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APPLICATION OF A PULSED NEUTRON TRANSMISSION METHOD TO A CULTURAL HERITAGE STUDY

Traditional neutron imaging has been used as one of important non-destructive methods and providing white beam transmission images. However, to obtain more detailed information, energy selective neutron images are recently becoming popular (Josic/Steuwer/Lehmann 2010). So far mainly steady neutron sources are used for neutron imaging. On the other hand pulsed neutron imaging can be used not only for white beam imaging and energy selective imaging, but also for spectroscopic imaging. In the spectroscopic imaging we can get position dependent physical quantities such as crystallographic structure, elements, magnetic field and so on (Kiyanagi et al. 2012; Sato/Kamiyama/Kiyangai 2011).
For the study of metal cultural heritages, this method will be useful to give new information nondestructively. Japanese swords are well known iron works made by a complicated method, and have different crystallographic characteristics depending on area and age where and when they were produced. So far, we measured fragments of swords (Kino et al. 2013; Nagashima et al. 2014). Here, we apply this method to analyze Japanese swords of different areas and ages. Furthermore, we performed a preliminary study on a coin from the Shin era in China to examine what kind of difference in the crystallographic structure is observed.

Principle of pulsed neutron imaging

In a pulsed neutron source, we measure the neutron energy, namely neutron wavelength by the time-of-flight method. Therefore, if we use a 2-dimensional position sensitive detector, we can get wavelength dependent transmission at each pixel of the detector. The transmission includes the crystallographic information and element information as shown in **figure 1**. In the wavelength dependent transmission Bragg edges appear at lower energy and their shape reflects the lattice spacing, preferred orientation, and crystallite size. On the other hand at higher energy dips due to resonance cross sections appear, and the energy of each resonance corresponds to the element. Therefore, we can obtain crystallographic and elemental information by using the pulsed neutron transmission.

Japanese swords

Difference due to area and age

First, we measured three fragments near the tang of the Japanese swords to study whether there are differences in crystallographic structures among them.

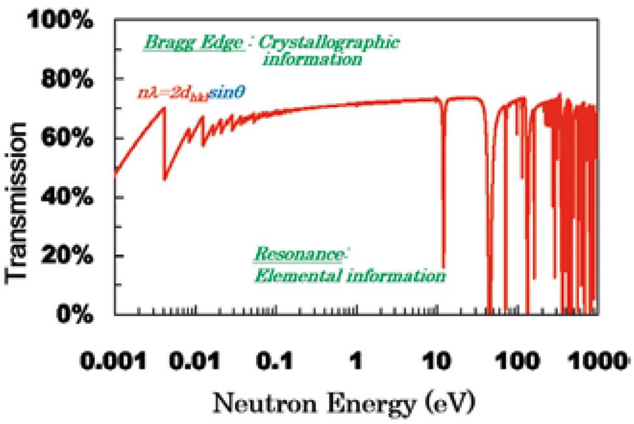

Fig. 1 Example of a transmission spectrum indicating the Bragg edges at low energy regions and resonance peaks at higher energy.

Fig. 2 Fragments of Japanese swords placed in front of the detector. From left to right: Bishu, Bizen and Sohshu (Mumei). – (Photo by the authors).

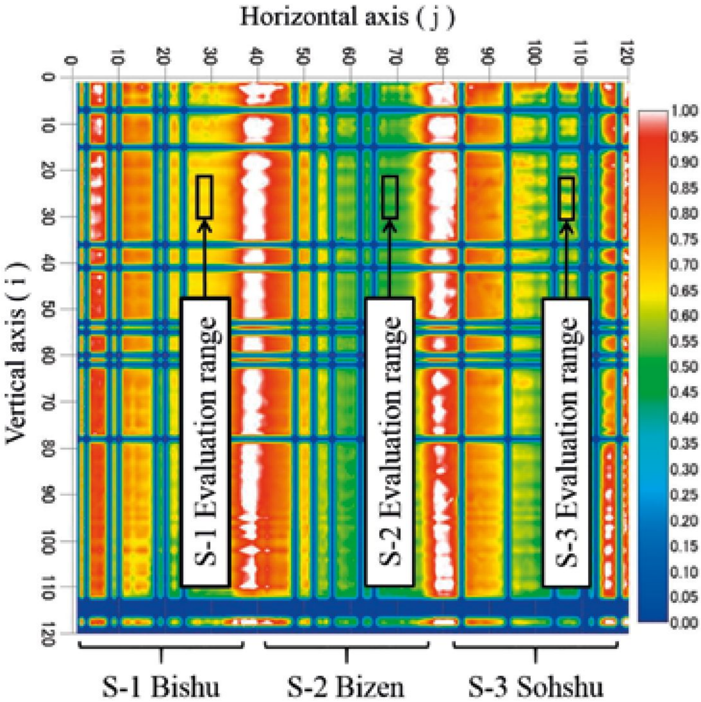

Fig. 3 Transmission image of each sword sample. Squares indicate the area those were used for evaluation of the transmissions. Axis i and j are pixel numbers with 0.8 mm pixel pitch.

