Application of $LaBr_3$ detector for neutron resonance densitometry

H. Tsuchiya[1], H. Harada[1], M. Koizumi[1], F. Kitatani[1], J. Takamine[1], M. Kureta[1], H. Iimura[1], B. Becker[2], S. Kopecky[2], P. Schillebeeckx[2]

[1]Japan Atomic Energy Agency (JAEA)
Tokai-mura, Naka-gun, Ibaraki 319-1195, Japan
E-mail: tsuchiya.harufumi@jaea.go.jp, harada.hideo@jaea.go.jp

[2]EC-JRC-IRMM
Unit D.4. Standards for Nuclear Safety, Security and Safeguards
Retieseweg 111, B-2440 Geel, Belgium
E-mail: peter.schillebeeckx@ec.europa.eu

Abstract:

A method to determine the amount of nuclear materials in melted fuel resulting from a nuclear accident such as the one occurred at the Fukushima Daiichi nuclear power plants has not yet been established. The problem is complex due to the expected presence of ^{10}B and other strong neutron absorbing impurities. For this reason, neutron resonance densitometry, combining neutron resonance transmission analysis and neutron capture analysis, is proposed and a feasibility study has been defined. In this contribution a method to account for the presence of ^{10}B is presented and investigated. The study includes GEANT4 simulations to study the performance of a new well type gamma-ray detector based on $LaBr_3$ scintillators. In the design of the detector the main emphasis was on the capability to separate the full energy peak corresponding to the 478-keV gamma ray resulting from the $^{10}B(n,\alpha\gamma)$ reaction from the contribution of the 662-keV gamma ray due to the decay of ^{137}Cs. In addition, experiments have been carried out at the time-of-flight facility GELINA of the EC-JRC-IRMM to test the capabilities of a $LaBr_3$ detector for NRCA applications, in particular to determine impurities present in the melted fuel. A neutron resonance capture gamma-ray experiment with a ^{nat}Se sample was performed using a $LaBr_3$ scintillator in parallel with a Ge-detector. The results of these measurements demonstrate that a $LaBr_3$ detector is suitable for NRCA as a part of neutron resonance densitometry.

Keywords: neutron resonance capture analysis; $LaBr_3$ detector; neutron time-of-flight; melted fuel

1. Introduction

On 2011 Mar 11, a huge earthquake and subsequent gigantic tsunami caused a loss of electricity of the Fukushima Daiichi nuclear power plant and the system cooling the nuclear fuel installed at the reactors Units 1−3 stopped its operation. As a consequence, the nuclear fuel melted in the pressure vessel with even the risk that some melted fuel penetrated through the pressure vessel. Consequently, the possibility exists that melted fuel (MF) was deposited on the concrete floor and solidified together with other materials, resulting in a contamination of the buildings. At present, plans are made to remove the MF from the reactors after a cooling time of at least 10 years.

From the viewpoint of nuclear safeguards and security, special nuclear materials (SNMs) of uranium and plutonium present in the MF should be quantified at the time of after its removal. One of the possible techniques for such a

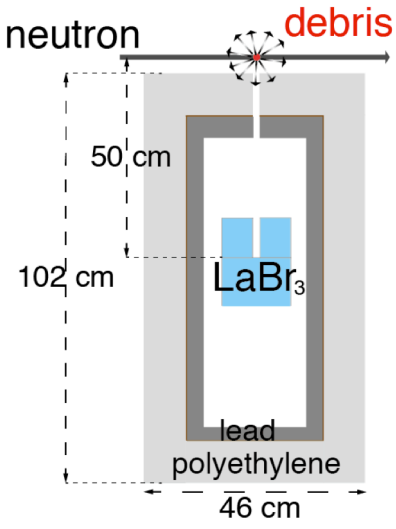

Figure 1: A schematic view of the $LaBr_3$ gamma-ray detector and its shields.

measurement is neutron resonance transmission analysis (NRTA). Bowman et al. [1] and Behrens et al. [2] successfully determined by NRTA the abundance of ^{239,240,242}Pu and ^{235,236,238}U in fresh and spent fuel pins with an accuracy better than 4%. Additionally, an extensive study based on Monte Carlo simulations was recently performed to investigate the feasibility of NRTA for the assay of plutonium in spent fuel from commercial light water reactors [3]. This study revealed the potential of NRTA to assay intact spent fuel assemblies. However, a method to measure SNMs in MF caused by a severe accident like the Fukushima case has not been established yet.

