JFE TECHNICAL REPORT
No. 22 (Mar. 2017)

Microstructural Evaluation Technique for Steel Using Neutron Beam†

NAKAMICHI Haruo[*1] SATO Kaoru[*2] SUEYOSHI Hitoshi[*3]

Abstract:

Structural analysis using neutrons is a unique technique from the viewpoint of its strong penetration ability in steel materials. An extremely wide range of neutron measurement techniques has been available. Focusing on microstructural analysis using neutron techniques, JFE Steel has performed a large number of analyses through participation in research activities of the Iron and Steel Institute of Japan and others. Among those activities, this paper introduces results of characterization of precipitates by small angle neutron scattering, estimation of residual stress in welds by diffraction and in-situ observation of the transformation process during deformation by the time-of-flight neutron beam diffraction.

1. Introduction

Neutrons are extremely promising for structural analysis of the interior and bulk of materials due to their strong penetration ability and have been the subject of numerous studies. Various efforts to apply neutron microstructural evaluation techniques to steel materials have already been carried out, including the "Basic Study for Application of Neutron Techniques on Steel Structural Analysis" (FY 2006–2008)[1)], "Application of Advanced Neutron Source for Research of Elemental Operation in Steel" (FY 2009–2012)[2)] and "Characterization of Microstructure in Steel by Compact Neutron Source" (FY2013-) in the projects of the Iron and Steel Institute of Japan (ISIJ). Accompanying progress in the operation of J-PARC (Japan Proton Accelerator Research Complex), which boasts the world's most powerful neutron beam, dynamic measurement observation technique of heat treatment and deformation processes have also been progressing. This technique is very important for the steel manufacturing process and development of steel product. Microstructural evaluation techniques utilizing a compact neutron source have also progressed. Based on these improvement of neutron techniques, it is expected that this analytical techniques on steels will be widely used near future. JFE Steel has actively participated in these research projects, etc. on neutron-based techniques since their inception, and is using or studying new characterization methods. Among the results of those research groups, this paper introduces an example of morphological evaluation of fine precipitates in steel by using small angle neutron scattering, measurement of the residual stress in welds of steel plates and *in-situ* observation of the tensile deformation behavior of TRIP (transformation induced plasticity) steel during deformation.

2. Evaluation of Precipitates in Steel by Small Angle Neutron Scattering[3)]

2.1 Background

Because fine precipitates in steel products have a large influence on material strength, evaluation of their morphology is important. Various evaluation techniques are used for this purpose, such as the transmission electron microscope[4)] (TEM) and the atom probe. However, it is difficult to obtain average information on precipitates by these techniques due to their limited observation region. On the other hand, in characterization by small

† Originally published in *JFE GIHO* No. 37 (Feb. 2016), p. 26–30

[*1] Dr. Eng.,
Senior Researcher Deputy General Manager,
Analysis & Characterization Research Dept.,
Steel Res. Lab.,
JFE Steel

[*2] Ph. D.,
Fellow,
Corporate Planning Dept.
JFE Techno-Research

[*3] Senior Researcher Deputy Manager,
Intellectual Property Dept.,
JFE Steel

angle neutron scattering (SANS), it is possible to obtain information on the size and shape of precipitates based on a comparison of the scattering spectra. Moreover, due to the high penetration ability of neutrons, it is also possible to obtain information on the average of a large volume irradiation region of the material. Precipitate morphologies are evaluated using TEM and SANS for understanding their behavior under different heat treatment.

2.2 Experimental Method

Ti-added low carbon steel was melted in vacuum in the laboratory and then rolled after solution heat treatment in the γ phase. The morphology of carbide precipitates was changed through the heat treatment condition of the cooling process after rolling in the range from 450˚C to 650˚C. Details of the sample treatment and properties are mentioned in ref. 3).

Neutron experiments were carried out using the SANS beam line of Japan Research Reactor-3 (JRR-3) of the Japan Atomic Energy Agency (JAEA, a National Research and Development Agency; Tokai-mura, Naka-gun, Ibaraki Prefecture). Samples with thickness of 2 mm grinded of the front and back surfaces were used in the neutron experiments. Measurements by SANS were performed with a neutron irradiation region of Φ 10 mm. Because ferrous materials possess magnetism, SANS measurements of steel materials are done under a magnetic field of 1 T for separating the nuclear and magnetic scattered component. The data analysis is performed using the nuclear scattering profile.