The experiment was performed at the Hokkaido University neutron source (HUNS). HUNS is a compact accelerator-driven neutron source with a neutron intensity of about 1.6×10^{12} n/s at 1 kW power. We used a solid methane cold moderator in a coupled geometry. The flight path length was 5.06 m and the measuring time of the swords was 23.3 h, and that of direct beam 14.6 h. A GEM type imaging detector (Uno et al. 2012) was used. The pixel size is about 0.8 mm and the active area is 100 × 100 mm.

Production areas and ages of the swords are as follows:

(S-1) Bishu Osahune Sukesada (Signature on the sword)
Bishu is name of an area of Okayama prefecture. It was made in mid 16th century.

(S-2) Bizen no Kuni no Ju Osahune Shibei No Jo Sukesada
Bizen is name of an area of Okayama prefecture. It was made in late 17th century.

(S-3) Mumei (No signature)
This was made in Sohshu corresponding to Kanagawa prefecture in mid 16th century.

Figure 2 shows these sword sets in front of the GEM detector. From left to right, S-1, S-2 and S-3 were placed.

A transmission image is shown in figure 3, where the transmission value was averaged over a wavelength range from 0.1 nm to 0.45 nm. The blue lines are dead pixels since at that time the data acquisition system was not perfect. We chose the area enclosed with a black square for each sword to compare the crystallographic information. The areas are along vertical orientation i = 21-30 for all swords and horizontal orientation j = 27-29 for Bishu, 67-69 for Bizen, and 106-108 for Sohshu. They are almost the same distance from the tang and from the back of the sword. The total neutron cross sections are compared in figure 4. As easily recognized, there are differences in the shape. The shapes around the Bragg cut-off around 0.4 nm are similar for Bizen and Sohshu, since they have a rather sharp top. The smaller value of Sohshu suggests larger crystallite size, since in a large crystallite the probability of a double diffraction increases and it causes a reduction of the effective scattering cross section. On the

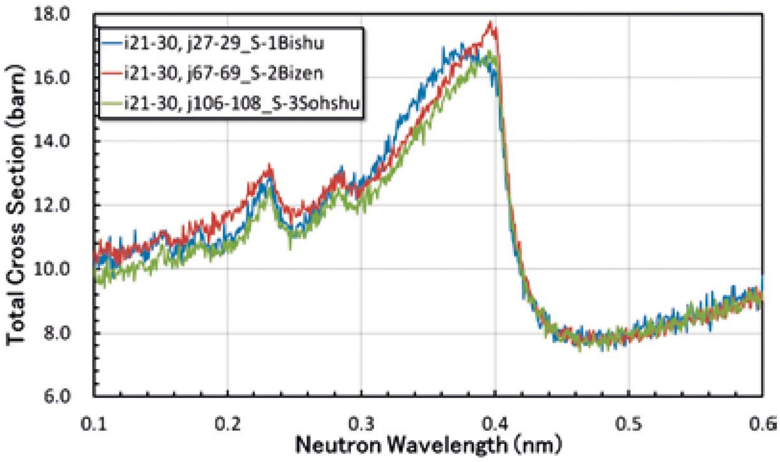

Fig. 4 Observed total neutron cross section of each sword.

other hand for Bishu the top is round, and it implies stronger texture compared with the other two. We analyzed these spectra by using the RITS code (Sato/Kamiyama/Kiyanagi 2011). The results are summarized in **table 1**. The crystallite size of Bishu and Bizen are almost the same and that of Sohshu is larger by more than 20 %. About the anisotropy we here used the March-Dollase factor. The factor is a measure of isotropy, and a value of 1 implies isotropic, and anisotropic nature increases with values diminishing from 1. Therefore, Bizen is almost isotropic and Bishu and Sohshu are rather anistropic.

The swords produced in the Okayama prefecture, Bishu and Bizen, have almost the same crystallite size, but are different in anisotropy. Sohshu has the largest crystallite size and medium anisotropy. However, it is not clear that these differences are caused by different areas and ages. For clarifying this situation, we need more data on the Japanese swords in these areas.

