Relation between Vickers Hardness and Bragg-Edge Broadening in Quenched Steel Rods Observed by Pulsed Neutron Transmission Imaging

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The width of crystal lattice plane spacing (d-spacing) distribution related to microscopic-strain and crystallite size in a martensite phase in a 2 cm thick quenched-ferritic steel sample was quantitatively mapped in real space by a Bragg-edge broadening analysis of spectral data from a pulsed neutron transmission experiment. This analysis was performed under the condition that the instrumental resolution parameters, determined from the data of ferrite in the same sample without microscopic-strain and crystallite size effects, were unchanged over the sample area, and assuming that the d-spacing was distributed according to a Gaussian function in the martensite area. As a result, the full width at half maximum (FWHM) of the Gaussian d-spacing distribution in the martensite was extracted at each position in a sample. Consequently, it was found that the real-space distribution of the FWHM of the d-spacing distribution is closely correlated with a real-space distribution of the Vickers hardness that corresponds to the quantity of martensite. Furthermore, it was indicated that the Vickers hardness was proportional to the FWHM of the d-spacing distribution. The results suggest that it will be possible to measure the Vickers hardness in the martensite non-destructively by using the Bragg-edge neutron transmission method. [doi:10.2320/matertrans.M2015049]

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1. Introduction

Bragg-edge transmission imaging using a pulsed neutron source is a new material characterization tool that makes it possible non-destructively to map crystalline microstructural information of a bulk material with spatial resolution around 100 µm over a large area (the order of 10 cm × 10 cm). The material properties obtained are crystal structure, crystalline phase, crystallographic texture, crystallite size and crystal lattice strain, which are deduced by using the Rietveld-type Bragg-edge analysis software, RITS (Rietveld Imaging of Transmission Spectra). Therefore, in one measurement, it can provide such information over a larger area than SEM (scanning electron microscopy)-EBSD (electron backscatter diffraction), X-ray/neutron scattering and synchrotron radiation phase-contrast microtomography.

Quenching is a popular method to harden the surface of the steel. It is well known that the micro-crystalline structure of the martensite in the quenched area is not simple since the crystal lattice plane spacing (d-spacing) depends on the carbon content and the content is not uniform. However, the availability of crystallographic information obtained by a non-destructive method is useful for inspection of the quenched region. For this purpose, we performed pulsed neutron transmission measurements on quenched ferritic steel rods (Fe-0.45%C, 2.6 cm diameter and 2 cm height) with 3, 5 and 7 mm expected quenched thicknesses. We deduced in the previous work the d-spacing with the use of RITS, and successfully assigned the quenched areas, coinciding with those expected. However, the average crystal lattice {110} plane spacing of 0.2042 nm in the quenched (martensite) region is much larger than the normal value of 0.2032 nm for martensite with a composition of Fe-0.45%C.

For more accurate evaluation of martensite phase information without such overestimation, the Bragg-edge broadening caused by microscopic-strain and crystallite size effects, which is equivalent to diffraction peak broadening, should be considered in the analysis. Therefore, the broadening will show the distribution of martensite and it relates to hardness. In the present study, the RITS code was revised in order to perform more reasonable analysis taking into account the Bragg-edge broadening, the width of d-spacing distribution was correctly deduced, and was compared with the Vickers hardness which correlates with the quantity of martensite. Then, the relation between the Vickers hardness and the Bragg-edge broadening was investigated in detail. This method will lead to a method to extract information of dislocation density from the Bragg-edge broadening as in a diffraction peak line-broadening analysis.

