An introduction to Neutron Resonance Densitometry

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Abstract:

Neutron resonance densitometry (NRD) is proposed as a non-destructive method to characterize particle like debris originating from severe nuclear accidents such as the one occurred at the Fukushima Daiichi nuclear power plants. NRD is based on Neutron Resonance Transmission Analysis (NRTA) and a combined use of Neutron Resonance Capture Analysis (NRCA) and prompt gamma-ray analysis (PGAA).

The basic principles are explained and systematic effects affecting the accuracy of the results are discussed, with a special emphasis on the impact of the variety in shape and size of the samples and the presence of strong neutron absorbing matrix materials.

Keywords: non-destructive assay; time-of-flight; resonance analysis; melted fuel; severe accidents; nuclear safeguards; transmission; capture; GELINA

1. Introduction

An accurate quantification of special nuclear materials (SNMs) contained in highly radioactive objects is a technical challenge, in particular when short counting times are aimed at. Such objects are debris that will be extracted from damaged reactors such as the Fukushima Daiichi nuclear power plant. According to the experience of TMI-II, they will include highly radioactive radionuclides and additional high neutron absorbing matrix materials. In addition, other characteristics such as size, shape and composition are almost all unknown. To quantify the amount of SNMs in such objects with a high accuracy in a relatively short time, a non-destructive method is required. To satisfy both these requirements an advanced non-destructive method referred to as neutron resonance densitometry (NRD) has been proposed [1,2]. NRD relies on neutron resonance transmission analysis (NRTA) to quantify the amount of SNM and neutron resonance capture analysis (NRCA) to identify the presence of unknown matrix materials.

The Japan Atomic Energy Agency (JAEA) and the Joint Research Centre, Institute for Reference Materials and Measurements (JRC-IRMM) started a collaboration to evaluate

the achievable accuracy of NRD for the characterization of debris samples [1,2]. In this paper the basic principles of NRTA and NRCA are discussed and the first experimental results obtained from measurements at GELINA are reported.

2. Principles of NRTA and NRCA

The probability that a neutron interacts with nuclei strongly depends on the energy of the neutron. This is shown in Fig. 1, which compares the total cross section as a function of the kinetic energy of the interacting neutron for several nuclides. The cross sections reveal the presence of resonance structures. These structures are related to excited states of the compound nucleus which is formed by the neutron and the target nucleus. Since resonances appear at energies that are specific for each nuclide, they can be used to determine the elemental and in some cases even the isotopic composition of materials and objects [3,4]. These resonance structures are the basis of Neutron Resonance Transmission (NRTA) and Neutron Resonance Capture Analysis (NRCA). They are non-destructive methods to determine the bulk composition, without the need of any sample preparation. NRTA and NRCA are applicable to almost all medium-weight and heavy elements. In the MeV region NRTA is also a valuable technique to analyse light elements such as hydrogen, carbon, nitrogen and oxygen and nuclides near closed shells [5,6]. Due to the difference in the experimental method, results obtained by NRTA will be more accurate compared to those resulting from NRCA [4]. In addition, NRTA is more applicable in case of radioactive samples. On the other hand, the detection limits for NRCA are mostly lower compared to those for NRTA. Hence, NRCA is preferred to determine the presence of impurities and trace elements.

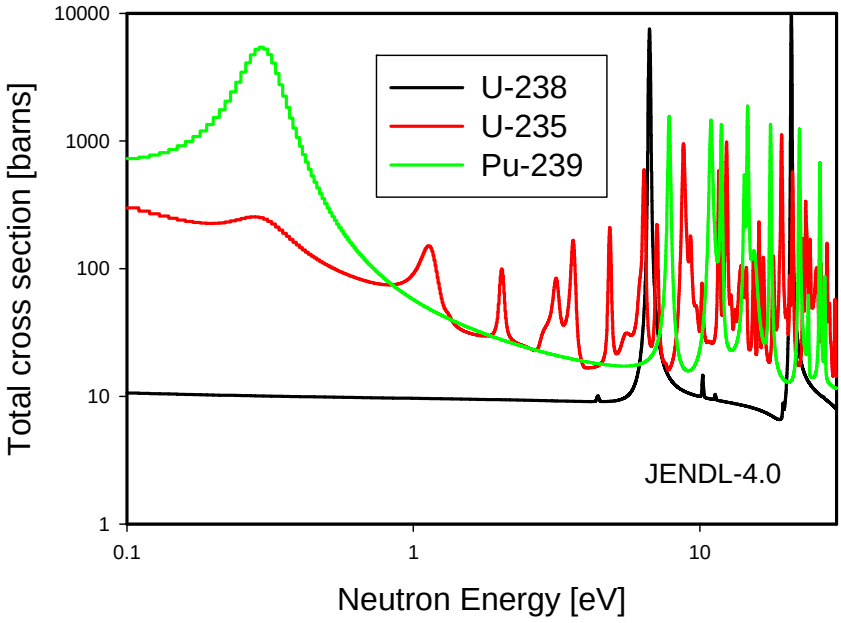

Fig. 1. *Total cross sections for neutron induced reactions in ^{235}U, ^{238}U and ^{239}Pu.*