	Bishu	Bizen	Sohshu
Crystallite size (μm)	2.99	2.93	3.70
Crystal orientation anisotropy	1.99	0.90	1.34

Tab. 1 Crystallite size and March-Dollase coefficient as a measure of anisotropy.

Imaging of the quenched area

Next, we studied the change of the crystal lattice spacing from edge to back since the edge area was quenched. In the quenched area, a martensitic phase appears and gives larger lattice spacing (Kiyanagi et al. 2012). In this study we used a sword produced in the Okayama prefecture, in which the signature of Bizen Osahune no Ju Norimitsu was written. We measured the sword at MLF (Material and Life Science Experimental Facility) at J-PARC at 300 kW power for 5.5 h. The flight path length was 14.2 m and a MCP detector (Tremsin et al. 2013) was used. The detector has a high spatial resolution of 55 μm and an active area of 28 mm.

Figure 5 shows a photo of the sword, and the circle indicates the area analyzed. **Figure 6** shows a traditional transmission image (**fig. 6a**) and mapping of the lattice spacing of {110} (**fig. 6b**). In the traditional image some contrast difference appears. It is clearly indicated in **figure 6b** that the area with larger lattice spacing appears around edge. This area corresponds to the quenched region.

From the results shown above, it is indicated that the pulsed neutron transmission method can give non-destructively position dependent crystallographic information, and the information is useful to characterize the Japanese swords. We should promote

Fig. 5 Japanese sword used for the martensitic phase analysis. A circle corresponds to the area where data analysis was performed. – (Photo by the authors).

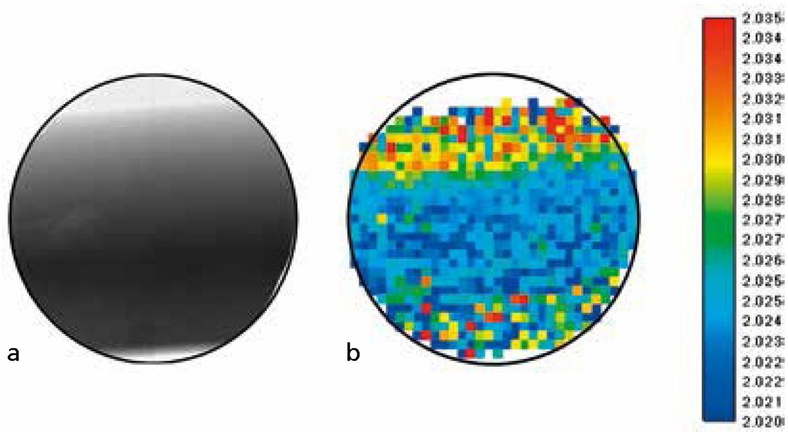

Fig. 6 Traditional transmission image (**a**) and bcc-Fe (110) lattice spacing mapping (**b**). The color indicator shows the lattice spacing in A unit.

Fig. 7 Coin made in China. – (Photo by the authors).

systematic research on the Japanese sword to clarify the crystallographic characteristics dependent on area and age.

China coin

So far, Bragg edge transmission experiments have not been performed for coins. To examine what kind of change of crystallographic characteristics appears in a coin, we measured a coin made in China. The sample is Kenryu Tsuhoh from the Shin period of 1736 to 1796. A photo is shown in **figure 7**, and the diameter is 23.6 mm. Components should be Cu and Zn from historical consideration. The measurement was performed at J-PARC at 300 kW power for 11.1 h. The sample was put before the MCP detector and the flight path length was 15.4 m. **Figure 8** shows traditional imaging. A contrast change was observed around the fringe, but the reason is unclear now. The squares indicated in the figure are the area where transmission data were obtained. **Figure 8** shows the transmissions depending on the wavelength for four areas of 100 × 100 pixels. Here, the size of each pixel is 55 μm, and wavelength dependent intensity correction has not been done in the transmission calculation. Therefore, the absolute value may not be correct, but relative comparison among the transmission spectra will be possible. The transmission spectra are different from each other, which indicates that there exists a position dependent change of anisotropy and crystallite size. As a preliminary analysis we calculated transmissions of three pure materials of Cu, Zn and Sn by using theoretical total cross sections. The results are shown in **figure 9**. The wavelength values at the Bragg edges of Cu correspond to the experimental ones although there is a difference in the absolute transmission value. On the other hand, those of Zn and Sn are not clearly observed.

Furthermore, we measured the resonance transmission to obtain the elemental information. **Figure 10** shows a transmission spectrum in which dips due to resonance transmission appear. For comparison we calculated the transmission intensity using resonance cross section data. The result is shown in **figure 11**, and it suggests that the Zn component is rather small. We are now developing the data analysis code for the resonance absorption, and after completion of the code we can evaluate the contents of the elements quantitatively.