2. Development of the Bragg-edge broadening analysis method

The new data analysis procedure developed using the RITS code is described as follows. For crystal lattice plane spacing and strain analysis, the profile fitting analysis of a Bragg-edge has to be carried out. In the RITS code, the Jorgensen-type edge-profile function $R_{hkl}(\lambda - 2d_{hkl}, \sigma_{hkl}, \alpha_{hkl}, \beta_{hkl})$ is used. Here, $\lambda$ is the neutron wavelength, and $d_{hkl}$ is the d-spacing of the crystal lattice plane $\{hkl\}$. This function is obtained from the convolution of the Heaviside step function and a Gaussian function (the standard deviation $\sigma_{hkl}$) with two back-to-back exponentials (the rise constant $\alpha_{hkl}$ and the decay constant $\beta_{hkl}$). The parameters $\sigma_{hkl}$, $\alpha_{hkl}$ and

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\( \beta_{hkl} \) express the instrumental resolution function at the Bragg-edge, which are in principle constant and independent of position in a sample. If the \( d \)-value is spatially uniform, this analysis is correct. However, for samples with the martensite phase, the \( d \)-value itself changes with content of the microstructure, and become to have a distribution. Effect of this phenomenon on the Bragg-edge is illustrated in Fig. 1 comparing with that on a diffraction peak. In macroscopic-strain case, the position of the Bragg-edge shifts uniformly. On the other hand, in microscopic-strain/crystallite-size case, the gradient of the Bragg-edge changes to gentle curve due to the broadening of the \( d \)-distribution. Therefore, especially in this case where the \( d \)-values are distributed, the instrumental resolution has to be constant independent of position, and we have to introduce another broadening parameter for Bragg-edge analysis.

In the new analysis, it was assumed that the distribution of \( d \)-spacing depending on the microscopic-strain and the crystallite size follows a Gaussian function. Therefore, the whole edge-profile Jorgensen-type function can be divided into the Jorgensen-type instrumental resolution function and the Gaussian \( d \)-spacing distribution function:

\[
R_{hkl}(\lambda - 2d_{hkl}, \sigma_{hkl}, \alpha_{hkl}) = R_{hkl}(\lambda - 2d_{hkl}, \sigma_{hkl}, \alpha_{hkl}) \odot G(\lambda - 2d_{hkl}, \sigma'_{hkl}),
\]

where \( G(\sigma) \) is a Gaussian function with a standard deviation \( \sigma \). Thus, since the convolution of two Gaussian functions can be expressed analytically, \( \sigma_{hkl} \) can be separated into two standard deviations \( \sigma_{0_{hkl}} \) and \( \sigma_{1_{hkl}} \) as follows:

\[
\sigma_{hkl}^2 = \sigma_{0_{hkl}}^2 + \sigma_{1_{hkl}}^2,
\]

where \( \sigma_{0_{hkl}} \) is the standard deviation of the Jorgensen-type edge-profile function, expressing the width of broadening of Bragg-edge due to the instrumental resolution. Now, \( \sigma_{1_{hkl}} \) is the standard deviation of the Gaussian function, representing the width of broadening of Bragg-edge due to the microscopic-strain and the crystallite size.

### 3. Experimental

Figure 2 shows a photograph of samples. The specimens were three ferritic steel rods (Fe-0.45% C, 2.6 cm diameter and 2 cm height) quenched from each rim surface by axial-symmetric induction hardening. The quenched depths from the surface were expected to be 3 mm, 5 mm and 7 mm, respectively. These different thicknesses were produced by changing the time of the induction hardening, and the actual thicknesses were evaluated by the Vickers hardness measurement as described later.