NRTA and NRCA are based on well-established principles and methods which are used for the determination of cross section data in the resonance region. These methods have been recently reviewed in Ref. [7]. To study resonance structures in neutron induced reaction cross sections the time-of-flight (TOF) technique is applied. This technique

relies on a measurement of the time t that a neutron needs to travel a distance L, which are directly related to the velocity v and kinetic energy E of the neutron by :

$$v = \frac{L}{t} \quad (1)$$

and

$$E = m\,c^2(\gamma - 1)\,, \quad (2)$$

respectively, where c denotes the speed of light, m the rest mass of the neutron and γ the Lorentz factor.

2.1 NRTA

NRTA is based on the analysis of characteristic dips in a transmission spectrum that is obtained from a measurement of the attenuation of a neutron beam by a sample. These dips are observed at TOF values that correspond to resonance energies. Their magnitudes can be used to quantify the relative amount of each nuclide in the sample.

The theoretical quantity of interest in a NRTA experiment is the transmission T, which is the fraction of the neutron beam that traverses the sample without any interaction. For a parallel neutron beam, which is perpendicular to a slab of material, the transmission is:

$$T = e^{-\sum_k n_k \bar{\sigma}_{tot,k}} \quad , \qquad (3)$$

where $\bar{\sigma}_{tot,k}$ is the Doppler broadened total cross section for neutron induced interactions with nuclide k and n_k is the number of atoms per unit area of nuclide k. The Doppler broadening accounts for the thermal motion of the nuclei in the sample.

The experimental transmission T_{exp} is obtained from the ratio of TOF-spectra resulting from a sample-in C_{in} and a sample-out measurement C_{out}, after subtraction of the background contributions B_{in} and B_{out}, respectively:

$$T_{exp} = \frac{C_{in} - B_{in}}{C_{out} - B_{out}}\,. \quad (4)$$

The experimental spectra in Eq. 2 are corrected for losses due to the dead time in the detector and electronics chain. All spectra in Eq. 2 are normalized to the same total neutron intensity. The background contribution is determined by an analytical expression applying the black resonance technique [7].

Eq. 2 reveals that the experimental transmission is deduced from a ratio of counting spectra. Therefore, NRTA is an absolute measurement and uncertainties due to systematic effects related to additional measurements to determine the detection efficiency or the incoming neutron flux are largely reduced. In addition, when the experiments are performed in an ideal transmission geometry the experimental transmission T_{exp} (Eq.2) is a direct measure of the theoretical transmission T (Eq. 1). The conditions for an ideal transmission geometry are fulfilled when [7]:

- the sample is placed perpendicular with respect to a parallel incoming neutron beam;
- all detected neutrons have passed through the sample; and

- neutrons scattered by the sample are not detected.

The conditions of an ideal or good transmission geometry can be achieved by a proper collimation of the neutron beam at the sample and detector position [7]. However, it requires a homogeneous sample which does not contain holes. For inhomogeneous samples a special procedure is required, as shown in Ref. [8] and discussed in section 3.1.

When the total cross sections of the nuclides present in the sample are known, their areal densities n_k can be derived from a fit to the experimental transmission. Such a least squares adjustment can be performed by a resonance shape analysis code like REFIT [9]. This code has been developed to parameterize cross section data in the resonance region in terms of resonance parameters. The code accounts for various experimental effects such as the response of the TOF-spectrometer, multiple interaction events, and Doppler broadening. For NRD applications on melted fuel debris a special module has been implemented, which will be discussed in section 3.1.

2.3 NRCA

The observable in a capture (or NRCA) experiment is the fraction of the incident neutron beam undergoing a capture reaction in the sample. The theoretical capture yield Y_γ resulting from a capture reaction can be expressed as a sum of primary $Y_{0,k}$ and multiple interaction events $Y_{m,k}$:

$$Y_\gamma = \sum_k \left(Y_{0,k} + Y_{m,k}\right) \quad . \quad (5)$$

The latter are due to a capture reaction after at least one neutron scattering event in the sample. For a parallel uniform neutron beam and a homogeneous slab of material perpendicular to the beam, the primary capture yield $Y_{0,k}$ resulting from a capture reaction by nuclide k is given by:

$$Y_{0,k} = \left(1 - e^{-\sum_j n_j \bar{\sigma}_{tot,j}}\right) \frac{n_k \bar{\sigma}_{\gamma,k}}{\sum_j n_j \bar{\sigma}_{tot,j}} \quad , \qquad (6)$$

where $\bar{\sigma}_{\gamma,k}$ is the Doppler broadened capture cross section. Only in case of very thin samples and/or small cross sections, the capture yield is directly proportional to the product of the areal density n_k and the capture cross section. For relative thick samples, multiple interaction events have a substantial contribution to the yield and complicate the analysis as demonstrated in Refs. [4,7].

