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Instructions for use

A new method to quantify multiple elements by pulsed epithermal neutron transmission spectroscopy

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ABSTRACT

To perform quantitative analysis of constituent elements, including light atoms, by time-of-flight (TOF) type neutron imaging, a new analysis method using epithermal neutrons excluding resonance dips has been proposed. The method is based on fitting the theoretical neutron transmission spectrum calculated from evaluated nuclear data to a measured neutron transmission spectrum. Compared to conventional imaging methods such as those using neutron radiography or neutron resonance analysis, it has the advantage of being able to evaluate multiple/light elements. We applied this technique to a

pulsed neutron transmission experiment at the Hokkaido University Neutron Source. A stack of iron and carbon plates was measured as a model sample consisting of heavy and light elements. The evaluated elemental densities show reasonable agreements within an error of 10%.

1. Introduction

Neutron imaging techniques have been utilized to evaluate elemental density/thickness distributions in a bulk object or a metal container. In particular, neutron radiography (NR) has been performed to qualitatively or quantitatively evaluate a light-element distribution, such as lithium in a Li-ion battery [1] and water in a fuel cell [2]. In order to quantify the density/thickness of a target element, NR needs a calibration curve that shows the relation between neutron transmission and density/thickness of the target element [3]. However, individual calibration curves are required for each element and therefore, multiple calibration curves are required for quantitative elemental analysis for a sample with multiple components. Thus, in reality, it is difficult for NR to quantify multiple elements in a complicated sample like a Li-ion battery.

In contrast, pulsed neutron transmission spectroscopy can apply to a sample with multiple

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components. Neutron resonance transmission analysis (NRTA), which is a pulsed neutron transmission method, identifies elements from the occurrence energies of resonance absorption, and give densities of each element by analyzing the area of each resonance dip [4]. When a two-dimensional (2D) detector with time-of-flight (TOF) measurement mode is used, the NRTA method can be extended to a mapping method for each element. However, neutron resonance imaging is difficult to apply to light elements because their resonance absorptions occur at a few MeV energy, and the neutrons are too fast for existing neutron imaging detectors to analyze via TOF.

In this study, we propose a new analysis method which can quantitatively evaluate multiple elements, including light elements, by pulsed neutron transmission spectroscopy. The new method is based on the idea that projected atomic number densities (PANDs) of each element can be quantified by fitting a theoretical neutron transmission spectrum calculated from the evaluated nuclear data (END) to an experimental neutron transmission spectrum in an appropriate neutron energy range, which occurs in the epithermal region as explained later.

To demonstrate this new method for evaluating PANDs of multiple elements, and to determine the appropriate fitting energy range, we carried out a pulsed neutron transmission experiment for three samples: a pure iron sample, a pure carbon sample and a stacked sample which consisted of the iron and carbon samples. Then, the proposed method was applied to the measured data, and we investigated two aspects of the proposed method as follows. Firstly, the appropriate fitting energy range was

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determined by comparing the measured total cross-sections of the iron sample and carbon sample to the END of Fe and C. Secondarily, the obtained PANDs of the three samples were compared with those calculated from the weight, volume, and thickness to discuss the performance of the new method.

2. Principle of the new multi-element quantification method

2.1 Conceptual basis

A neutron transmission spectrum is obtained by a pulsed neutron transmission spectroscopy. If a sample consists of one element, the neutron transmission spectrum, $Tr(E)$, is theoretically expressed by Equation (1).

$$Tr(E) = \frac{I(E)}{I_0(E)} = \exp(-N\sigma(E)). \tag{1}$$

Here E is the neutron energy, $I(E)$ is the neutron spectrum with a sample, $I_0(E)$ is the neutron spectrum without a sample, N is the PAND [cm^{-2}] of the element and $\sigma(E)$ is the neutron microscopic total cross-section of the element [cm^2]. On the other hand, if a sample consists of multiple elements, the neutron transmission spectrum is expressed by Equation (2).

$$Tr(E) = \prod_i Tr_i(E) = \exp(-\sum_i N_i \sigma_i(E)). \tag{2}$$

Here i means the number of elements included in a sample.

When the constituent elements of a sample are known, each $\sigma_i(E)$ is known from the END such as ENDF/B-VIII [5], JENDL-4.0 [6]. Thus, PANDs of each element, N_i, are estimated by fitting Eq. (2)

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(a theoretical transmission spectrum with the END) to an experimental transmission spectrum by means of non-linear least squares methods. Note that the energy range where the theoretical transmission spectrum agrees with an experimental transmission spectrum must be properly selected in the data analysis.

