Proposal of Neutron Resonance Densitometry for Particle Like Debris of Melted Fuel using NRTA and NRCA

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Abstract:

Neutron resonance densitometry (NRD) has been proposed to quantify nuclear materials in particle-like debris of melted fuel formed in a severe accident of nuclear reactors such as the Fukushima Daiichi nuclear power plants. NRD is a method that combines NRTA (neutron resonance transmission analysis) and NRCA (neutron resonance capture analysis) using a pulsed neutron generator and the TOF (time of flight) technique. NRTA is used to quantify the amount of Pu and U isotopes. NRCA is used to identify matrix materials, such as B and Fe, which are present in the melted fuel. A special gamma-ray spectrometer has been designed to apply NRCA in the presence of highly radioactive materials. The applicability of the NRD method has been studied using Monte Carlo simulations and neutron TOF experiments at the GELINA facility of the EC-JRC-IRMM. We conclude that NRD has a potential to determine the quantities of Pu and U isotopes in particle-like debris of melted fuel with counting statistics uncertainties less than 1%, even in the presence of 2.5 w% ^{nat}B and 9 w% ^{56}Fe.

Keywords: neutron resonance densitometry; neutron time-of-flight; melted fuel; debris; NRTA; NRCA

1. Introduction

An innovative non-destructive analysis technique, referred to as neutron resonance densitometry (NRD), is presented in this paper. This method was proposed to quantify nuclear materials in particle-like debris of melted fuel formed in a severe accident of nuclear reactors such as the Fukushima Daiichi nuclear power plants. NRD is a method combining NRTA (neutron resonance transmission analysis) and NRCA (neutron resonance capture analysis).

The NRTA technique was applied in the past to quantify nuclear materials in irradiated nuclear fuel pellets by Priesmeyer and Harz [1] using a fast chopper time-of-flight (TOF) spectrometer and by Behrens *et al* [2] using a pulsed neutron TOF spectrometer. Recently, Sterbentz and Chichester have proposed NRTA as a non-destructive assay technique for the next generation safeguards initiative's plutonium assay challenge [3].

Behrens *et al* [2] have analysed neutron transmission spectra covering a neutron energy range between 1 and 40 eV. The abundances of $^{234-236}U$ and $^{238-242}Pu$ isotopes relative to ^{238}U were deduced with uncertainties ranging from better than 1 % for ^{235}U to 20 % for ^{238}Pu. In the analysis, they have taken into account 16 nuclides (11 actinides and 5 fission products) with a resonance structured cross section and oxygen as matrix of the sample and the steel end cap. Both, the oxygen and steel, have a slowly varying cross section without resonance in this energy region.

On the contrary, a detailed elemental and isotopic composition of melted debris is not known. Very likely, the samples will contain water, boron, structural materials, and concrete materials. However, no information about their relative amounts is available. It is important to identify which components are included in the melted debris in advance of the NRTA measurement to optimize the analysis of the data. NRCA is used for identifying possible contaminants, such as ^{10}B and ^{56}Fe. A well-type gamma ray spectrometer made of a LaBr$_3$ scintillation detector was proposed and designed. This detector is optimized to discriminate against the intense γ rays emitted from the debris of melted fuels due to the decay of ^{134,137}Cs.

Both NRTA and NRCA have been developed and applied at the GELINA facility of the EC-JRC-IRMM as non-destructive analysis methods [4]. The Japan Atomic Energy Agency (JAEA) and EC-JRC-IRMM have started a collaboration on the feasibility study and the development of NRD since 2012 JFY.

In this paper, the principles of NRD are given in chapter 2; the experimental study in chapter 3; the design of a practical facility in chapter 4; an evaluation of uncertainties in chapter 5. A part of the concept of the NRD is described in refs. [5, 6].

2. Principles

The total neutron cross section is one of the most accurately determined nuclear data, and neutron resonance parameters of major actinides are well known in the resolved resonance region. Therefore, NRTA is expected to be one of the most accurate non-destructive analysis techniques. Fundamental principles of NRTA and NRCA have been well described in ref. [4]. In the proposed NRD, resolved resonances are used to identify and quantify the Pu and U isotopes, whose resonance energies are less than 50 eV. To resolve the resonances in this energy region, a short neutron flight path length of about 5 m can be applied as discussed in ref. [7].