Figure 3 shows a schematic layout of setup of pulsed neutron transmission imaging experiments. The pulsed neutron transmission experiments were carried out at the neutron engineering test port “NOBORU”9) at the BL10 neutron beam-line of MLF (Materials and Life Science Experimental Facility) at J-PARC (Japan Proton Accelerator
Research Complex). The accelerator beam power was 120 kW during this experiment, and it corresponded to a cold neutron flux of about $5.8 \times 10^6$ n/cm²/s at the sample/detector position. The neutron wavelength resolution is 0.34\% at the detector position of 14 m from the decoupled-type para-H₂ neutron moderator. The beam angular divergence was ± 3 mrad. The two-dimensional neutron time-of-flight detector used was a neutron ¹⁰B-MCP (Micro Channel Plate) detector.¹⁰ The pixel size was 55 μm × 55 μm, and the detection area was 1.4 cm × 1.4 cm. The binning width of the neutron TOF spectroscopy was 4 µs; the binning width of the neutron wavelength was 1.13 Å or 10⁻⁴ nm which was significantly smaller than the neutron wavelength resolution in the measured wavelength region. The measurement time for the incident neutron beam in the absence of a sample was 3.0 h for the 3 mm-depth quenched rod, and 4.0 h for the 5 mm- and 7 mm-depth quenched rods. The measurement time for transmitted beam when samples were present was 9.0 h for the 3 mm-depth quenched rod, 8.0 h for the 5 mm-depth quenched rod, and 5.0 h for the 7 mm-depth quenched rod, respectively. The neutron flight path length from the moderator to the detector was 14.04 m for the 3 mm-depth sample, and 14.42 m for the 5 mm- and 7 mm-depth samples. The samples were set closely in front of the detector. Neutrons were transmitted along the axial direction of the sample cylinder, and the neutron transmission data contain the crystalline microstructural information averaged over the axial direction of the specimen (2 cm thickness).

4. Results and Discussion

4.1 Real-space mapping of the width of crystal lattice plane spacing distribution

Figure 4 shows Bragg-edge neutron transmission spectra of the center region and the rim region of the 7 mm-depth quenched rod. In the analysis, firstly, we analyzed the data of the center region of the cylindrical specimen, which consists mainly of ferrite. In this first-stage analysis, we derived the parameters $\sigma_{0,hkl}$, $\kappa_{hkl}$ and $\beta_{hkl}$ describing the instrumental resolution function, under the condition that $\sigma_{1,hkl}$ is zero (no microstructure caused by the martensite). Secondly, we determined the crystal lattice plane spacing $d_{hkl}$ and the microscopic-strain/crystallite-size parameter $\sigma_{1,hkl}$ by analyzing the data over whole area of the specimen, under the condition that the instrumental resolution parameters $\sigma_{0,hkl}$, $\kappa_{hkl}$ and $\beta_{hkl}$ obtained in the first-stage analysis, were constant. Figure 4(b) shows experimental Bragg-edge {110} data with the profile fitting curves obtained by the developed data analysis procedure.
It was shown that a Bragg-edge broadened by martensite-induced microstructure (in the rim region) has been profiled by this data analysis procedure, indicating that the procedure for the determination of the instrumental resolution function by using a sharp ferrite Bragg-edge is acceptable. From this fitting, two values have been obtained. First one is the \( d \)-value, and second one is the standard deviation of \( d \)-value distribution. First, it was found that the extracted \( d \)-values were reasonable as expected from the carbon content, Fe-0.45% C (as 0.2026 nm for ferrite (in the center region) and 0.2032 nm for martensite (in the rim region)), which indicates that an overestimation of the average \( d \)-spacing in the martensite region (0.2042 nm)\(^{39}\) was corrected by this analysis method. Next, a standard deviation \( \sigma_{110} \) of the Gaussian \( d \)-spacing distribution due to the microscopic-strain and the crystallite size was estimated in a value of the order of 10\(^{-3}\) nm from the data in the martensite region. The meaning of this new parameter is discussed later.

Figure 5 shows results of mapping of crystal lattice plane spacing \( d_{110} \) (a), (b) and (c) and \( w_{110} \) (d), (e) and (f), of the 7 mm-, 5 mm- and 3 mm-depth quenched ferritic steel rods. Here, \( w_{110} \) is the FWHM (full width at half maximum) of a Gaussian \( d \)-spacing distribution, and is defined as:

\[
  w_{110} = 2\sqrt{2}\ln 2\sigma_{110}. \tag{3}
\]

The data in this figure indicates broadening of the Bragg-edge \{110\} only due to the microstructure of martensite. From Fig. 5(a)~(c), it is found that the reasonable values around 0.2026 nm for ferrite region and also those around 0.2032 nm for martensite region are obtained, which agree with the results indicated in Fig. 4(b). From Fig. 5(d)~(f), it is recognized at the mere sight of these that the area having the value of \( w_{110} \) of the order of 10\(^{-3}\) nm almost corresponds to the quenched depth (7 mm, 5 mm or 3 mm) from the surface.

