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Present status of research on pulsed neutron imaging in Japan

Y. Kiyanagi\textsuperscript{a*}, T. Shinohara\textsuperscript{b}, T. Kai\textsuperscript{b}, T. Kamiyama\textsuperscript{a}, H. Sato\textsuperscript{a}, K. Kino\textsuperscript{a}, K. Aizawa\textsuperscript{b}, M. Arai\textsuperscript{b}, M. Harada\textsuperscript{b}, K. Sakai\textsuperscript{b}, K. Oikawa\textsuperscript{b}, M. Ooi\textsuperscript{b}, F. Maekawa\textsuperscript{b}, H. Iikura\textsuperscript{b}, T. Sakai\textsuperscript{b}, M. Matsubayashi\textsuperscript{b}, M. Segawa\textsuperscript{b}, and M. Kureta\textsuperscript{b}

\textsuperscript{a}Graduate School of Engineering, Hokkaido University, Kita 13 Nishi 8, Kita-ku, Sapporo, Hokkaido 060-8628, Japan
\textsuperscript{b}Japan Atomic Energy Agency, 2-4 Shirakata-shirane, Tokai-mura, Naka-gun, Ibaraki, 319-1195, Japan

Abstract

Pulsed neutron imaging methods provide information on crystallographic structure, magnetic field, elemental composition, hydrogen bound state and other material features. Such methods have been expected to be a powerful complement to the traditional imaging method. Data analysis codes, detectors and new applications are being developed in Japan, and a new imaging beam line is being constructed at J-PARC. Here, recent progress in Japanese research on pulsed neutron imaging is presented, and a design for construction of the new imaging beam line is reported.

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1. Introduction

Energy-resolved imaging has proved to be useful since it can provide additional information to that of traditional imaging, allowing for the development of new applications. Although energy-resolved imaging can be performed at reactor sources, a pulsed neutron source gives much higher energy resolution, enabling researchers to perform spectroscopic analysis of the time-of-flight spectrum at each pixel of an imaging detector. Therefore, pulsed neutron imaging is attracting attention. Pulsed neutron imaging provides physical data such as crystallographic structure, magnetic field, elemental composition, hydrogen bound state and others [1-7]. The method has been intensively studied in Japan using the J-PARC neutron
source and the Hokkaido University neutron source (HUNS). A new imaging beam line at J-PARC has just started its construction phase. A detailed design study is now being performed. Planning for the experimental device, data analysis, and model experiments to indicate the feasibility of this method, as well as research on realistic applications are indispensable to its development.

A data analysis code, RITS, was developed [8]. It has been proved to be a very powerful tool for analyzing the Bragg edge transmission. However, its applicability needs to be expanded to handle various kinds of crystal structures. The detectors are one of the key issues for the pulsed neutron imaging. Pixel type detectors with 8×8 pixels or 16×16 pixels work fairly well, but the spatial resolution is not so good, about 2-3 mm [9]. We are working on new neutron detectors, for example, a neutron colour image intensifier combined with a high-speed camera, a GEM detector, an MCP detector and so on, and are planning to apply them to pulsed neutron imaging. To expand the applications of pulsed neutron imaging we have carried out magnetic field imaging and succeeded in measuring the vector components of the magnetic field in a thin foil [7]. Hydrogen is one of the important elements for neutron imaging. The gradient of the neutron cross section of the hydrogenous materials is one of the measures of its dynamic character [7]. Therefore, we are now trying to image the hydrogen dynamic state in the material. Furthermore, as one real-world application, we have started crystallographic structural analysis of cultural heritage samples. Building upon such development in various fields required for establishing pulsed neutron imaging, we have just started construction of the imaging beam line.

Here, we report recent progress on pulsed neutron imaging in Japan and also a design study of the imaging beam line at J-PARC for constructing the instrument during the three years up until the end of fiscal year 2014.

2. Recent progress on pulsed neutron imaging in Japan

For developing pulsed neutron imaging, various kinds of improvements and developments both in hardware and software are necessary. A Bragg edge analysis code, RITS, was developed. However, the crystal structures treated by the RITS code were limited. Therefore, the code has been improved to treat 230 space groups and multiphase [10]. Thereby, the applicability of the RITS code has been expanded. Additional improvements are in progress for more precise analysis.

As one of the detector developments, a system consisting of a neutron imaging intensifier coupled with a fast camera is being developed [11] since it is expected to be used even at high neutron intensity fields and to give high spatial resolution of about several 10 μm. In our system the measurement is performed continuously without interruption to transfer the image data. This method minimizes the loss-time required for the data saving. Using this detector system we successfully measured time-of-flight spectra and observed the Bragg edge transmission clearly. However, since the time bin is still not sufficiently short for various envisioned experiments. Therefore, we are still developing a new type of a fast camera.