For water, boron, structural materials and materials used in concrete, there are no resonances present in the energy region below 50 eV. However, most of the light and medium elements present in these materials emit discrete prompt γ rays with significant intensities via neutron capture reactions. By detecting these discrete prompt γ rays, these elements can be identified. In **Table 1**, energies of prominent prompt γ rays are summarized for some elements expected to be included in the debris.

Except ^{10}B, all of the listed isotopes emit γ rays of energies much larger than the 661-keV γ ray following the decay of ^{137}Cs. Therefore, measurements of these high energy γ rays do not suffer from a Compton background due to 661-keV γ rays.

Nucleus	Reaction	Energy of Prominent Prompt γ ray	Energy of 1st Neutron Resonance
^{1}H	^{1}H (n, γ) ^{2}H	2223 keV	—
^{10}B	^{10}B (n, αγ) ^{7}Li	478 keV	170 keV
^{28}Si	^{28}Si (n, γ) ^{29}Si	3539, 4934 keV	31.7 keV
^{56}Fe	^{56}Fe (n, γ) ^{57}Fe	7631, 7646 keV	1.1 keV
^{53}Cr	^{53}Cr (n, γ) ^{54}Cr	835, 8885 keV	4.2 keV
^{58}Ni	^{58}Ni (n, γ) ^{59}Ni	465, 8999 keV	6.9 keV

Table 1: Energies of prominent prompt γ rays and 1st neutron resonances for light and medium elements.

The γ ray spectrometer used for NRCA requires besides a high-energy resolution also a fast-timing response. The latter is needed since it is in an extreme γ ray background originating from the presence of ^{137}Cs in the debris. A study of the radioactivity in melted fuel of the TMI-2 accident by Uetsuka et al.

[8], reveals that the strongest activity originates from ^{137}Cs, with a specific activity of ranging from 10^6 to 3×10^8 Bq/g.

Furthermore, it is required to have a very high peak-to-Compton ratio, since the Compton edge energy for 661-keV γ rays is very close to the energy of γ rays induced by the [10]B(n, $\alpha\gamma$)[7]Li reaction. To satisfy these requirements, a well-type spectrometer made of LaBr$_3$ detectors was designed and is currently under development. A study based on Monte Carlo simulations of a well-type LaBr$_3$ spectrometer showed that the contribution of the Compton edge could be reduced by a factor by adding a back catcher detector. Such a reduction enables the identification of [10]B even in the presence of a high background due to the decay of [137]Cs [9].

3. Experimental Study

As a first experimental feasibility study of NRD, a normal shaped cylindrical LaBr$_3$ detector, whose crystal is 7.6 cm in diameter and 7.6 cm in length, has been utilized in NRCA experiments at the Geel electron linear accelerator (GELINA) facility of the EC-JRC-IRMM. The measurements have been performed with a [10]B sample in the beam, with and without a standard ^{137}Cs source attached to the detector.