In a capture (or NRCA) experiment the prompt γ-rays, which are emitted after a neutron capture reaction in the sample are detected. The experimental quantity that can be obtained from such an experiment and related to the theoretical capture yield, is the experimental yield Y_{exp}. This yield is derived from:

$$Y_{exp} = N \frac{C_\gamma - B_\gamma}{\varphi} \quad , \qquad (7)$$

where C_γ and B_γ are the observed dead time corrected sample and background spectra, respectively, and φ is the incident neutron flux. The factor N is a normalization factor that accounts for the effective area of the sample seen by the neutron beam; the probability that the prompt γ-rays escape from the sample; the solid angle between sample and detector and the probability to detect at least one γ-ray created in the capture event. To estimate the background, additional measurements without a sample in the beam and with a pure scattering sample (e.g. a carbon or ^{208}Pb sample, which have a low capture cross section) are performed. A detailed discussion on the background determination is given in Ref. [7].

Eq. 7 reveals that the experimental observable in a NRCA experiment is more complicated compared to the one obtained from a NRTA experiment. The yield Y_{exp} can only be derived from the observed response once the incoming neutron flux and quantities which are related to the detection of the prompt γ-rays are known. Moreover, in most cases only the solid angle and effective area are independent of the energy of the incident neutron beam. The energy dependent neutron flux can be determined by measurement of a neutron standard reaction [10]. The efficiency to detect at least one γ-ray depends on the technique that is applied to measure the prompt γ-rays. Ideally, a detection system is used with an efficiency that is independent of the γ-ray cascade, i.e. independent of multiplicity and energy spectrum. Such a system can be realized by a total absorption detector with an almost 100 % efficiency or by applying the total energy detection principle, so that the detection efficiency becomes proportional to the total γ-ray energy produced in the capture event. More details about such systems can be found in Ref. [7].

An analysis based on a full methodological approach involving a resonance shape analysis, is not evident in case of NRCA. Besides accurate nuclear data, it also requires detailed information about the neutron beam characteristics, detection system and even some sample properties. Therefore, a more empirical approach relying on the use of calibration samples with known composition is often applied. The experimental signature for such an approach is the net area of a resonance peak. A detailed discussion on the calibration approach is given in Ref. [3,4].

3. Validation experiments at GELINA

The potential of NRTA for the characterization of fresh and spent fuel pins has already been demonstrated in Ref. [11] and [12]. The method has also been applied successfully to determine the elemental and isotopic composition of a waste sample that was extracted from high radioactive nuclear liquid waste originating from the reprocessing facility in La Hague. [13]. However, the samples analysed in these references were all homogeneous samples. Also the feasibility study of Sterbentz and Chichester [14], which is fully based on Monte Carlo simulations, considers NRTA only for the characterization of intact fresh and spent fuel assemblies.

The analysis of particle like debris samples of melted fuel produced in a severe nuclear accident will be more complex and challenging. This is mainly due to the specific characteristics of the samples, such as the radioactivity, sample temperature, presence of unknown matrix material including strong neutron absorbers and the diversity in shape and size of the particle like debris samples. In Ref. [2,4] problems related to the

sample characteristics have been studied based on theoretical transmissions through 2.5-cm thick spent fuel samples. The results of this study suggest that heterogeneous samples, even in the presence of strong absorbing matrix materials, can be characterized by NRTA. This statement was verified by results of measurements performed at the time-of-flight spectrometer GELINA, which is described in detail in Ref. [15]. Experiments were performed at a 25 m transmission station using a NE905 Li-glass scintillator as neutron detector. The TOF of a neutron traversing the sample was determined by the time difference between the start signal, given at each electron burst, and the stop signal from the scintillator.

3.1 Characterization of a heterogeneous W sample by NRTA

One of the main difficulties for a correct interpretation of the result of a transmission measurement is to account for any heterogeneity of the sample. The attenuation of a neutron flux in a heterogeneous sample can differ significantly from the attenuation in a homogenized medium. In case $\sum_j n_j \bar{\sigma}_{tot,j}$ is not very small a substantial bias will be introduced when the heterogeneity of the sample is not taken into account.