Counting type detectors are proven ones for time-of-flight measurements, but the defect of this kind of detector is its low count rate. To improve this point, the GEM detector has been equipped with a high-speed data-acquisition system. We examined the performance at J-PARC and found that it achieved a counting rate of 1.85 MHz. A new method for improving the counting rate is now being developed. The spatial resolution of the GEM detector is about 1 mm. As a higher-spatial-resolution detector, a μ-PIC detector is being used. Its spatial resolution is about 200 μm, and its counting rate is about 0.15 MHz. Therefore, this detector will be suitable for relatively low intensity condition.

One of the applications performed recently was on a hydrogen storage material. The principle of the measurement is to use the characteristic change of the gradient of hydrogen neutron cross sections in the long-wavelength region. We measured three metal hydrates with different hydrogen contents. The material was Ti0.45Cr0.25Mo0.30 and the hydrogen-to-metal concentrations were 1.60, 0.62 and 0. The gradients of the cross sections at the long-wavelength region were almost the same in the 0.62 and 0
content samples but much larger for the 1.60 sample. This suggests that the hydrogen moves as a hydride in the low-concentration sample but moves more freely in the high-concentration sample. We also measured the structure of the cross section around 0.15eV to look at the vibrational mode, in which the typical structure of a metal hydride appears as a hump. However, we could not observe the hump in two samples. This indicates that the structure of this hydride did not cause simple hydrogen vibration. As a Bragg edge application we tried to measure cultural heritage samples, Japanese swords [12]. We measured three kinds of Japanese swords made in different ages and different places. They showed different-shaped Bragg edge transmissions, indicating their different crystallographic textural structures. A fine crystallite size was observed near the sword edge and larger ones toward the back, which is a similar trend to those observed by destructive methods. As a resonance absorption application, a measurement on a decoupler material for the J-PARC neutron source was performed to check homogenous distribution of indium in an Au-Cd-In alloy. The concentration of In was 0.5%, so it was very difficult to check its homogeneous distribution by other methods. The resonance peak of In was selected by the time-of-flight method and its spatial distribution was clearly visualized. From this result we confirmed that the decoupler was made successfully [13]. Magnetic field imaging has also been performed to visualize the vector components of magnetic fields [9].

We are promoting the development of various kinds of devices and applications to help establish the method of pulsed neutron imaging. In parallel, a design study for constructing the imaging beam line at J-PARC is ongoing [14] and we are now at the construction phase. Here, we introduce the recent design study of ERNIS (Energy-Resolved pulsed Neutron Imaging System).

3. Imaging beam line, ERNIS, at J-PARC

3.1. Imaging functions required for ERNIS

The imaging instrument ERNIS is the first dedicated imaging machine constructed for high-power pulsed neutron sources in the world. Near J-PARC there is JRR-3, in which TNRF (a thermal neutron radiography facility) exists. ERNIS should take full advantage of the pulsed nature of the neutron source, so we considered spectroscopic imaging first. Bragg edge imaging, resonance absorption imaging and polarized neutron imaging are indispensable, and other applications using the characteristics of the neutron total cross section are given priority over traditional imaging. However, L/D values achieved at neutron imaging facilities in Japan are less than about 200, and there is no effective cold neutron beam line, since CNRF (the cold neutron radiography facility) at JRR-3 does not have enough space for various applications. From these considerations, we concluded that ERNIS should also cover high L/D and cold neutron applications.

3.2. Consideration of performance for pulsed neutron imaging at J-PARC

Here, we discuss the required performance for Bragg edge imaging, resonance absorption imaging and polarized neutron imaging. The required performance for the Bragg edge imaging is as follows.
* Wavelength resolution: $\delta\lambda/\lambda$ of 0.2 % since strain measurements need high wavelength resolution.
* Band width: $\lambda > 5$ Å to cover 1st Bragg edge.
* Beam size: $> 100 \times 100$ mm$^2$ since industrial sample may be large.
* Spatial resolution: 10 µm ~ 1 mm depending on the needs and the sample size.

In the resonance absorption imaging, we perform elemental analysis and temperature measurement. For the former experiment a wide energy range is desired and for the latter, high wavelength resolution is required. Therefore, the required performance is as follows.
* Energy range: around $1 < E_n < 1000$ eV
* Energy resolution: $\Delta E_n/E_n < 1\%$ ($\Delta t/t < 1\%$) for getting 0.1\% resolution by fitting the resonance peaks.
* Peak separation power: requiring variable flight path length depending on materials.
* Beam size: $> 100 \times 100$ mm$^2$ since industrial samples may be large.
* Spatial resolution: 10$\mu$m – 1mm depending on the needs and the sample size.

The requirements for the polarized neutron are more complicated than others since it has to take into account the limitations of various devices, and the wavelength resolution and the wavelength region depend on the strength of the magnetic field. The requirements or realistic choices are summarized as follows:
* Beam size: 50×50 mm$^2$ since the cross section of polarizing devices is smaller than about 100 mm.
* Spatial resolution: 100 $\mu$m is realistic due to the long distance between the sample and the detector.
* Wavelength band: shorter wavelength for strong field and longer for weak field.
* Wavelength resolution: Weak magnetic fields, > 1\% is enough and strong magnetic fields, e.g., magnetic field inside ferromagnetic, < 0.2\% are necessary.