Figure 1 shows a pulse-height spectrum measured with the LaBr$_3$ detector for 478-keV γ rays produced by the reaction [10]B(n, $\alpha\gamma$)[7]Li (shown in black colour). Figure 1 also shows a pulse-height spectrum (shown in red colour) for 661-keV γ rays measured by the same LaBr$_3$ detector. The latter was deduced by subtracting from the spectrum recorded with both the [10]B sample and the ^{137}Cs source the one obtained with only the [10]B sample present. It is shown that the 661-keV γ ray peak due to the ^{137}Cs decay is clearly separated from the peak corresponding to 478-keV γ rays resulting from the reaction [10]B(n,$\alpha\gamma$)[7]Li. However, the Compton edge of the 661-keV γ ray is very close to the 478-keV γ ray peak. Therefore, the combination with the back catcher detector as discussed in chapter 2 is expected to have a significant role to identify the boron content in debris containing high radioactive [137]Cs.
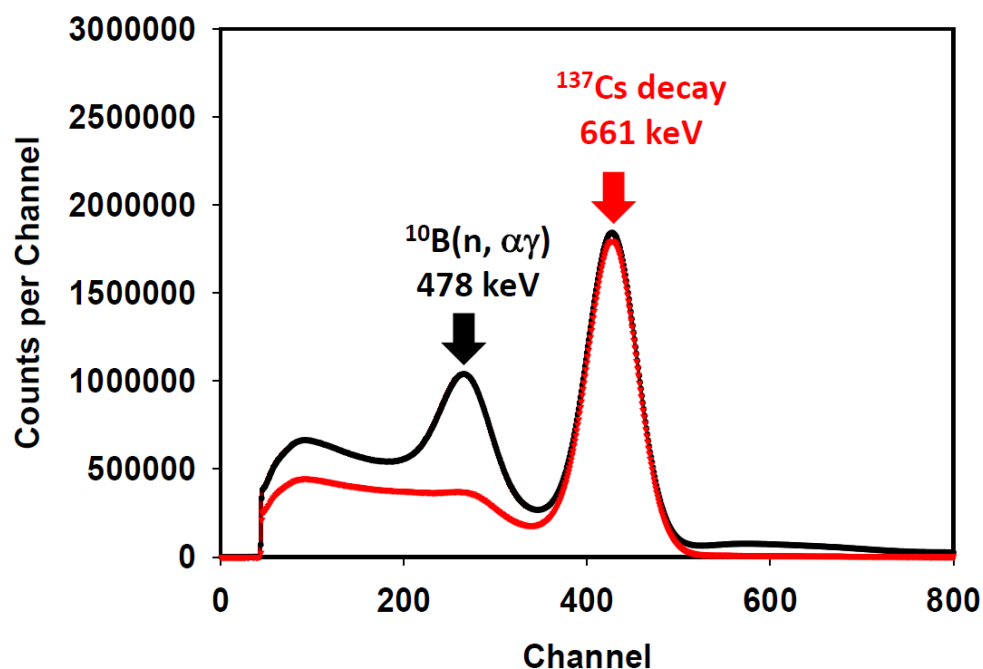

Figure 1: γ ray pulse-height spectrum measured by a LaBr$_3$ detector

4. Design of Practical Facility

Based on a simulation study, the parameters required to design a NRD facility have been investigated. The minimum neutron flight path for NRTA to resolve resonances below 50 eV is 5 m. This parameter mainly determines the size of the facility. The size of a sample for NRTA is 10-30 cm in diameter and 1-2 cm in thickness.

A shorter neutron flight path is applied for NRCA, since elements are mainly identified by the energies of the prompt γ rays. Three beam lines are planned to be installed for NRCA, since the sample size for NRCA is smaller than that for NRTA. The size of a sample for NRCA is 1-2 cm in diameter and 1-2 cm in thickness.