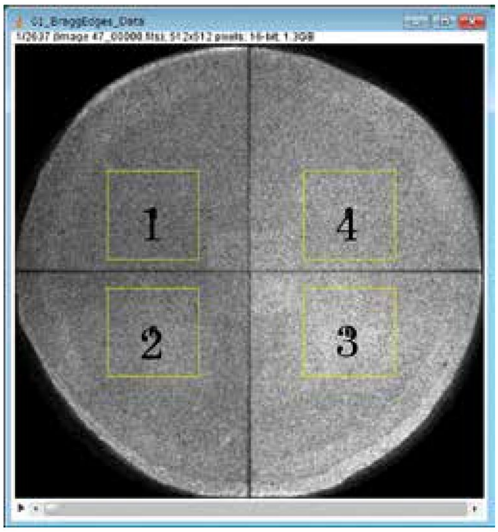

Fig. 8 Traditional transmission image of the coin. The squares are areas the transmission spectra were calculated and the numbers correspond to the transmission spectra shown in **fig. 9**.

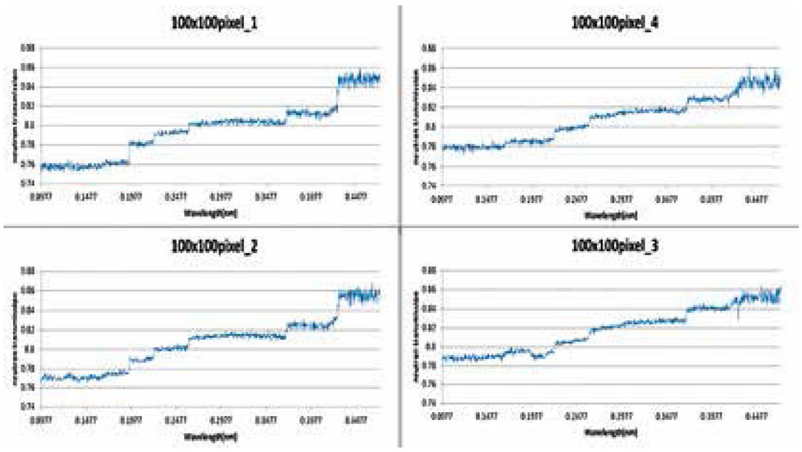

Fig. 9 Wavelength dependent transmissions at four different positions.

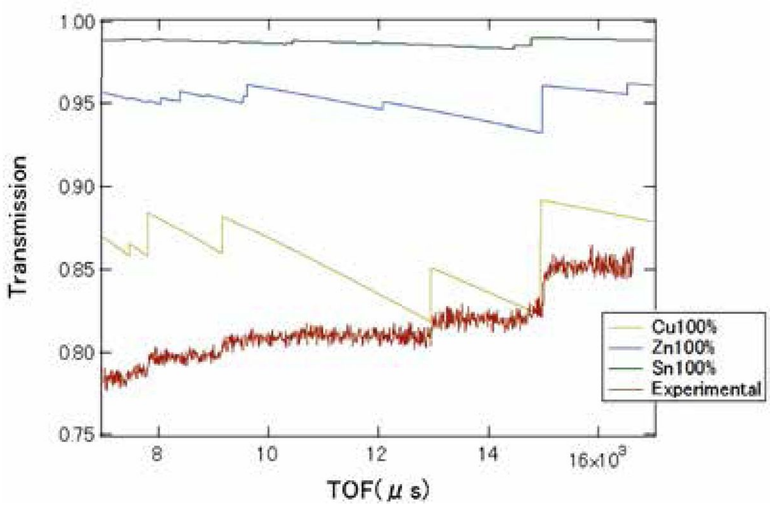

Fig. 10 Comparison of Bragg edge positions of the experimental transmission with those of pure Cu, Zn and Sn calculated by theoretical cross sections.