Unlike conventional commercial spent fuel, it is expected that the MF contains a substantial amount of impurities such as B, Si, Ni, and Fe. At present, no information is available about their relative abundance in the MF resulting from the Fukushima accident. In addition, particle-like debris of MF was formed due to e.g. a steam explosion [4]. Debris will also be produced during the process of removing the MF from the side. Such particle-like debris of MF will have a wide variety of size and composition, which complicates the measurements. Therefore, a method is needed which takes into account these difficulties. For this reason, neutron resonance densitometry (NRD) [5,6], which is based on NRTA combined with a kind of neutron resonance capture analysis (NRCA), is under development.

NRTA utilizes an intense pulsed white neutron source as interrogating radiation. The probability that a neutron beam is transmitted through the sample under investigation is studied as a function of neutron energy. The transmitted spectrum has characteristic dips. These dips are the result of the resonance structures in neutron induced reaction cross sections. The position and magnitude of these dips strongly depend on the content of the MF, i.e. on the relative amounts of the fission products and U- and Pu-isotopes present in the sample. Unfortunately, some of the impurities present in the MF have large total cross section for neutron interactions. As a consequence, their presence will result in a substantial reduction of the neutron flux and dilute the resonance profiles in the transmission spectrum. Among them, ^{10}B present in both the control rods and the water that was poured for cooling has a large absorption cross section in the energy range of interest. It has been shown in Ref. [6] that the presence of ^{10}B has an impact on the accuracy of the results obtained with NRTA. The accuracy can be improved when the amount of high absorbing impurities can be quantified. Since light elements like ^{10}B do not have resonance in the low energy region, their relative abundance cannot be determined by NRTA or NRCA. Therefore, NRTA used for the analysis of SNM will be complemented by measurements of prompt gamma rays following neutron capture to determine impurities which do not have large resonances in the low energy region. Consequently, the full NRD system is based on NRTA and a combined use of NRCA and prompt gamma-ray activation analysis (PGAA). A description about NRD is given in Ref. [5], and in the contribution of Harada et al. [7] at this conference.

In this paper, the focus is on a combined use of NRCA and PGAA, referred to as neutron capture analysis, as a part of the NRD development. First, the role of neutron capture analysis in NRD is presented. Then, results of Monte Carlo simulations are discussed which demonstrate the capabilities of a well type $LaBr_3$ gamma-ray detector. This detector is especially developed to quantify the relative abundance of light elements such as ^{10}B in the presence of a high background due to ^{137}Cs. Finally, results of NRCA experiments performed with a cylindrical $LaBr_3$ at the neutron time-of-flight (TOF) facility GELINA of the EC-JRC-IRMM are presented.

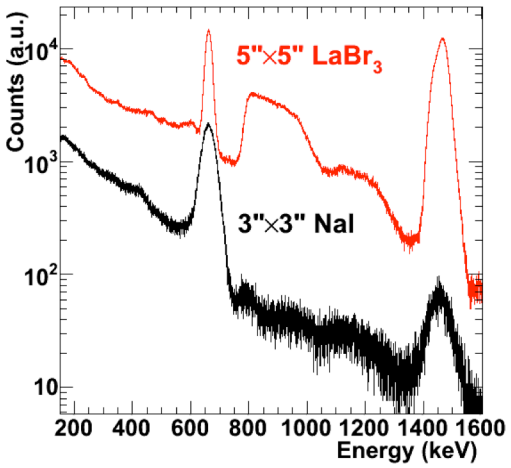

Figure 2: Energy spectra in 1000s, obtained by cylindrical $LaBr_3$ and NaI scintillators. The vertical axis indicates an arbitrary unit, because the distances from the ^{137}Cs source to the two detectors are not the same.

