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# Progress on Three-dimensional Observation and *in situ* Observation Using Scanning Ion Microscopy

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## Abstract

***The progress on the focused ion beam fabrication method provides us several kinds of advanced techniques for the sample preparation of transmission electron microscopy. With improvement of the beam convergence and the increase of the ion accelerating voltage, the ability of its scanning ion microscope becomes glade up with a good image resolution. The scanning ion microscope is now powerful tool to investigate the three dimensional microstructure to cut and view in any places under the observation and recently the FIB serial sectioning method is developed to one of the method to reconstruct the 3D image in the computer using a series of sliced cross sectioning images, in the research filed of steel microstructure. As another application filed of the scanning ion microscopy, the high temperature in situ observation technique has been developed, and the melting behavior of metal particles have been observed.***

## 1. Introduction

Electrolytic polishing is basic technology used to prepare specimens for transmission electron microscopy (TEM). In view of the growing need for TEM observation of cross-section structures of steel, the focused ion beam (FIB) method was introduced as a new cross-sectioning method with ion-beam technology. Since then, micro-sampling technology has been developed, which permits cutting out cross-sections or plane structures of suitable parts of steel materials using a manipulator operation with pinpoint accuracy while observing the microstructure through its secondary electron images. In particular, in the case of magnetic materials, such as steel, it is necessary to make the specimen as small as possible in order to avoid adverse effects of the strong magnetic field within the TEM. The FIB and micro-sampling methods are the most effective techniques for preparing TEM specimens in steels.[1] Nippon Steel Corporation introduced micro-sampling technology[2] ahead of other steelmakers, and reported on it in a special issue on steel technology and nanotechnology in 2004[3] as regards the pinpoint microstructure analysis technique. The company is carefully and constantly monitoring the progress of FIB technology.[4]

In fact, FIB technology has evolved rapidly, in which the acceleration voltage has been increased to 40 kV from 30 kV to shorten the time required to fabricate TEM specimens, and the beam of ions has been made so narrow that the resolution of the scanning ion microscope (SIM) images and the accuracy of specimen preparation have improved dramatically. As a result, the scope of applications for scanning ion microscopy has expanded. Interlocked with the latest 3D imaging technology for microstructures, SIM observation is revealing new aspects about complex microstructure in various materials. The principle behind scanning ion microscopy is as follows. When a beam of Ga ions is irradiated onto a metallic material, sec-

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ondary electrons are generated at the surface of those areas in the same manner as the case of irradiation electrons. By detecting those secondary electrons and synchronizing them with the scanning ion beam, it is possible to obtain images of the microstructure of a material.

One characteristic of these SIM images is that when the specimen is ferrous, the ion beam can only penetrate approximately 30 nm into the specimen because the Ga ion is more than 10,000 times heavier than an electron. Although the depth of penetration of both the ion beam and the electron beam differs slightly according to the grain orientation, the influence of the relative difference in grain orientation manifests itself markedly with the ion beam. Thus, with a scanning ion microscope, an image of higher contrast can be obtained even when the specimen orientation changes by only 0.1 degree. Scanning ion microscopy is very useful for observing grain boundaries and the microstructures of bainite, martensite, etc. which consist of a fine microstructure with only slight orientation difference.

In this paper, we describe the development of a new three-dimensional observation technique and FIB serial sectioning technique for scanning ion microscopy, which is very useful in observing the microstructures of steel materials. Concerning the development of hot, *in-situ* observation technology that has been continually pursued, we introduce an example of observation of the melting of fine metallic particles, etc.

## 2. Three-Dimensional Structural Analysis Using Scanning Ion Microscopy

### 2.1 Cross section observation of bainite lath structure of low carbon steel

In order to develop high quality steel with excellent strength and ductility, it is important to design the optimum alloy composition and determine the ultimate microstructure required of steel performance, in which the lower bainite microstructure becomes utilized. Since the bainite microstructure has been strongly affected by the water cooling condition in a rolling process, for large steel products, it is important to control the structure across the thickness of the slab.

**Photo 1** shows SIM images of a bainite structure observed by cross section view. In the ordinary SEM observation, only the surface structure can be observed. With SIM, by contrast, cutting out cross-section structures by means of a Ga ion beam as shown in Photo 1 enables one to observe the bainite microstructure from various angles. Observation from two oblique directions as in Photo 1 (a) reveals that the lath structure resembles plate-shaped crystal domains. The regions of the same contrast are considered to have the same orientation. As the next step, the microstructure observed after several layers were cut out from the specimen end is shown in Photo 1 (b). It can be seen that black and white plate-shaped parts overlap each other in a complicated manner at the center. In this simple way, it is possible to estimate easily the three-dimensional microstructure of lath bainite, which comprises the principal structure of low carbon bainite steel.