The impact of the inhomogeneity of the sample was studied at EC-JRC-IRMM by means of numerical simulations and measurements [8, 16]. Different analytical models were analysed in terms of their capability to take into account the impact of the powder characteristics in an adjustment procedure of the areal density. To validate the models they have been implemented in the REFIT code [17]. Transmissions through heterogeneous powder samples were calculated by Monte Carlo simulations to study the performance of the analytical models for NRTA on particle like debris samples. In these simulations stochastic heterogeneous samples were generated using a discrete element modelling code from Sandia National Laboratory [19]. The best results were obtained using the model developed by Levermore et al. [18], which is often referred to as the LP-model. This Markovian model assumes that the chord length probability distribution of particle rays passing through a grain can be described by an exponential. The model is in particular suited for mixtures with irregular shaped grains such as powder samples. Since the model is fully dimensionless, it can be applied for all sizes of grains as long as stochasticity of the mixture is given. Fig. 2 shows a photomicrograph of a tungsten powder illustrating the expected irregular shapes of powder grains.

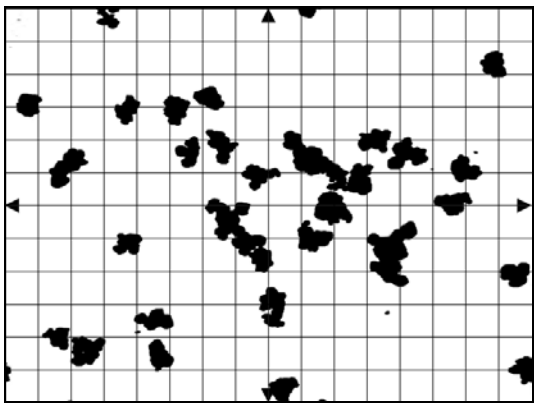

Fig. 2 *Photomicrograph of tungsten grains (grid mesh size of 150 µm).*

The performance of the LP-model was verified by experiments at GELINA using a sample that consisted of a mixture of sulphur and tungsten powder [8]. The photomicrograph of the tungsten grains in Fig.2 was taken before the preparation of the mixture. The experimental transmission and the results of an analysis assuming a homogeneous sample and a heterogeneous sample based on the LP-model are shown in Fig. 3. The lower part of the figure shows the residuals, defined as:

$$R = \frac{T_{exp} - T}{u_{T_{exp}}}, \qquad (10)$$

with $u_{T_{exp}}$ the uncertainty resulting from a propagation of the uncorrelated uncertainty components due to counting statistics. The large differences between T_{exp} and T in the region of strong resonances are due to the limitations of the homogeneous assumption. The residuals resulting from an analysis with the LP-model are flat in the whole energy region and demonstrate that the quality of the fit is significantly improved by applying this model. Using the LP-model the average areal density of tungsten in the sample derived from the transmission profile is $n_W = (1.058 \pm 0.003)\ 10^{-3}$ at/b. This value is in very good agreement with the expected value, which is $n_W = (1.036 \pm 0.026)\ 10^{-3}$ at/b. Supposing a homogeneous sample the average areal density is underestimated by almost 10%.

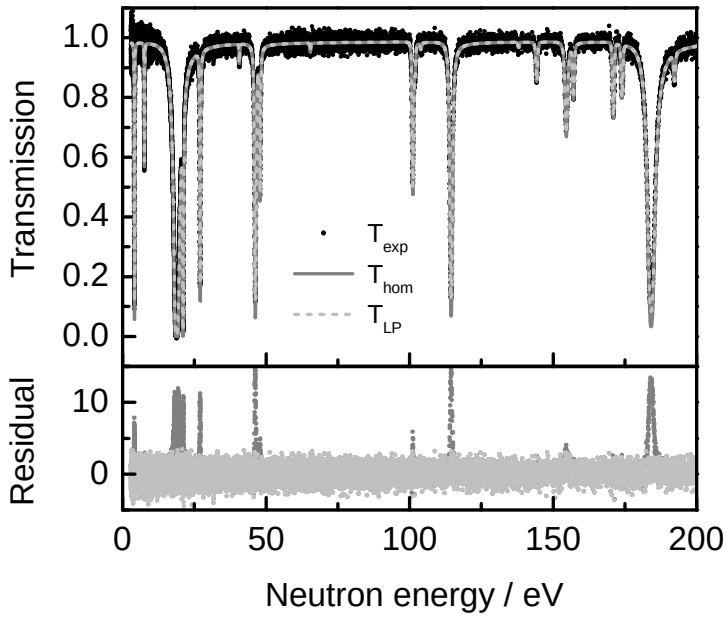

Fig. 3 *Transmission through a sample consisting of a mixture of tungsten and sulphur powder. The experimental data (T_{exp}) are compared with the fitted transmission based on a homogeneous sample (T_{hom}) assumption and on the LP-model (T_{LP}) [8].*

