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Reference samples for NRA applications

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Reference samples for NRA applications

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Contents

Abstract

Reference materials for Neutron Resonance Analysis (NRA) applications are discussed. This non-destructive analysis technique can be applied for a variety of applications, such as: validation of nuclear data, characterisation of reference samples and complex nuclear materials and archaeological and cultural heritage studies. The technique requires a pulsed white neutron source combined with time-of-flight measurements. The characteristics and use of reference samples that are available at JRC Geel to calibrate NRA systems and assess their performance are described.

1 Introduction

The presence of resonance structures in neutron interaction cross sections can be used to study materials and objects by Neutron Resonance Analysis (NRA). Resonances appear at energies that are specific for each nuclide. Therefore, they are ideal fingerprints to determine the elemental and isotopic composition of materials. This is the basis of Neutron Resonance Transmission Analysis (NRTA) and Neutron Resonance Capture Analysis (NRCA) [1]. NRTA and NRCA are Non-Destructive Analysis (NDA) techniques to determine the elemental and isotopic composition of materials and objects. Both methods are non-invasive and do not require any sample taking or preparation. They require a pulsed white neutron source combined with the Time-Of-Flight (TOF) technique and rely on well-established methodologies that are applied for neutron interaction cross section measurements [2].

At present NRTA and NRCA are applied at relatively large neutron TOF facilities such as the GELINA facility of the Joint Research Centre (JRC) in Geel (Belgium) [3] and the ANNRI measurement station installed at the J-PARC/MLF facility of the Japan Atomic Energy Agency (JAEA) in Tokai-mura (Japan) [4]. These facilities have primarily been constructed to produce nuclear data for nuclear technology applications. They are not foreseen for routine characterisation of materials and objects by NRTA or NRCA. For such routine applications a more compact facility can be designed. Specific requirements and design specifications of a compact NRA system can be found in Ref. [5] and [6]. A compact facility can be built based on a 30 MeV linear accelerator producing a pulsed electron beam with a width of less than 100 ns and average current of about 10 μA combined with flight path lengths of about 8 m. Once such a facility is in operation, its performance has to be assessed by reference materials. In this report reference materials that can be used for a performance assessment and calibration of NRTA and NRCA systems are described. Examples of reference materials available at the JRC Geel together with their use at GELINA are given.

2 NRTA and NRCA techniques

NRTA and NRCA are based on methodologies that are applied for studying neutron induced interaction cross sections in the resonance neutron energy region [2]. NRCA refers to an analysis of the resonance structures in the spectra obtained by recording prompt γ-rays that are emitted after a neutron capture reaction in the sample. NRTA is based on the detection of neutrons that are transmitted through a sample without any interaction in the sample. A detailed description of NRTA and NRCA can be found in Ref. [1] and [7].

Important advantages of NRCA and NRTA are that:

- they are non-invasive and determine the bulk composition;
- they do not require to take or prepare samples as other methods like Neutron Activation Analysis (NAA) or atomic spectroscopy methods;
- the induced radioactivity is low and mostly well below the limit for free release after a short waiting period; and
- relatively large objects can be tested.

When transmission measurements are carried out the areal density and mass ratios are obtained in an absolute way, without the need of any calibration measurement [1]. In addition, for an analysis of the data only total cross sections are required and uncertainties of total cross sections are in general considerably smaller compared to those of reaction cross sections. Therefore, NRTA can be considered as one of the most accurate NDA methods. However, for the majority of elements NRCA has a more favorable detection limit compared to NRTA. The difference is roughly a factor ten. Therefore, NRCA is more suitable to identify and quantify impurities and trace elements compared to NRTA.

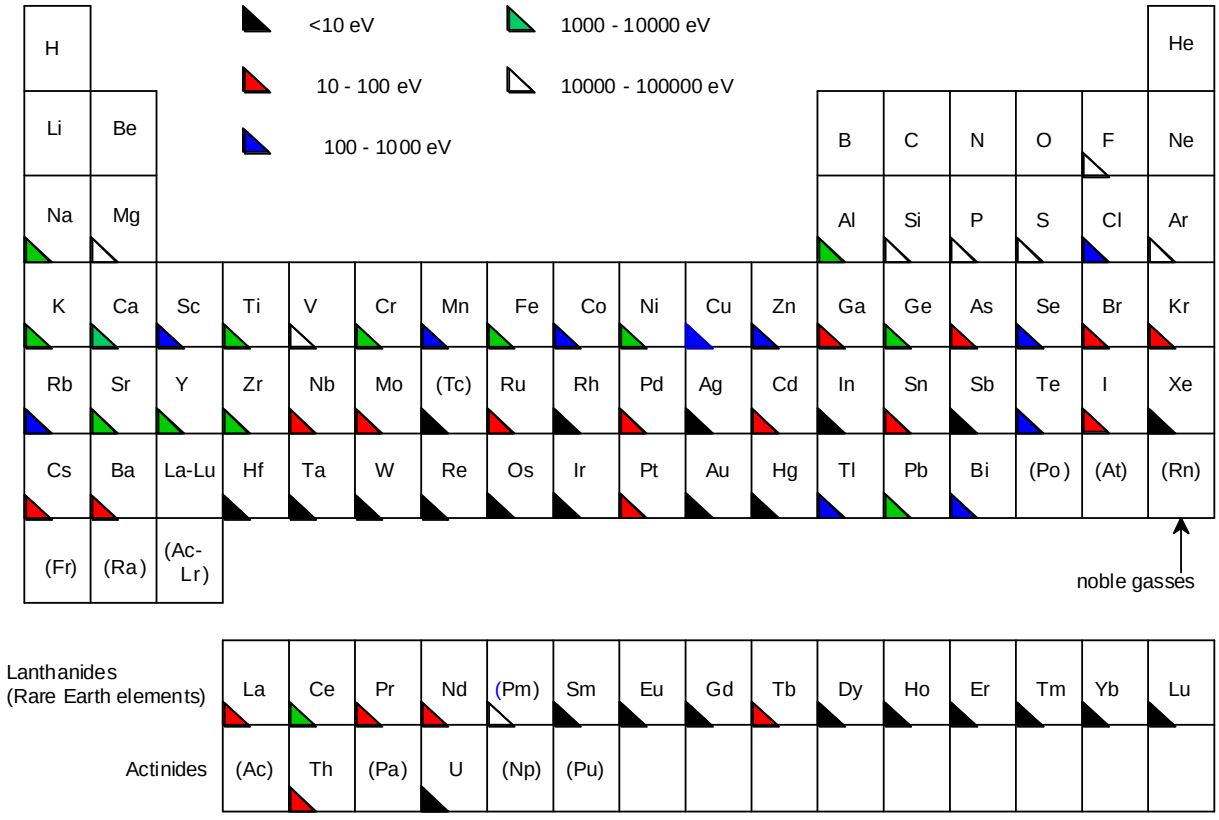

Figure 1. Periodic table with an indication of the elemental sensitivity for an analysis by NRA. The sensitivity is based on the resonance strength: black for elements with resonances below 10 eV, grey for elements with resonances between 10 and 100 eV etc. (figure taken from Ref. [7]).

