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Characterization of microstructure using Bragg edge and energy-resolved small-angle neutron scattering

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Abstract: The combined use of neutron diffraction (ND), small-angle neutron scattering (SANS), Bragg edge transmission and energy-resolved SANS (ESANS) is useful to characterize microstructures in steels. ND and Bragg edge transmission give crystallographic information, while SANS and ESANS are suitable for analyzing precipitates. The simultaneous measurement of conventional SANS and Bragg edge transmission can be carried out easily by using time-of-flight (TOF) SANS instruments. The combined analysis of Bragg edge and ESANS needs only measurements of neutron transmission spectra. This can be applied to neutron imaging experiments, although it is difficult to separate the contributions of magnetic and nuclear scattering. Moreover, synchrotron X-ray diffraction (XRD) and small-angle X-ray scattering (SAXS) are useful especially for time-resolved experiments, although the gauge volume for synchrotron X-ray studies is lower than that of neutron. It is necessary to select a suitable combination of these techniques based on the purpose of the experiment.

1. INTRODUCTION

Neutron scattering techniques are useful for characterizing microstructures in steels and are frequently used for *in-situ* measurements due to access to a large gauge volume and high penetration power. In current steel research, neutron diffraction (ND) and small-angle neutron scattering (SANS) are used widely [1-4]. ND provides crystallographic information about the steel matrix, while SANS provides morphological information about precipitates and inclusions. The total characterization of microstructures using both ND and SANS is needed for further research and developments on advanced steels. However, in neutron scattering experiments, sample environment equipment such as tensile testing machine and furnace often shadow the detector, thereby limiting the accessible angular range of the neutron measurements.

Recent developments in neutron transmission analyses have created an opportunity to solve this issue. The wavelength dependence (spectrum) of neutron transmission contains the contributions of neutron absorption as well as scattering. Bragg edge transmission analysis can be used to obtain the contribution of diffraction to neutron transmission and provide information basically identical to that obtained using ND [5]. As a method to analyze the SANS contribution, energy-resolved SANS (ESANS), which can provide morphological information about the precipitates, has been proposed recently [6]. Since these neutron transmission measurements require only a transmission monitor, the sample environmental equipment need only have small windows for the incident and transmitted neutron beams. In addition, the neutron transmission spectra can be measured easily using time-of-flight (TOF) analysis at new-generation pulsed neutron facilities such as J-PARC and SNS.

By contrast, conventional ND and SANS remain beneficial because they can be used to characterize details of any crystallographic orientation dependence, which are averaged in the neutron transmission spectra. Proper combination of ND, SANS, Bragg edge transmission analysis, and ESANS must be selected based on the purpose of the experiments. In this manuscript, several

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applications of the simultaneous measurements of ND, SANS, Bragg edge transmission analysis, and ESANS in steel research are shown. This improves the flexibility of the experiments and enables the total quantitative characterization of the precipitates and the steel matrix.

2. BRAGG EDGE TRANSMISSION AND SANS

The first example is the combined use of Bragg edge transmission analysis and conventional SANS. The advantage of this technique is that the Bragg edge can be measured easily by using the neutron transmission monitor equipped in most TOF-SANS instruments, thus eliminating the need for an additional device (Fig. 1) [7]. This allows for the simultaneous characterization of the precipitates and the steel matrix by using TOF-SANS. The first combined measurement of Bragg edge and SANS was conducted using the small- and wide-angle neutron scattering instrument BL15 TAIKAN installed at J-PARC [7,8]. The sample was carbon steel with an additive. Nano-sized precipitates were formed by tempering at 873 K [9,10]. The matrix was subsequently transformed to ferrite by reheating at 1273 K followed by furnace cooling. The results confirmed that the Bragg edge of steels can be obtained on a TOF-SANS instrument. SANS characterized the size distribution of the precipitates. The obtained Bragg edge could be excellently interpreted by the analysis technique used in neutron imaging experiments. It provided the degree of crystallographic texture and the crystallite size of the ferrite matrix. The pending issues pertaining to this technique are improvements of the statistical accuracy of neutron transmission and the resolution of lattice spacing. Most SANS instruments are more concerned with higher intensity rather than higher resolution of lattice spacing. However, these improvements enable precise analysis of lattice strains and dislocation density which are valuable for microstructural analysis.