The required intensity of the neutron source is in the order of 10^{12} 1/s. Such an intensity can be produced by a 1 kW electron beam with a kinetic energy larger than 30 MeV [11]. High energy neutrons produced by photonuclear reactions are moderated to epithermal neutrons by a moderator surrounding a neutron generation target. Neutron collimators are installed along the beam line for NRTA and the beam lines for NRCA. **Figure 2** shows a schematic view of the practical NRD facility including an accelerator room.
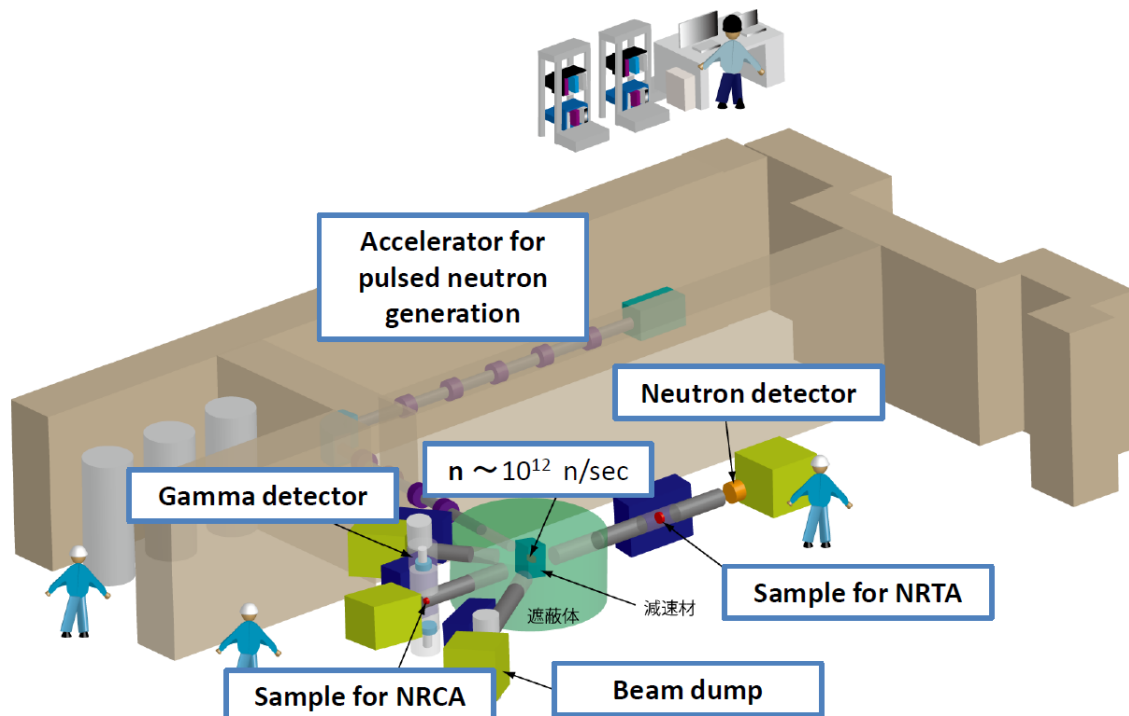

Figure 2: A Schematic View of the Practical NRD Facility

5. Evaluation of Uncertainties

The achievable uncertainty due to only counting statistics in case of NRTA was examined in Ref. [10]. The sample used in this study contained not only nuclear fuel materials but also matrix elements, such as B and Fe. The composition of the spent nuclear fuel was based on a burn-up of 40 GWd/t. **Figure 3** shows the achievable statistical uncertainty for each Pu- and U-isotope for a measurement period of 24 hours, a beam repetition rate of 100 Hz, and a neutron source intensity of 10^6 1/pulse (shown by red colour) or 10^9 1/pulse (shown by blue colour). In this case study 9 w% ^{56}Fe and 2.5 w% ^{nat}B were included in the sample. Th sample thickness was 1 cm and the diameter 30 cm.

By extrapolating the results shown in Figure 3, an uncertainty below 1% on the atomic number density due to only counting statistics can be achieved for the Pu and U isotopes with a measurement period of 20 min and a neutron source intensity above 10^{12} 1/s. This even under the condition that 9 w% ^{56}Fe and 2.5 w% ^{nat}B is present the sample.

A systematic study varying the sample thickness and the ratio of contamination materials has been carried out in ref. [10]. It was shown that the optimal sample thickness for NRTA depends strongly on the amount of contamination materials.
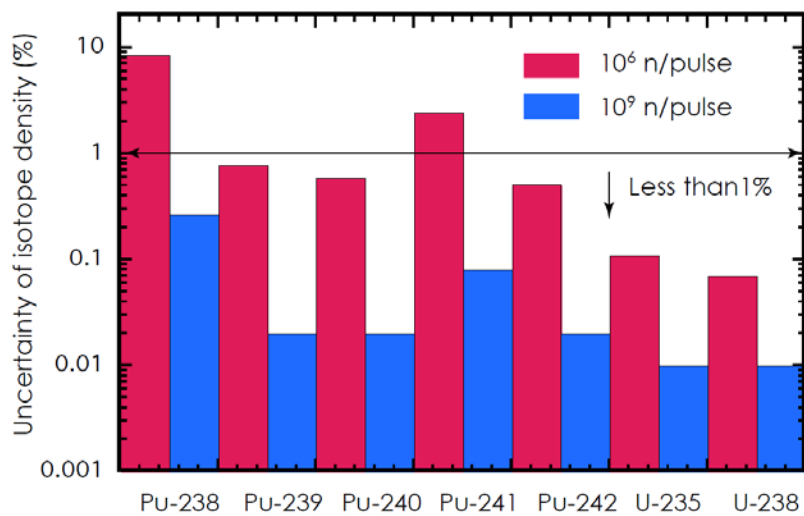

