論文の内容の要旨

Thesis Summary

論文題目 Study on Melted Fuel Debris Material Analysis by Portable X-band Linac Neutron Source

(可搬型 X バンドライナック中性子源による溶融燃料デブリ成分分析の研究)

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This research focused on short-distance neutron time-of-flight (TOF) measurement system for limited neutron resonance transmission analysis (NRTA) in nuclear fuel debris at Fukushima Daiichi nuclear reactor core. After the accident, as part of the decommissioning project, removal of nuclear fuel debris is necessary as they are subject of safeguards and criticality safety. From the investigations and work regarding the current condition of Fukushima Daiichi reactor core unit 1, 2, and 3, the strategy to retrieve the nuclear fuel debris will be accessing it from the side through the leaking point of the PCV in a condition with minimum water amount. According to the roadmap for Fukushima Daiichi reactor fuel retrieval by Nuclear Damage Compensation and Decommissioning Facilitation Corporation (NDF) and Ministry of Economy, Trade and Industry (METI), nuclear fuel debris trial extraction from unit 1 and 2 is scheduled to be started from 2021.

Before a thorough debris removal plan is made, the condition of the nuclear debris distribution inside the PCV must be investigated through nuclear debris activity mapping and debris storage criticality control, which is a collaboration project between JAEA, The University of Tokyo, The University of Sheffield and The University of Bristol. The data for the mapping is gathered from on-site nuclear debris screening activity, consisting of two different methods: polychromatic X-ray CT and short-distance TOF NRTA. The latter is a method to identify isotopes inside a material using the compact, mobile version of pulsed neutron source, that complements the former in terms of confirming the existence of uranium and/or plutonium contained in nuclear debris. While measurement using polychromatic X-ray CT can differ materials with large gap atomic number, it is unable to identify between uranium and plutonium which have very close atomic number. Identify these two elements is important because plutonium's microscopic cross-section is 200 times bigger than uranium, leading it to reach critical condition faster. This aspect of U/Pu detection can be covered by NRTA method, which is able to detect isotopes contained in a material through observation of neutron absorption energy that is unique to each isotope. The flow of on-site debris screening is shown in figure 1.

As the current available neutron source capable to perform NRTA are all the stationary type at relatively far nuclear facilities from Fukushima Daiichi, the need for a mobile pulsed neutron source to increase the efficiency of nuclear debris analysis activity arise. Therefore, the focus of this research is to develop a short-distance neutron TOF NRTA that is capable to identify isotopes of uranium and plutonium. The short distance is the most necessary feature since the total of the system should be compact and mobile for on-site use, particularly around the Fukushima Daiichi nuclear reactor area. Approach used by means to achieve this objective is through the utilization of low-energy electron linac-based neutron source as the pulsed neutron source necessary for neutron TOF measurement. Pulsed neutron source system is usually consisted of pulse electron accelerator coupled with material that can emits neutron when hit by fast electrons. In order to make a compact and portable pulsed neutron source is constructed from 3.95 MeV X-band electron linac, with tungsten and copper layer as electron-to-photon converter to generate Bremsstrahlung X-ray, and beryllium as photon-to-neutron converter to generate neutron.



+ Information on size (1 mm accuracy) and weight (100 mg accuracy)

Figure 1: Schematic of on-site nuclear debris screening activity flow according to combined detection system using polychromatic X-ray CT and NRTA.

Beryllium has low photonuclear energy threshold of 1.67 MeV that makes it possible to generate neutron from photonuclear reaction with 3.95 MeV bremsstrahlung X-ray energy, and is easier to handle when it comes to compact and mobile use compared to D_2O . Monte Carlo simulation result of this neutron source shows that it can generate pulsed neutron with flux value of 3.86×10^5 n/cm²/sec, which rounds it up to around 10^7 n/sec. This flux number is enough to perform neutron TOF measurement in short distance in regards to its compact size and mobility. Schematic of compact 3.95 MeV X-band electron linac-based neutron source is shown in figure 2, with total size for the system is estimated to be below 5 m², including the TOF path. Aside from its compact size, the advantage of this neutron source is its short pulse that is generated by X-band type electron linac with the highest RF frequency, resulting in nanosecond pulse as short as 500 ns. Short pulse is beneficial in detecting isotopes with higher neutron energy, because of its lower value of measurement uncertainty.

The detailed experimental setup for measuring neutron TOF can be seen on figure 3. It is divided into 3 main parts: neutron source part, signal amplifier part, and digital signal processing part. In neutron source part, X-band electron linac-based neutron source emits pulsed neutron. The neutron absorption process happened as the neutron passed through the sample material, which later will be translated as resonance peak in the plotted neutron TOF spectrum. The transmitted neutron is detected by the ³He neutron detector, and the analog signal from detector is transmitted to preamplifier to convert the current signal into voltage signal. It also provides an optimized coupling between the output of the detector and the rest of the counting system to minimize any sources of noise that may change the signal. Later, amplifier will convert the signal at the output of the preamplifier into a form suitable for the measurement desired, in other words, shaping the signal. For the case of 3.95 MeV X-ray source, the gamma burst from it will create a long dead time for the detector. Therefore, bipolar shaping is better than unipolar because it can quickly recover the signal baseline to zero, getting the measurement starts back again in short time. The amplified signal is sent to digital signal processor which consists of comparator, voltage divider, and field-programmable gate array (FPGA). Comparator converts the sinusoidal signal into square-shaped, and voltage divider match the voltage between the signal of accelerator and detector. Lastly, FPGA calculate neutron TOF and record it. The recorded TOF data is later converted to energy axis and plotted into neutron energy spectrum, where the resonance peak can be observed in order to identify the isotopes contained in the sample material.



