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Neutron Resonance Analysis System Requirements

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Neutron Resonance Analysis

System Requirements

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Table of contents

Abstract

Requirements to construct a compact Neutron Resonance Analysis system for the characterisation of nuclear materials are described. The design of the system is focussed on the characterisation of melted fuel formed in a severe nuclear accident. The requirements are based on results of previous experimental studies and simulations and results of validation measurements at a transmission station with a short flight path that was installed and commissioned at GELINA. In addition the potential to apply this technique for other nuclear applications is discussed.

1 Introduction

The presence of resonance structures in neutron induced reaction cross sections is the basis for Neutron Resonance Transmission Analysis (NRTA) and Neutron Resonance Capture Analysis (NRCA) [1]. NRTA and NRCA are Non-Destructive Analysis (NDA) methods to determine the elemental and isotopic composition of materials and object. Both methods are non-invasive and do not require any sample preparation. They require a pulsed white neutron source combined with Time-Of-Flight (TOF) measurements and rely on well-established methodologies for neutron induced reaction cross section measurements [2].

NRTA has been proposed as a method to quantify Special Nuclear Materials (SNM), i.e. Pu-isotopes and ^{235}U, in debris of melted fuel formed in a severe nuclear accident [3], such as the one that occurred at the Fukushima Daiichi power plants. NRTA is an absolute NDA method, that does not require any additional calibration measurements using reference samples that are representative for the material under investigation [1]. The accuracy of the results relies on the quality of the nuclear data, in particular, the total neutron cross sections of the nuclides present in the sample. Since total cross sections for neutron interactions are one of the most accurate nuclear data, NRTA can be considered as one of the most accurate absolute NDA methods for the characterisation of materials.

The potential of NRTA for the characterisation of fresh and spent nuclear fuel has already been demonstrated by Priesmeyer and Harz [4] and Behrens et al. [5]. NRTA has been applied by Noguere et al. [6] at the TOF-facility GELINA [7] to characterise a solution sample that originated from nuclear waste of a reprocessing facility. The samples used in Refs. [4], [5] and [6] were homogeneous samples with a regular shape. An analysis of particle- and powder-like debris samples of melted fuel will be far more complex and challenging due to their characteristics, in particular [8]:

- the diversity in shape and size of the particle- and powder-like debris samples,
- the presence of neutron absorbing matrix material without low energy resonances (e.g. ^{10}B),
- the complexity of the transmission spectra due to the presence of fission products, and
- the high temperature of the debris samples.

Most of these problems have been studied and solved as part of a collaborative effort of the Japan Atomic Energy Agency (JAEA) and the Joint Research Centre (JRC) at Geel. The impact of the diversity in shape and size of the samples was investigated by means of simulations and measurements by Becker et al. [9], [10]. Various analytical models were studied in terms of their capability to take into account the powder characteristics. Several numerical benchmarks were produced by creating stochastic geometries using Monte Carlo methods [9]. The best results were obtained using an empirical model proposed by Kopecky et al. [11] and the so-called LP-model of Levermore-Pomraning [12]. Both models were implemented in the Resonance Shape Analysis (RSA) code REFIT [13] and validated in Ref. [10] by results of experiments at a 25 m transmission station of GELINA. A method to account for the contribution of strong neutron absorbing light materials without resonances in the low energy region was proposed and validated in Ref. [1]. A full quantitative validation was obtained from measurements at a 25 m station of GELINA using a U_3O_8 reference sample, with reference CBNM 446 [1]. The areal density of ^{235}U and ^{238}U was reproduced within 1% of the declared value by the NRTA results. This was within the quoted uncertainties due to counting statistics.

2 Compact transmission station at GELINA

The studies and validation experiments mentioned in the Introduction were primarily based on relatively high resolution measurements at a 25 m or 50 m transmission station of GELINA. For industrial, routine application a compact NRTA system with a short flight path is required. To investigate the potential of NRTA under such conditions and to define requirements for an industrial system experiments were performed at a new transmission station of GELINA [14],[15]. This station with a nominal flight path length of 10 m was optimised for cross section measurements in the low energy region. It was installed at flight path 13 of GELINA viewing a moderated neutron beam. This flight path forms an angle of 18^o with respect to the normal of the moderator face viewing the flight path.

A schematic representation of the experimental set-up is shown in Figure 1. The moderated neutrons are collimated into the flight path through evacuated aluminium pipes of 50 cm diameter with annular collimators. An automatic sample changer is positioned at about 7.7 m from the neutron producing target, allowing automated sample-in and sample-out measurement sequences. A second sample changer at the same position is used to mount black-resonance and anti-overlap filters. Neutrons passing through the sample and the filters are further collimated and detected by a 6.35 mm x 76 mm x 76 mm Li-glass scintillator, which is placed at 10.9 m distance from the neutron target. The Li-glass is enriched to 95% in ^{6}Li.