Figure 3: Achievable uncertainty due to counting statistics for U- and Pu isotopes for a measurement period of 24 hour, a beam repetition rate of 100 Hz, and neutron source intensity of 10^6 1/pulse (shown by red colour) or 10^9 1/pulse (shown by blue colour)

In order to evaluate the achievable accuracy and uncertainty of NRD, the next effects were identified to have a strong influence on the final results;

i)   Particle size
ii)   Sample thickness
iii)   Presence of contaminated materials
iv)   Sample temperature
v)   The response of the TOF-spectrometer

To study the impact of these effects quantitatively, the resonance shape analysis code REFIT will be used. The code will be adapted to the needs of the NRD. Evidently, to quantify the amount of the Pu and U isotopes present in particle-like debris, the final accuracy will strongly depend on the quality of the resonance parameters used in the analysis, in particular, the parameters for the relevant Pu and U isotopes. Hence, a survey of the total cross sections is an important issue together with a measurement of some reference spectra.

5. Summary

We have proposed neutron resonance densitometry (NRD) to quantify nuclear materials in particle-like debris of melted fuel formed in a severe accident of nuclear reactors such as the Fukushima Daiichi nuclear power plants. NRD is a method that combines NRTA (neutron resonance transmission analysis) and NRCA (neutron resonance capture analysis) using a pulsed neutron generator and a neutron TOF (time of flight) technique.

One of the advantages of NRD to other non-destructive methods is its applicability to quantifying nuclear materials in a sample containing unknown components including strong neutron absorbing materials such as boron. NRCA is used to identify these elements included in melted fuel components. The other advantage of NRD is because NRTA, which is an absolute method, is applied to quantify the main elements (i.e. U and Pu). However, it requires a special treatment in case of inhomogeneous samples. A study based on simulations for melted samples containing ^{nat}B and ^{56}Fe showed that the

main Pu and U isotopes can be quantified in 20 minutes with a counting statistics uncertainty less than 1 % by using a neutron source of 10^{12} 1/s. This neutron intensity could be generated by a 1 kW electron beam having a kinetic energy larger than 30 MeV, which is produced using a small size electron linear accelerator.

A well-type gamma-ray spectrometer made of $LaBr_3$ detector has been designed for applying the NRCA under the circumstance of highly radioactive material mixture. Monte Carlo simulations showed that the identification of ^{10}B is possible within a 1 hour measurement even under the condition of strong background by ^{137}Cs using a pulsed neutron source with a source strength of 10^{12} 1/s. As initial experimental efforts, a normal cylindrical $LaBr_3$ detector has been used for NRCA experiments using a ^{10}B sample at the GELINA facility of EC-JRC-IRMM. It was demonstrated that the 478-keV γ rays induced by the reaction $^{10}B(n, \alpha\gamma)^7Li$ can be clearly observed even in the presence of a strong γ ray background due to the decay of ^{137}Cs.

Further systematic studies are planned or under investigation for evaluating achievable accuracy accounting for uncertainties due to systematic effects such as the particle size, sample thickness, contaminated materials, and sample temperature and to demonstrate the effectiveness of NRD experimentally at the GELINA facility of EC-JRC-IRMM. Some recent advancements are also reported in this ESARDA meeting [9, 12, 13].

6. Acknowledgements

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