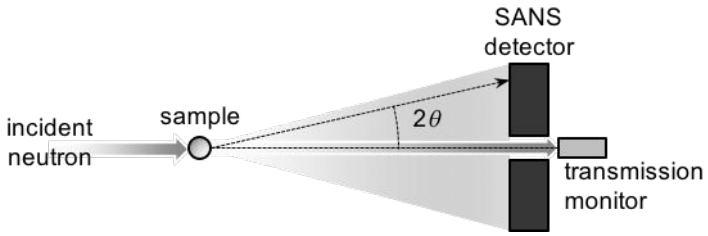

Fig. 1 Schematic illustration of a TOF-SANS instrument.

3. BRAGG EDGE TRANSMISSION AND ESANS

Similarly, Bragg edge transmission and ESANS can be combined easily. In this case, since both contributions appear in the neutron transmission spectra, only a transmission monitor is required. Fig. 2 shows the neutron transmission of ferritic steel with precipitates of vanadium carbide obtained using TAIKAN [6]. The experimental result is well explained by the summation of Bragg edge, ESANS, and extra contributions. The extra contributions include absorption, elastic incoherent scattering, inelastic coherent scattering, and inelastic incoherent scattering. The value of the extra contribution can be simply calculated from the mass density and the chemical composition of the sample [5,11]. The effect of the microstructures to the extra contributions can be ignored. The ESANS and Bragg edge contributions contain information about the microstructures. The Bragg edge appears at the wavelengths shorter than the Bragg cut-off, which is the jump corresponding to the largest lattice spacing and is equal to 0.4 nm in the case of the ferrite matrix. Only the ESANS contribution can be analyzed at wavelengths longer than 0.4 nm. The dashed line in Fig. 2 denotes the calculated ESANS contribution from the conventional SANS profile measured on the SANS instrument QUOKKA installed at the Australian Nuclear Science and Technology Organisation (ANSTO) [6,12]. The sum of the calculated ESANS and extra contributions agree well with the experimentally observed neutron transmission. At wavelengths shorter than 0.4 nm, the total sum of the ESANS, extra, and Bragg edge contributions can explain the experimental results.

The combination of Bragg edge transmission and ESANS has the potential for application to neutron transmission imaging in a manner similar to Bragg edge imaging [5]. Furthermore, the Bragg edge can be characterized more accurately with ESANS analysis because the ESANS contribution overlaps with the Bragg edge contribution in the neutron transmission spectra. However, it is difficult to separate the magnetic and nuclear scattering contributions from a single neutron transmission

measurement [6]. ESANS is sensitive to the summation of the magnetic and nuclear scattering contributions, whereas conventional SANS can separate them from their azimuthal anisotropy. Although the magnetic scattering contribution makes it difficult to precisely analyze the precipitates, information from ESANS is beneficial from the viewpoint of qualitative analysis. Two measurements with different directions of a magnetic field would probably be adequate to separate the magnetic scattering contribution.

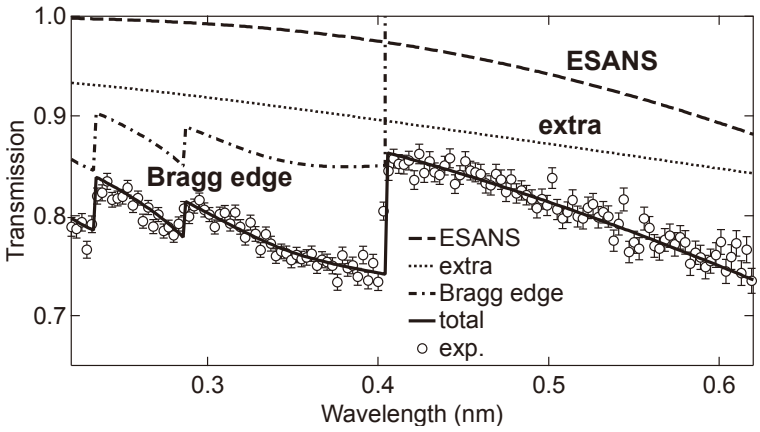

Fig. 2 Neutron transmission spectra of ferritic steel with vanadium carbide. Plots are experimental results; dashed, dotted, dashed-dotted, and solid lines are ESANS, extra, Bragg edge contributions, and their summation.