3.2 Characterization of a uranium sample by NRTA

A validation of NRTA applied for the characterization of special nuclear material in the presence of strong absorbing matrix material is reported in Ref. [4]. Measurements were performed with a U_3O_8 sample that was enriched to 4.514 at% in ^{235}U. This sample, which is part of the EU nuclear reference material 171 set [20] with reference CBNM 446, was produced as a reference sample for γ-ray spectroscopy. It is not ideal for transmission measurements. The significant EPOXY content of the ultrasonic identifier,

which is placed in the plug of the sample container, resulted in a strong attenuation of the incident neutron beam. On the other hand, the results of these measurements provide an ideal basis for a validation of the procedure proposed in Ref.[4] to account for the presence of matrix materials that cannot be identified and quantified by resonances.

The dead time corrected sample-in and sample-out TOF-spectra together with the background contributions are shown in Fig. 2. The resulting transmission through the CBNM 446 sample is plotted as a function of energy in Fig. 3. The strong reduction in neutron flux resulting in a transmission baseline below 0.01 is primarily due to the presence of the neutron absorbing matrix materials. As suggested in Ref. 4 the influence of the matrix materials can be taken into account by lumping their contribution to the observed transmission using a single total cross section $\sigma_{tot,X}$ of a dummy element X with areal density n_X. The energy dependence of this cross section is expressed as a sum of a constant and a 1/v contribution:

$$n_X \sigma_{tot,X} = a_X + \frac{b_X}{v} . \quad (11)$$

The parameters a_X and b_X are adjusted in a fit to the experimental data together with the areal densities of the materials of interest, i.e. ^{235}U and ^{238}U. To apply this procedure the REFIT code was used and the required nuclear data (resonance parameters) were taken from the JENDL-4.0 library [21]. The resulting areal densities are listed in Table 1. The quoted uncertainties are only due to a propagation of uncorrelated components due to counting statistics. They do not include the uncertainty due to nuclear data or other systematic effects. The areal densities of ^{235}U and ^{238}U obtained from a NRTA analysis are within these uncertainties in agreement with the reference values. The resulting isotopic ratio $^{235}U/^{238}U = 0.0475 \pm 0.0008$ is in very good agreement with the certified value, which is $^{235}U/^{238}U = 0.047290 \pm 0.000003$.

Table 1. *Areal densities of ^{235}U and ^{238}U resulting from a fit to the experimental transmission compared with the reference values. The uncertainties are standard uncertainties resulting from the propagation of uncorrelated components due to counting statistics.*

	Areal density	
	NRTA	Reference
^{235}U	$(5.063 \pm 0.090)\ 10^{-4}$ at/b	$(5.0326 \pm 0.0080)\ 10^{-4}$ at/b
^{238}U	$(1.062 \pm 0.010)\ 10^{-2}$ at/b	$(1.0628 \pm 0.0015)\ 10^{-2}$ at/b

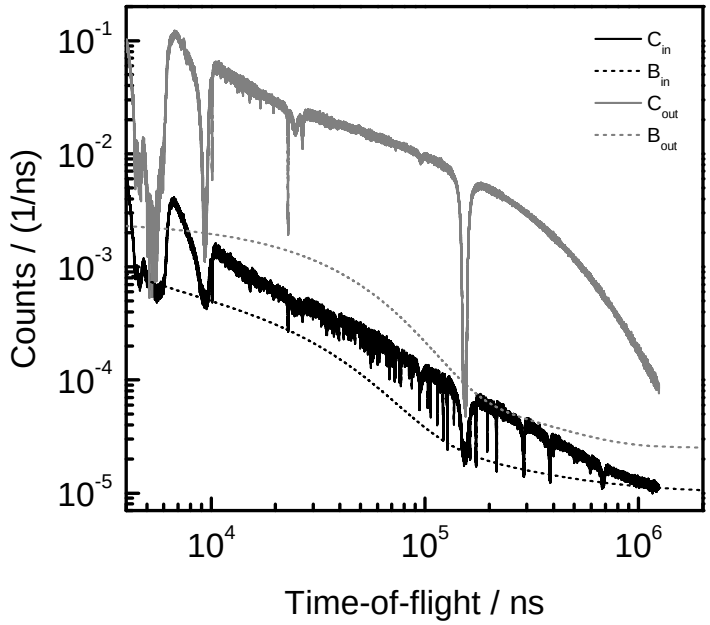

Fig. 4 *TOF-spectra resulting from transmission measurements at a 25-m station of GELINA. The sample-out (C_{out}) spectrum is compared with the sample-in (C_{in}) spectrum obtained with the CBNM 446 reference sample. The background contributions B_{out} and B_{in} are also shown.*