2. Neutron Capture Analysis

2.1. Prompt gamma-ray emission and gamma-ray background in MF

Almost no information is available about the elemental and isotopic composition of MF after a severe nuclear accident. Very likely it will consists of a mixture of

SNMs, fission products, structural materials, steel and concrete. Thus, the presence of impurities such as B, Si, Cr, Ni, Fe, and Zr are to be expected. Neutron capture reactions in these impurities result in the emission of prompt gamma rays with energies ranging from a few hundred keV to about 10 MeV. In particular prompt gamma rays with an energy of 478 keV, 3534 keV, 7939 keV, and 1205 keV are emitted after neutron capture in ^{10}B, ^{28}Si, ^{52}Cr, and ^{90}Zr, respectively. Thus, an identification and quantification of these nuclides, and in particular of ^{10}B, can be done by a prompt gamma-ray measurement.

Unfortunately, the detection of the prompt gamma rays and especially the 478 keV gamma ray resulting from the ^{10}B(n,$\gamma\alpha$) reaction will be hampered by a high gamma-ray background due to the presence of radioactive fission products like ^{137}Cs. The radioactivity of typical debris can be estimated from gamma-ray spectroscopic measurements of eight samples of debris originating from the Three Mile Island-2 accident [8]. More than 15 years after the accident, these debris samples are still radioactive mainly due to the presence of ^{137}Cs (30.2 y), ^{134}Cs (2.1 y), ^{154}Eu (8.6 y), and ^{60}Co (5.3 y). The number in parentheses indicates the half-life. From an analysis of the gamma-ray spectra the specific activity at the time of the accident were derived. The results are: $9.4\times10^{5}-3.4\times10^{8}$, $2.5\times10^{5}-8.5\times10^{7}$, $1.9\times10^{6}-2.9\times10^{6}$, and $7.8\times10^{4}-2.8\times10^{6}$ Bq/g, for ^{137}Cs, ^{134}Cs, ^{154}Eu and ^{60}Co, respectively. These values indicate that the main background affecting the background for neutron capture analysis is caused by the decay of ^{137}Cs.

2.2. Neutron capture analysis detection system

To determine the quantity of impurities by neutron capture analysis in the presence of a high background due to^{137}Cs, a gamma-ray detector is required with a fast time response and a good energy resolution. The fast response is needed to avoid problems due to dead time, while the energy resolution is essential to separate in the pulse height spectra the prompt gamma rays resulting from neutron capture from the background due to the presence of radioactive material. Furthermore, the data acquisition system has to be optimized in order to process detector pulses in high count rate conditions. As data acquisition system the VME module V1720 of CAEN, which is a 8 channel 12 bit 250 MHz flash ADC waveform digitizer, is under investigation. For these studies measurements have been carried out with NaI and $LaBr_3$ scintillators at a neutron TOF facility of the Japan Proton Accelerator Research Complex (JPARC). The analysis of the data is in progress and the results will be published later.

