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Characterization of nuclear material by Neutron Resonance Transmission Analysis

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Summary. — The use of Neutron Resonance Transmission Analysis for the characterization of nuclear materials is discussed. The method, which relies on resonance structures in neutron-induced reaction cross sections, can be applied as a non-destructive method to characterise complex nuclear materials such as melted fuel resulting from a severe nuclear accident. Results of a demonstration experiment at the GELINA facility reveal that accurate data can be obtained at a compact facility even in the case of strong overlapping resonances.

PACS 81.70.-q – Methods of materials testing and analysis.
PACS 24.30.-v – Resonance reactions.
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1. – Introduction

Neutron Resonance Transmission Analysis (NRTA) and Capture Analysis (NRCA) are non-destructive methods to determine the elemental and isotopic composition without the need of any sample preparation [1, 2]. They are based on the presence of resonance structures in neutron induced reaction cross sections [2] and rely on well established methodologies for neutron induced reaction cross section measurements [3].

NRCA has been used extensively at the time-of-flight facility GELINA of Joint Research Centre at Geel (BE) [4] to study objects of cultural heritage interest [1, 3].

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The use of NRCA for the characterisation of reference samples including nuclear material is discussed in [5]. The potential of NRTA as a NDA method for the quantification of Special Nuclear Material (SNM), *i.e.* Pu-isotopes and ^{235}U, in fresh and spent fuel has been demonstrated by Priesmeyer and Harz [6] and Behrens *et al.* [7]. Noguere *et al.* [8] applied NRTA to characterize a PbI_2 sample that was produced from a solution of nuclear waste from the reprocessing facility in Le Hague. An extensive overview of NRTA and NRCA applications is given in [2].

Neutron Resonance Densitometry (NRD) is being developed as a method to quantify rock- and particle-like debris of melted fuel formed in severe nuclear accidents [9, 10]. NRD is based on NRTA combined with NRCA. The quantification of SNM is based on NRTA, while NRCA using a detector with a good γ-ray energy resolution is applied to determine the amount of impurities. A conceptual design of a NRD facility has been presented by Harada *et al.* [9]. The analysis of rock- and particle-like debris samples is challenging due to the characteristics of the samples: *i.e.* the diversity in shape and size of the particle like samples, the radioactivity and temperature of the sample, presence of unknown matrix material including strong neutron absorbers and overlapping of resonances [10]. The impact of the diversity in particle size has been investigated by Becker *et al.* [11]. They implemented the Levermore-Pomraning model [12] in the resonance shape analysis (RSA) code REFIT [13] and validated this method for NRTA by Monte Carlo simulations and experiments at GELINA [11]. A method to account for the contribution of strong absorbing light matrix materials that do not have resonances in the low energy region has been proposed and validated in [3].

For industrial applications a compact NRTA system is considered. In this contribution the performance of NRTA as an absolute NDA method for the characterization of complex nuclear materials at a short flight path station is demonstrated.

2. – Basic principles of NRTA

The quantity of interest for NRTA is the fraction of the neutron beam traversing the sample without any interaction. For a parallel neutron beam that is perpendicular to a slab of material, this fraction or transmission T is given by

$$T = \exp\left[-\sum_k n_k \overline{\sigma}_{\mathrm{tot},k}\right], \tag{1}$$

where $\overline{\sigma}_{\mathrm{tot},k}$ is the Doppler broadened total cross section and n_k is the number of atoms per unit area of nuclide k. When the total cross sections are known, a measurement of the transmission can be used for elemental and isotopic analysis.

The experimental transmission T_{exp} is derived from the ratio of the counts of a sample-in measurement and a sample-out measurement, both corrected for losses due to the dead time in the detector and electronics chain and their background contributions. Hence, the experimental transmission is independent of the detector efficiency and incoming neutron flux and no additional calibration measurements are needed. In addition, the experimental transmission is a direct measure of the theoretical transmission eq. (1) if the measurements are performed in a good transmission geometry, which can be achieved by a proper collimation of the neutron beam at both the sample and detector position [3]. A good transmission geometry results in a small solid angle subtended by the sample and neutron detector such that the background due to the radioactivity of the sample and

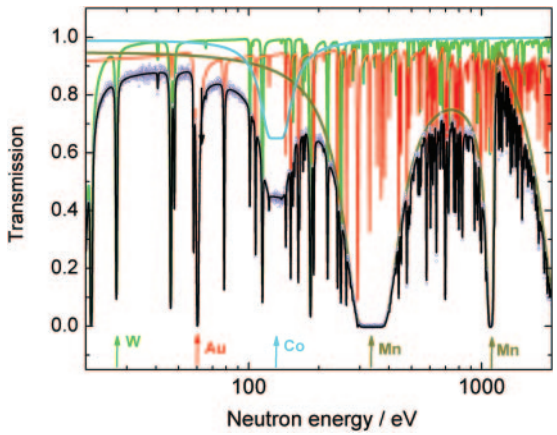

Fig. 1. – Transmission through a blind sample with the composition given in table I. The experimental transmission is compared with the result of a RSA. The contribution due to the different elements present in the sample is also shown.

fission neutrons produced by (n, f) reactions in the sample can be neglected. Finally, the areal density of the nuclides present in the sample can be derived by a least squares fit to the experimental data. Such a fit relies on a theoretical estimate of the transmission which is derived from a folding of the theoretical transmission in eq. (1) with the response function of the TOF-spectrometer [3].

3. – Results

To investigate the potential of NRTA for complex nuclear materials at a compact transmission station a demonstration experiment was organized at a 10 m station of GELINA. An inventory of 18 different samples of medium-weight (Cu, Co, Mn, Nb and Rh) and heavy elements (W and Au) in the form of metallic discs were made available. These elements were chosen to mimic the resonance structures of elements present in nuclear fuel assemblies. In addition, strong neutron absorbing B_4C samples were made available. A sample box was constructed that could host up to 8 samples. A nuclear inspector from DG-ENER and a staff member of ORNL (US) were asked to assemble a test sample. The box was closed and sealed by the DG-ENER inspector.

After a one night measurement the transmission data were analysed by REFIT using

TABLE I. – *Results of a NRTA analysis of a blind sample. The measurements were performed at a 10 m GELINA station. The uncertainties on the NRTA data are only due to counting statistics.*

Element	Areal number density (at/b)		Ratio
	Declared (C)	NRTA (E)	(C/E)
Mn	$1.901(0.002) \times 10^{-2}$	$1.886(0.002) \times 10^{-2}$	1.008(0.002)
Co	$4.585(0.005) \times 10^{-3}$	$4.550(0.066) \times 10^{-3}$	1.008(0.015)
W	$1.337(0.001) \times 10^{-2}$	$1.334(0.002) \times 10^{-2}$	1.002(0.002)
Au	$6.844(0.007) \times 10^{-3}$	$6.862(0.005) \times 10^{-2}$	0.997(0.001)

the cross section data in the JEFF-3.2 library. The result of a least squares fit to the data is shown fig. 1. A comparison of the areal density derived by NRTA and the declared values is shown in table I. These data reveal that the amount of the elements present in the sample was predicted within 1%. Evidently the accuracy of NRTA strongly depends on the quality of the nuclear data [3,14]. It should be noted, that for the analysis of NRTA data only total cross sections are required and the uncertainties on total cross sections are mostly considerably smaller compared to uncertainties on partial cross sections. Since NRTA can also be considered as an absolute method, it is one of the most accurate NDA methods for the characterization of nuclear materials.

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