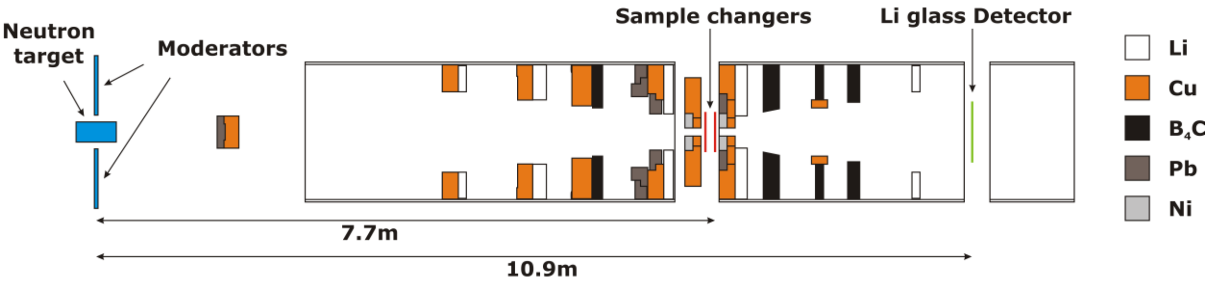

Figure 1 Schematic representation of the transmission set-up at flight path 13 of GELINA. The vertical scale is a factor 10 larger compared to the horizontal scale.

To assure a good transmission geometry, i.e. to guarantee that all neutrons reaching the detector have passed through the sample and that neutrons which are scattered by the sample are not detected, great care was taken in defining and aligning the collimation. A set of Pb, Cu and Ni collimators with decreasing diameter were installed between the neutron producing target and the sample changer, reducing the neutron beam size to about 10 mm diameter at the sample position. Additional Li and B_4C collimators were installed to absorb neutrons that are scattered by the collimators. Similarly collimators with increasing diameter were positioned between the sample changer and the neutron detector, to prevent scattered neutrons from reaching the detector. The collimation is modular and can be adapted to the sample size. To further minimize the background from scattered neutrons, including those from neighbouring beam lines, the detector was placed inside a shielding structure composed of lead, boron-oxide, wax and borated polyethylene. The set-up was commissioned in 2015 [14],[15]. The commissioning included an evaluation of the background conditions using different overlap filters (Cd and ^{10}B) and various configurations of black resonance filters. The transmission dips due to the presence of these black resonance filters were used to approximate the background contribution by an analytical expression, as described in Ref. [2]. Results of the background measurements are shown in Figure 2. The TOF-spectra without sample in the beam are shown and compared with the total background. These figures reveal that a background level of less than 10% can be reached. A detailed description of the various background components in TOF transmission measurements can be found in Refs. [1], [2] and [16].

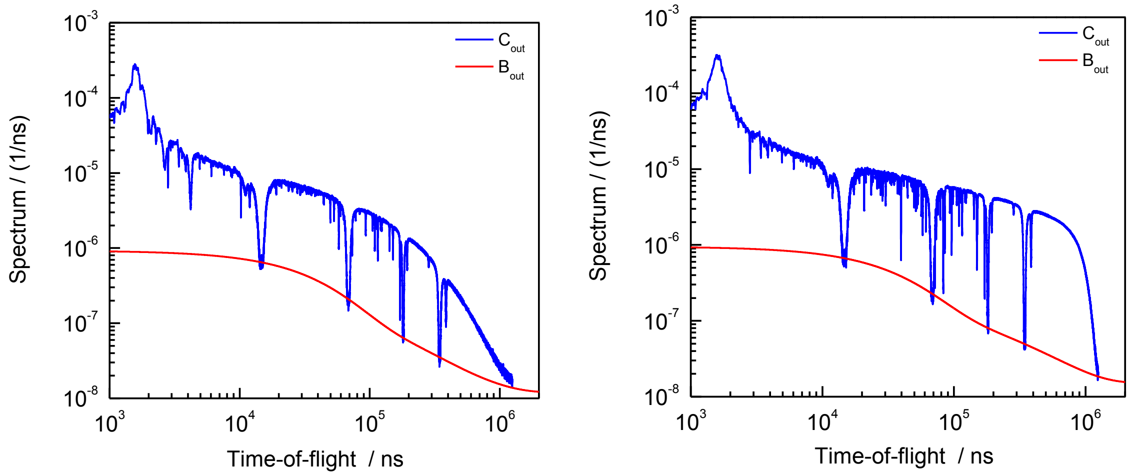

Figure 2 TOF-spectrum without a sample in the beam (C_{out}) together with the corresponding total background (B_{out}). The measurements were obtained with a B_4C (left) and a Cd (right) overlap filter and Na, Co, W and Ag background filters in the beam.

