total sixty B-10 detectors were examined their ability to detect a neutron with D-T and ^{252}Cf neutron source, and exhibited the expected performance. Figure 4 shows an example of simulation results of the neutron detector test measurements of DDA. The short flight path of NRTA is highly affected by a background neutron because of the short distance from the D-T neutron source.

The results of Monte Carlo simulations for the short flight path show that the scattered neutrons with the wall of experimental room are the origin of the main neutron background (See Fig. 5). The experimental results also supported the simulation results. Additional neutron shields were adopted to decrease the neutron background. Consequently, the neutron background efficiently decreased low enough for NRTA measurements (See Fig. 6).

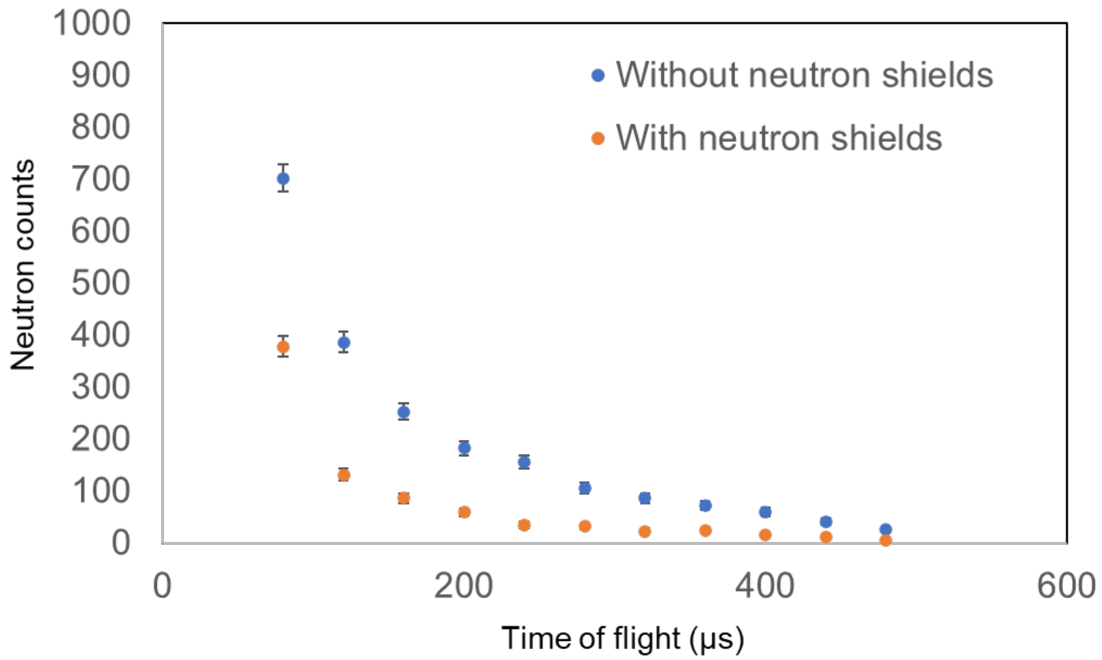

Figure 6. The experimental results of the effect of the additional neutron shields for NRTA.

CONCLUSIONS

The Japan Atomic Energy Agency (JAEA) and the Joint Research Centre (JRC) of the European Commission are collaborating to develop an active neutron NDA system for nuclear non-proliferation and nuclear security. The second phase of AS-7 for the joint collaboration launched in 2018. It aims to establish the active neutron NDA system for the quantification of SNM and MA in highly radioactive nuclear materials, such as spent fuel, MA transmutation fuel, nuclear fuel debris. Four active neutron NDA techniques, namely DDA, PGA(NRCA), NRTA and DGA have been studied and improved. The new Active-N, which consists of DDA, PGA and NRTA systems, was developed and installed at NUCEF in the JAEA Tokai-site. The high-density polyethylene including Li-6 instead of boron and lead is adopted as the shields of the new Active-N to reduce background neutrons and gamma-rays. We have begun experimental studies with the new Active-N in 2021. The primary tests of the new Active-N system were carried out and obtained expected performance.

ACKNOWLEDGMENTS

This research was implemented under the subsidy for nuclear security promotion of MEXT: Ministry of Education, Culture, Sports, Science and Technology.

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