2.2 Choice of energy range

If a sample has crystalline structure, since coherent elastic scattering is caused by crystal structure, the characteristic zigzag shapes, Bragg-edges [7, 8], appear in a neutron transmission spectrum in the energy range from cold to thermal. As an example, the neutron transmission spectrum of a 1 cm thick sheet of molybdenum is shown in Figure 1. Since the total cross-section of the Bragg-edges is not included in the END, a theoretical transmission spectrum does not agree with an experimental transmission spectrum and thus, PANDs cannot be reasonably evaluated in this energy range. Additionally, in the energy range of epithermal neutrons, neutron resonance dips generally appear. Since the total cross-section of resonance dips changes depending on the sample temperature due to the Doppler effect, a theoretical neutron transmission spectrum does not agree with an experimental one and thus, it is difficult to use the occurrence energy of resonance dips. Therefore, the energy range used for the new method is that of epithermal neutrons outside the energy region where resonance dips occur.
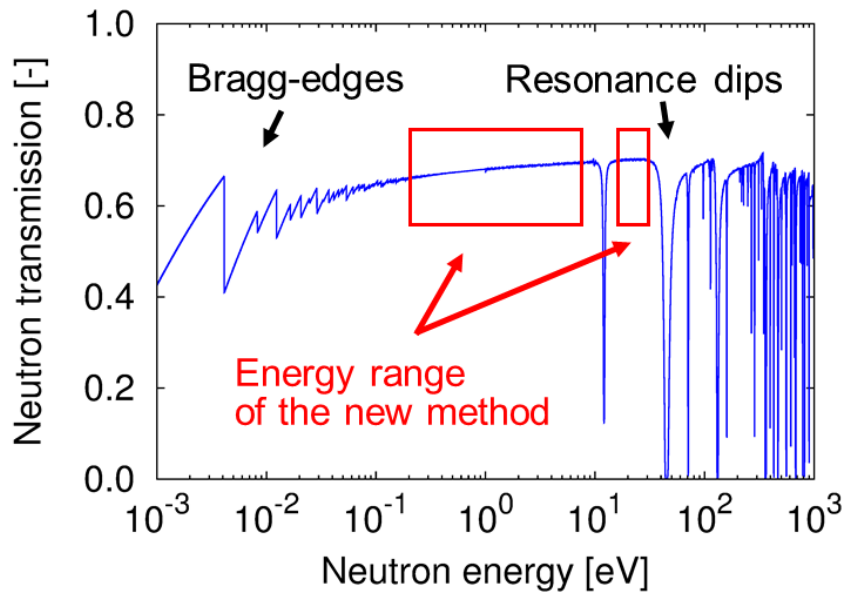
Fig. 1 An example of the energy range used for the new method in the neutron transmission spectrum of molybdenum with a thickness of 1 cm.

3. Experimental

3.1 Samples

Three samples, an iron plate sample, a carbon plate sample and a two-piece stacked sample of the iron and carbon plates were prepared for demonstration of the new analysis method. The iron sample is representative of a heavy element sample, and the carbon sample is representative a light element sample. The sizes of iron and carbon plate samples were $100 \times 100 \times 5$ mm^3. The size of the stacked sample was $100 \times 100 \times 10$ mm^3. The purity of the iron and carbon plate samples were 99.99% and 99.95%, respectively.

True PANDs of the two plates were measured from their weight, volume and thickness for comparison of the PANDs evaluated by the analysis. The true PANDs of the iron and carbon plate

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samples were 4.35×10^{-2} [atoms/barn] and 4.51×10^{-2} [atoms/barn], respectively.

3.2 Experimental setup

The pulsed neutron transmission spectroscopy experiment was carried out at the thermal neutron

port in Hokkaido University Neutron Source (HUNS) at the Hokkaido University electron LINAC [9].