Another important characteristic of a transmission station for NRTA applications is the TOF-response function. The response function R(t,E) expresses the probability that a neutron with energy E is observed with a time-of-flight t. For TOF measurements at a moderated neutron beam one of the main components determining the broadening of the observed resonance profile is the neutron transport in the target-moderator assembly [2]. This component is best expressed in terms of an equivalent distance, as discussed in detail in Refs. [1] and [2]. The equivalent distance L_t is defined by $L_t = v\, t_t$, where t_t is the time difference between the moment the neutron leaves the target-moderator assembly and its time of creation. Response functions $R(L_t,E)$ for flight path 13 are shown in Figure 3. These functions, which were obtained from Monte Carlo simulations, show that response functions expressed as a function of equivalent distance L_t do not strongly depend on the energy of the neutron escaping from the moderator. The Full Width at Half Maximum (FWHM) of the distributions in Figure 3 is about $\Delta L_t \approx$ 3.5 cm. For a flight path forming an angle of 0^o with respect to the normal of the moderator surface facing the flight path this FWHM reduces to about $\Delta L_t \approx 2$ cm.

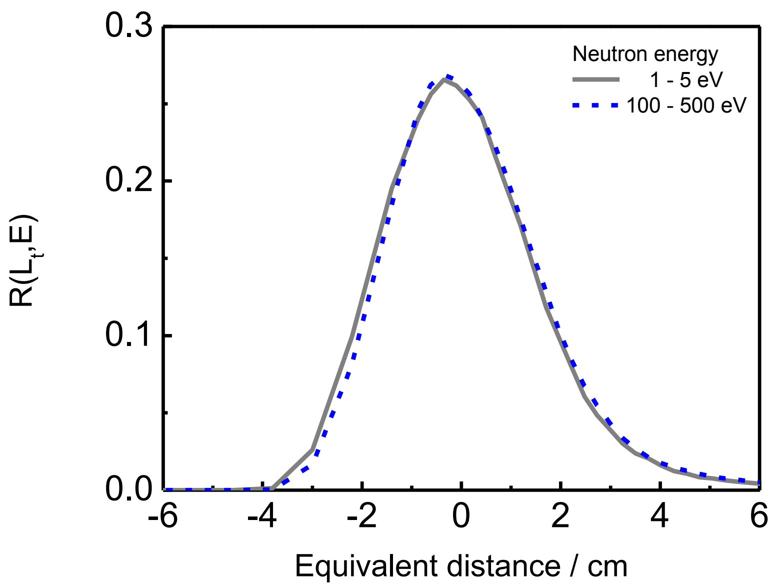

Figure 3 Probability distribution $R(L_t,E)$ of the equivalent distance L_t, which reflects the time a neutron spends in the moderator. The distribution is given for two different energy ranges for the neutron leaving the moderator.

3 Results

The 10 m transmission station at flight path 13 of GELINA was used to validate the results of previous studies [1],[9],[10]. The latter were mainly based on simulations and measurements at relatively long flight paths. Additional experiments were carried out to verify the potential of NRTA for nuclear safeguards and security applications and for the characterisation of nuclear spent fuel pellets.

3.1 Complex transmission spectra

The performance of NRTA for an analysis of complex transmission spectra obtained at a relatively short flight path was verified by measurements of complex samples consisting of B_4C, Au, Co, Mn, Nb, Rh and W [17],[18]. Examples of the transmission through such samples together with the results of a RSA analysis with REFIT [13] to determine the sample composition are shown in Figure 4 and Figure 5. The results for the corresponding areal densities of the samples are given in Table 1 and Table 2, respectively.

The sample reported in Table 1 with the transmission shown in Figure 4 contained a Co disc with a hole of about 5 mm diameter. The presence of the hole results in a black resonance transmission profile around 132 eV which does not reach the zero level. Even with the presence of this hole the areal density of the Co material was estimated within less than 1%. The effect of the presence of B_4C is illustrated in Figure 5. The influence of such matrix material can be taken into account by attributing their contribution to a dummy element with a cross section consisting of a constant and a term that is inversely proportional to the velocity of the incoming neutron, as shown in Ref. [1].

The results of these measurements demonstrate that a 10 m transmission station of GELINA can be used to characterise complex materials by NRTA. They also reveal that for an accurate analysis of such complex samples a relative energy resolution of about $\Delta E/E \approx 0.008$ is required and the spectrum should cover an energy range from about 0.5 eV to about 500 eV.