The conventional scheme of the bainite structure is as shown in **Fig. 1** (a). However, we consider that the actual bainite structure is not like this. Certainly, the prior $\gamma$ grain boundary, which is the nucleation site, remains the same, but, at the part where the growth fronts of bainite laths encounter each other, the laths interlace as shown in Photo 1. Therefore, it seems that the packet interface, which is defined as a boundary between groups of different orientation blocks,—is not always a uniform boundary structure, but that the scheme thereof is as shown in Fig. 1 (b).

During the bainite transformation of low carbon steel, lath microstructures can be observed even under a TEM. Since a single block is formed while those lath microstructures are formed continuously, the above idea suggests that the growth front of each block can hardly form a simple uniform interface. If a certain block interface becomes the origin of growth of a new bainite lath, it can become a uniform interface the same as a grain boundary. However, this does not occur very frequently. Together with the results of *in-situ* observations of bainite transformation, in the future, the microstructure of a packet boundary will be discussed in more detail, concerning the fracture origin in steel.

Since the bainite transformation accompanies the diffusion of carbon, the transformation stops halfway, when the concentration of carbon in the transformation front region increases and the austenite stabilizes. As a result, the austenite phase may remain in the neighborhood of the packet interface where laths interlace in a complicated manner. In order to confirm this, it is necessary to analyze the phenomenon using the electron back-scanning diffraction (EBSD) method or some other suitable method, since scanning ion microscopy cannot be used to analyze crystal structures. In the future, it will become necessary to make a quantitative evaluation of three-dimensional forms in such complicated packet microstructure regions and discuss the strength and fracture characteristics in the neighborhood of the interfaces.

### 2.2 Three-dimensional analysis using FIB serial sectioning method

By making a stereoscopic observation of various parts of any structure using the FIB method and microscopy, the observer can

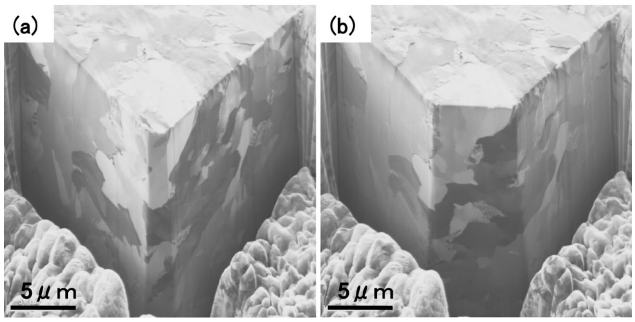


**Photo 1 Cross section views of bainite lasth microstructure using FIB, (b) is further fabricated at the front area from (a).**

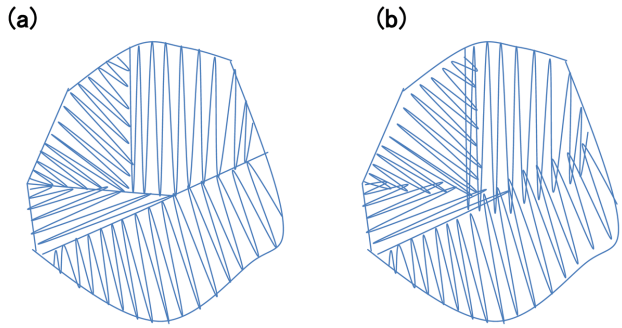


**Fig. 1 Schematic diagram of bainite lath microstructure; (a) is conventional aspect and (b) is a proposed microstructure aspect.**

grasp a three-dimensional image of the structure fairly well. However, from the standpoint of allowing the observer to share the knowledge obtained with other persons or quantify the structure, it is of course necessary for the observer to obtain accurate three-dimensional images of the structure.

Technology that enables an accurate, three-dimensional analysis of structures, from nanometer to macro levels, has been progressing rapidly in recent years. These include, in ascending order of object size, electron tomography, FIB serial sectioning that combines FIB, SEM/SIM images and EBSD orientation image mapping, and polishing serial sectioning that repeats mechanical polishing and optical microscopy using a robot that carries the specimen automatically.

In this section, we present an example of FIB serial sectioning that utilizes scanning ion microscopy. As mentioned earlier, in a heat affected zone of low carbon steel in welding, sometimes the bainite transformation stops halfway and the austenite is retained while the steel continues to be cooled. In such cases, the austenite undergoes martensite transformation to form a region harder than the surrounding bainite phase and often causes the low-temperature toughness of the steel to deteriorate. In order to determine whether it will become an origin of steel fracture, it is necessary to grasp the three-dimensional form of the hard region such as the small martensite phase around the bainite microstructure in steel.

