

Nondestructive technique for identifying nuclides using neutron resonance transmission analysis at CSNS Back-n

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Abstract

Nondestructive and noninvasive neutron assays are essential applications of neutron techniques. Neutron resonance transmission analysis (NRTA) is a powerful nondestructive method for investigating the elemental composition of an object. The back-streaming neutron line (Back-n) is a newly built time-of-flight facility at the China Spallation Neutron Source (CSNS) that provides neutrons in the eV to 300 MeV range. A feasibility study of the NRTA method for nuclide identification was conducted at the CSNS Back-n via two test experiments. The results demonstrate that it is feasible to identify different elements and isotopes in samples using the NRTA method at Back-n. This study reveals its potential future applications.

Full Text

Preamble

Nondestructive Technique for Identifying Nuclides Using Neutron Resonance Transmission Analysis at CSNS Back-n

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Abstract: Nondestructive and noninvasive neutron assays represent essential applications of neutron techniques. Neutron resonance transmission analysis (NRTA) is a powerful nondestructive method for investigating the elemental composition of objects. The back-streaming neutron line (Back-n) is a newly constructed time-of-flight facility at the China Spallation Neutron Source (CSNS) that provides neutrons in the eV to 300 MeV range. A feasibility study of the NRTA method for nuclide identification was conducted at CSNS Back-n through two test experiments. The results demonstrate that identifying different elements and isotopes in samples using the NRTA method at Back-n is feasible. This study reveals its potential for future applications.

Key words: CSNS, white neutron beam, NRTA, nuclide identification, nondestructive method

1. Introduction

Neutrons are unique probes for analyzing the interior properties of materials in a nondestructive manner due to their strong penetration power. Various neutron techniques based on interactions between neutrons and materials have been successfully developed and applied across different fields. Neutron resonance analysis (NRA) is a technique that utilizes neutron resonance absorption to identify and quantify elements and isotopes in samples [?, ?, ?]. Its basic principle is that the absorption of neutrons by nuclei as a function of neutron energy exhibits specific resonance peaks and dips, based on which the elemental composition of the material can be determined [?]. Neutron resonance transmission analysis (NRTA) is an NRA method based on the transmission of a neutron beam through a sample, in which resonances are observed as dips as a function of neutron energy.

The first demonstration of NRTA as a nondestructive method for determining the isotopic abundance of spent nuclear fuel pins was conducted by Priesmeyer and Harz [?]. Their study obtained $^{235,238}\text{U}$ and ^{239}Pu contents. Subsequently, NRTA for isotopic assay was employed as a nondestructive technique to characterize special nuclear materials [?, ?, ?]. Results obtained using NRTA were consistent with those from mass spectrometry and neutron activation methods [?]. NRTA has also been applied in other fields, such as the characterization of reference materials for nuclear data measurement [?] and cultural heritage objects and artifacts [?]. Moreover, NRTA has been combined with neutron imaging, known as neutron resonance transmission imaging, at the J-PARC facility [?], ISIS spallation neutron source [?], and KURRI-LINAC [?]. The NRTA method was investigated extensively by a group from EC-JRC Geel at the GELINA facility [?, ?, ?], which is a white neutron source providing neutrons from 10 eV to 20 MeV [?]. A white neutron beam covering the energy range from eV to MeV offers significant advantages over thermal neutron beams because a broad energy range covers almost all resonance regions of the elements, whereas thermal beams are mainly used for heavy elements. The back-streaming neutron line (Back-n) at the China Spallation Neutron Source (CSNS) is a newly constructed white neutron beam covering the range from 0.5 eV to 300 MeV [?, ?]. The time-of-flight (TOF) technique was used at Back-n to accurately determine neutron energy. The energy resolution at Back-n end station 2 (ES#2) was lower than ~2% for neutrons below 1 MeV. Due to these favorable beam properties, Back-n is suitable for studying and applying the NRTA technique.

This paper describes the first test experiments using the NRTA method at the CSNS Back-n facility. Experimental results for two samples containing light or medium elements are presented. Data analysis shows that element/isotope identification using the NRTA method is feasible at Back-n.