Element	n_{NRTA}	n_{REF}	n_{NRTA}/n_{REF}
Mn	$(1.886 \pm 0.002) \times 10^{-2}$	$(1.901 \pm 0.002) \times 10^{-2}$	0.992 ± 0.002
Co	$(4.550 \pm 0.066) \times 10^{-3}$	$(4.585 \pm 0.005) \times 10^{-3}$	0.992 ± 0.015
W	$(1.334 \pm 0.002) \times 10^{-3}$	$(1.337 \pm 0.001) \times 10^{-3}$	0.998 ± 0.002
Au	$(6.862 \pm 0.005) \times 10^{-3}$	$(6.884 \pm 0.007) \times 10^{-3}$	1.003 ± 0.001

Table 1 Comparison of the areal density derived by NRTA (n_{NRTA}) and the reference value (n_{REF}) for a complex sample used in a demonstration experiment reported in Ref. [17]. The uncertainties on the NRTA data are only due to counting statistics. The experimental and theoretical transmissions are compared in Figure 4.

Element	n_{NRTA}	n_{REF}	n_{NRTA}/n_{REF}
Mn	$(1.928 \pm 0.003) \times 10^{-2}$	$(1.901 \pm 0.002) \times 10^{-2}$	1.014 ± 0.002
Co	$(4.509 \pm 0.015) \times 10^{-3}$	$(4.583 \pm 0.005) \times 10^{-3}$	0.984 ± 0.003
Nb	$(5.382 \pm 0.010) \times 10^{-3}$	$(5.485 \pm 0.006) \times 10^{-3}$	0.981 ± 0.002
Rh	$(1.891 \pm 0.003) \times 10^{-3}$	$(1.856 \pm 0.002) \times 10^{-3}$	1.019 ± 0.002
W	$(2.250 \pm 0.002) \times 10^{-3}$	$(2.269 \pm 0.001) \times 10^{-3}$	0.992 ± 0.001

Table 2 Comparison of the areal density derived by NRTA (n_{NRTA}) and the reference value (n_{REF}) for a complex sample used in a demonstration experiment reported in Ref. [17]. The uncertainties on the NRTA data are only due to counting statistics. The experimental and theoretical transmissions are compared in Figure 5.

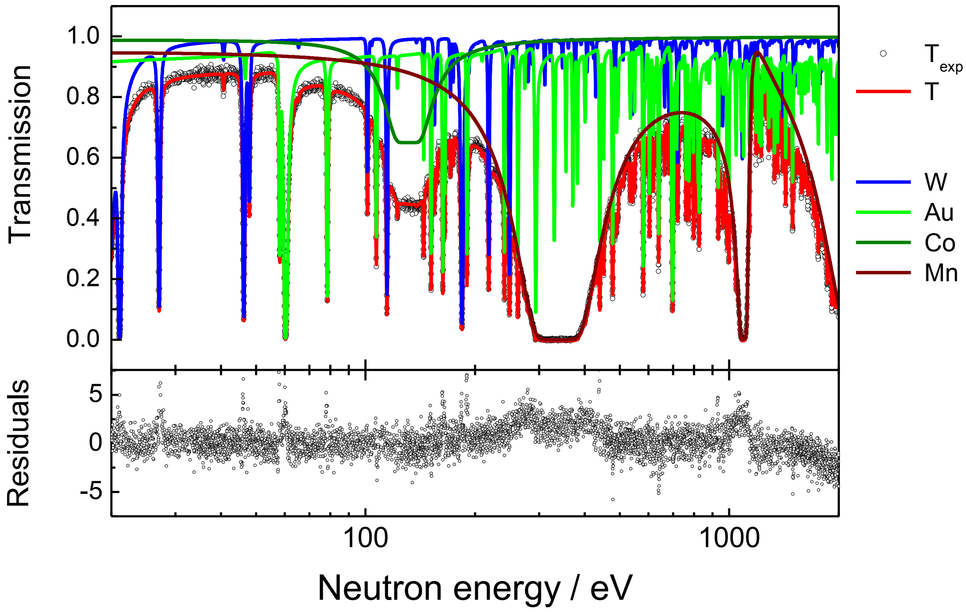

Figure 4 Results of a transmission experiment with a sample consisting of Au, Co, Mn and W. The experimental transmission (T_{exp}) is compared with the result of a RSA analysis with REFIT (T) to determine the composition of the sample. The effect of the presence of Au, Co, Mn, and W to the total transmission is also shown. The composition of the sample is given in Table 1. The Co sample had a hole of about 5 mm diameter.