2.3 Experimental Results and Discussion

Figure 1 shows the small angle scattering profiles for different heat treatment conditions. It is found that the scattering spectra change in the range of values of the scattering vector, q, from 0.1 to 1 nm^{-1}, for indicating the precipitate morphology changing depending on the heat treatment condition. It is possible to evaluate the size and shape of precipitates from the profiles of these scattering spectra. Based on the TEM observation results a disk shape (major axis: Diameter, disk thickness: Thickness) precipitates are assumed for the small angle scattering profiles calculation of the particle size, density and aspect ratio. The results of the heat treatment dependency of particle size are shown in **Fig. 2**. From this figure, it can be understood that the diameter and thickness of the precipitates increase with increasing heat treatment temperature. The average size of the precipitates measured by TEM is also shown similarly in the figure. The increasing tendency of the average size measured by TEM shows good agreement with the results of the SANS measurement. The TEM measuring results at intermediate positions between the diameter and thickness by SANS, is showing good agreement with both evaluation techniques.

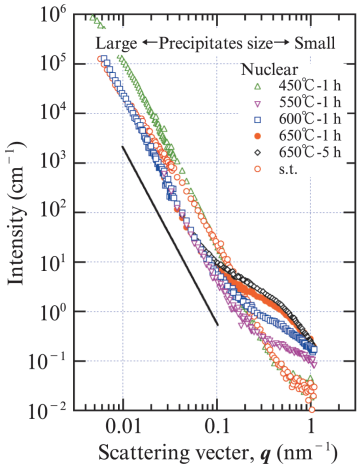

Fig. 1 Small angle scattering profiles[3)]

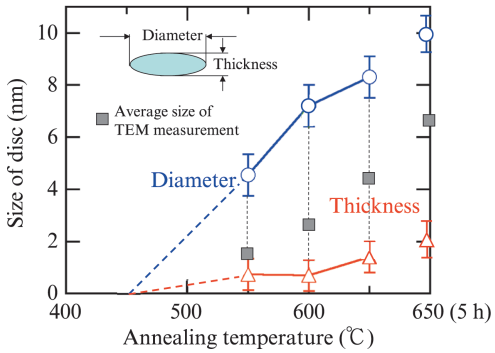

Fig. 2 Size changes of precipitates measured by small angle scattering (Values of transmission electron microscope (TEM) experiments are shown in together.)[3)]

The TEM results are obtained by local observation, whereas the neutron analysis is average information for the irradiation area (Φ 10 mm) x specimen thickness (2 mm). Thus, it was found that information on fine precipitates can be evaluated systematically by performing complementary analyses and much useful information can be obtained in this manner.

3. Measurement of Residual Stress of Welds Metal[5,6)]

3.1 Background

In higher strength steel products, sensitivity to delayed fracture due to hydrogen absorption also generally increases. At the welding parts, the risk of weld cracks due to residual stress by welding and absorption of hydrogen is increasing. For this reason, information on residual stress in welds metal is important for preventing delayed fracture and evaluating the safety of welds. Neutron measurement of residual stress is widely used due to the distinctive feature that detailed measure-

ment of a wide irradiation region to a depth of a few centimeters in the interior is possible because of the high penetration ability of neutrons[5–8]. The following sections 3.2 and 3.3 introduce measurement of the distribution of residual stress in a welded joint of high strength steel.

3.2 Experimental Method

Welded test pieces were prepared by using 980 MPa class steel plates with the thickness of 25 mm. Welds were prepared by 1-pass MIG (metal inert gas) welding in a Y-shaped slit in the center at the welding heat input of 16–17 kJ/cm using a commercial weld metal[5].

In the residual stress measurements, the REsidual Stress Analyzer, RESA[9], developed by the JAEA was used. Diffraction experiments were performed in the three directions of the weld, namely, the plate transverse direction (T), the fusion line direction (L) and the plate thickness direction (N) by using a neutron beam having the wavelength of 0.1590 nm. For the T direction and L direction, the diffraction line of (211) transmission were used, and for the N direction, the diffraction line of (211) reflection was used. The slit had dimensions of 2 mm in width and 15 mm in height for the T and N directions and dimensions of 2 mm in width and 4 mm in height for the L direction. A no-strain sample (D0) of the same sample was also prepared for comparison purposes by cutting out the weld part and releasing stress by making a slit with a wire cutter.