**Photo 2** is a series of SIM images indicating the procedure of FIB serial sectioning method containing a hard region formed as a result of martensite transformation. In many cases, the retained austenite shows 0.4% or more carbon enrichment and the martensite after the transformation contains twins as internal defects. In the SIM image, therefore, the martensite phase can be identified as the twined region where the black and white contrast changes strikingly. First, as shown in Photo 2 (a), with the structure containing such a region left out, decide a target region while observing the surface layer structure and cross-section structure where appropriate. Then, protect the topmost layer of the target region by chemical deposition of a layer of amorphous tungsten. This can be performed inside the FIB fabrication system. Next, as shown in Photo 2 (b), enclose the tungsten deposition layer in the form of a thin layer by the FIB fabrication. By previously enclosing the part whose 3D images are needed, it is possible to prevent the field of vision from shifting during subsequent positioning or FIB slicing work.

Now, as shown in Photo 2 (c), incline the specimen to 45° and obtain an SIM image of its cross section. Then, return the specimen surface to right angles to the ion beam and subject it to FIB slicing. After that, incline the specimen to 45° again and obtain an SIM image of its cross section. Repeat the work continuously and obtain about one hundred SIM images of the slices. Ordinarily, the slicing interval is approximately 50 nm to 100 nm, although it can be shortened to a minimum of 30 nm. Photo 2 (d) shows an example of trimming of a cross-section image to obtain a three-dimensional image. By making the X-Y plane 10 $\mu$m × 10 $\mu$m and taking 100 cross-section images at a slicing interval of 100 nm, it is possible to construct a three-dimensional image measuring 10 $\mu$m × 10 $\mu$m × 10 $\mu$m.

### 2.3 Reconstruction of three-dimensional image for martensite-austenite (M-A) hard layer

Using the method described in the preceding section, obtain about 100 slices SIM images of cross sections of the specimen containing the M-A hard layer. Next, using a suitable analytical computer program, position each of those slice images. First, decide several positions on a slice image as shown by the dotted-line circles in **Fig. 2**. Then, at each of those positions, obtain the product sum of pixel intensities for each of the two adjoining slice images and determine the minimum value of the relative position between those two slice images from the amount of variation of the product sum. In this way, any deviation in the relative position between two adjoining slice images is minimized automatically. Actually, however, the relative position of the slice images often changes in a complicated manner. In order to improve the precision of positioning, therefore, it is necessary to develop a new and advanced technique.

Incidentally, by previously making the pixel size in the two-dimensional plane the same as the pixel size in the direction of the slice thickness while creating a three-dimensional image, it is possible to obtain isotropic cubic voxel during 3D imaging. This method is important since it dramatically improves the accuracy of subsequent analysis of 3D images. As illustrated in Fig. 2, when the magnification is such that there are 373 pixels within a 10.5 $\mu$m-wide plane and the interval between slices is 0.03 $\mu$m, it is possible to obtain cubic voxel about 30 nm/pixel in size in the three x, y and z axes.

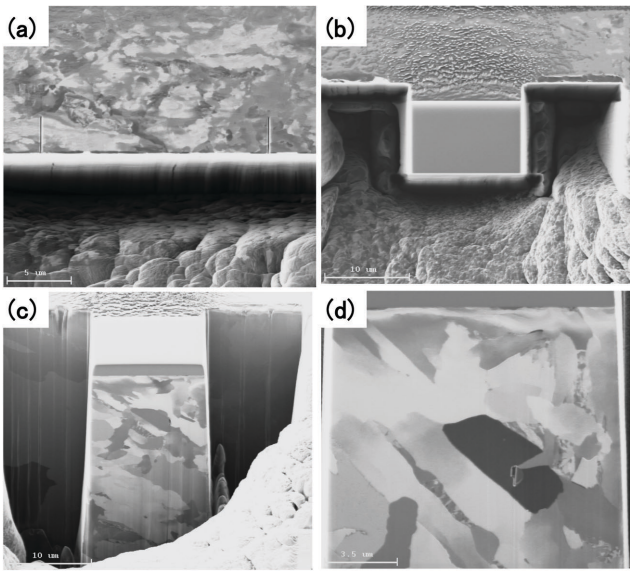


**Photo 2 Procedure of FIB serial sectioning method; (a) Observation, (b) Pre-fabrication around specific area, (c) Slice and view of cross section image, (d) Trimming of cross section image**