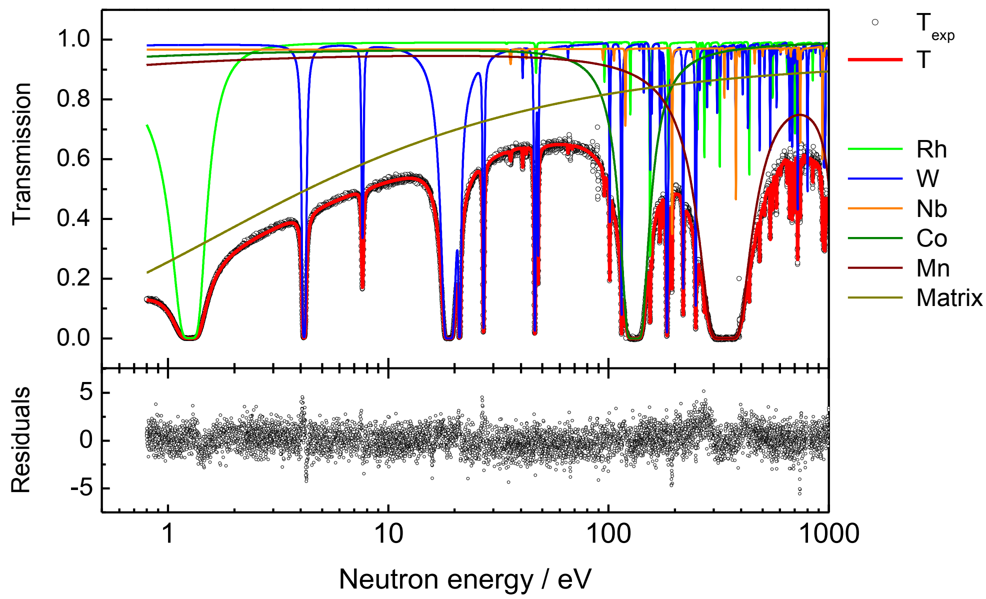

Figure 5 Results of a transmission experiment of a sample consisting of B_4C, Co, Mn, Nb, Rh and W. The experimental transmission (T_{exp}) is compared with the result of a RSA analysis with REFIT (T) to determine the composition of the sample. The effect of the presence of Co, Mn, Nb, Rh and W and matrix materials without low energy resonances, i.e. < 100 eV, is also shown. The composition of the sample is given in Table 2.

3.2 Nuclear safeguards

As shown in Ref. [1], NRTA is one of the most accurate NDA methods. Hence, it can be considered as a complementary or even alternative analytical technique to Destructive Analysis (DA). The latter requires a complex and time consuming chemical analysis of the material. To demonstrate the potential of NRTA for nuclear safeguards applications a set of U_3O_8 (CBNM NRM 171) and PuO_2 (CBNM NRM 271) reference materials were characterized by NRTA at the 10 m station of GELINA. These samples were produced and characterised at the JRC Geel. The transmission measurements are finalised and part of the data has been processed. An example of a transmission through a PuO_2 reference sample that is enriched to 63 % in ^{239}Pu is shown in Figure 6. The result of a RSA analysis with REFIT to determine the areal density of the Pu-isotopes and ^{241}Am is also shown. A comparison of the areal density determined by NRTA and the declared values is reported in Table 3. The nuclear data were taken from the JEFF-3.2 data library. Most of the differences between the values determined by NRTA and the declared ones are within the uncertainties due to counting statistics. These results demonstrate that the amount of SNM can be determined accurately by NRTA. It should be noted that the areal densities were derived without any additional adjustment of the nuclear data. Hence, systematic deviations between declared and NRTA values might be due to limitations in the nuclear data.

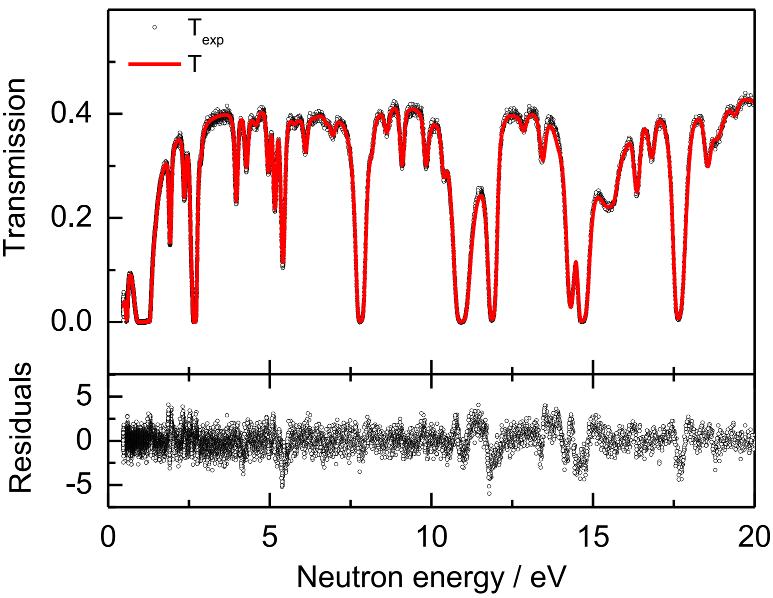

Figure 6 Transmission trough a PuO_2 sample enriched to 63% in ^{239}Pu. The experimental (T_{exp}) and theoretical transmission (T) obtained from a RSA analysis are compared.