3.3 Experimental Results and Discussion

Figure 3 shows the diffraction intensity patterns for the T direction near the weld root and near the surface of weld metal together with results for the comparison no-strain sample. Although the peak of the curve for the weld surface is positioned slightly to the low angle side, the peak of the curve for the weld root has shifted significantly to the low angle side and the lattice spacing has expanded. **Figure 4** shows the strain distribution map for the T direction at various positions on the weld when residual strain (ε) was obtained by using the deviation of the lattice constant from the no-strain sample.

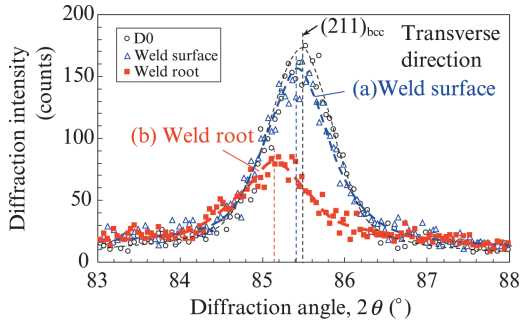

Fig. 3 Neutron diffraction patterns for root and surface of welding together with no strain sample (D0)[5,6]

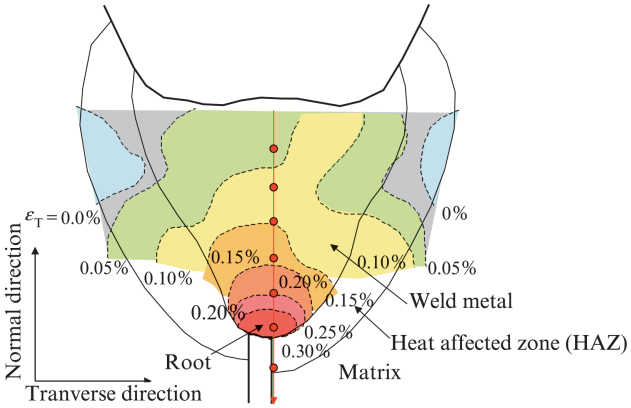

Fig. 4 Contour plot of strain in the tranverse-direction[5,6]

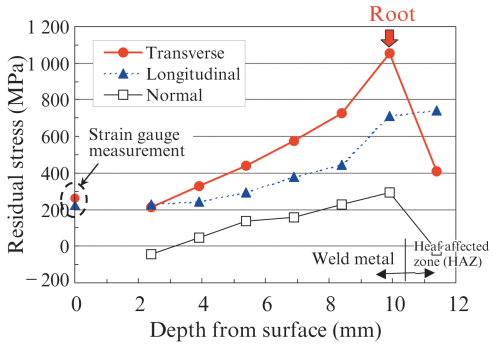

Fig. 5 Distribution of residual stress along weld center line[5,6]

Residual stress can be obtained from the lattice strain in the three perpendicular directions. The distribution of residual stress along the weld center line is shown in **Fig. 5**. Tensile residual stress increases from the vicinity of the surface to positions closer to the root. Residual stress is highest in the T direction, and high tensile stress of 1 000 MPa or more remains in the root part. Tensile stress exists at the root part in both the L direction and the N direction at this time, showing that a condition of high triaxial stress exists. The figure also shows the residual stress of the weld metal surface measured with a strain gauge. Since the value is substantially the same as the value of the surface residual stress value measured by neutron diffraction, it can be said that residual stress measurement by neutron diffraction evaluated residual stress with good accuracy.

Evaluation of the distribution of residual stress in thick materials to the interior of a weld was possible by utilizing the high penetration ability of neutrons, and effective knowledge on the characteristics of the distribution of residual stress during welding by position could be obtained. Moreover, it is also expected to be possible to clarify many points concerning control of weld cracking by investigating the relationship between cracking and hydrogen absorbed during welding[5].