2.2.1 Gamma-ray detector

A schematic representation of the gamma-ray detection system for neutron capture analysis is given in Figure 1. In the figure the location of the debris samples is also indicated. The gamma-ray detector (Fig. 1) has a well type shape to reduce the Compton background resulting from the 662-keV gamma rays emitted by ^{137}Cs. This reduction is indispensable for the detection of the 478 keV gamma ray from neutron capture in ^{10}B. The gamma-ray detector is made out of $LaBr_3$ scintillators. They are in the form of a tube or a cylinder, with a diameter of 12.7 cm. The height is 10 cm for the tube and 12.7 cm for the cylinder. The tube scintillator has a hole with a diameter of 2 cm. Each scintillator is packed in an Al container with a 0.5 mm wall

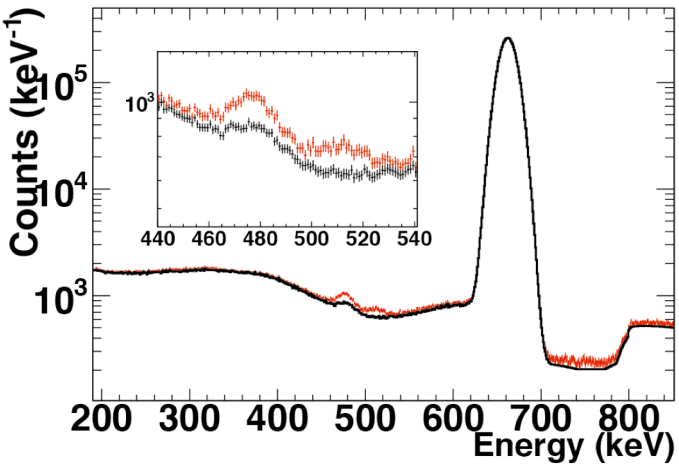

Figure 3: Pulse height spectra of the well type detector assuming 1h measurement time, calculated by the GEANT4 simulation. Red and black histograms correspond to an expected spectrum and background one, respectively. An inset shows the spectra at energies of $440-540$ keV. Errors are statistical ones.

thickness. To reduce the gamma ray and neutron background, the central gamma-ray detector is shielded with lead, silicon rubbers containing B_4C (40 wt%), and polyethylene. They act as a collimator for the gamma rays emitted by the debris. The diameter of the polyethylene hole is 2 cm, while for the other shielding materials the diameter is 1.5 cm.

A $LaBr_3$ scintillator has a fast timing with a rise time of about 30 ns [9]. In addition, the energy resolution is better compared to the one of other inorganic scintillators. Figure 2 compares an experimental pulse height spectrum collected with a $LaBr_3$ scintillator (12.7 diameter and 12.7 cm height) with the one collected with a NaI scintillator (7.62 cm diameter and 7.62 cm height) in case of an isotropic irradiation by a ^{137}Cs source. The background due to environmental (e.g. ^{40}K) and internal radioactivity (^{138}La for the $LaBr_3$ scintillator) are not subtracted. From the spectra (Fig. 2) an energy resolution at 662 keV of 3.7% for the $LaBr_3$ and 7.4 % for the NaI scintillator was computed. The resolution of a Ge detector, about 0.5% at 662 keV, is much better. However, they cannot be used because their time response is not fast enough to perform measurement in a high gamma-ray background.

2.2.2 Results of Monte Carlo simulations

To verify the performance of the detection system (Fig.1), Monte Carlo simulations have been carried out using GEANT4. The detector response for a 478-keV gamma ray emitted from a debris sample was simulated in a 662 keV gamma ray background equivalent to a ^{137}Cs amount of 10^8 Bq/g. This specific activity was derived from the TMI-2 debris measurements in Ref. [8]. The debris sample, with a mass of about 8 g, was contained in a cylinder with a 1 cm diameter and 1 cm height, corresponding to a density of 10.1 g/cm^3. The debris consisted of ^{nat}B, ^{235}U and ^{238}U with a relative abundance of 10 wt%, 1wt% and 84wt%. The remaining of the sample was filled with 1wt% Pu and 4wt% O. This composition was based on typical spent fuel of a boiling water reactor with a burn-up of 40 GWd/t [10]. For a neutron capture analysis of the debris sample a neutron beam with an intensity of 10^{12} /s was assumed. The neutron energy spectrum was calculated with MCNP5. The results in Figure 3 reveal that the peak from the 478 keV gamma ray is superimposed on a high background resulting from the decay of ^{137}Cs and ^{138}La. From an analysis of the spectrum one concludes that a 1h measurement is needed to obtain a net peak area of the 478 keV full energy peak with an 8% uncertainty only due to counting statistics.