The sensitivity to detect elements by NRCA is discussed in Ref. [7] and represented in Figure 1. This sensitivity depends on both the energy dependence of the cross section and neutron fluence rate. For elements with resonances between 1 eV and 10 eV the detection limit is in the range of parts per million (ppm). When resonances have energies between 10 eV and 500 eV the lowest detectable mass fraction is between 10^{-3} and 10^{-5}. Detection limits for resonances in the keV region are in the order of one per cent. Figure 1 indicates for each element the energy region in which resonances are present that are suitable for NRA. The schematic representation in Figure 1 reveals that NRA is not very suitable to detect small quantities of light elements and of elements with a proton number close to a magic shell. For such elements Prompt Gamma-ray Analysis (PGA) is a better alternative. In general PGA and NRA are very complementary, as discussed in Ref. [8]. If one of the elements present in the sample has a dominating capture cross section, NRCA offers a considerable advantage compared to PGA. The γ-ray spectrum observed in a PGA experiment will be dominated by the presence of the element with the largest capture cross section. In the TOF-spectrum obtained from a NRCA measurement it is possible to select regions where the elements with a small contribution are not hampered by resonances of other elements with a large capture yield.

Moxon et al. [9] were able to quantify the presence of Ta and W impurities in a Hf sample by NRA at a ~100 ppm and ~500 ppm level, respectively. Due to the large thermal capture cross section of Hf isotopes such impurities cannot be determined by PGA. Similarly, using the 1.45 eV resonance of indium this element has been observed in bronze artefacts down to a few 10's of ppm by Postma et al. [10], as illustrated in Figure 2. This figure shows a TOF-spectrum obtained with a bronze vessel at a 12.8 m capture station of GELINA. In the TOF-region between 700 μs and 850 μs the detected events originate predominantly from the 1.45 eV resonance of ^{115}In. In this TOF-region there is a negligible contribution from neutron capture of Cu, which is the element with the largest abundance in the object.

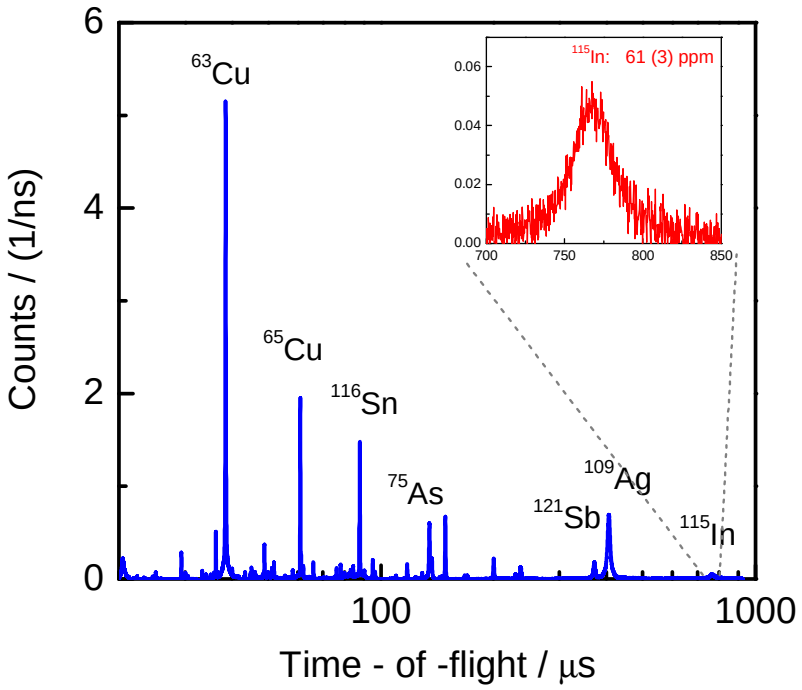

Figure 2 TOF-spectrum obtained from capture measurements of a cauldron sample at a 12.8 m capture station of GELINA. The relative amount of In 61 (3) ppm was derived from the resonance at 1.45 eV (figure based on data from Ref. [10]).

3 NRTA and NRCA applications

NRTA and NRCA can be applied for a variety of applications. They range from characterisation of reference materials and complex nuclear materials, archaeological studies, nuclear safeguards, combatting illicit traffic of nuclear material to the study of fundamental properties of materials [1]. In this section examples of NRTA and NRCA applications are discussed to better understand the need and requirements of reference samples for NRTA and NRCA applications.

Most of the results are derived from a Resonance Shape Analysis (RSA) using the REFIT code [11]. This code, which is based on the Reich-Moore approximation [12] of the R-matrix formalism [13], accounts for various experimental effects such as Doppler broadening, neutron self-shielding, multiple interaction events, sample and detector characteristics and the response function of the TOF-spectrometer. In the analysis the sample characteristics are considered as free parameters and adjusted to the experimental data.

3.1 Characterisation of reference materials for cross section measurements

Accurate cross section data require measurements with homogeneous samples for which the areal density of the nuclides present in the sample is accurately known. Evidently, NRCA is a suitable technique to identify and quantify impurities with a relatively high neutron capture cross section in the resonance region, in particular when capture cross section data with a low uncertainty are requested.

For example, the thermal capture cross section for ^{103}Rh is requested with an uncertainty of less than 2% [14]. This requires capture experiments with ^{103}Rh samples that are very well characterised for the presence of impurities with a large capture cross section. Therefore, NRCA measurements were carried out to identify and quantify impurities that could contribute to the response of the capture detection system [15]. Results of such an analysis by NRCA are summarised in Table 1. The impurities listed in Table 1 contribute for about 0.5 % to the observed experimental yield at an incident neutron energy of 25.3 meV.

Table 1. Results of the characterisation of a ^{103}Rh metal disc by NRCA. The weight ratios (w_X/w_{Rh}) for ^{181}Ta, ^{191,192}Ir, ^{182,183,186}W and ^{197}Au relative to the one of ^{103}Rh are given together with the contribution of each nuclide to the experimental yield Y_{exp} at 25.3 meV relative to the one of ^{103}Rh. The uncertainties result from propagating only uncorrelated components due to counting statistics (the results are taken from Ref. [15]).