Nuclide	n_{NRTA}	n_{REF}	n_{NRTA}/n_{REF}
^{238}Pu	0.0098 ± 0.0002	0.00950 ± 0.00002	1.031 ± 0.020
^{239}Pu	0.6250 ± 0.0010	0.62602 ± 0.00028	0.999 ± 0.002
^{240}Pu	0.2630 ± 0.0002	0.25272 ± 0.00024	1.039 ± 0.001
^{241}Pu	0.0157 ± 0.0001	0.01549 ± 0.00002	1.016 ± 0.005
^{242}Pu	0.0398 ± 0.0001	0.04149 ± 0.00006	0.960 ± 0.002
^{241}Am	0.0632 ± 0.0001	0.06300 ± 0.00063	1.003 ± 0.001

Table 3 Areal density of ^{238,239,240,241,242}Pu and ^{241}Am in a CBNM NRM 271 sample that is enriched to 63% in ^{239}Pu. The areal densities derived from a NRTA analysis are compared with the reference value. The uncertainties on the NRTA data are only due to counting statistics.

3.3 Characterisation of spent nuclear fuel

The nuclide vector of spent nuclear fuel is a crucial input to verify the performance of theoretical models for the prediction of the decay heat, reactivity and neutron and gamma-ray emission rates of spent fuel assemblies before temporary or final disposal. Using NRTA to determine the nuclide vector of spent nuclear fuel pellets would avoid the need of expensive and time consuming chemical analyses. The main problem for NRTA measurements is the cylindrical geometry of spent fuel samples. Such a geometry results in a neutron track length within the sample that varies as a function of the entrance point. To have equal track lengths of the transmitted neutrons a pencilled neuron beam would be required. This is not possible from practical point of view. Therefore, analytical models were investigated to analyse data from transmission measurements through homogeneous samples with an irregular shape [19].

To validate the analytical models transmission experiments were carried using Cu samples with different geometries. Cupper was chosen since it is a material that can be easily machined. In addition, the resonance parameters of Cu for resonances below 2 keV have been recently reviewed [20]. Since Cu has resonances above 200 eV, the measurements were carried out at a 50 m station to rule out systematic effects due to the TOF-response. For the sample configuration shown in Figure 7a the measurement data have been processed and analysed. The experimental transmission through this sample is shown in Figure 8 and compared with theoretical transmission for the different configurations shown in Figure 7. The transmission T_1 reflects the geometry of the experiment. This configuration results in a rectangular neutron track length distribution, ranging from 0 to $\sqrt{2}\,L$. To reproduce the experimental transmission one also needs to account for the part of the neutron beam that does not pass through the sample. A comparison between the experimental and theoretical transmission illustrates that the experimental data can be reproduced based on an analytical expression for the track length distribution. A comparison of the theoretical transmissions for the configuration in a) and b), denoted by T_1 and T_2, respectively, illustrates the impact of the part of the beam that is not covered by the sample. A comparison of T_2 and T_3 shows the difference in transmission resulting from a rectangular track length distribution and a fixed track length. The analysis of cylindrical pellets is in progress.