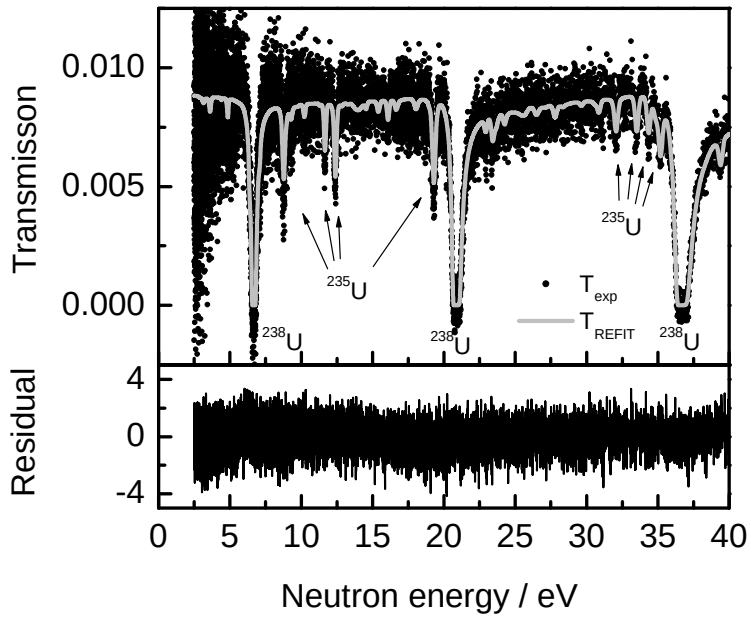

Fig. 5 *Transmission through* the CBNM 446 reference sample *as a function of neutron energy. The experimental transmission(T_{exp}) is compared with the result of a fit (T_{REFIT}). The residual is shown in the lower part of the figure.*

It should be noted that the results in Table 1 and those obtained in section 3.1 are derived from absolute measurements without the need of any additional calibration measurement involving representative reference samples. Therefore, they confirm the statement of Ref. [4] that NRTA can be considered as one of the most accurate non-destructive methods for the characterization of materials. Evidently the results strongly depend on the quality of the nuclear data involved [4,22]. However, uncertainties on

total cross sections are mostly considerably smaller compared to uncertainties on capture and scattering cross sections [7].

The areal densities of ^{235}U and ^{238}U in Table 1 were obtained from a 30 hour measurement at a 25-m station with a 35-mm effective sample diameter using an electron beam with an average energy of 100 MeV and an average electron current of 40 μA. This resulted in a 1.6 % and 0.6 % uncertainty due to counting statistics for the areal density of ^{235}U and ^{238}U, respectively. A similar counting statistics uncertainty can be obtained in about 1800 s at a 10-m station using samples with a 30-cm diameter and a 1 kW electron beam. This confirms the results of preliminary calculations presented in Ref. [1]. These authors performed calculations for a 1-cm thick and 30-mm diameter spent fuel sample with an additional amount of 2.5 wt % ^{10}B and 9 wt% Fe to simulate the presence of absorbing and structural material. The calculations were done for a neutron source intensity that can be produced by a 1 kW electron beam with a kinetic energy of 40 MeV, a current of 25 μA, an operating frequency of 250 Hz and a pulse width of 400 ns [23].

4. Detector development for NRCA

One of the challenges in measurements of debris samples is due to the presence of matrix material or impurities with relatively large total cross sections, e.g. H, B, Cl, Fe. Due to the interaction of the neutron beam with these materials, their presence will reduce the intensity of the neutron, reducing the counting statistics, and distort the transmission spectra. Unfortunately for most of these light elements no resonance structure is present in the energy region below 100 eV. Hence, their amount cannot be determined by NRTA from an analysis of resonance dips taken with a compact TOF system with a short flight path. Fig. 6 illustrates that the amount of strong neutron absorbing matrix material such as ^{10}B has a strong impact on the uncertainty due to counting statistics. An optimum sample thickness can be defined for a given boron concentration.