Nuclide	Weight ratio $100 \times w_X/w_{Rh}$	Relative Y_{exp} at 25.3 meV $100 \times Y_X/Y_{Rh}$
^{103}Rh	100	100
^{181}Ta	0.034 (3)	0.003
^{191}Ir	0.099 (4)	0.310
^{193}Ir	0.170 (9)	0.060
^{182}W	0.063 (3)	0.004
^{183}W	0.034 (3)	0.001
^{186}W	0.069 (3)	0.009
^{197}Au	0.006 (1)	0.002

3.2 Validation of nuclear data

With the areal density as the derived integral quantity, NRTA is a valuable tool to assess the quality of cross section data in the Resolved Resonance Region (RRR) that are recommended in evaluated nuclear data libraries. The experimental observable is the transmission through a homogeneous sample that is characterised for the areal density of each nuclide present in the sample. When a metallic sample can be produced for an element with a well-known and stable natural isotopic composition, the reference areal density can be derived from its weight and area with an uncertainty ≤ 0.1%. Since a transmission measurement is an absolute measurement [2], NRTA using such samples is one of the most accurate integral experiments to validate resonance parameters.

Examples of such a validation of resonance parameters are reported by Becker et al. [16] and Tsuchiya et al. [17]. Becker et al. [16] performed transmission measurements on a 0.15 mm thick metal disk of ^{nat}W at a 25 m station of GELINA. Their results are summarised in A similar exercise to verify the quality of resonance parameters for Cu has been carried out by Tsuchiya et al. [17]. They performed transmission measurements at a 25 m station of GELINA using metallic discs of ^{nat}Cu with different thicknesses (i.e. 0.125 mm, 0.25 mm, 0.7 mm and 20 mm). The results indicate clear shortcomings in the resonance parameter files recommended by e.g. Mughabghab [19] and Sobes et al. [20].

Table 2. The table shows a comparison of the areal densities obtained from an adjustment to the experimental transmission using resonance parameters recommended in the main evaluated data libraries. Note that the resonance parameters in ENDF/B-VII.1 are taken from JENDL-3.3. They are also adapted in JENDL-4.0. The ratio of the fitted areal density of ^{183}W and the reference value is given together with the parameters used for the fit. The uncertainty of the fitted areal density obtained with the JEFF-3.2, ENDF/B-VI.8 and JENDL-3.3 parameters is due to propagating only uncorrelated components due to counting statistics. For the result obtained with ENDF/B-VII.1 the covariance matrix of the resonance parameter recommended in ENDF/B-VII.1 (see Trkov et al. [18]) was propagated. The results in A similar exercise to verify the quality of resonance parameters for Cu has been carried out by Tsuchiya et al. [17]. They performed transmission measurements at a 25 m station of GELINA using metallic discs of ^{nat}Cu with different thicknesses (i.e. 0.125 mm, 0.25 mm, 0.7 mm and 20 mm). The results indicate clear shortcomings in the resonance parameter files recommended by e.g. Mughabghab [19] and Sobes et al. [20].

Table 2 reveal that only using the parameters recommended in JEFF-3.2 there is a good agreement between the areal density derived by NRTA and the reference value. The difference between the reference value and the one derived by NRTA using the ENDF/B-VII.1 parameters is not covered by the uncertainty of the resonance parameters. Hence, both the covariance matrix reported by Trkov et al. [18] are not reliable.

A similar exercise to verify the quality of resonance parameters for Cu has been carried out by Tsuchiya et al. [17]. They performed transmission measurements at a 25 m station of GELINA using metallic discs of ^{nat}Cu with different thicknesses (i.e. 0.125 mm, 0.25 mm, 0.7 mm and 20 mm). The results indicate clear shortcomings in the resonance parameter files recommended by e.g. Mughabghab [19] and Sobes et al. [20].

Table 2. Radiative (Γ_γ) and neutron (Γ_n) widths of the 46.62 eV (1) and 47.8 eV (2) resonances of ^{183}W recommended in evaluated data libraries are given together with the ratio of the areal density n_{NRTA} derived by NRTA when using each library and the reference value (n_{ref} = 9.145 x 10^{-4} at/b) (the data are taken from Ref. [16]).

Library	Γ_n (1)	Γ_γ (1)	Γ_n (2)	Γ_γ (2)	100 x n_{NRTA}/n_{ref}	
JEFF-3.2	163.4	75.3	120.8	61.5	100.2	(5)
ENDF/B-VI.8	154.0	69.0	115.0	78.0	109.7	(5)
JENDL-3.3	154.0	46.0	119.0	81.0	111.3	(5)

ENDF/B-VII.1	154.0 (8)	46.0 (21)	119.0 (12)	81.0 (51)	111.3 (11)

3.3 Characterisation of nuclear waste

The composition of a PbI_2 pressed powder pellet was characterised by NRTA at GELINA [21]. This pellet was produced from a solution of radioactive nuclear waste originating from the French reprocessing facility at La Hague [22]. Representative samples were also analysed by mass spectrometry and Neutron Activation Analysis (NAA). The data reported in Table 3 show that there is a very good agreement between the results obtained with the different techniques. They also illustrate that by applying NRTA the isotopic composition of I and Pb was derived. In addition, NRTA shows to be very useful for the determination of the relative amount of Na, O, S and Pb.

Schrack [23][24] applied NRTA to determine the amount of ^{235}U in simulated nuclear waste that consisted of incinerator ash as matrix material and small concentrations of ^{235}U (between 4.8 x 10^{-4} g/cm^3 and 4.6 x 10^{-3} g/cm^3). The technique was demonstrated on sample sizes ranging from 2 l bottles to 200 l drums.

Table 3. Elemental and isotopic composition of a PbI_2 sample that was produced from radioactive waste originating from the reprocessing facility at La Hague (F). The results obtained by NRTA, mass spectrometry and NAA are compared (the data are taken from Ref. [21]).

Element	Isotope	Abundance (wt%)					
		NRTA		Mass spectrometry		NAA	
I		20.24	(7)	19.86	(9)		
	^{127}I	3.44	(5)	3.36	(8)	3.35	(10)
	^{129}I	16.8	(4)	16.5	(4)		
Pb		52.3	(14)	59.47	(18)	51.1	(18)
	^{204}Pb	0.8					
	^{206}Pb	12.8	(5)				
	^{207}Pb	11.5	(1)				
	^{208}Pb	27.1	(17)				
O		13.92	(5)	14.5	(15)		
S	^{32}S	5.17	(3)				
Na	^{23}Na	0.72	(2)	1.00	(15)		
N				1.2	(4)		
H		< 0.13		0.020	(2)		

3.4 Characterisation of nuclear fuel

The potential of NRTA for a quantitative non-destructive analysis of fresh and spent nuclear fuel has already been demonstrated by Priesmeyer et al. [25]. Low- and high-enriched MTR fuel plates with different burnups were characterised by transmission measurements. These measurements were carried out at a fast-chopper TOF-spectrometer installed at a research reactor. The areal densities of ^{131}Xe, ^{133}Cs, ^{152}Sm, $^{235,238}U$ and ^{239}Pu were derived from transmission profiles for neutron energies below 20 eV.

