

Chapter 2

Recent Progress in Research and Development in Neutron Resonance Densitometry (NRD) for Quantification of Nuclear Materials in Particle-Like Debris

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Abstract To quantify special nuclear materials (SNM) in particle-like debris, a technique named neutron resonance densitometry (NRD) has been proposed. This method is a combination of neutron resonance transmission analysis (NRTA) and neutron resonance capture analysis (NRCA) or prompt gamma-ray analysis (PGA). In NRTA, neutron transmission rate is measured as a function of neutron energy with a short flight path time-of-flight (TOF) system. Characteristic neutron transmission dips of Pu and U isotopes are used for their quantification. Materials in the samples (H, B, Cl, Fe, etc.) are measured by the NRCA/PGA method. For the NRD measurements, a compact TOF facility is designed. The statistical uncertainties of the obtained quantities of the SNMs in a sample are estimated. A high-energy-resolution and high-S/N γ -ray spectrometer is under development for NRCA/PGA. Experimental studies of systematic uncertainties concerning the sample properties, such as thickness and uniformity, are in progress at the TOF facility GELINA of European Commission (EC), Joint Research Centre (JRC), Institute for Reference Materials and Measurements (IRMM).

Keywords Capture • Fukushima • GELINA • Neutron resonance densitometry • NRD • Nuclear security • Severe accident • Transmission

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2.1 Introduction

Quantifying nuclear materials (NM) in the debris of melted fuel (MF) formed in a severe accident is considered to be difficult because of their variety of size, shape, unknown composition, and strong radioactivity. Although techniques of non-destructive assay (NDA) are indispensable for the evaluation of NM in debris, quantification methods have not been established so far [1]. In the cases of TMI-2 or Chernobyl-4, accounting for the NM was based on some estimations.

We have proposed a technique called neutron resonance densitometry (NRD) [2, 3] to quantify NM in particle-like debris that is assumed to be produced in the rapid cooling processes of a severe accident [4]. Small pieces are also produced when MF are cut or broken down to be taken out of the damaged reactors [1].

To examine the NRD method, studies have begun. Some experiments were carried out at the time-of-flight (TOF) facility GELINA [5] of EC-JRC-IRMM under the agreement between JAEA and EURATOM in the field of nuclear materials safeguards research and development.

In this chapter, we briefly describe the concept of NRD, give an overview of the development of NRD, and explain some parts of the recent progress.

2.2 Neutron Resonance Densitometry

2.2.1 *The Concept of NRD*

Neutron resonance densitometry is a method of a combination of neutron resonance transmission analysis (NRTA) and neutron resonance capture analysis (NRCA) or prompt gamma-ray analysis (PGA). The fundamental principles of NRTA and NRCA are described by Postma and Schillebeeckx [6].

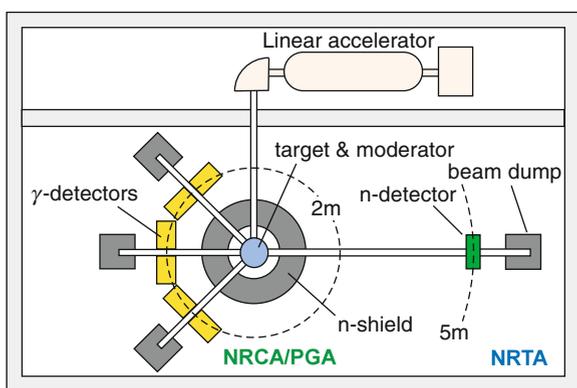
In NRTA, neutron transmission is measured as a function of neutron energy with a TOF technique. Characteristic neutron transmission dips of Pu and U isotopes are observed in the neutron energy in the range of 1–50 eV [7, 8]. Measurements of these transmission spectra can be carried out with a short-flight path TOF system [9, 10].