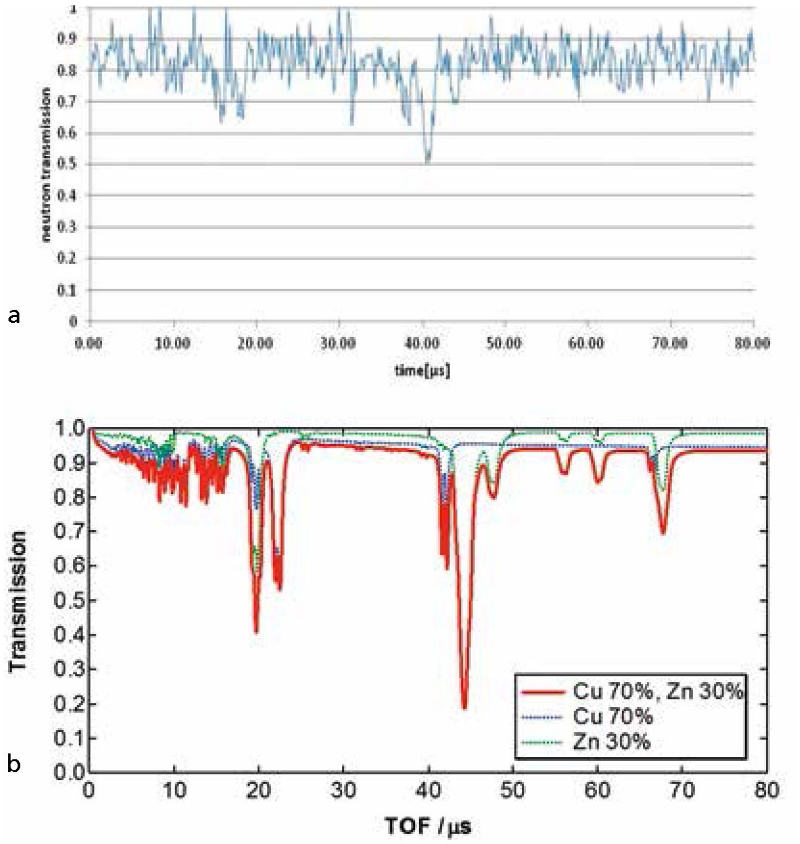

Fig. 11 Wavelength dependent spectrum of resonance transmission: **a** experimental result. – **b** simulation calculation.

Conclusion

We applied the pulsed neutron transmission method to the Japanese swords and the coin at J-PARC and HUNS, and obtained the crystallographic information and elemental information. Therefore, it is concluded that the method is useful for the study of metal cultural heritages not only at high power neutron sources but also compact neutron sources. However, materials with a complicated shape are at present not so easy to analyze. We have to improve the data analysis method so as to treat the complicated shape samples.

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Summary / Zusammenfassung

Application of a Pulsed Neutron Transmission Method to a Cultural Heritage Study

Pulsed neutron transmission imaging is a unique method that can give spatially dependent information on physical values of materials, such as crystallographic characteristics, elements, magnetic field, and so on. These data are obtained by analyzing the transmission spectra dependent on the neutron wavelength observed at each pixel of a 2-D position sensitive detector. We applied this method to cultural heritage samples. As test samples, we have studied Japanese swords. Three of them are fragments of swords with different areas and ages and the fourth is a full size one. We found that there were differences in texture and crystallite size among fragment samples, and the quenched area was clearly demonstrated in the full size sword. Furthermore, a Chinese coin was studied, and major elements and differences in crystallographic characteristics in the coin were indicated. The results indicated that the pulsed neutron transmission method can nondestructively give information that cannot be easily obtained by other methods.

Anwendung einer Transmissionsmethode mit gepulsten Neutronen für eine Studie zum Kulturerbe

Gepulstes Neutronen-Transmissions-Imaging ist eine einzigartige Methode, die ortsaufgelöste Information zu physikalischen Eigenschaften von Materialien geben kann, wie z. B. kristallographische Charakteristika, Elemente, magnetische Felder usw. Diese Daten erhält man, indem die Transmissionsspektren in Abhängigkeit von der Neutronenwellenlänge in jedem Pixel eines zweidimensional ortsauflösenden Detektors untersucht werden. Wir haben diese Methode auf Proben des kulturellen Erbes angewandt und zwar auf japanische Schwerter. Drei dieser Proben waren Fragmente von Schwertern mit verschiedenen Flächen und unterschiedlichem Alter, die vierte war ein Schwert von voller Größe. Wir fanden in den Fragmentproben Unterschiede in der Textur und in der Größe der Kristallite, und der gequenchte Bereich ließ sich im Schwert von voller Größe klar nachweisen. Weiterhin wurde eine chinesische Münze untersucht und die wichtigsten Elemente und Unterschiede in den kristallographischen Charakteristika in der Münze aufgezeigt. Die Ergebnisse zeigten, dass die gepulste Neutronen-Transmissions-Methode zerstörungsfrei Informationen liefern kann, die sich mit anderen Methoden nicht leicht gewinnen lassen.

Keywords

neutron imaging / pulsed neutron / energy dependent / Japanese sword / coin