The expected transmission through a 2.5 cm thick spent fuel sample is shown in Figure 3 as a function of the incident neutron energy. The sample composition is representative for BWR fuel initially enriched to 3.2 wt% in ^{235}U with a burn up of 25 GWd/MTU. The contribution of the different nuclides present in the sample is illustrated by plotting

separately the transmission due to the presence of only the fission products (i.e. ^{99}Tc, ^{131}Xe, ^{133}Cs, ^{145}Nd, ^{152}Sm), ^{241,243}Am, ^{234,236,238}U, ^{235}U, ^{240,242}Pu and ^{239,241}Pu. This figure illustrates that the transmission for such materials is complex due to overlapping resonance profiles resulting from the presence of fission products and main and minor actinides. Hence, the accuracy of the results will strongly depend on the quality of the resonance parameters of the overlapping resonances and the resolution of the TOF spectrometer.

When fresh or irradiated fuel pellets have to be characterised, the basic Lambert-Beer law, which is valid for samples with a constant thickness seen by the neutron beam, is not valid due to the cylindrical shape of the pellets. In addition, it will be difficult to design a geometry that avoids that part of the detected neutrons pass next to the pellet. This part is also referred to as the holes fraction. Therefore, not all conditions for a good transmission geometry will be fulfilled. For the analysis of transmission measurements using pellet samples the Lambert-Beer law has to be replaced by an expression that accounts for the holes fraction and the variation in neutron track length in the sample is [26]:

$$T(E) = f_h + (1 - f_h) \int_0^1 e^{-x \, \Sigma_k n'_k \bar{\sigma}_{tot,k}(E)} \frac{x}{\sqrt{1-x^2}} dx, \qquad (1),$$

with $\bar{\sigma}_{tot,k}(E)$ the Doppler broadened total cross section for neutron interactions with nuclide k, f_h the holes fraction, n'_k the volume number density of nuclide k multiplied by the pellet diameter, i.e. the areal number density corresponding to the maximum pellet thickness.

In the analysis, the quantities f_h and n'_k are the adjustable parameters. The accuracy of the fitted parameters will strongly depend on the resolution of the TOF spectrometer and the background conditions. To avoid bias effects due to the holes fraction an analysis of saturated resonance dips is required, preferably by comparing such a dip due to a strong neutron absorber in the sample and due to the presence of a black resonance filter that covers the full beam. To test the performance of the model reference pellets made from an element with well-known resonance parameters are required. Therefore, cylindrical Cu rods were produced and used to validate the expression in Eq. 1 by Ma et al. [26].

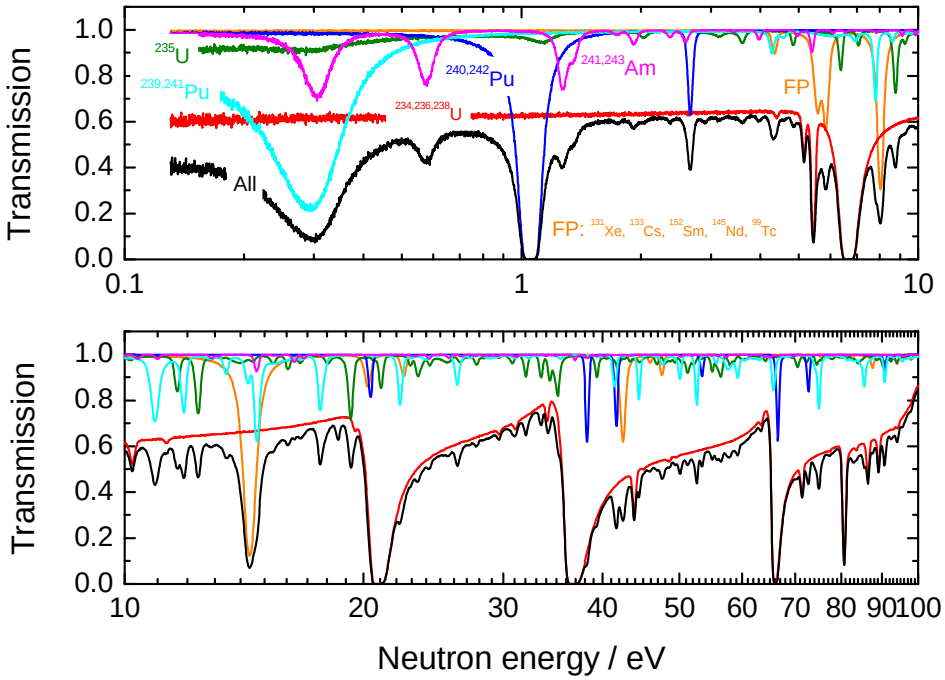

Figure 3. Calculated transmission through a 2.5 cm thick spent fuel sample with a temperature T = 400 K. The composition is representative for BWR fuel initially enriched to 3.2 wt% in ^{235}U with a burnup of 25 GWd/MTU. The contribution of the different nuclides present in the sample is

illustrated by plotting separately the transmission due to the presence of only the fission products (FP), ^{241,243}Am, ^{234,236,238}U, ^{235}U, ^{240,242}Pu and ^{239,241}Pu (the data are taken from Ref. [1]).

3.5 Characterisation of melted fuel

NRTA is being developed as a method to quantify special nuclear material in particle-like debris of melted fuel formed in severe nuclear accidents [27]. The objective is to derive the amount of fissile material, i.e. ^{235}U and ^{239}Pu, with a 2% standard uncertainty at a 68% confidence limit. To realise this target value is challenging due to the characteristics of such samples.

The analysis of the transmission profiles will be complicated due to [26]:

- the variety in shape and size of the particle like debris samples;
- the presence of neutron absorbing impurities which cannot be quantified by low energy resonances (e.g. ^{10}B);
- the radioactivity and temperature of the sample; and
- the overlapping of resonance profiles.

To study the impact of these effects experiments were carried out at a transmission station with a 10 m flight path installed at GELINA [28]. In addition, models to account for the sample characteristics and presence of matrix material were developed and validated using reference samples available at JRC Geel [1][29][30].