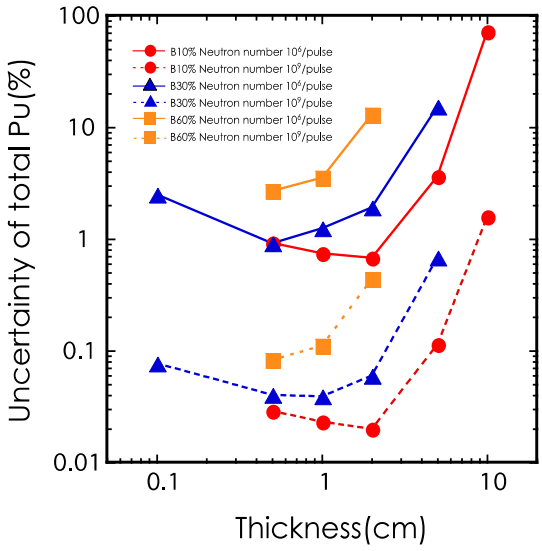

Fig. 6. *Counting statistics uncertainty on the total Pu amount in the sample as a function of the sample thickness for different boron concentrations.*

To optimize the measurement conditions, i.e. sample thickness, and improve the accuracy of NRTA, a combination of NRCA and PGA is proposed to determine the amount of strong neutron absorbing material, in particular ^{10}B, in an extreme gamma-ray background due to the presence of ^{137}Cs. For such measurements a detection system is required with both a good time resolution and a high energy resolution that can be operated in high count rate conditions. Therefore a γ-ray spectrometer consisting of $LaBr_3$ scintillation detectors and a dedicated data acquisition (DAQ) system are under development.

A well-type γ-ray spectrometer consisting of a main $LaBr_3$ scintillation detector and four square $LaBr_3$ scintillation detectors has been designed and constructed. Due to their short decay time of about 20 ns such scintillators have a fast recovery time and good time resolution. The energy resolution of the individual $LaBr_3$ scintillation detector, which is 3.6% at 662 keV, should be sufficient to separate the peak resulting from the detection of the 478 keV γ-rays induced by the $^{10}B(n,\alpha\gamma)\ ^{7}Li$ reaction from the Compton background due to detection of 661-keV γ-rays following the decay of ^{137}Cs. The detection system will be connected to DAQ system based on digitizers which will be able to record events at a rate of 500 kHz for each of the 8-channel inputs, i.e. in total a 4 MHz event rate.

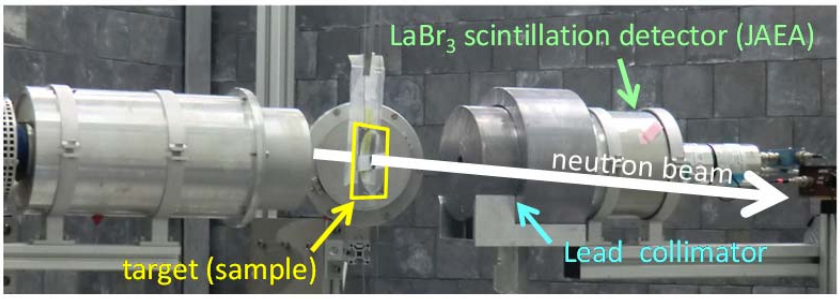

Fig. 7 *A setup of experiments at GELINA to test the performance of a $LaBr_3$ scintillator in the presence of ^{137}Cs.*

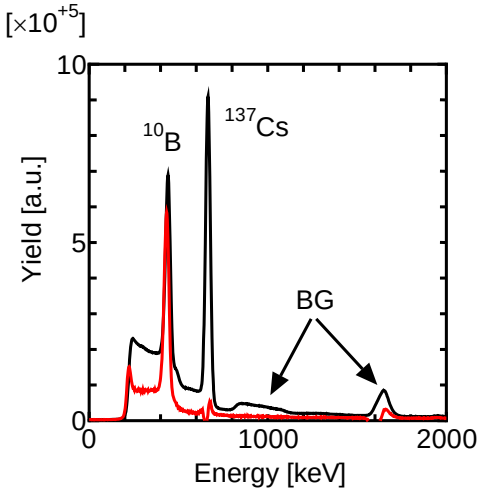

Fig. 8 *Pulse-height spectra (black line) measured by a $LaBr_3$ detector for gamma-rays from the $^{10}B(n, \alpha\gamma)$ reaction and ^{137}Cs source. The red solid line, which is a background subtracted pulse-height spectrum, illustrated that the peak due to the 478-keV gamma-ray line can be analysed.*

Experiments were carried out at GELINA to test the performance of the main $LaBr_3$ scintillation detector. The set-up is shown in Fig. 7. A pulse height spectrum obtained by placing a boron sample in the beam is shown in Fig. 8. A comparison of this spectrum with a spectrum resulting from measurements taken with the boron sample and a ^{137}Cs source illustrates that the resolution of a $LaBr_3$ based spectrometer is good enough to determine the relative amount of ^{10}B from an analysis of the g-ray peak due to ^{10}B(n, ag) reaction. Further development is in progress and NRD demonstration experiments are planned in 2015 at a short neutron flight path of GELINA.