3. Test experiment using a $LaBr_3$ detector at GELINA

To investigate the applicability of NRD, the Japan Atomic Energy Agency and the Institute for Reference Materials and Measurements of the Joint Research Centre (EC-JRC-IRMM) started collaboration in 2012. Within the collaboration various experiments are scheduled at the time-of-flight (TOF) facility GELINA of the EC-JRC-IRMM [11,12]. The experiments concentrate mainly on the impact of the characteristics of the debris on results of NRD, in particular the influence of the distribution of the particle size, the presence of neutron absorbing impurities, and the radioactivity of the sample.

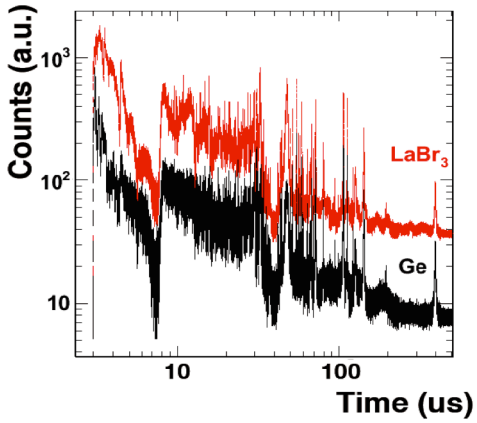

Figure 4: TOF spectra for the Se sample. Vertical axis represents an arbitrary unit. These are preliminary results.

Within this collaboration the performance of a $LaBr_3$ scintillation detector for neutron capture analysis applications was studied at GELINA. At this facility neutrons are produced via a photonuclear reaction by accelerated electrons impinging on a rotating neutron target consisting of a U-Mo alloy. After being moderated by light water, neutrons travel through vacuum flight paths to measurement stations in which detectors and samples are arranged. There are 10 flight paths with a length ranging from 10 m up to 400 m. Several measurement stations are installed at various nominal distances of 10, 30, 50, 60, 100, 200, 300, and 400 m. A detailed description of this TOF-facility can be found in Ref. [13].

The experiments to test the $LaBr_3$ scintillator were carried out at a 30 m flight path station. They started in November 2012 and finished in February 2013. Together with the $LaBr_3$ and a Ge detector was used. The $LaBr_3$ and Ge detectors were placed at 125° and 150° with respect to the incoming neutron beam, respectively. The $LaBr_3$ detector was cylindrical in shape with a 7.62 cm diameter and a length of 7.62 cm. The Ge detector was a coaxial detector with a 7.75 cm diameter and 7.65 cm length. To calibrate both detectors in energy and to verify their energy resolution measurements with radioactive sources of ^{137}Cs, ^{60}Co, Th, and a Pu-C mixture were performed.

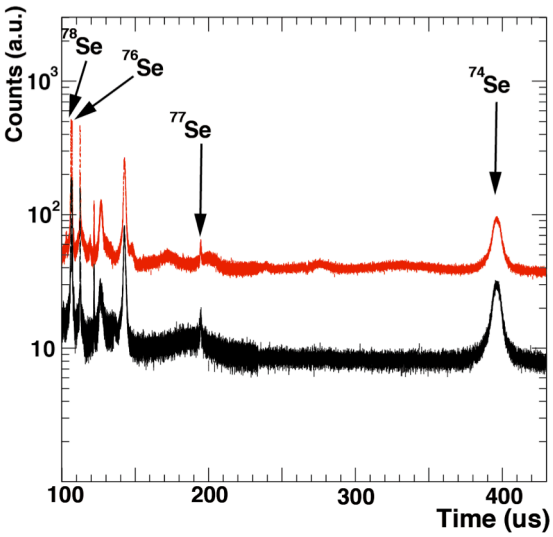

Figure 5: Same as Figure 4, but spectra between 100 – 430 us.