Becker et al. [29][30] implemented the Levermore-Pomraning model [31] in REFIT [11] to account for the diversity in shape and size of the samples. The model was validated by Monte Carlo simulations [29] and experiments at GELINA [30]. A method to account for the contribution of strong absorbing light matrix materials that do not have resonances in the low energy region was proposed in Ref. [1]. This method was validated by experiments using the CBNM U_3O_8 reference samples presented in section 4.2.

To investigate the potential of NRTA for complex nuclear materials with strong overlapping resonances at a compact facility, demonstration experiments were organised 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. One of the Co samples contained a hole. These samples were chosen to mimic the resonance structures of elements present in nuclear fuel assemblies. A sample box was constructed that could host up to 8 samples. Representatives of DG-ENER, IAEA, DOE and ORNL were asked to assemble two test samples which were closed and sealed. The content of the sealed boxes was determined by NRTA. The results of these demonstration experiments are given in Table 4. The areal densities derived by NRTA and the declared values are compared. The data in Table 4 reveal that the areal densities are reproduced within 2%. In addition, from an analysis of the transmission close to the 132 eV resonance of ^{59}Co it was concluded that the second box contained a Co sample with a hole. The results of these blind tests are discussed in detail in Ref. [32] and [33].

Table 4 Comparison of the reference sample composition (n_{ref}) and the composition derived from NRTA (n_{NRTA}). The ratio between the two values is given in the last column. The uncertainties on the NRTA data are only due to the propagation of uncorrelated uncertainties due to counting statistics. All uncertainties are quoted at 1 standard deviation (the data are taken from Ref. [32] and [33]).

	Element	Areal number density (at/b)		n_{NRTA}/n_{ref}
		Declared (n_{ref})	By NRTA (n_{NRTA})	
Sample 1	Mn	1.901 (2) x 10^{-2}	1.928 (3) x 10^{-2}	1.014 (2)
	Co	4.583 (5) x 10^{-3}	4.509 (15) x 10^{-3}	0.984 (3)

	Nb	5.485 (6) x 10^{-3}	5.382 (10) x 10^{-3}	0.981 (2)
	Rh	1.856 (2) x 10^{-3}	1.891 (3) x 10^{-3}	1.019 (2)
	W	2.269 (1) x 10^{-3}	2.250 (2) x 10^{-3}	0.992 (1)
Sample 2	Mn	1.901 (2) x 10^{-2}	1.886 (2) x 10^{-2}	0.992 (2)
	Co	4.585 (5) x 10^{-3}	4.550 (66) x 10^{-3}	0.992 (15)
	W	1.337 (1) x 10^{-2}	1.334 (2) x 10^{-2}	0.998 (2)
	Au	6.844 (7) x 10^{-3}	6.862 (5) x 10^{-3}	1.003 (1)

3.6 Archaeology and cultural heritage

In the course of several years NRCA has been applied at GELINA to study objects of archaeological and cultural heritage interest [7]. Most of the objects were produced from a copper-alloy. Apart from Cu, they contain Sn or Zn as other major elements, and As, Ag, Sb, Co, Fe, In and Pb as minor or trace elements. Examples of bronze objects of different origins that were studied are: Etruscan statuettes [34], prehistoric bronze axes [35][36], Roman metal objects like parts from water taps [37] and Bronze-Age swords [38][39]. NRCA and Neutron Diffraction (ND), which are two complementary NDA techniques, are often combined to study the elemental composition together with the structure of the objects and to provide information about the origin of the objects, the fabrication methods, trade relations and the usage of the objects (see e.g. Ref. [37] and [39]). A detailed overview of archaeological applications is given in Ref. [7].

In general, an analysis of NRCA data is far more complicated compared to the analysis of NRTA data. A full methodological approach based on a RSA starting from resonance parameters is not always evident, in particular in case of objects with an irregular shape. At present the elemental composition for complex objects is mostly derived from ratios of observed resonance areas combined with results of calibration measurements [7]. Calibration factors are determined from measurements with representative calibration samples with known elemental composition. These factors account for the nuclear data and experimental conditions such as the detection efficiency and neutron fluence rate. For example, for archaeological applications calibration samples have been made from melts of known quantities of different elements or by stacking sets of thin metallic foils or discs of the components [7].

4 Reference samples

An overview of reference materials that are available at JRC Geel and have been used for specific NRTA and NRCA applications is given. The NRTA data result from transmission measurements at 10 m, 25 m and 50 m stations as described in Refs. [33], [17] and [40], respectively. The NRCA results were mainly obtained from experiments at the 12.5 m capture station described in e.g. Ref. [41].

4.1 Metallic discs of stable elements

Becker et al. [16] and Tsuchiya et al. [17] have shown that NRTA is a valuable method to validate resonance parameters in the Resolved Resonance Region (RRR) by comparing the declared areal density and the one that is derived by NRTA. To avoid problems related to the declared value a homogeneous sample is needed for which the areal density can be accurately determined. In addition, bias effects on the areal density derived by NRTA should be avoided. Therefore, homogeneous metallic samples or alloys are recommended. The best results are obtained when the measurements are performed in good transmission geometry.

Such reference materials can be realised for elements with a well-known and stable natural isotopic composition and for which samples in metallic form can be made using base material with a purity level better than 99.95%. Under these conditions the areal density of the element of interest can be determined with an uncertainty of less than 0.1%. Recommended values for the isotopic composition of stable elements are given in [42]. The optimum thickness should be chosen such that the transmission in the main energy region of interest is between 0.3 and 0.8. This will reduce the impact of systematic effects due to the background.

The sample used by Becker et al. [16] to validate the resonance parameters of tungsten was a 1 mm thick metallic disc made of ^{nat}W. The areal density was derived from the weight and the area. The latter was determined by an optical surface inspection with a microscopic based measurement system from Mitutoyo, i.e. the Quick-Scope QS200Z [43]. The information sheet of this sample, which is given in Appendix A, can be taken as an example for the characterisation requirements. The measurements were carried out at a 25 m transmission station of GELINA using a ^{6}Li glass scintillator as neutron detector. To reduce bias effects due to e.g. dead time and background, the measurement and data reduction procedures recommended in Ref. [2] were followed. To produce the full covariance information the AGS (Analysis of Geel Spectra) system [44] was used. The experimental conditions and data reduction and analysis procedures together with the full covariance information in the AGS format are documented in Ref. [45], following the recommendations that resulted from a consultant's meeting organised by the Nuclear Data Section of the IAEA [46]. These transmission data, which have been submitted to the EXFOR data base, can be used to validate new recommended resonance parameters for ^{nat}W. In general, a sample with characteristics as given above, together with the measurement and data reduction procedures described in the associated report [45], are a good example of how experimental data to validate evaluated nuclear data in the resonance region should be produced.