3.3. Structure of the imaging instrument, ERNIS

In addition to the requirements for each method, an experimental area as large as possible is desired, and the maximum field of view of 300 mm × 300 mm was chosen considering industrial applications.

![Fig. 1 Imaging instrument area at BL22 MLF in J-PARC](image-url)
characteristics at the epithermal region are almost the same. We have chosen the decoupled moderator since we have to satisfy the requirements on both intensity and wavelength resolution [14]. There were two candidates for beam lines, BL22 and BL23. The available flight path length at BL23 is shorter than BL22; therefore, we chose BL22 and designed beam lines accordingly. A new design of the imaging instrument area at the 2nd experimental hall in MLF (Materials and Life Science experimental Facility) is shown in Fig. 1 although there may be slight changes. The structure of the beam line is shown in Fig. 2.

The shortest sample position is about 18 m and the longest one is about 23 m. The neutron time-of-flight intensity at 23 m is shown in Fig. 3 and the wavelength resolution in Fig. 4. The maximum intensity was about $9.8 \times 10^7$ n/cm$^2$/sec at 18 m and $5.8 \times 10^7$ n/cm$^2$/sec at 23 m at 1 MW accelerator power. The wavelength resolution was 0.26% at 18 m and 0.2% at 23 m. The single-frame band width was $\lambda < 8.8$ Å at 18 m and $\lambda < 6.9$ Å at 23 m. These band widths are usually enough for Bragg edge imaging but they are a little bit shorter for hydrogen-bound-state imaging and some Bragg edge imaging. In such case we can use a second frame by adjusting the band width chopper.

We will set a wide range of L/D values to cover the large L/D values that are not available at other facilities in Japan. To attain a large L/D we put collimators a short distance from the moderator. Table 1 shows the possible L/D values at present; the largest one is 7500. These data formed the basis for our decision about the L/D values.
We will use various detectors for Bragg edge imaging and resonance absorption imaging. The detectors now available are a GEM detector, \( \mu \)-PIC detector, and high-speed camera combined with a neutron scintillator or a neutron color I.I. An MCP detector [15] is also a good candidate. However, we need further development and improvement to achieve a high counting rate to overcome the very high instantaneous neutron intensity around a peak of the time-of-flight spectrum. In the transmission measurement we place a suitable detector at a proper position. On the other hand, for polarized neutron imaging we set up polarized neutron equipment for measurement as shown schematically in Fig. 5.

![Fig. 5 Polarized neutron equipment setup](image)

<table>
<thead>
<tr>
<th>Collimator</th>
<th>100x100 at 3m</th>
<th>47.8mm( \varphi ) at 3m</th>
<th>25.2mm( \varphi ) at 4.3m</th>
<th>15mm( \varphi ) at 8m</th>
<th>5mm( \varphi ) at 8m</th>
<th>2mm at 8m</th>
</tr>
</thead>
<tbody>
<tr>
<td>L/D at 19m</td>
<td>190</td>
<td>335</td>
<td>583</td>
<td>733</td>
<td>2200</td>
<td>5500</td>
</tr>
<tr>
<td>L/D at 23m</td>
<td>230</td>
<td>418</td>
<td>742</td>
<td>1000</td>
<td>3000</td>
<td>7500</td>
</tr>
<tr>
<td>Intensity</td>
<td>100%</td>
<td>30%</td>
<td>9.6%</td>
<td>5.3%</td>
<td>0.59%</td>
<td>0.09%</td>
</tr>
<tr>
<td>Umbra at 18m/23m</td>
<td>100 mm</td>
<td>213 mm</td>
<td>213 mm</td>
<td>91 mm</td>
<td>113 mm</td>
<td>120 mm</td>
</tr>
<tr>
<td></td>
<td>100 mm</td>
<td>300 mm</td>
<td>300 mm</td>
<td>144 mm</td>
<td>173 mm</td>
<td>181 mm</td>
</tr>
</tbody>
</table>

Table 1 Candidates of L/D values

At the first stage of the construction we did not consider the devices for a phase contrast method and a grating method although we promote test experiments about such methods, too.

At the pulsed neutron imaging we used the neutron energy range from cold to fast neutrons and big samples. Therefore, shielding is one of most serious problems since the thickness of the shield places a limitation on the space of the experimental area. Figure 6 shows a preliminary result of the shielding calculations of a horizontal view. We confirmed that the total dose of neutrons and gamma rays was within the limitation of JAEA, and detailed calculation is now underway.
4. Conclusions

Test experiments and application to actual materials have been performed. Characteristic information by this method was obtained. Construction of a new imaging instrument ERNIS has been launched as a three-year project, and a design study for the construction is being executed. Bragg edge imaging, resonance absorption imaging and polarized neutron imaging are the first priorities. A large L/D will be covered to compensate for the lack of large L/D facilities in Japan. The first beam is expected by the end of the fiscal year 2013 and construction will be completed by the end of the fiscal year 2014.

Acknowledgements

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