4.2 Comparison with the Vickers hardness

The significance of the obtained parameter, \( w_{110} \), is discussed in terms of the quantity of martensite and also microscopic-strain by using the Vickers hardness data. At first, we calculated the radial dependent data of \( w_{110} \) of each rod by averaging over a circular region of data shown in Fig. 5(d)~(f). The center of the circular average corresponds to the center of a cylinder axis. Figure 6(a) shows radial changes of \( w_{110} \) of the 7 mm-, 5 mm- and 3 mm-depth quenched rods. Continuously, we fitted the sigmoid functions to the radial dependences of \( w_{110} \) (curved lines in Fig. 6(a)), and derived the inflection point of each curve (vertical dashed lines in Fig. 6(a)). As a result, the inflection points derived are 6.5 mm, 5.2 mm and 3.3 mm for 7, 5 and 3 mm-depth quenched rod, respectively. These inflection points seem to correspond to the actual depths of quenching.

To verify this assumption, the result of Fig. 6(a) was compared with the radial dependence of the Vickers hardness, which is correlated to the quantity of ferrite/martensite. In particular, the Vickers hardness 450 (Hv 450) is well known as the hardness boundary between ferrite and martensite. Figure 6(b) shows the radial dependence of the Vickers hardness of each quenched rod. Note that the hardness data were obtained just along one radial direction path, not by circular-averaging. From this figure, it is found that the shapes of the radial dependences of \( w_{110} \) are quite close to those of the Vickers hardness for each quenched rod. Additionally, the point of Hv 450 also corresponds to the
inflection point of the \( w_{110} \) real-space distribution for each quenched rod. Therefore, this result indicates that the \( w_{110} \) depicts the martensite/ferrite concentration ratio in a ferritic material.

Finally, we deduced the relation between the Vickers hardness value and the \( w_{110} \) value by comparing both values at the same position. Figure 7 shows the plots of the Vickers hardness as a function of \( w_{110} \), with the fitting lines. This figure shows that the Vickers hardness is linearly proportional to the FWHM of the distribution of crystal lattice plane spacing, which also indicates that the width is linearly proportional to the quantity of ferrite/martensite. Furthermore, it was shown that quantitative mapping of such information can be carried out with high spatial resolution over a large area owing to the Bragg-edge neutron transmission imaging.

5. Conclusion

By using the pulsed neutron Bragg-edge transmission imaging combined with the revised RTS code, the broadening of a Bragg-edge of martensite in a bulk ferritic material was analyzed as a Gaussian \( d \)-spacing distribution, over a large area with high spatial resolution. With this method, reasonable \( d \)-spacing information and new information related to the microscopic-strain and the crystallite size can be extracted quantitatively. Quantitative mapping results of the Bragg-edge broadening due to the microscopic-strain and the crystallite size can reproduce the Vickers hardness real-space distribution very well, showing that quantitative crystalline phase imaging of martensite in a ferritic material is possible in spite of their small differences in crystal structure. Furthermore, since it is found that the Vickers hardness is linearly proportional to the FWHM of the \( d \)-spacing distribution, an estimation of the material hardness in a ferrite/martensite steel may be carried out by using this method.

Furthermore, this study also provides the first detailed quantitative result that the broadening of a Bragg-edge is equivalent to the broadening of a diffraction peak. And, their results show the possibility of non-destructive measurement of the hardness distribution of steel materials with a martensite structure by Bragg-edge analysis. The Bragg-edge profile analysis can be indeed based on the same principle as the diffraction peak profile analysis. As a result, in the future, information on dislocation density and crystallite size can be non-destructively visualized by further improvement of the Bragg-edge analysis.

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