5. Generalization of NRD for objects with an irregular shape

In Ref. 4 a detailed study of the performance of NRTA is presented. These results, which were partly presented in section 3, demonstrate its potential to quantify SNM in case of homogeneous materials and even for the characterisation of samples consisting of particles which vary in shape and size. Recently, it was pointed out that NRTA can also be applied to analyse samples with an irregular shape [24]. **Fig. 9** (lower) shows how the transmission of a neutron beam changes due to the difference in geometrical shape of an object in a box. The transmission is given for three extreme shapes which are shown in the upper part of Fig. 8: (a) a flat shape geometry, (b) a triangle shape, and (c) a half-open shape including a 50% fraction of void. The objects in the samples (a), (b) and (c) consist of ^{238}U with a 1 g/cm^2 average areal density. Although the average areal density is the same, the resulting transmission strongly depends on the shape of the ^{238}U object.

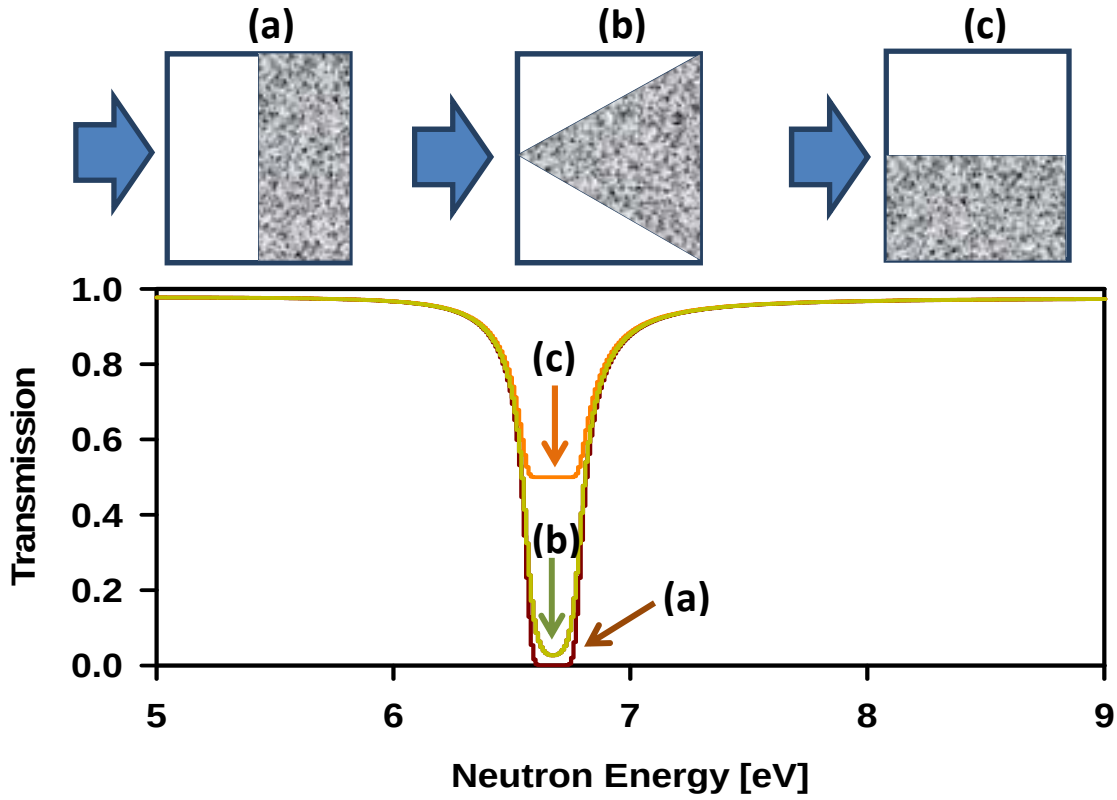

Figure 9: Neutron transmission for different shapes of a ^{238}U object in a box: (a) a flat shape representing a homogeneous geometry (brown line), (b) a triangle shape (green line), and (c) a half-open shape with a 50% fraction of void (Orange line). The average areal density of ^{238}U is 1 g/cm^2 for the three objects.

The shape of the object has on impact on both the dip and form of the transmission profile. Therefore, it is expected that information about the shape of the object can be deduced from a resonance shape analysis of the observed transmission. An analytical model to parameterize irregular objects was presented and studied in Ref. [25]. The results of these studies showed that applying this analytical model NRTA can also be used to quantify SNM in samples made of large rocks with an irregular shape.

6. Future perspectives

The capabilities of Neutron Resonance Densitometry, which is based on a combination of NRTA and NRCA, have been presented. The method relies on the appearance of resonance structures in neutron induced reaction cross section. It was demonstrated that a combination of NRTA and NRCA, referred to as Neutron Resonance Densitometry, can be applied to characterize particle like debris samples of melted fuel and that an accuracy of 2% on the areal density of ^{235}U and ^{239}Pu can be reached.

7. References

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