Four samples containing natural Se, S, Fe, and B_4C were measured. The samples were placed at a 29 m distance from the neutron target, and located at a 13.5 cm and 25 cm distance from the $LaBr_3$ and Ge detector, respectively. The diameter of the beam at the sample position was 75 mm. In Table 1 a summary of the experiments is given together with some characteristics of the samples. The results of the measurements with the sulfur sample are used to correct the TOF-spectra taken with the Fe and Se sample for background due to neutron scattered by the sample. The measurements with the natural Fe and Se sample were carried out to verify the performance of a $LaBr_3$ detector for NRCA, i.e. using the resonance structures in the TOF spectra for elemental analysis. The measurements with the B_4C sample were performed to verify the use of a $LaBr_3$ for the determination of the amount of ^{10}B by detecting the 478-keV gamma ray emitted in the $^{10}B(n,\gamma\alpha)$ reaction. A part of these measurements were carried out with a ^{137}Cs source placed in front of the $LaBr_3$ detector.

The analysis of the data is still in progress. In this paper, the potential of using a $LaBr_3$ detector for NRCA is demonstrated. For the Se sample the TOF spectra obtained with the $LaBr_3$ (red) and Ge (black) detector are compared in Figure 4. Even if the spectra are not yet corrected for the various background contributions, they exhibit the same resonance structures. The dips that appear in the TOF-region around 7 – 8 μs and 30 – 40 μs are due to the presence of the S and Na black resonance filters in the beam. Both spectra show also the same resonance structures resulting from neutron capture in the Se sample. This is emphasized in Figure 5 where the spectra are compared for TOF-values between 100 μs and 430 μs, corresponding to 430 eV and 23 eV, respectively. The TOF-spectra obtained with both the $LaBr_3$ (red) and Ge (black) detector clearly reveal the low energy resonances of ^{74}Se at 27.1 eV, ^{77}Se at 112 eV, ^{76}Se at 378 eV and ^{78}Se at 383 eV, corresponding to a TOF of 395 μs, 195 μs, 107 μs and 106 μs, respectively.

Sample	Thickness (mm)	Diameter (mm)	Mass (g)	Background filters
Se	8.0	80	66	^{10}B, Na, S
S	5.3	80	27	^{10}B, Na, S
Fe	1.0	80	40	^{10}B, Na, S, Ag, W
B_4C	2.3	70	21	^{10}B, Na, S, Ag, W

Table 1: Summary of the test NRCA experiments.

4. Summary

In order to quantify SNMs in MF caused by the Fukushima accident, a method called Neutron Resonance Densitometry is under development. NRD combines neutron resonance transmission analysis and neutron capture analysis. One of activities within the R&D programme of NRD concerns the determination of the amount of ^{10}B to account for the flux attenuation in NRTA spectra used to quantify the amount of SNMs. In this paper a method relying on the detection the 478-keV gamma-rays produced in the $^{10}B(n,\alpha\gamma)^7Li$ reaction is proposed. To optimize the detection system for

measurements in a high gamma-ray background a well type gamma-ray detector consisting of $LaBr_3$ scintillators was designed. The performance of the well type detector was studied by results of Monte Carlo simulations. In the calculations the contribution of the background due to the radioactivity of ^{137}Cs was calculated using characteristics of debris samples originating from the TMI-2 nuclear accident. It was shown that a 1h measurement is needed to deduce the amount of ^{10}B from the net peak area of the 478-keV gamma ray with a 8% uncertainty resulting from only counting statistics. In addition to this study, time-of-flight experiments were carried out at the GELINA facility of the EC-JRC-IRMM as part of collaboration between the JAEA and EC-JRC-IRMM. The capabilities of a cylindrical 3"×3" $LaBr_3$ detector for NRCA applications were demonstrated by a comparing its performance with the results of TOF-measurements using a Ge-detector.