Figure 2: Schematic of the compact X-band electron linac-based neutron source.



Figure 3: Neutron energy spectrum obtained from 3.95 MeV X-band electron linac-based neutron source.

The first experiment is to measure the neutron energy range of this compact neutron source. By using TOF path of 2.5 meters, 2.5 microseconds pulse width, 2 cm polyethylene neutron moderator and 1-hour measurement time, 3.95 MeV X-band electron linac-based neutron source can generate neutron energy spectrum up to 100 eV, with the NRTA resonance peak observation area at 1-50 eV. It has been planned that the number of isotopes this system can measure will be limited due to its short range of neutron energy spectrum. And by using this experiment result, the isotopes targeted for the measurement by short-distance NRTA are decided to be ²³⁵U, ²³⁸U, ²³⁹Pu, ²⁴⁰Pu and ²⁴²Pu. A Monte Carlo simulation of the NRTA of

uranium and plutonium mix sample using this compact neutron source system shows that the resonance peaks of these isotopes are observable. The plot of the simulation result can be seen in figure 4.



NRTA simulation spectrum of Uranium-Plutonium mix sample

Figure 4: NRTA simulation result for a material with different contents. The resonance peaks represent specific isotopes.

Based on that result, short-distance NRTA experiments using 3.95 MeV X-band electron linac-based neutron source are conducted with non-radioactive materials as dummy sample, which isotopes having the similar neutron energy absorption value as the selected U/Pu isotopes. These samples are sheet-shaped with dimension of $20 \times 80 \times 0.3 \text{ mm}^3$. The first experiment using indium sample which isotope (¹¹⁵In) simulates ²⁴⁰Pu shows a clear resonance peak at 1.5 eV in neutron energy spectrum for only 1 hour of measurement. This is consistent with ¹¹⁵In neutron absorption energy reference from JENDL. The similar result was found in the next experiment using tungsten sample, which isotopes simulate ²³⁸U. In the experiment with mixed indium and tungsten sample with dimension of $20 \times 80 \times 0.6 \text{ mm}^3$, multiple different isotopes can be simultaneously observed, as shown in figure 5. Experiments with different sample size confirmed that the smallest sample size this system can measure is $20 \times 20 \times 1.2 \text{ mm}^3$ with 2 hours of measurement time, shown in figure 6. It proves that this system is feasible to detect uranium and plutonium inside nuclear debris, through the observation of neutron energy absorption by limited U/Pu isotopes of ²³⁵U, ²³⁸U, ²³⁹Pu, ²⁴⁰Pu and ²⁴²Pu.

The main objective of this research is to make a meaningful contribution to criticality calculations for nuclear debris containers, via the determination of uranium and plutonium material density. In the practical application of nuclear debris extraction and U/Pu identification, due to its limited sample shape for measurement and long measuring time, this short-distance NRTA will be used at the test extraction step for the purpose of system calibration. Rapid line detector system of X-ray CT will be used to estimate the size and weight of heavy atomic number material content in the debris, which is necessary for material density calculation. After that, the existence of uranium and/or plutonium will be confirmed through short-distance NRTA method, to correct the X-ray screening result. The material density calculation result can estimate the concentration of U/Pu content inside the corresponding nuclear debris, which will determine the category of nuclear debris storage type to prevent criticality of the storage cans and canisters.



Figure 5: Neutron energy spectrum obtained from 1-hour measurement of indium and tungsten combined sample, using ³He neutron detector for 20×80×0.6 mm³ sample size.



Figure 6: Neutron energy spectrum obtained from 2-hour measurement of indium and tungsten combined sample, using ³He neutron detector for 20×20×1.2 mm³ sample size.

With its compact size and short TOF path, this advantage will make the system able to be implemented in on-site nuclear debris screening system and perform isotope identification system at Fukushima Daiichi nuclear reactor, as a complementary to increase the accuracy of polychromatic X-ray CT imaging method. Combination of these two measurement methods will be used to obtain nuclear debris composition data necessary for nuclear debris storage criticality control of Fukushima Daiichi reactor core area, which will later contribute to nuclear debris activity mapping.

Future developments of short-distance NRTA system with compact neutron source include measurement with realistic model of nuclear fuel debris, as well as optimization of experiment parameters/settings. In effort to increase the neutron intensity, a possibility of using 30 MeV X-band linac is considered too, as it could improve the neutron intensity by four order $(3.86 \times 10^7 \text{ to } 8 \times 10^{11} \text{ n/s})$, enabling NRTA measurement to be done within order of ten seconds.