2. Experimental Setup of NRTA at CSNS Back-n

Located in Dongguan, Guangdong, the multi-disciplinary CSNS facility has been operational since 2018. It generates neutrons via a spallation reaction by impinging 1.6 GeV protons onto a massive tungsten target. The nominal proton beam power of CSNS Phase I is 100 kW. In the forthcoming CSNS Phase II upgrade project, the beam power will be increased to 500 kW, and several additional spectrometers [?, ?] and experimental stations [?] will be constructed. Although most neutron beamlines at CSNS deliver thermal neutrons for neutron scattering applications, the Back-n beamline delivers neutrons across a very broad energy range (from 0.5 eV to 300 MeV) primarily for nuclear data measurements [?, ?, ?] and nuclear technology applications [?, ?].

Since its inception, the Back-n beam's time characteristics have been a focal point of investigation [?] due to their crucial role in determining neutron energy and influencing energy resolution. The energy resolution of a TOF facility, considering the relativistic effect, is calculated as $\Delta E = \gamma(\gamma + 1) \frac{v}{c} \Delta T$, (1) (2) where γ is the Lorentz factor, v is the neutron velocity, and c is the speed of light. T and L are the neutron flight time and flight path length, respectively, and ΔT and ΔL are their respective uncertainties. ΔT originates primarily from the incident proton pulse width, whose full-width at half-maximum was 60 ns during the measurements in this study. ΔL is caused by neutron moderation and scattering in the spallation target before neutrons enter the neutron tube. The moderation process has been investigated via Monte Carlo simulations [?, ?], and the uncertainties of the moderation distance (ΔL) at different energies were obtained. Table 1 lists the ΔL values at different energies and the energy resolutions calculated using Eq. (1), which indicates that the energy resolution is a function of neutron energy.

Table 1 Energy resolutions of CSNS Back-n at ES#2

In this study, we implemented NRTA measurements using a neutron total cross-section spectrometer. This spectrometer comprised a multilayer fast fission chamber (FIXM) and a sample changer [?]. The FIXM is a neutron detector that uses ^{235}U and ^{238}U samples as neutron converters. Fig. 1 [Figure 1: see original paper] shows the typical setup for neutron total cross-section measurements. The samples to be measured were placed on a sample changer at end station 1 (ES#1), and the detector was set up at ES#2. The beam spot at the sample position is typically tuned to $\Phi 60$ mm, whereas that at the detector position is typically configured to be approximately $\Phi 40$ mm. This configuration ensures that the neutron beam at ES#2 is entirely covered by a neutron converter with a diameter of $\Phi 50$ mm. Total cross-section measurements typically include sample-out and sample-in measurements. Sample-out measurements involve characterizing a neutron beam without any sample in the beam, whereas sample-in measurements involve placing the sample in the beam at ES#1. The long distance (approximately 20 m) and the collimator between the sample and detector minimally suppress the multiscattering neutron

background. The transmission spectrum is the ratio of the normalized neutron counts of the sample-in to the sample-out measurements, from which the total cross-section can be determined. The neutron total cross-sections of ^{nat}C [?], ^{nat}Li [?], ^9Be , ^{27}Al [?], ^{nat}Fe , ^{nat}Pb , ^{nat}Cr , and ^{209}Bi [?] have been measured using this setup at Back-n.

The NRTA experiment at Back-n utilized the same setup as that used for total cross-section measurements. Photographs of the sample changer and neutron detector are shown in Fig. 2 [Figure 2: see original paper]. The resonances were observed as absorption dips in the transmission spectrum as a function of neutron energy, from which the nuclides in the sample could be identified because the resonances of each nuclide are as unique as fingerprints. The energy positions of the resonance dips in the transmission spectrum provide qualitative information about the composition, whereas the profiles of the resonance dips present quantitative details about the composition. In our first NRTA experiments, we resolved the qualitative information about the samples. Quantification of the elemental composition is more complicated and will be addressed in subsequent work.