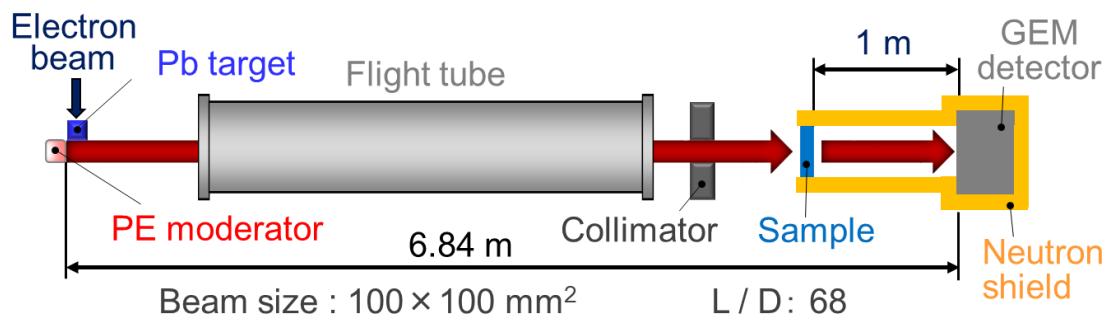The energy of an electron beam from LINAC was 34 MeV, the beam current was 36 μA, the pulse

Fig. 2 Experimental setup of the pulsed neutron transmission spectroscopy in HUNS.

repetition rate was 50 Hz, and the pulse width was 3 μs. Lead (Pb) was used for a neutron production

target. Neutrons were produced by photonuclear reaction after bremsstrahlung. High-density

polyethylene (PE) was used for a neutron moderator

Figure 2 shows the experimental setup. The distance between the moderator surface and the position

of the neutron detector was 6.84 m. The neutron beam from the PE moderator goes through an

evacuated flight tube, and was narrowed at a collimator with the size of 100×100 mm^2. The size of PE

moderator was $100 \times 100 \times 50$ mm^3. The collimation ratio was 68 with natural collimation. The distance

between the sample position and the neutron detector was kept to 1 m to prevent contamination by

neutrons scattered from a sample. The detector, sample and beam-line between them were surrounded

by neutron shields to prevent contamination by environmental neutrons.

As a neutron detector, a GEM (gas electron multiplier) detector [10] was used for TOF measurement.

The width of each TOF channel was 2 μs, and the number of TOF channels was 4000. The energy

range of measured neutron was from 0.004 eV to 25000 eV. The neutron count rate in the measurement

without a sample was 0.12×10^4 events/cm^2/sec, which was the sum of all TOF channels. The

measurement time for a single element sample was 3 hours. That for the stacked sample was 5 hours,

while that for an open beam was 2.5 hours.

4. Results and discussion

4.1 Determination of an appropriate energy range for the fitting analysis

In the new analysis method, PANDs are reasonably deduced by fitting in an appropriate energy

range, in which a calculated neutron transmission spectrum from Eq. (2) reasonably agrees with a

measured neutron transmission spectrum as mentioned in Sec. 2. Therefore, in this section, the

appropriate fitting energy range was determined by comparing the END and measured microscopic

total cross-section.

Figure 3 shows the results of comparisons between the ENDs of Fe and C in JENDL-4.0 and

measured total cross-sections of the iron and carbon samples. The measured total cross-sections were

obtained from measured transmission spectra and their true PANDs using Eq. (1). In Fig. 3, below 0.3

eV, the ENDs did not agree with the measured total cross-sections due to the Bragg-edges. Additionally,

in the energy range above 20 eV, it was shown that the measured total cross-section of the iron sample

was smaller than the END of Fe, and that the measured total cross-section became smaller as the

neutron energy became higher. Although the magnitude of the decrease was small, a similar trend was

observed in the data from the carbon sample.

It is possible that the reason for the decrease was contamination made up of background neutrons

caused by environmental neutrons which penetrate the neutron shield. If there is contamination by

background neutrons, the neutron transmission value can be estimated by Equation (4).

$$Tr_{\mathrm{exp}} = \frac{I + I_{\mathrm{B}}}{I_0 + I_{\mathrm{B}}} > \frac{I}{I_0} = Tr_{\mathrm{true}} . \tag{4}$$

Here Tr_{exp} is the measured neutron transmission value, I is the ideal neutron intensity with a sample,

I_0 is the ideal neutron intensity without a sample, I_{B} is the intensity of background neutrons, and Tr_{true}

is the ideal neutron transmission value. In accordance with Eq. (4), Tr_{exp} is always overestimated if

there is contamination by background neutrons. Conversely, the total cross-section is always

underestimated in such a case.

Thus, we determined that the appropriate fitting energy range was from 0.3 eV to 20 eV in this

experiment. It is likely that the appropriate fitting energy range changes in accordance with samples

and experimental setup, and thus, the new method needs a determination procedure of the energy range

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for fitting under different conditions.