4. *In-situ* Observation of Behavior of γ Phase Transformation during Deformation Process of TRIP Steel Sheet[6)]

4.1 Background

TRIP steel sheets with high strength and high ductility have been developed as automotive steel sheets. Because TRIP steel sheets achieve a combination of high strength and high elongation by utilizing the martensite transformation from retained austenite (γ) during the deformation process. For this reason, a knowledge of that deformation behavior is extremely important. Analyses of phase transformation behavior are also performed by scanning electron microscopy and X-ray diffraction, however, the effects of specimen surface are disturbing the precise observation with using these technique. Neutron measurement is effective for evaluation of the bulk as a whole[10)], because of their high penetration ability. Therefore, the γ to martensite transformation behavior during deformation was evaluated by using the time-of-flight neutron diffraction method.

4.2 Experimental Method

Transformation behavior during tensile test was measured using a TRIP steel with the sheet thickness of 1.5 mm. An *in-situ* neutron diffraction experiment during tensile deformation was carried out with the J-PARC beam line #19 of Engineering Materials Diffractometer "TAKUMI"[11)]. In the neutron diffraction experiment under tensile deformation, it was possible to obtain multiple diffraction peaks simultaneously by the time-of-flight (TOF) method using pulse neutrons having a wide wavelength band. **Figure 6** shows a diffraction intensity profile obtained by the TOF method. As can be understood from the figure, peaks from the α and γ phases are observed simultaneously in a wide region. For tensile *in-situ* observation, a flat sheet tensile test piece having a parallel portion of 50 mm, width of 5 mm and thickness of 1.2 mm was set for adjusting the direction of the tension axis inclined 45° relative to the incident neutron beam. Two detectors were set at directions of 90°, respectively, to the incident beam, and the diffraction intensities of the lattice plane parallel and perpendicular to the direction of the tension axis were measured.

4.3 Experimental Results and Discussion

The change of the γ (200) diffraction peaks during tensile deformation is shown in **Fig. 7**. With tensile deformation, a decrease in diffraction peak intensity, change in the position of the diffraction peaks and increase in the lattice plane spacing can be observed. Since the peak height after unloading was smaller in comparison with the initial height, it can be found that the dynamic behavior of retained γ in the deformation process has been detected. The change in lattice strain and the amount of retained γ in the transformation behavior of retained γ during tensile deformation were analyzed by a Rietveld analysis. Peak fitting of the peak of the α phase at that time was also performed, and the peak of the α phase was separated into the peaks of ferrite (bcc) and martensite (bct). **Figure 8** shows the changes in the lattice strains of the respective ferrite, martensite and retained γ phases during tensile deformation when calculated in this manner. From Fig. 8, in the stage when the ferrite phase yielded, the retained γ phase was increasing lattice strain to bear the strain; then, with yielding of the retained γ, the martensite phase was increasing in lattice strain and bore the strain. Moreover, from the measurement of retained γ, a decrease in the amount of retained γ was observed together with yielding, and a further decrease in retained γ was also observed accompanying the increase in work hardening due to plastic deformation. Summarizing the transformation behavior of retained γ, from the analysis described above, it was found that the stress on the retained γ increased with yielding of ferrite, and a stress induced transformation occurred, and strain induced transformation then proceeded accompanying the plastic deformation of the retained γ in the subsequent work-hardening stage.

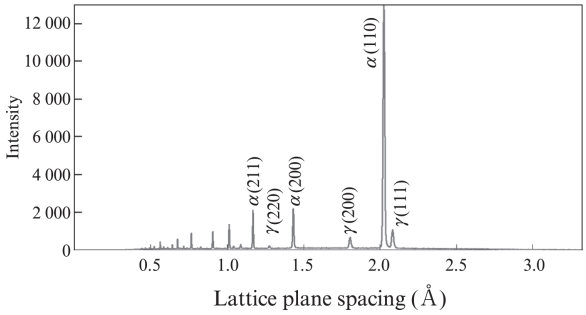

Fig. 6 Time-of-flight neutron diffraction profile of transformation induced plasticity (TRIP) steel[6)]

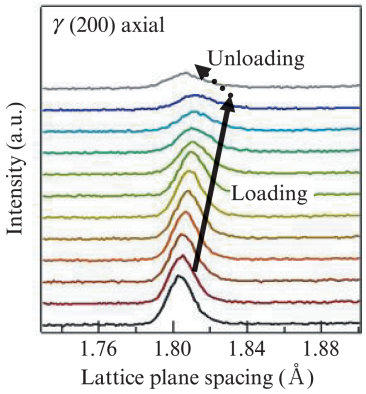

Fig. 7 Changing of Neutron diffraction peaks during tensile test[6)]