Metallic discs of natural elements were used by Paradela et al. [32] [33] to demonstrate the potential of NRTA to analyse materials containing nuclides with strongly overlapping resonances, as discussed in Section 3.4.

In case of strong resonances an optimum transmission, i.e. larger than 0.3, can only be obtained by the use of extreme thin samples. In addition, to ensure a good transmission geometry the samples should be homogeneous and not contain holes. This can be realised by metallic alloys consisting of the element/nuclide of interest mixed with a matrix material with a small total cross section. To validate the resonance parameters of gadolinium in the low energy region a set of high purity Mg-Gd alloys with different thickness were prepared and characterised at the Montanuniversität Leoben (AT). They

were all produced from the same batch with a nominal Gd concentration of 0.25 wt%. The characteristics of the samples are summarised in Table 5. For each sample, cut-offs were analysed as representative samples by X-Ray Fluorescence (XRF) and Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES). The weight and area of the samples were determined at JRC Geel. One of the samples, i.e. the one with Id = 2.0XRF2, was fully dissolved and analysed by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) after capture and transmission measurements with this sample were carried out at GELINA. A report describing the production of the samples and the results of the XRF, ICP-OES and ICP-MS measurements is in progress.

Table 5. Characteristics of a set of Gd-Mg discs made of the same batch of material with a nominal Gd concentration of 0.25 wt%. The discs were produced at the Montanuniversität Leoben (AT).

Sample Id	Weight g	Area mm^2	Thick mm	wt.% Gd ICP-OES	wt.% Gd ICP-MS
0.8XRF1	2.56293 (15)	1789.7 (10)	0.8	0.241 (3)	
0.8XRF4	2.46396 (15)	1791.9 (10)	0.8	0.241 (3)	
0.8XRF5	2.60943 (16)	1791.1 (10)	0.8		
0.8XRF6	2.55666 (115)	1790.7 (10)	0.8	0.239 (3)	
1.5XRF1	4.85681 (29)	1788.6 (10)	1.5		
1.5XRF2	4.89241 (29)	1789.0 (10)	1.5	0.236 (3)	
2.0XRF1	6.1089 (4)	1788.1 (10)	2.0		
2.0XRF2	6.1530 (10)	1791.1 (10)	2.0	0.252 (3)	0.2522 (54)
3.0XRF1	9.4794 (6)	1790.4 (10)	3.0	0.238 (3)	

4.2 CBNM-171 reference materials

The Nuclear Reference Material 171 (referred to as CBNM-NRM-171 or EC-NRM-171) is a set of low-enriched uranium standards which was jointly prepared and certified by the former Central Bureau of Nuclear Measurements (CBNM) (presently JRC Geel) and the former National Bureau of Standards (NBS) of the US (presently National Institute for Science and Technology (NIST)). A CBNM-NRM-171 set consists of five sealed aluminium cylindrical cans each containing U_3O_8 with a different ^{235}U abundance. They are labelled according to their nominal relative amount of ^{235}U.

They have been designed, produced and certified to calibrate measurement systems used to determine the ^{235}U enrichment in nuclear material by γ-ray measurements. The material is certified for the isotopic composition. The total amount of material, chemical composition and impurities, and internal diameter were also determined, however, not certified. A detailed description of the design specifications and production and characterisation procedures is given in Ref. [47]. The report includes a detailed description of the aluminium container in view of γ-ray measurements. A cross-sectional view of the samples is shown in Figure 4. The cans have an external diameter of 80 mm and a height of 90 mm. Each can contains 200.0 (2) g U_3O_8 as a pressed powder layer with an internal diameter of 69.9 (1) mm and a thickness of about 20 mm. The isotopic

composition and the relative uranium content are reported in Table 6. From these data and the weight and internal diameter the areal density of the U-isotopes can be calculated. The samples can be used to assess the $^{235}U/^{238}U$ ratio and the areal densities for ^{235}U and ^{238}U derived by NRTA.

Figure 4 reveals that the sample contains a substantial amount of matrix material, that is, other material besides U_3O_8. These samples were used in Ref. [1] to validate a model that accounts for the presence of matrix materials without resonances in the low energy region. The results of this exercise illustrate that only 1% of the absorption in the sample is due to the presence of uranium. The other 99% is due to matrix materials, which do not contain nuclides with strong resonances in the observed energy region. To account for the absorption by these nuclides a dummy element is introduced with a cross section that is proportional to the sum of a constant and a term that is inversely proportional to the neutron velocity.

Table 6. Isotopic composition of the CBNM-NRM-171 reference material together with the relative amount of uranium. The quoted uncertainties are expanded uncertainties with a 95 % confidence limit.

Sample ID	$^{234}U/U$ at%	$^{235}U/U$ at%	$^{236}U/U$ at%	$^{238}U/U$ at%	U/U_3O_8 wt%
CBNM-031	0.0020 (2)	0.3206 (2)	0.0147 (3)	99.6627 (4)	84.56 (15)
CBNM-071	0.0053 (2)	0.7209 (5)	< 0.00002	99.2738 (4)	84.53 (15)
CBNM-194	0.0174 (2)	1.9664 (14)	0.0003 (1)	98.0159 (18)	84.56 (15)
CBNM-295	0.0284 (4)	2.9857 (21)	0.0033 (2)	96.9826 (29)	84.52 (15)
CBNM-446	0.0365 (3)	4.5168 (32)	0.0069 (2)	95.4398 (32)	84.66 (15)

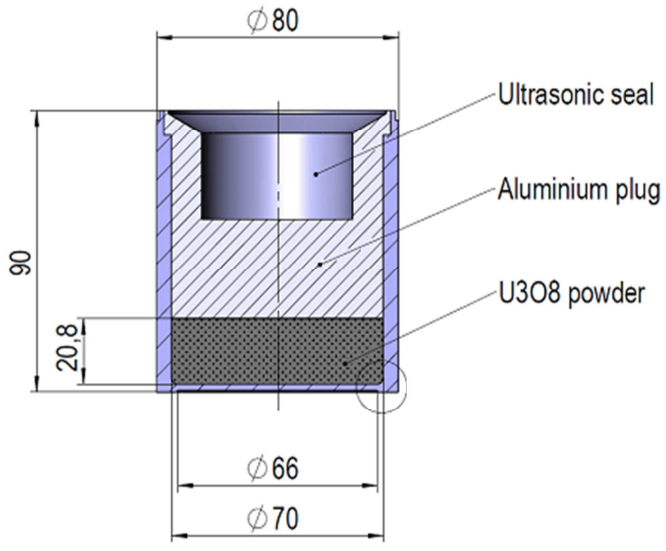

Figure 4 Cross-sectional view of a CBNM-NRM-171 reference sample.