5. Acknowledgements

This work was done under the agreement between JAEA and EURATOM in the field of nuclear materials safeguards research and development. This work is supported by JSGO/MEXT. We are very grateful for the technical assistance of J.C. Drohe, D. Vendelbo, and R. Wynants during the measurements at GELINA.

[1] Bowman C.D., Schrack R.A., Behrens J.W., and Johnson R.G.; *Neutron resonance transmission analysis of reactor spent fuel assemblies; Neut. Radiography; 1983; p 503-511.*

[2] Behrens J.W., Johnson R.G., and Schrack R.A.; *Neutron resonance transmission analysis of reactor fuel samples*; *Nuclear technology* 67; 1984; p 162-168.

[3] Strebentz J.W. and Chichester D.L.; *Neutron resonance transmission analysis (NRTA): A non-destructive assay technique for the next generation safeguards initiative's plutonium assay Challenge*; INL/EXT-10-20620; 2010.

[4] Nuclear Emergency Response Headquarters; *Report of Japanese Government to IAEA Ministerial Conference on Nuclear Safety - Accident at TEPCO's Fukushima Nuclear Power Stations*; 2011; available at http://www.iaea.org/newscenter/focus/fukushima/japan-report/

[5] Koizumi M., Kitatani F. Harada H., Takamine, J., Kureta M., Seya M., Tsuchiya H., and Imura H.; *Proposal of neutron resonance densitometry for particle like debris of melted fuel using NRTA and NRCA*; *Proc. of Ins. Nucl. Mat. Manag. 53th Annual Meeting*; 2012

[6] Kitatani F., Harada H. Takamine J., Kureta M., and Seya M.; *Nondestructive analysis of spent nuclear fuels by Neutron Resonance Tranmission Analysis; examination by linear absorption model*; *submitted to J. Nucl. Sci. Technol.*; 2013.

[7] Harada H., Kitatani F., Koizumi M., Tsuchiya H., Takamine J., Kureta M., Iimura H., Seya M., Becker B., Kopecky S., Schillebeeckx P.; *Proposal of Neutron Resonance Densitometry for Particle Like Debris of Melted Fuel using NRTA and NRCA*; Proc. of ESARDA35 (2013) presented in this meeting

[8] Uetsuka H., Nagase F., and Suzuki T.; *Gamma Spectrometry of TMI-2 Debris*; *JAERI-Research* 95-084; 1995;

[9] Stevanato L., Cester D., Nebbia G., Viesti G., Neri F., Petrucci S., Selmi S., and Tintori,C.: *High-rate read-out of LaBr(Ce) scintillator with a fast digitizer*; *Nucl. Inst. Meth. A* 678; 2012; p 83-87.

[10] Ando Y., and Takano H.; *Estimation of LWR Spent fuel Composition*; *JAERI-Research* 99-004; 1999

[11] Schillebeeckx P., Abousahl S., Becker B., Borella A., Harada H., Kauwenberghs K., Kitatani F., Koizumi M., Kopecky S., Moens A., Sibbens G., Tsuchiya H.; *Development of Neutron Resonance Densitometry at the GELINA TOF Facility;* Proc. of ESARDA35 (2013) presented in this meeting

[12] Becker B., Harada H., Kauwenberghs K., Kitatani F., Koizumi M., Kopecky S., Moens A., Schillebeeckx P., Sibbens G., Tsuchiya H.; *Particle Size Inhomogeneity Effect on Neutron Resonance Densitometry*; Proc. of ESARDA35 (2013) presented in this meeting

[13] Mondelaers W. and Schillebeecks P.; *GELINA, a neutron time-of-flight facility for high-resolution neutron data measuremnets*; *Notiziario*; 11, No 2; 2006; p 19-25.