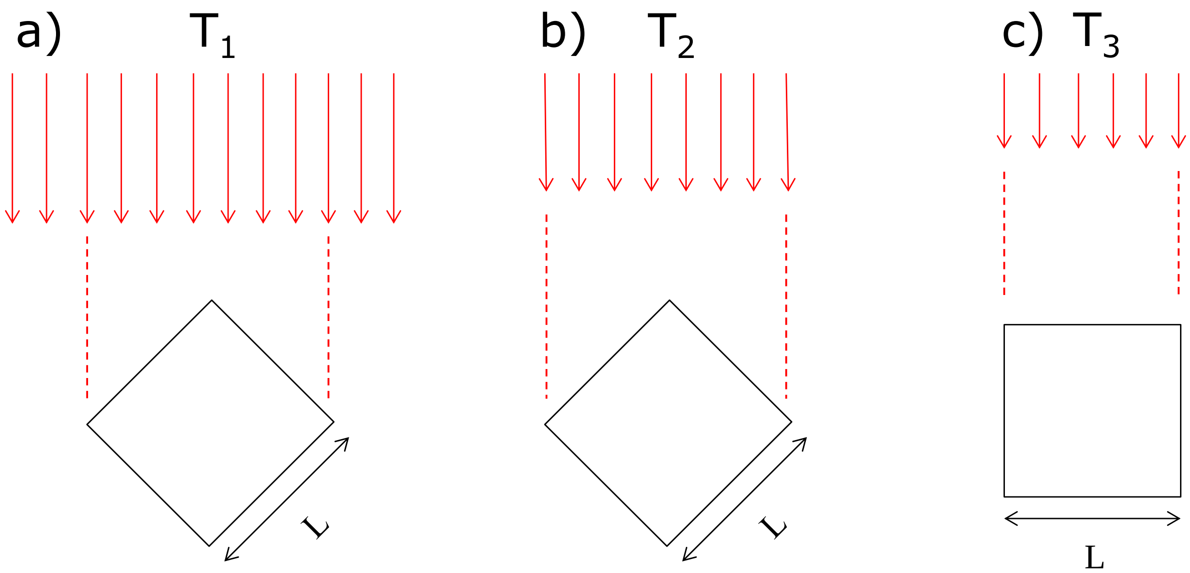

Figure 7 Geometries used for the transmission data that are shown in Figure 8

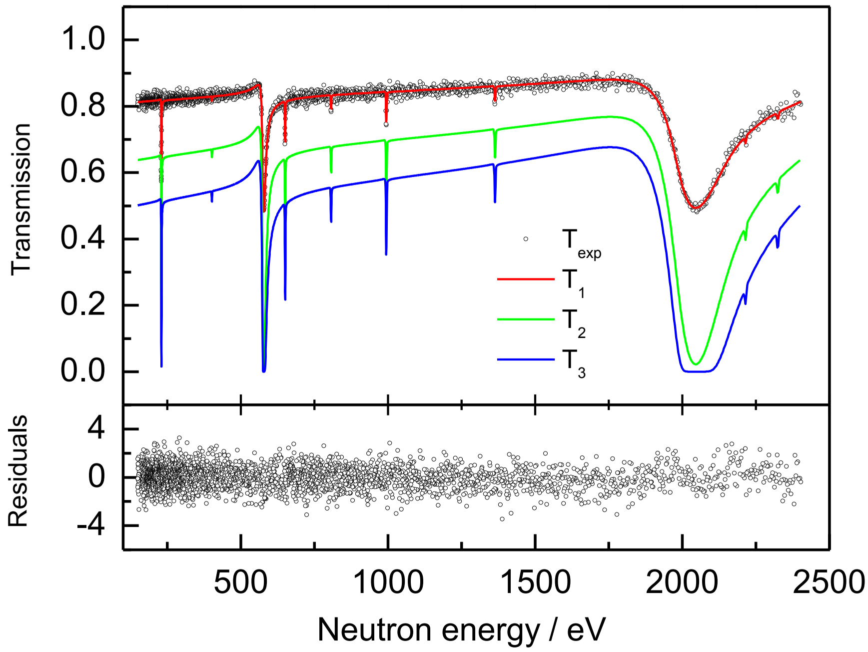

Figure 8 Experimental transmission (T_{exp}) through the sample configuration that is shown in Figure 7a. The experiments were carried out at a 50 m station of GELINA. The theoretical transmissions through the three configurations of Figure 7 are also shown.

4 System requirements

The experiments at GELINA discussed in section 3 together with the studies in Refs. [2] and [10] demonstrate that the composition of complex samples of nuclear material can be determined at a transmission station with a relatively short flight path of about 10 m and with an accelerator operating at 800 Hz. The following conclusions can be drawn:

- to analyse complex transmission data with strong overlapping resonances and to account for matrix materials without low energy resonances the spectrum should cover at least an energy region between 0.5 and 500 eV,
- the resolution of the measurements at GELINA ($\Delta E/E \approx 0.008$) was sufficient to analyse complex transmission data below 500 eV,
- the background in the whole region of interest should be less than 10%, and there should be enough shielding between the sample and detector such that neutrons scattered from the sample do not reach the detector.

Ideally the broadening of the resonance due to the resolution of the TOF spectrometer is smaller compared to the one due to the Doppler effect and the total resonance width Γ. In most cases the Doppler width (ΔE_D) is dominating (i.e. $\Delta E_D > \Gamma$). Therefore, the resolution requirements are best defined by comparing the Doppler width and the width due to the TOF-response. The Doppler width ΔE_D, expressed in FWHM, is given by:

$$\Delta E_D = 2\sqrt{\ln 2}\sqrt{\frac{4Ek_BT}{M/m}}$$

where E is the neutron energy, k_B is the Boltzman constant, T is the sample temperature and M and m denote the rest mass of the target nucleus and neutron, respectively. The relative Doppler width ($\Delta E_D/E$) at room temperature for a sample consisting of an element with mass number 100 is shown in Figure 9. This width is compared with the resolution due to the neutron moderator (ΔL_t) and due to the width of the initial pulsed beam (Δt_0) for measurements at 10 m. The resolution due to the moderator was taken as $\Delta E/E = (2 \times \Delta L_t/L) \approx 0.008$. A resolution of $\Delta E/E = 0.008$ corresponds almost to the resolution for measurements at flight path 13 of GELINA.