4.3 CBNM-271 reference materials

The Nuclear Reference Material 271 (CBNM-NRM-271 or EC-NRM-271) is a set of plutonium standards that was produced and certified at the CBNM [48]. They have been designed, produced and certified as reference materials for non-destructive γ-ray measurements to determine the isotopic composition of plutonium containing samples. A reference set consists of four sealed stainless steel containers each containing a sintered

pellet of about 6.6 g of PuO_2. The containers have an external diameter of 40 mm and a thickness of 20 mm. They are labelled according to their nominal relative amount of ^{239}Pu. A schematic drawing of a sample is shown in Figure 5 and a photo of a set in Figure 6. The container is in accordance with the requirements of "Special Form Radioactive Material" as defined in the IAEA "Regulation for the Safe Transport of Radioactive Material". The samples are certified for the Pu isotopic composition and the $^{241}Am/Pu$ fraction [48]. The relative abundances together with their uncertainties as of 20 June 1986 are given in Table 7. Nominal characteristics of a pellet, together with the specific data for the samples of set 3 are specified in Table 8.

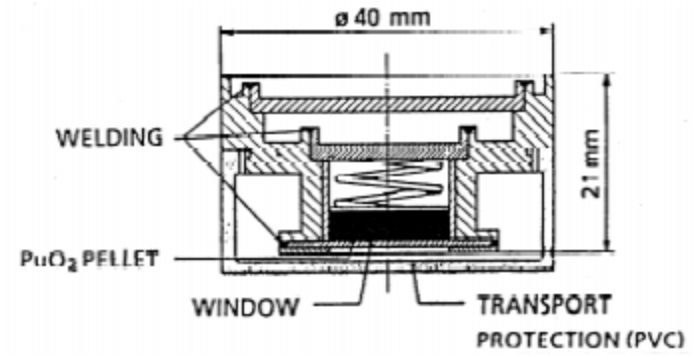

Figure 5 Schematic drawing of a CBNM-NRP-271 PuO_2 reference sample

Figure 6 Picture of a CBNM-NRM-271 set of PuO_2 reference samples

The samples of set 3 were used by Paradela et al. [49] to assess the performance of NRTA for the characterisation of Pu samples. The measurements were carried out at a 10 m transmission station of GELINA. A preliminary analysis of these measurements reveals that these data can also be used to validate cross section data for Pu isotopes and ^{241}Am in the resolved resonance region.

Table 7. Pu isotopic composition of the CBNM-NRM-271 reference material together with the relative amount of ^{241}Am. The uncertainties are expanded uncertainties at a 95% confidence interval.

	^{238}Pu/Pu	^{239}Pu/Pu	^{240}Pu/Pu	^{241}Pu/Pu	^{242}Pu/Pu	^{241}Am/Pu
	at%	at%	at%	at%	at%	at%
CBNM-Pu93	0.0117 (2)	93.4392 (40)	6.2886 (39)	0.2215 (4)	0.0390 (3)	0.1039 (21)
CBNM-Pu84	0.0706 (6)	84.3985 (84)	14.1578 (85)	1.0197 (18)	0.3534 (10)	0.2157 (22)
CBNM-Pu70	0.8506 (13)	73.4248 (98)	18.2445 (87)	5.4257 (34)	2.0544 (23)	1.1624 (116)
CBNM-Pu61	1.2045 (25)	62.6562 (293)	25.3526 (241)	6.6376 (87)	4.1491 (64)	1.4362 (144)

Table 8. Nominal characteristics of a pellet together with the specific values for set 3, indicated by O/3, which is available at JRC Geel. The uncertainties on the nominal values are given as maximum deviations. The uncertainties of the data for set 3 are not given in the certificate.

	Nominal	93 O/3	84 O/3	70 O/3	61 O/3
Disc mass /g	6.65 (6)	6.616	6.687	6.687	6.620
Disc height / mm	3.75 (14)	3.80	3.80	3.66	3.65
Disc diameter / mm	14.77 (21)	14.91	14.81	14.59	14.84
Area density / (g/cm^2)	3.87 (12)	3.79	3.88	4.00	3.83

4.4 Reference pellet

Two cylindrical rods with a diameter of 4.98 (1) and 10.01 (1) mm were prepared from the same batch of metallic natural Cu. The volume density of 8.913 (3) g/cm^3, which was determined from the weight and volume of a representative sample is close to the nominal volume density of 8.94 g/cm^3. These samples were used in Ref. [26] to validate the analytical expression in Eq. 1.

4.5 BCR-691 reference materials

A series of reference bronze discs, in composition representative of ancient alloys, were prepared by modern powder metallurgical techniques. The production and characterisation processes were co-ordinated by JRC Geel [50]. These samples, referred to as BCR-691, were especially produced for XRF applications. The samples were certified for the mass fractions of the main alloying elements As, Pb, Sn and Zn. They are available as a set of five alloys in the form of polished discs. The discs have a nominal diameter of 35 mm and thickness of 2 mm.

The certified weight fractions of As, Pb, Sn and Zn are reported in Table 9. This table also specifies the relative amounts of Fe, Mn, Ni, S and Sb. These values are reported without uncertainties in Ref. [50]. The certification report does not provide any information about the weight and area. The weight and area of the samples that are available at JRC Geel are specified in Table 10. The area was obtained with a Quick-Scope QS200Z from Mitutoyo [43]. These samples can be used to verify the performance of NRCA and NRTA for archaeological applications. Given the information provided in the certification report,

the samples should only be used to determine the certified relative abundances of As, Pb, Sn and Zn.

Table 9. Characteristics of the BCR-691 reference samples of the set that is available at JRC Geel. The quoted uncertainties are expanded uncertainties corresponding to a 95% confidence level. The samples are only certified fo the As, Pb, Sn and Zn abundance.

Element	Relative abundance in wt%									
	Quaternary bronze		Brass		Arsenic bronze		Lead bronze		Tin bronze	
As	0.194	(10)	0.099	(10)	4.60	(27)	0.285	(22)	0.194	(20)
Pb	7.9	(7)	0.39	(4)	0.175	(14)	9.2	(17)	0.204	(18)
Sn	7.16	(21)	2.06	(7)	0.202	(29)	10.1	(8)	7.0	(6)
Zn	6.02	(22)	14.8	(5)	0.055	(5)	0.148	(24)	0.157	(25)
Fe	0.2		0.5		0.2		0.3		0.1	
Mn	0.2		0.4		0.2		0.3		0.1	
Ni	0.1		0.2				0.5		0.3	
S	0.3		0.3		0.3		0.5			
Sb	0.5				0.5		0.7		0.5	

Table 10 Weight and area of the BCR-691 set that is available at JRC Geel.

