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Analysis of crystallographic structure of a Japanese sword by the pulsed neutron transmission method

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Abstract

We measured two-dimensional transmission spectra of pulsed neutron beams for a Japanese sword sample. Atom density, crystalline size, and preferred orientation of crystals were obtained using the RITS code. The position dependence of the atomic density is consistent with the shape of the sample. The crystalline size is very small and shows position dependence, which is understood by the unique structure of Japanese swords. The preferred orientation has strong position dependence. Our study shows the usefulness of the pulsed neutron transmission method for cultural metal artifacts.

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Keywords: pulsed neutron; transmission; crystallographic structure; Japanese sword

1. Introduction

Japanese swords have been recognized as very high quality iron products because of their properties: they do not break, not bend, and are very sharp. Sword manufactory has a long history in Japan, and the unique manufacturing process that lead to these properties have been developed as early as 700 years ago.

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[1,2] The studies of Japanese swords from metallurgical point of view are focused on their crystallographic structure and the perfect combination of the different phases of carbon steel. Scanning electron microscopy (SEM) and the electron backscattering pattern (EBSP) have been applied to analyzing the metallurgy of the object. However, these methods are confined in scope to near the sample surface because of the limited penetration of the electron beams. Therefore, Japanese swords have to be destructively sampled to obtain internal crystallographic information, and this is often not acceptable, especially for the highest quality objects. In this case, neutron beam is a better probe because of its high penetration into materials with a high atomic number. The most popular technique using a neutron beam is the diffraction, which has been applied for the average characterization of Japanese sword fragments [3]. However, diffraction requires surveying over a wide area of a sample to get position dependence of the crystallographic structure. Recently, a method for analyzing the position dependent crystallographic structure has been developed, which uses the pulsed neutron-transmission method combined with a data analysis code RITS (Rietveld Imaging of Transmission Spectra) [4-6]. The RITS code provides quantitative data of the crystallographic structure by fitting the theoretical calculation on the Bragg edge structures seen on the wavelength spectra of transmitted neutrons. Spatially averaged crystallographic structure of the external and internal parts of the samples can be obtained without destructoying them. Therefore, we applied this new method on a Japanese sword fragment already analysed through neutron diffraction [see ref. 3].

2. Experiment and Analysis

We performed a neutron transmission measurement at the ISIS facility at the Rutherford Appleton Laboratory, UK. The N2 beam line (ROTAX) was used. The moderator is the 95 K methane. The proton beam was about 140 μA. A GEM (Gaseous Electron Multiplier) type two-dimensional neutron detector was set at 16.0 m from the moderator. This detector mainly consists of an aluminum cathode plate coated with 2μm thick boron-10, two GEM plates of 50 μm thick, and readout strips. The sensitive area is 100×100 mm^2 and the position resolution is 0.76 mm at FWHM. The neutron detection efficiency is about 6% for neutrons with a 0.18 nm wavelength. Figure 1 is a picture of the detector and sample setup. The Japanese sword sample was set just in front of the detector. The sample, which was provided by one of the collaborators (F. Grazzi), was made in the middle of 16th century in Okayama prefecture, Japan. The available beam spot size was about 43 mm × 27 mm. The number of pixels in this area is 1836 (54×34). Therefore, the area inside the dashed light blue line in Fig. 1 was analysed.

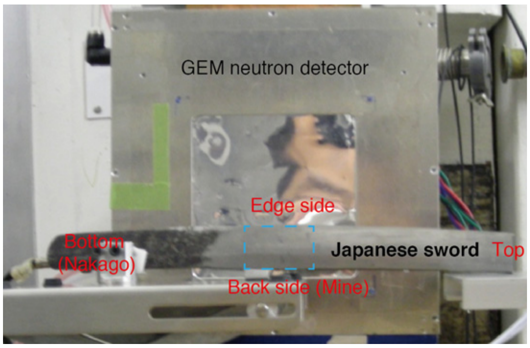

Fig. 1. Experimental setup. The area in the dashed light blue line was analyzed.

We obtained two data sets, one with the sample in the beam and another one without the sample. We normalized both data sets using the beam flux provided by a beam monitor set upstream the sample. The transmission spectra of all 1836 pixels were analysed using the RITS code. Figure 2 is an example of the data and fit results. There are some well visible Bragg edges, which are consistent with α-phase iron. The green curve is the fit result under the ideal condition that the crystalline size is zero and there is no preferred orientation of the crystallographic grains. This curve fails to represent the measured spectrum especially for the {110} Bragg reflection. The red curve is the fit obtained by parameterizing both the crystalline size and the preferred orientation, and represents the data much better than the green one.

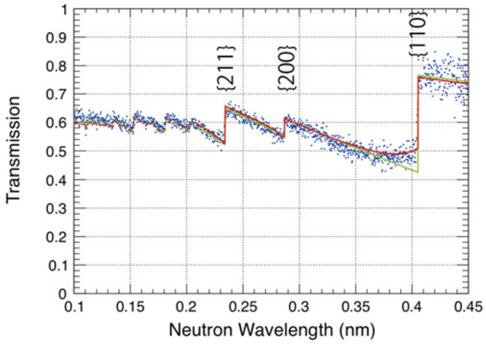

Fig. 2. An example of the fits. The green curve is the fit under the ideal condition that the crystalline size is zero and there is no preferred orientation of the crystalline grains. The red curve is the fit result with parameterizing the crystalline size and preferred orientation.