4.2 Evaluation results of PANDs

To demonstrate the new method, the deduced PANDs were compared to the true PANDs. The plots

in Figure 4 show measured transmission spectra and fitted transmission spectra of the three samples.

The sub-tables of Fig. 4 show the deduced PANDs, the true PANDs and the differences between them.

Figs. 4(a), 4(b) and 4(c) show the results of the iron plate sample, the carbon plate sample and the

stacked sample, respectively. In the fitting analysis, the END of Fe and C in JENDL-4.0 were used,

and the fitting energy range was from 0.3 eV to 20 eV as mentioned in Sec. 4.1.

In the one-element samples, the difference in the iron sample was -1.4 %, and the difference in the

carbon sample was -0.5%. Namely, the PANDs of both one-element samples were evaluated within a

difference of ~1% from the true PANDs. In the two-element sample, the differences of Fe and C were

+3.4% and -7.5%, respectively. Hence, the PANDs of each element were evaluated within a difference

of 10% in the two-element sample. Although the differences of the deduced PANDs of the stacked

sample were rather bigger than those of one-element samples, the difference of 10% is comparable

with that of conventional neutron imaging techniques. In the NR, the difference of deduced thickness

ranges from a few percent to ten percent depending on the experimental setup and samples [11, 12].

In the NRTA, the difference of deduced PANDs ranges from a fractions of percent to ten percent [13].

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Therefore, it was confirmed that the new method can reasonably evaluate the PANDs of more than
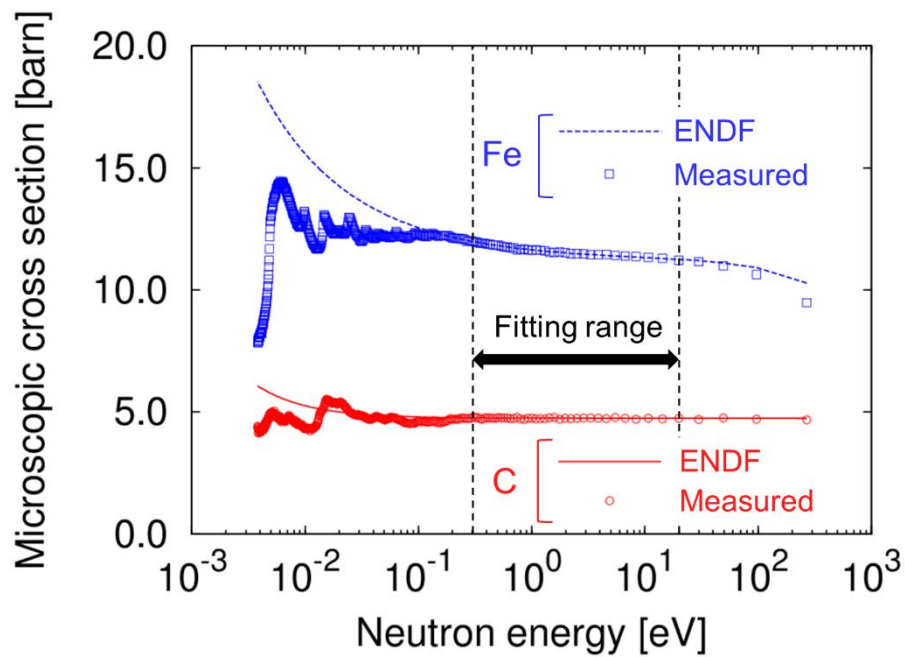

Fig. 3   Results of comparisons between nuclear data of Fe and C in JENDL-4.0 and measured total microscopic cross-sections of the iron and carbon samples. Lines show the nuclear data and plots shows measured cross-sections.