Although strong γ -ray radiation from MF samples does not interfere with NRTA measurements, reduction of neutron flux caused by nuclei with large total cross section (such as H, B, Cl, Fe) makes accurate NM quantification difficult. Nevertheless, the quantities of these contained nuclei could not be determined by NRTA only, because these nuclei do not resonantly interact with neutrons in this energy range. To identify and to quantify the composing isotopes, the NRCA/PGA method is required. Characteristic prompt γ rays were utilized. Table 2.1 shows prompt γ -rays emitted from nuclei after neutron capture reaction. Most of these discrete prompt γ -rays have significant intensities. The information obtained by NRCA/PGA enables us to determine the appropriate sample thickness and measurement time. This information also supports NRTA analysis.

Table 2.1 Energies of prominent prompt γ -rays and the first neutron resonances of nuclei

Nucleus	Reaction	Prompt γ rays (KeV)	First resonance (KeV)
^1H	$^1\text{H} (n, \gamma) ^2\text{H}$	2,223	–
^{10}B	$^{10}\text{B} (n, \alpha\gamma) ^7\text{Li}$	478	170
^{27}Al	$^{27}\text{Al} (n, \gamma) ^{28}\text{Al}$	3,034, 7,724	5.9
^{28}Si	$^{28}\text{Si} (n, \gamma) ^{29}\text{Si}$	3,539, 4,934	31.7
^{53}Cr	$^{53}\text{Cr} (n, \gamma) ^{54}\text{Cr}$	835, 8,885	4.2
^{56}Fe	$^{56}\text{Fe} (n, \gamma) ^{57}\text{Fe}$	7,631, 7,646	1.1
^{59}Co	$^{59}\text{Co} (n, \gamma) ^{60}\text{Co}$	230, 6,877	0.132
^{58}Ni	$^{58}\text{Ni} (n, \gamma) ^{59}\text{Ni}$	465, 8,999	6.9

Fig. 2.1 A rough draft of an NRD facility. The neutron flight path length for NRTA is 5 m and that for NRCA/PGA is 2 m



2.2.2 A Rough Draft of an NRD Facility

For practical application, the scale of an NRD facility should be minimized. Figure 2.1 shows a rough draft of the NRD facility. An electron linear accelerator with a power of 1 kW and acceleration voltage of 30 MeV is assumed [11]. High-energy neutrons are generated in the order of 10^{12} n/s by photonuclear reactions following Bremsstrahlung at the electron target. The generated neutrons are slowed down to epithermal energy by collisions in a moderator surrounding the target. Neutrons from the moderator are collimated to supply for NRTA and for NRCA/PGA.

The length of the flight path is important to design a TOF system, because the longer flight path reduces the neutron flux whereas it increases the energy resolution of the system. It may require at least a 5-m flight path to achieve a good enough resolution to resolve resonances of NMs below 50 eV in NRTA [9, 10]. A shorter neutron flight path is feasible for NRCA/PGA because the nuclei in Table 2.1 are identified by the prompt γ -ray energies. We consider that a 2-m flight path is sufficient for NRCA/PGA. The beam line lengths mainly determine the scale of

Table 2.2 Estimated statistical uncertainty of quantities of U and Pu isotopes in a sample

Nucleus	Concentration in a fuel (kg/tHM)	Statistic error (%)
^{238}Pu	0.19	0.85
^{239}Pu	5.25	0.074
^{240}Pu	2.13	0.051
^{241}Pu	1.23	0.23
^{242}Pu	0.48	0.069
^{235}U	14.6	0.049
^{238}U	928	0.010

The measurements are assumed to be carried out for 40 min with a 10^{12} n/s neutron source

the facility. One beam line for NRTA and three beam lines for NRCA/PGA are placed as shown in Fig. 2.1. The sample size for NRTA is assumed to be 10–30 cm in diameter and 1–2 cm in thickness. In comparison, the sample size for NRCA/PGA is smaller; the diameter is 1–2 cm, and the thickness is 1–2 cm. A collimator is placed between the NRCA/PGA sample and the γ -ray detector to reduce the background γ -rays from the sample. Because optimal sample thickness for NRTA strongly depends on the amount of impurities or matrix material, the quantity of the interfering nuclei in debris has to be measured roughly by NRCA/PGA preceding NRTA measurements [12].