3. Discussion

Figure 3 shows the reduced χ^2 value distribution of all the fits. The reduced χ^2 values of most pixels are within 1.0 to 1.8 and this result indicates that the fits are reasonable. The reduced χ^2 values are larger near the backside of the sample and are smaller near the edge. The reason is probably that the Bragg edge structure is clearer as the sample is thicker parallel to the neutron beam. In any case, there is no large position dependence on the fitting accuracy.

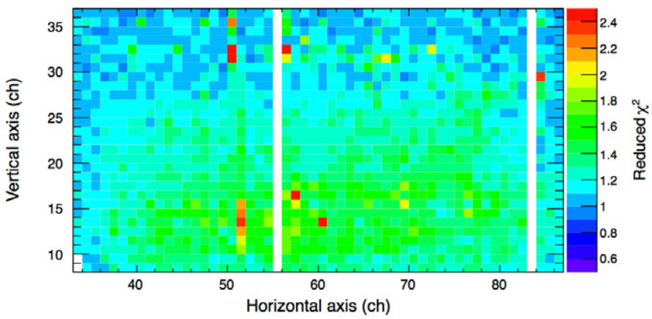

Fig. 3. The distribution of reduced χ^2 values for the all fits.

Figure 4 shows the distribution of the iron atoms density per unit area. This value is smaller near the edge. However, the densest area is not the back but a little bit inside the back. This is fully consistent with the shape of the Japanese sword sample studied.

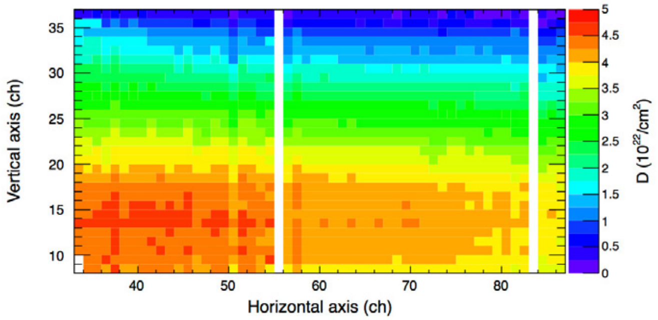

Fig. 4. The distribution of number of iron atoms per unit area (D) for the all fits.

Figure 5 shows crystalline size distribution. The sizes in most of the area are smaller than 5 μm and much smaller than the majority of common iron products. This is because Japanese swords are made of heavily worked steel. This feature makes Japanese swords difficult to bend and very sharp. The crystalline size is larger near the backside. This could be understood by the structure of Japanese swords. Low carbon steel with large crystalline size is wrapped on the sides and the edge by highly worked high carbon steel. Low carbon steel with large crystalline size is capable of absorbing strikes coming from the external high carbon layers and makes Japanese swords very difficult to break. In the backside of the sample in comparison with the edge side, the ratio of ferrite with a large crystalline size to that with a small size is larger. This explains the position dependence of the crystalline size in Fig. 5.

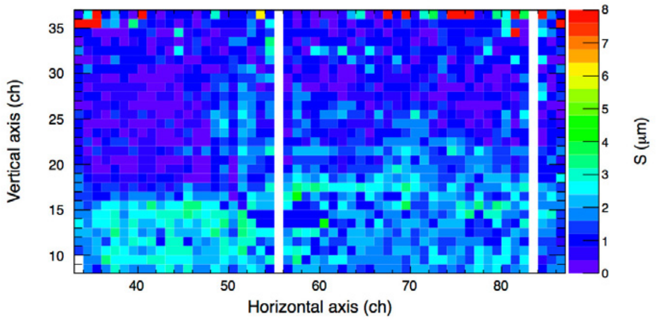

Fig. 5. The distribution of crystalline size for the all fits.

Figure 6 shows the distribution of the degree of the preferred orientation of crystals. The value 1.0 corresponds to the completely uniform orientation. If the value deviates from 1.0, the preferred orientation becomes non-uniform. Figure 6 indicates that the degree of the preferred orientation has a large dependence on the position. Furthermore, this dependence is much different from that of the crystalline size. This result might be due to the non-uniformity of the manufacturing process.

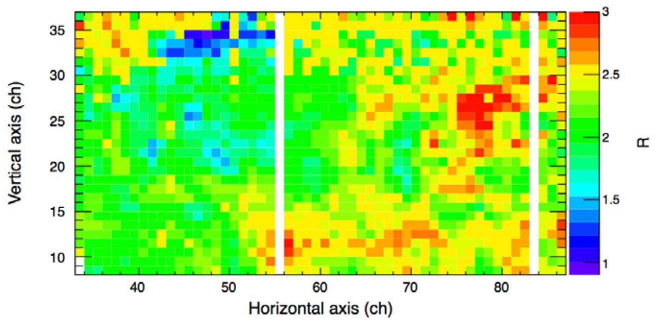

Fig. 6. The distribution of the degree of the preferred orientation of crystals (R) for the all fits.

4. Summary

We measured the transmission spectra of a Japanese sword using the pulsed neutron beam at ISIS. These spectra are analysed by the RITS code. The distributions of the number of iron atoms per unit area, crystalline size, and the degree of the preferred orientation were obtained. The position dependence of the number of atoms was consistent with the sample shape. The crystalline size was very small compared to that of common steel products. The position dependence could be understood by the unique structure of Japanese swords. The degree of the preferred orientation showed non-uniform distribution.

We extracted the internal crystallographic structure of a Japanese sword in a non-destructive way by using the pulsed neutron transmission method. Our successful result showed the usefulness of this method for studying crystallographic structure of cultural metal artifacts.

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