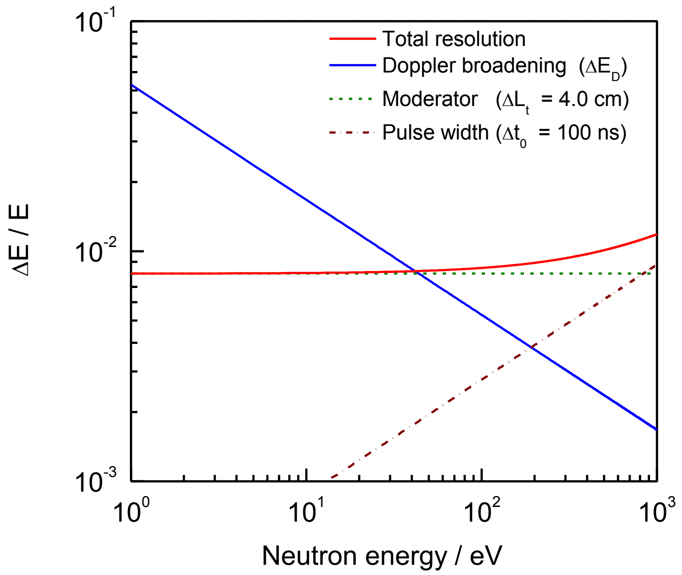

Figure 9 Comparison of different components contributing to the observed width of a resonance profile for measurements at a 10 m flight path distance. The total resolution due to the neutron transport in the moderator ($\Delta L_t = 4.0$ cm) and the pulse width ($\Delta t_0 = 100$ ns) is shown together with the Doppler broadening (ΔE_D).

One can conclude that the overall relative resolution should be less than 0.008. To fulfil this condition at a 10 m station for energies below 500 eV, the contribution of the moderator to the resolution (ΔL_t) should be less than 4.0 cm and the corresponding pulse width (Δt_0) should be less than 100 ns. For a 5 m station these conditions become: $\Delta L_t \leq 2.0$ cm and $\Delta t_0 \leq 50$ ns. These conditions can be summarised by:

$$\Delta L_t < 0.004\,L \text{ and } \Delta t_0 < \frac{L}{v_0} 0.004$$

with v_0 corresponding to the velocity of a 500 eV neutron.

An additional requirement is related to the operating frequency of the accelerator. This frequency should be adapted to the flight path length such that overlap neutrons can be removed from the beam with a Cd filter. This condition can be formulated by:

$$f_M < \frac{1}{L}\sqrt{\frac{2E_{Cd}}{m}}$$

with f_M the maximum frequency, L the flight path distance, E_{Cd} the Cd-cut off energy ($E_{Cd} \approx 0.5$ eV) and m the neutron mass.

Evidently, the beam intensity should be as high as possible. For the measurements at GELINA the neutron flux at 10 m from the target for an average current of 50 μA was about 5000 s^{-1} cm^{-2} eV^{-1} at 1 eV. The counting statistics uncertainties quoted in Table 1 and Table 2 correspond to a 12h measurement time in these conditions. Hence, these data can be used to estimate uncertainties due to counting statistics. To optimise a system in terms of beam intensity, one needs to consider that the beam intensity is inversely proportional to the square of the flight path distance and mostly directly proportional to both the frequency and pulse width. Given the requirements for the pulse width and frequency their impact on the intensity will compensate. Therefore, the intensity will be primarily determined by the distance that is chosen to fulfil the condition $\Delta L_t < 0.004$ L.

5 Summary and conclusions

Requirements for a compact measurement station for NRTA on complex nuclear materials have been defined. Some of these requirements result from experiments that were carried out previously at a 25 m and 50 m station of GELINA. For a final definition and validation of the requirements measurements were performed at a 10 m station of GELINA. The results of these measurements revealed that a characterisation of complex materials is feasible at a relatively short flight path, provided that the total relative energy resolution is better than $\Delta E/E \approx 0.008$. In addition, it was demonstrated that NRTA can be considered as a non-destructive, absolute and accurate analytical technique to determine the amount of SNM for nuclear safeguards and security applications and for the characterisation of spent nuclear fuel material.

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List of abbreviations and definitions

EC	European Commission
CBNM	Central Bureau of Nuclear Measurements
DA	Destructive Analysis
GELINA	Geel Electron LINear Accelerator
JAEA	Japan Atomic Energy Agency
JRC	Joint Research Centre
NDA	Non-Destructive Analysis
NRCA	Neutron Resonance Capture Analysis
NRM	Nuclear Reference Material
NRTA	Neutron Resonance Transmission Analysis
RSA	Resonance Shape Analysis
SNM	Special Nuclear Material
TOF	Time-Of-Flight

List of figures

List of tables

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