Fig. 1 Layout for neutron total cross-section measurements

Fig. 2 Sample changer at Back-n ES#1 (left) and detector at ES#2 (right)

3. Results and Discussion

The NRTA method centers on measuring and analyzing neutron transmission spectra. The TOF technique was used to determine neutron energy. Further details of the TOF method at Back-n can be found in our previous publications [?, ?]. Another crucial aspect of neutron energy determination is the double-bunch effect, in which two identical proton bunches have well-defined intervals (410 ns) in an incident pulse. Double-bunch unfolding is necessary for energy regions higher than 10 keV, as the 410 ns uncertainty is no longer negligible. An unfolding code based on Bayes' theorem was developed to unfold the TOF spectrum [?, ?]. The systematic uncertainty introduced by the unfolding process can be estimated using the unfolding code, which is highly dependent on statistical uncertainties. The uncertainty of the unfolding is typically 1.5–2.5 times that of the statistical uncertainty.

3.1 Experiment Using Simulated Lunar Soil

As shown in Fig. 3 [Figure 3: see original paper], an artificially simulated lunar soil sample was measured to verify the feasibility of NRTA at Back-n. The sample was a cylinder with a diameter of 27 mm and a height of 21 mm, comprising uniformly distributed elements. The details of its composition are listed in Table 2.

Fig. 3 Photograph of simulated lunar soil sample

Table 2 Composition of simulated lunar soil sample

Fig. 4 [Figure 4: see original paper] compares the measured fission rate spectra for sample-in and sample-out measurements. The sample-in spectrum (the red curve in Fig. 4) was attenuated by scattering and absorption from the sample. Fig. 5 [Figure 5: see original paper] shows the transmission of the neutron beam through the sample, which is the ratio of sample-in to sample-out measurements. Resonance absorption dips were observed in the transmission spectrum and could be used to determine the isotopes present in the sample.

Fig. 4 Comparison of sample-out and sample-in measurements for simulated lunar soil

Fig. 5 Transmission spectrum of simulated lunar soil

Fig. 6 [Figure 6: see original paper] shows the transmission spectra in different energy regions to provide clear profiles of the absorption dips. All results are presented in 100 bins per decade (bpd), and the bin center is adopted as the energy of each bin. The resonances of sodium, aluminum, magnesium, iron, silicon, and oxygen were identified. Table 3 presents a comparison of the measured resonance energy values and the corresponding resonance energy values in the ENDF/B-VIII.0 library [?]. The differences between the measured and library values, along with the energy resolution at each energy level, are also presented. The differences at most points are comparable to the energy resolution, except at 88.11 keV, which is assumed to be the overlap of the resonances of ^{24}Mg and ^{56}Fe at 83.45 and 84.84 keV, respectively. The overlap of these two resonances may increase the deviation between measured and evaluated data. The differences vary with energy because, as discussed in Section 2, the energy resolution is a function of neutron energy. Binning is another reason for the differences in resonance energy positions between measured and ENDF/B-VIII.0 library values. The energy bin center depends on the bin width, and smaller energy binning should provide more accurate resonance energy; however, it increases statistical uncertainties. This issue can be addressed through long beam-time measurements in the future. In this study, 100 bpd was used as the final option. In addition, although the unfolding method was properly applied for the region above 10 keV, the double-bunch commission inevitably introduced systematic uncertainties.

Fig. 6 Expanded transmission spectra of simulated lunar soil in different energy regions

Table 3 Resonance energy of simulated soil sample

Through qualitative analysis, most elements listed in Table 2 were identified, except for calcium, potassium, and titanium. Their absorption was not evident, probably due to significant statistical fluctuations and their low abundance. In the above analysis, we only selected apparent dips and did not attempt to determine all resonances of each nuclide. This approach is feasible for qualitative study of the sample.

3.2 Experiment Using Underwater Relic from South China Sea

NRTA at Back-n was also used to investigate an underwater relic found in the South China Sea. Fig. 7 [Figure 7: see original paper] shows a block of silicate and calcium carbonate embedded with many copper coins. It had an irregular shape, with approximate dimensions of $25\text{ cm} \times 10\text{ cm} \times 10\text{ cm}$. The experimental setup was the same as that used to measure the simulated lunar soil. However, this object contained nonuniformly distributed elements and had an irregular shape. An associated experiment using a CMOS camera was also conducted to capture transmission images of the sample using total transmission and resonance-selected methods [?].