The statistical uncertainties of NMs quantified by NRTA were estimated [12]. The size of a MF sample is assumed to be 1 cm in thickness and 30 cm in diameter. The weight of the sample becomes about 4 kg: it consisted of nuclear fuel (64 vol.%), $^{\text{nat}}\text{Fe}$ (8 vol.%), $^{\text{nat}}\text{B}$ (8 vol.%), and 20 vol.% of vacancy. The composition of the nuclear fuel was taken from Ando and Takano [13] [a fuel of 40 GWd/t burn-up in a boiling water reactor (BWR)]. The measurement was assumed to be carried out for 40 min, in which 20 min was for sample and 20 min for background. Table 2.2 shows the estimated statistical uncertainties of quantified Pu and U isotopes in the sample. The achieved statistical uncertainties are less than 1 %.

With the measurement cycle given here, about 0.15 ton of debris can be handled in a day; this enables us to measure 30 tons of debris in a year (200 working-days are assumed). This amount can be increased with the number of NRTA beam lines.

2.3 Development of a γ -Ray Spectrometer for NRCA/PGA

The γ -ray background from debris is expected to be strong. The strongest radioactive isotope in a MF of the TMI-2 accident was ^{137}Cs , which ranged from 10^6 to 3×10^8 Bq/g [14]. The energies of the prominent γ rays from nuclei listed in Table 2.1 is larger than the 661 keV γ -rays from ^{137}Cs , except for ^{10}B . Accordingly, most of the measurements of the NRCA/PGA will not have interference with the γ -rays from ^{137}Cs . On the other hand, the Compton edge of the 661 keV γ -rays surely overlaps with the 478-keV γ -ray peak originating the $^{10}\text{B}(n, \alpha\gamma)^7\text{Li}$ reaction.

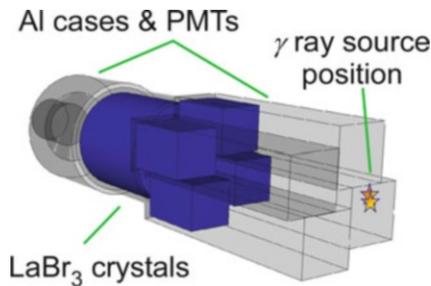


Fig. 2.2 Design of a prototype LaBr_3 γ -ray spectrometer, which consists of a cylindrical detector and four square pillar detectors. Collimated γ -rays pass through the square opening of the spectrometer to reach the cylindrical detector

The γ -ray spectrometer used for NRCA/PGA, therefore, requires properties of not only high-energy resolution and fast timing response, but also a high peak-to-Compton ratio.

To satisfy these requirements, a well-type spectrometer made of LaBr_3 crystal has been proposed [2, 15]. In a study based on Monte Carlo simulations of a well-type LaBr_3 spectrometer [15, 16], the Compton edge was successfully reduced by an order of magnitude. Such reduction enables us to roughly quantify ^{10}B in a sample, even in the presence of high background γ -rays from ^{137}Cs .

A prototype LaBr_3 γ -ray spectrometer has been designed (Fig. 2.2). Because of the technical difficulty of producing a crystal with a well, the spectrometer is made of several detectors: a cylindrical detector and four square pillar detectors. The cylindrical crystal is 120 mm in diameter and 127 mm in length; each square pillar crystal is $50.8 \times 50.8 \times 76.2$ mm. An arrangement of the detector pillars opens a square channel of 20×20 mm for the passage of collimated γ -rays from the samples. This spectrometer will be tested soon.

2.4 Experiments for NRD Developments

To evaluate systematic uncertainties in NRD, we have started experimental study at GELINA [5] under the collaboration between JAEA and EC-JRC-IRMM. The items to be studied are as follows: (1) particle size, (2) sample thickness, (3) presence of contaminated materials, (4) sample temperature, and (5) the response of the TOF spectrometer [3, 17]. Some experiments has been performed at GELINA [18–21]. A resonance shape analysis code, REFIT [22], has been adopted for the data analysis.