one element of widely differing atomic masses, in this case, Fe and C, but also can be extended to
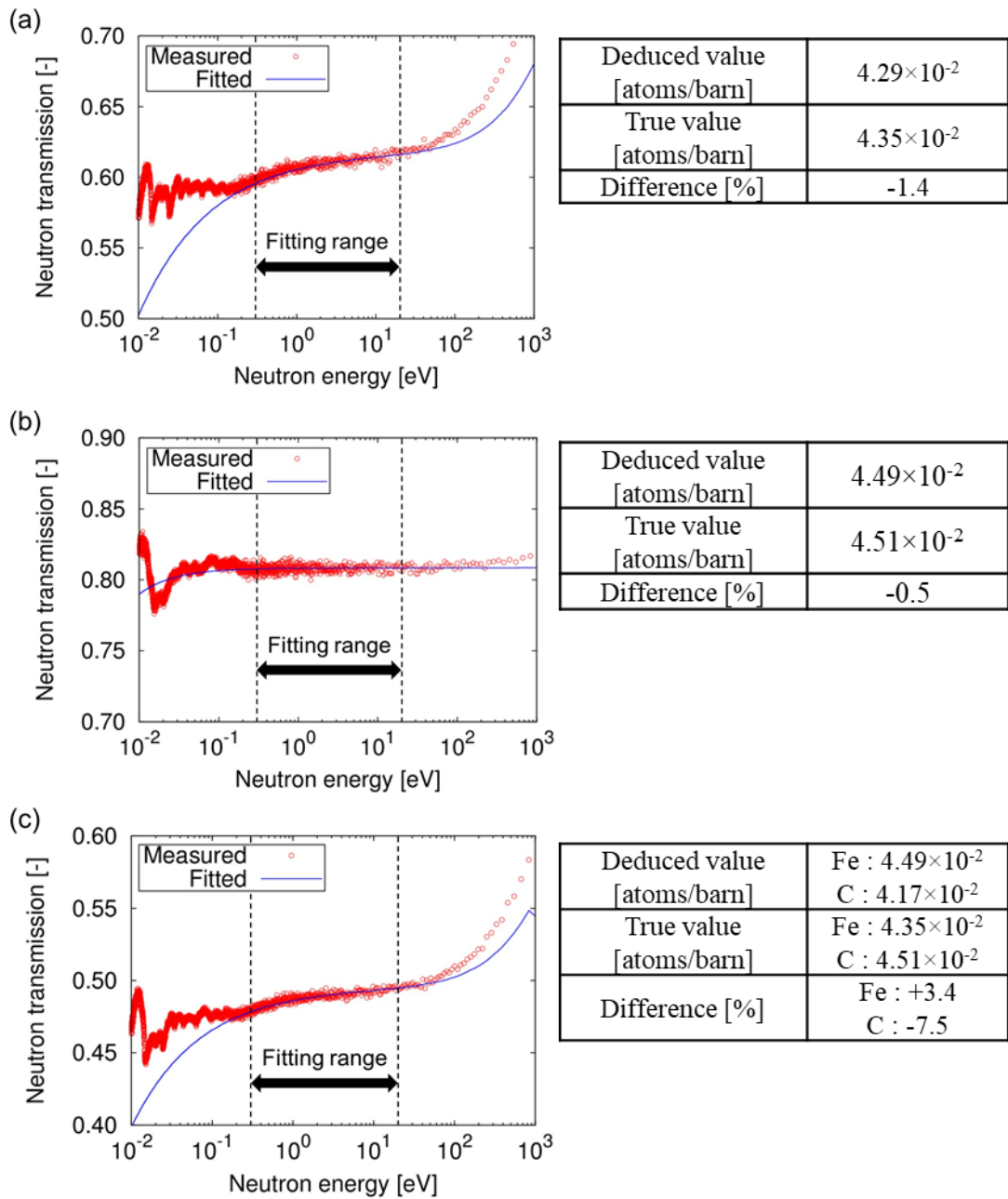
multiple element.

Fig. 4 Measured neutron transmission spectra and fitted neutron transmission spectra of (a) iron plate sample, (b) carbon plate sample and (c) stacked sample. Sub tables show the obtained and true PANDs, and the differences between them in the three samples.

5. Conclusion

We have proposed a new analysis method for quantification of constituent elements by pulsed neutron transmission spectroscopy with epithermal neutrons in an energy region outside resonances. In this method, reasonable PANDs are obtained by fitting a theoretical transmission spectrum calculated from the END to an experimental transmission spectrum in an appropriate fitting energy range. By comparing the END of Fe and C to the measured microscopic total cross-sections of the iron and carbon samples, the appropriate fitting energy range was determined to be from 0.3 eV to 20 eV in the demonstration case. The appropriate fitting energy range must be determined in accordance with experimental setup and samples. As a result of the fitting analysis of the stacked sample, the deduced PANDs of Fe and C were within 10% of the true PANDs, which is comparable with result from the conventional neutron imaging techniques. Therefore, it was confirmed that the new method can reasonably evaluate PANDs of multiple elements including a light element. This method has advantages of being able to evaluate multiple/light elements, compared to conventional neutron imaging techniques.

As future works, we are planning to extend the proposed method to imaging experiments, and apply it to practical uses, e.g., a Li-ion battery.

Acknowledgements

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