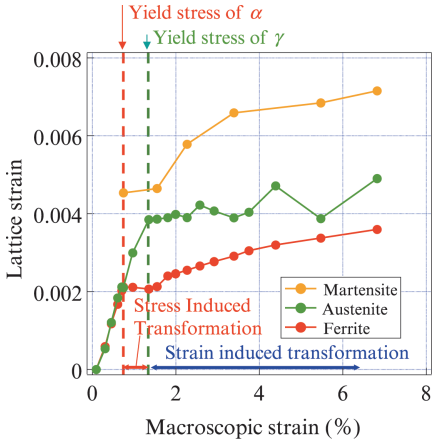

Fig. 8 Lattice strain changes of each phases during tensile test[6)]

5. Conclusion

Because of the high penetration ability of neutrons, neutron-based analytical techniques are extremely promising for structural analysis of steels. In addition to J-PARC and other large-scale facilities, analytical techniques utilizing compact neutron beam sources have also progressed in recent years. As great progress is also foreseen in the future, JFE Steel has adopted a policy of performing structural analyses actively utilizing neutron techniques through participation in research groups, etc.

This research was carried out through research groups such as the Industry-Originated Iron and Steel Research for Project Development and public invitation type (C-type) research groups of the Iron and Steel Institute of Japan, “Fundamental Studies on Technologies for Steel Materials with Enhanced Strength and Functions” of the New Energy and Industrial Technology Development Organization (NEDO), the Neutron Application Transfer Promoting Program of the Ministry of Education, Culture, Sports, Science & Technology (MEXT) and others. We received generous technical support from Prof. Yo Tomota of Ibaraki University (now affiliated with the National Institute for Materials Science (NIMS)), Prof. Masato Ohnuma of Hokkaido University, Dr. Yuji Suzuki, Dr. Koichi Akita and Dr. Stefanus Harjo of the Japan Atomic Energy Agency and Dr. Jun-ichi Suzuki of the Comprehensive Research Organization for Science and Society (CROSS) in the neutron experiments in this research. We also received the cooperation of many other persons, beginning with the members of the respective research groups, in the planning and execution of experiments applying neutron techniques. We wish to take this opportunity to express our sincere appreciation to all those concerned.

References

1) Tomota, Y. et. al. Proceeding of Basic Study for Application of Neutron Techniques on Steel Structural Analysis. Industry-Originated Iron and Steel Research for Project Development of ISIJ, 2009.
2) Ohnuma, M. et al. New era of neutron scattering and diffraction as a tool for the steel research, CAMP-ISIJ. 2010, vol. 23 p. 735–776.
3) Yasuhara, H.; Sato, K.; Toji, Y.; Ohnuma, M.; Suzuki, J.; Tomota, Y. Tetsu-to-Hagané. 2010, vol. 96, no. 9, p. 545.
4) ex. Sato, K.; Nakamcihi, H.; Yamada, K. Kenbikyo. 2005, vol.. 40, no. 3, p. 183.
5) Ishikawa, N.; Sueyoshi, H.; Suzuki, H.; Akita, K. Quarterly J. the Japan Welding Society, 2011, vol. 29, p. 218.
6) Sueyoshi, H.; Ishikawa, N.; Yamada, K.; Sato, K.; T. Nakagaito, T.; Matsuda, H.; Arakaki, Y.; Tomota, Y. Hamon (Bulletin of the Japanese Society for Neutron Science) 2014, vol. 23, p. 34.
7) Tanaka, K.; Akiniwa, Y.; Hayashi, M. Mater. Sci. Res. Int. 2002, vol. 8, no. 4, p. 165.
8) Kim, S-H.; Kim, J-B.; Lee, W-J. J Mat Process Tech. 2009, vol. 209, no. 8, p. 3905.
9) Suzuki, H.; Moriai, A.; Shimojo, Y.; P. G. Xu,; Shiota, Y.; Akita, K. Materials & Mechanics conference. 2010, p. 333.
10) ex. Tomota, Y. Ferrum. Bulletin of the ISIJ, 2010, vol. 15, no. 1, p. 17.
11) S. Harjo. J. of the Crystallographic Society of Japan. 2008, vol. 50, p. 40.