Fig. 7 Photographs of underwater relic found in South China Sea

Fig. 8 [Figure 8: see original paper] displays the spectra for both sample-in and sample-out measurements. The sample-in measurements were significantly attenuated due to the large sample size. Fig. 9 [Figure 9: see original paper] shows the transmission of the neutron beam through the sample, from which the isotopes in the sample were examined. Fig. 10 [Figure 10: see original paper] shows zoomed-in graphs of the transmission spectra in different energy regions. The resonances of copper, calcium, sodium, silicon, and iron were observed. These identified isotopes were consistent with our expectations. ^{63}Cu and ^{65}Cu originated from the coins in the object. ^{28}Si , ^{40}Ca , ^{56}Fe , and ^{16}O originated from silicate and calcium carbonate, which are the main components of the object. The presence of ^{23}Na is normal because sodium is abundant in the sea.

Fig. 8 Comparison of sample-out and sample-in measurement results for underwater relic

Fig. 9 Transmission spectrum of underwater relic

Fig. 10 Expanded transmission spectra of underwater relic in different energy regions

We compared the measured resonance energy positions with corresponding values in the ENDF/B-VIII.0 library (Table 4). The energy binning was 100 bpd. The differences in resonance energy positions between measurement and library were comparable to the energy resolution. In addition to the reasons mentioned in Section 3.1 for such differences, the irregular shape and massive volume of the sample could also increase the deviation between measured and library values.

Table 4 Resonance energy of underwater relic sample

4. Conclusions and Prospect

In 2018, CSNS introduced a new white-neutron beamline, Back-n. Its beam characteristics (high neutron flux, broad energy range, and good energy resolution) motivated us to launch a feasibility study of NRTA for nuclide identification. Experiments on two different samples—a simulated lunar soil sample and an underwater relic—were performed for the first time at Back-n. The existence of ^{16}O , ^{28}Si , ^{27}Al , $^{54,56}\text{Fe}$, ^{24}Mg , and ^{23}Na in the simulated lunar soil sample

and $^{63,65}\text{Cu}$, ^{16}O , ^{28}Si , ^{56}Fe , ^{40}Ca , and ^{23}Na in the underwater relic were confirmed using the NRTA method. A simple qualitative analysis demonstrated that CSNS Back-n can identify nuclides using the NRTA method. The beam characterization and results of this study demonstrate that CSNS Back-n is a promising platform for nuclide identification.

FIXM serves as the primary neutron detector for transmission measurements at Back-n. This helps monitor the flux based on $^{235}\text{U}(\text{n}, \text{f})$ and $^{238}\text{U}(\text{n}, \text{f})$ reactions. However, the resonances of the $^{235}\text{U}(\text{n}, \text{f})$ cross-section below the $\sim\text{keV}$ region are somewhat intense, which might induce fluctuations in the transmission spectrum. Lithium glass can be a valuable complement to the fission chamber in the low-energy region because of the large and smooth cross-section of the $^6\text{Li}(\text{n}, \text{t})$ reaction [?]. A lithium-glass monitor is planned for future transmission measurements at Back-n, characterized by its quick response, high efficiency, and smooth transmission spectrum. However, because the lithium glass scintillator is sensitive to γ -rays, n/γ discrimination [?] and γ -ray background must be investigated thoroughly.

In this paper, we present a simple qualitative analysis focused on confirming the presence of specific isotopes. Accurate quantitative analysis for resolving elemental abundance in a sample is significantly more complicated. Quantification analysis is feasible by performing sophisticated resonance analysis that must reflect experimental effects, resolution function, sample properties, and detector characteristics. This is the subject of our subsequent study.

Author Contributions

All authors contributed to the study conception and design. Material preparation, data collection, and analysis were performed by Sheng-Da Tang, Yong-Hao Chen, Jing-Yu Tang, Rui-Rui Fan, Qiang Li, Gong Li, Dong Liu, Zheng-Yao Jin, Xing-Zhu Cui, Tian-Xiang Chen, Yi-Wei Yang, Rong Liu, Han Yi, Yang Li, and Zhen Yang. The first draft of the manuscript was written by Yonghao Chen, and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

Data Availability Statement

The data that support the findings of this study are openly available in Science Data Bank at <https://www.doi.org/10.57760/sciencedb.j00186.00352> and <https://cstr.cn/31253.11.sciencedb.j00186.00352>.

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Note: Figure translations are in progress. See original paper for figures.

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