Experiments on sample thickness were carried out at the 25-m TOF neutron beam line of GELINA. Cu plates with various thicknesses were measured with an NRTA method. Peaks at the 579 eV resonance of ^{63}Cu were analyzed with the REFIT program. The evaluated areal densities are compared with the declared values, which were derived from measurements of the weight and the area of the

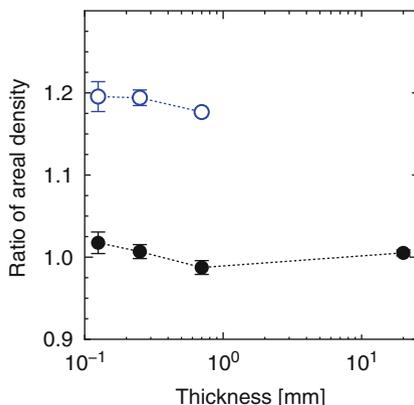


Fig. 2.3 Ratios of evaluated and declared areal densities. The 579 eV transmission peaks of ^{63}Cu were analyzed with REFIT. *Open circles* indicate the results analyzed with the resonance parameter values taken from Mughabghab [23] (#6 in Fig. 2.4), and *closed circles* represent tentatively introduced values (#7 in Fig. 2.4), which reproduce the areal densities of Cu plates better. The lines are guides for the eye. *Note:* We also analyzed the transmission spectrum of a 2-cm-thick copper sample with the parameter #6. The obtained fitted curve, however, did not reproduce the peak shape at all. Thus, the misleading *open circle* data point was removed

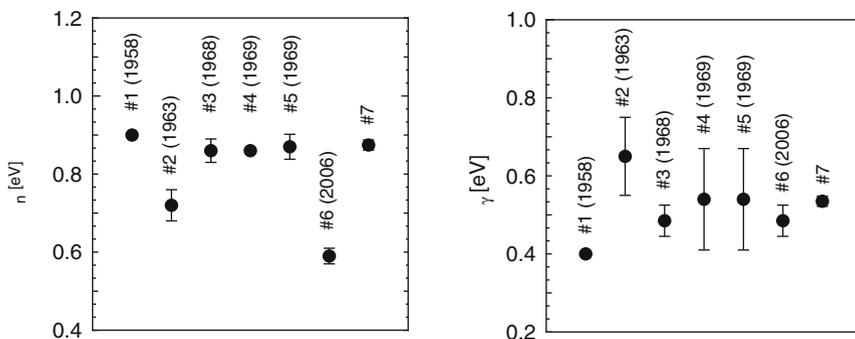


Fig. 2.4 Experimentally obtained 579 eV ^{63}Cu resonance parameters. Each data point is taken from different references (#1 [24], #2 [25], #3 [26], #4 [27], #5 [28], and #6 [23]). The data of #6 were utilized by REFIT originally; the data of #7 are tentatively introduced to reproduce the experimental transmission dips

samples. Figure 2.3 shows the results. The abscissa is the thickness of the Cu plates and the ordinate is the ratio of evaluated and declared areal densities. The open circles are the results analyzed with the resonance parameter values taken from Mughabghab [23] (#6 of Fig. 2.4); the closed circles are the results analyzed with the tentatively introduced values (#7 of Fig. 2.4), which reproduced the areal density of Cu plates better. Figure 2.4 shows measured ^{63}Cu resonance parameters. The 579-eV resonance parameters of ^{63}Cu may require being reevaluated. It should be emphasized that survey of the total cross sections of Pu and U isotopes is quite important to quantify NM.

2.5 Summary

We have proposed NRD for measurements of NM in particle-like debris of MF. The NRD system utilizes a compact neutron TOF system equipped with a neutron detector for NRTA and high-energy-resolution and high-S/N γ -ray detectors for NRCA/PGA. The rough design of a NRD facility is given. The capacity of NM measurements in the facility has been shown. Experiments on systematical uncertainties caused by sample properties, such as sample thickness and uniformity, are in progress under the collaboration between JAEA and EC-JRC-IRMM. The importance of confirmation of nuclear data has been shown in the case of Cu thickness measurements by NRTA.

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