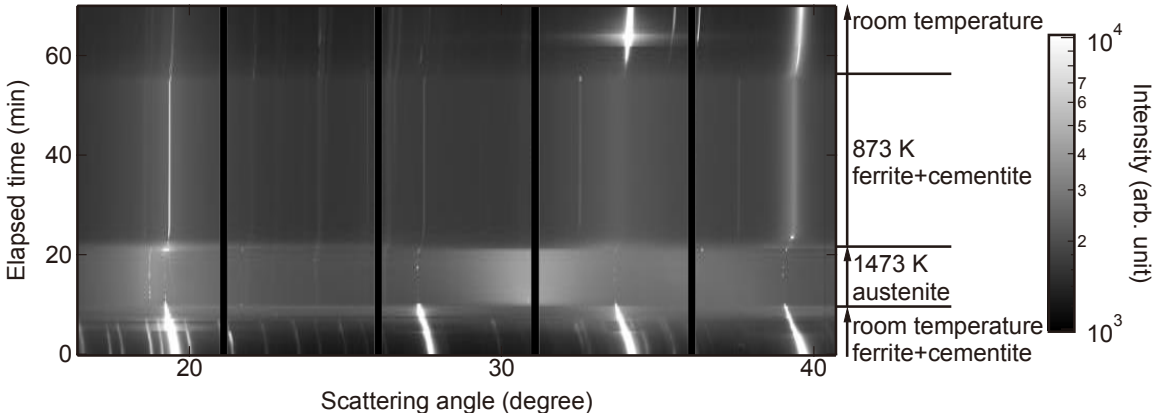

Fig. 3 Time dependence of XRD profiles during heat treatment of medium carbon steel. Dark vertical lines at the scattering angles = 21, 26, 31, and 36° are insensitive areas of detector.

4. X-RAY DIFFRACTION AND SMALL-ANGLE X-RAY SCATTERING

The combined measurements can also be carried out using X-ray. X-ray diffraction (XRD) and small-angle X-ray scattering (SAXS) provide information similar to ND and SANS, respectively. In addition, synchrotron X-ray enables access to higher speed time-resolved measurements compared to neutron. Another feature of the synchrotron X-ray is a small beam size compared to neutron. This is useful to characterize local microstructures, whereas the gauge volume of X-ray scattering is lower than that of neutron scattering. Fig. 3 shows the time dependence of XRD profiles during heat treatment of medium carbon steel measured at the beamline BL46XU at SPring-8 using a one-dimensional position sensitive detector (1D-PSD). The heat treatment condition is shown on the right side in Fig. 3. The energy of the incident X-ray was 18 keV. The thickness of the specimen was about 40 μm to enable sufficient X-ray transmission. The measurement time of each XRD profile is 20 seconds. Before heating, the Bragg peaks of ferrite and cementite are observed. During holding at 1473 K, all Bragg peaks almost disappear. Although this indicates the phase transition to austenite, the corresponding Bragg peaks are scarcely observed. When the temperature decreases to 873 K, the Bragg peaks of ferrite and cementite reappear. The few Bragg peaks in austenite can probably be ascribed to the very large grain size of the austenite phase. The beam size used in this experiment was about 0.2 mm in height and width. Since this value will be close to the austenite grain size, only few

austenite grains lie within the beam path. In such a case, the diffracted X-ray turns to spots rather than Debye rings; it is consequently difficult to observe the Bragg peaks of austenite using 1D-PSD.

5. SUMMARY

The recent development of neutron scattering techniques provides the potential for several new experiments that are useful to characterize the microstructures in steels. Total characterization of the steel matrix and the precipitates can be performed using simultaneous measurements of Bragg edge transmission and SANS or Bragg edge transmission and ESANS. Other combinations, for example, ND and SANS or ND and ESANS, are possible as well. Although the combination of XRD and SAXS is useful for time-resolved measurements, the limitation of small gauge volume needs to be considered.

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