	Quaternary bronze	Brass	Arsenic bronze	Lead bronze	Tin bronze
Weight / g	17.608 (1)	15.666 (1)	16.719 (1)	18.432 (1)	17.707 (1)
Area/mm^2	963.4 (2)	969.2 (2)	967.7 (2)	989.6 (2)	967.1 (2)
Diameter/mm	35.03 (2)	35.13 (2)	35.11 (2)	35.50 (2)	35.09 (2)

4.6 Aluminium alloys

Aluminium discs containing small amounts of Ag, In, Sb and Ho were produced at the Montanuniversität Leoben (AT). The discs with a nominal diameter of 80 mm and nominal thicknesses of 0.5 mm, 1.0 mm, 2.0 mm and 3.0 mm were made starting from the same batch of material. The relative amounts of Ag, In, Sb and Ho are 10.8 (4) ppm, 10.1 (3) ppm, 55.7 (14) ppm and 56.1 (9) ppm, respectively. These values reflect the mean and standard deviation resulting from an analysis of 40 representative samples by ICP-MS. The weight and area of the discs were determined at JRC Geel. The characteristics of the discs are reported in Table 11. These samples are suitable to calibrate TOF systems for NRCA and to verify their performance in particular detection limits for samples containing trace elements or impurities with similar resonance properties as Ag, In, Sb and Ho.

Table 11. Characteristics of Al discs containing small amounts of Ag, In, Sb and Ho. The discs have a nominal diameter of 80 mm. The values for the thickness are nominal values.

Sample Id	Weight g	Area mm^2	Thickness mm
SN3S-2018-11-01	7. 2471 (1)	5022. 68 (6)	0.5
SN3S-2018-11-02	7.2834 (1)	5024.17 (2)	0.5
SN3S-2018-11-03	7.3632 (1)	5024.24 (6)	0.5
SN3S-2018-11-04	7.3584 (1)	5024. 55 (6)	0.5
SN3S-2018-11-05	13.0006 (1)	5023.3 (2)	1.0
SN3S-2018-11-06	13.0261 (1)	5023.3 (3)	1.0
SN3S-2018-11-07	13.0626 (1)	5022.8 (4)	1.0
SN3S-2018-11-08	13.0319 (1)	5023.2 (3)	1.0
SN3S-2018-11-09	27.6563 (1)	5019.3 (12)	2.0
SN3S-2018-11-10	27.6597 (1)	5019.4 (12)	2.0
SN3S-2018-11-11	27.6721 (1)	5019.1 (27)	2.0
SN3S-2018-11-12	27.7048 (1)	5019.9 (10)	2.0
SN3S-2018-11-13	40.4184 (1)	5021.3 (15)	3.0
SN3S-2018-11-14	40.4122 (1)	5019.4 (4)	3.0
SN3S-2018-11-15	40.4200 (1)	5019.4 (7)	3.0
SN3S-2018-11-16	40.3800 (1)	5019.7 (4)	3.0

5 Summary

The use of Neutron Resonance Analysis (NRA) as a non-destructive analysis technique was reviewed. The review covered applications ranging from archaeological studies, characterisation of reference and complex nuclear materials to the validation of nuclear data. To support these methods a set of reference materials that are available at JRC Geel were described. Their use to calibrate NRA systems and assess their performance was discussed. Based on the material presented in this work we can conclude that a combination of NRTA and NRCA can be considered as an analytical NDA method to determine the elemental and isotopic composition for a variety of applications.

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Appendix A

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18.02.2008

INFORMATION SHEET
Mr.Peter Schillebeeckx
LINAC (NP)
TP-NP 08/08

1. **Description of the sample**

   W disc ± Ø 80.02 x 0.218mm

2. **Preparation method**

   Punched by use of PW30 lab press. Press Force: 50kN

3. **Chemical analyses**

   Origin Material : Linac

4. **Metrological information**

   Total mass: 20.5345g ± 0.0001g
   Ø 80.02 ± 0.0093mm / Area: 50,2907cm^2
   Thickness: 0.218mm (Stdev.0.00365mm) / 0.40832g/cm^2 / 0.00134at/b
   At.mass ^{Nat}W: 183.84179 (Ref.) Wapstra 2003 tables
   Density (Theoretically): 19.250g/cm^3
   Density (in Practice): 18.730g/cm^3

Diameter and Area determination
Mitutoyo Vision Measuring Machine
TP-NP 08-08
W-disc : Ø80 x 0,218mm

Date: 19/11/2012
Operator: RW

1) Sample Reference TP-NP 08-08

2) Input parameters

Trace length: 160
Scan interval: 200
Zoom: 2,00
Stage light: 25%

3) Results

Measurement No	Area/mm²	Diameter/mm
1	5030,418	80,0332
2	5030,507	80,0339
3	5030,347	80,0327
4	5030,423	80,0333
5	5030,412	
Average	5030,421	**80,0333**
Stdev	0,057	**0,000**
Rel Stdev	0,001%	**0,001%**

4) Remarks

Measurements done with glass plate and Cu-weight on top.

List of abbreviations and definitions

ANNRI	Accurate Neutron-Nucleus Reaction Measurement Instrument
BWR	Boiling Water Reactor
CBNM	Central Bureau of Nuclear Measurements
DG-ENER	Directorate General - Energy
DOE	Department Of Energy
EC	European Commission
ENDF	Evaluated Nuclear Data File
DA	Destructive Analysis
FP	Fission Product
FWHM	Full Width at Half Maximum
GELINA	Geel Electron LINear Accelerator
GWd/MTU	GigaWatt Day per Ton Metal Uranium
IAEA	International Atomic Energy Agency
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
ICP-OES	Inductively Coupled Plasma Optical Emission Spectrometry
JAEA	Japan Atomic Energy Agency
JEFF	Joint Evaluated Fission and Fusion File
JENDL	Japanese Evaluated Nuclear Data Library
J-PARC	Japan Proton Accelerator Research Complex
JRC	Joint Research Centre
MTR	Materials Test Reactor
NAA	Neutron Activation Analysis
NBS	National Bureau of Standards
ND	Neutron Diffraction
NDA	Non-Destructive Analysis
NRA	Neutron Resonance Analysis
NRCA	Neutron Resonance Capture Analysis
NRM	Nuclear Reference Material
NRTA	Neutron Resonance Transmission Analysis
ORNL	Oak Ridge National Laboratory
PGA	Prompt Gamma-ray Analysis
RRR	Resolved Resonance Region
RSA	Resonance Shape Analysis
SNM	Special Nuclear Material
TOF	Time-Of-Flight
XRF	X-Ray Fluorescence

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