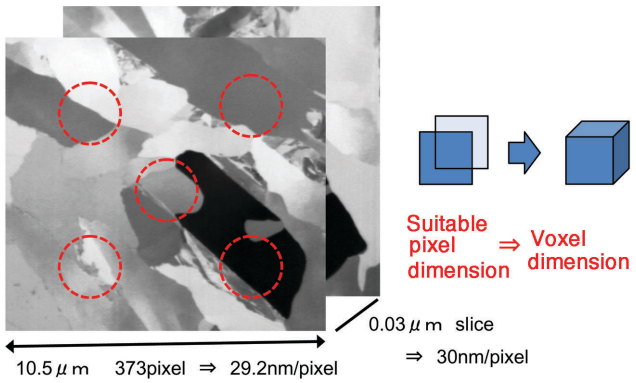


**Fig. 2 Alignment method of each slice imaging position and an example of data size dimension**

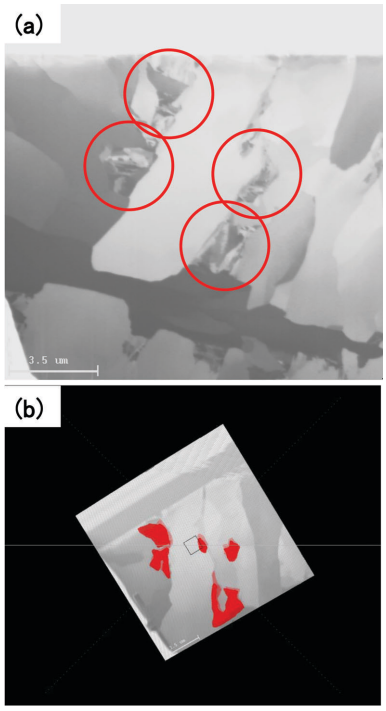


**Photo 3 Scanning ion micrograph showing M-A distribution between lath boundary (a) and its reconstructed 3D image (b) M-A regions are painted by red areas in 3D image.**

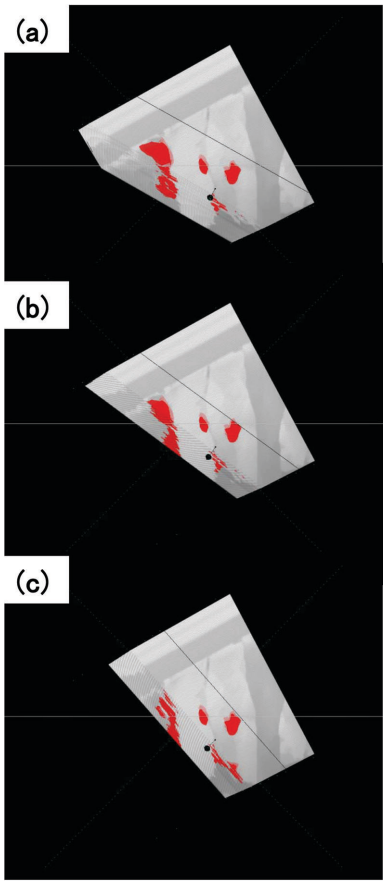


**Photo 4 A series of different cross section images of 3D imaging showing distribution of M-A by red areas**

The most difficult thing in reconstructing a 3D image is segmentation; that is, the task of extracting the target element. In the case of an SIM image of gray contrast, in particular, segmentation is often extremely difficult because regions having the same gradation are not always meaningful ones. Therefore, as the most rudimentary method of segmentation, we manually colored the noted regions in each of the slice images. In **Photo 3** (a), the parts enclosed in red circles are the noted hard M-A phases. These are regions that show a sharp change in black and white contrast, which is characteristic of a martensite structure. Although they can easily be recognized by the naked eye, the computer can hardly recognize them. Photo 3 (b) shows a 3D image obtained by coloring those M-A regions in each slice image in red and overlaying 100 slice images one upon another. Since the coloring of M-A regions was performed by hand, their sizes are not very accurate. Even so, the continuity of the structure to better discern the form of the M-A regions is well maintained.

Next, in order to study the form of the M-A phase (red-colored segment) in the specimen, we cut the reconstructed 3D image at arbitrary sections as shown in **Photo 4**. Characteristic parts are shown in Photos 4 (a) to (c). Actually, it is possible to observe various cross-section shapes by performing the cutting work continuously by computer. The above work revealed that the M-A structure in the specimen was connected continuously. In other words, it was found that the distribution of the hard phase observed in a two-dimensional image of a cross section was significantly different from that in an actual three-dimensional crystal. We consider that in future it will become possible to make clear how to associate the hard phase with the macro deformation of a specimen on the basis of the above results accompanied with the means of simulations.

## 3. High-temperature *in situ* Observation

In addition to the three-dimensional imaging method using scanning ion microscopy (SIM), a high-temperature *in situ* observation technique that utilizes the excellent channeling characteristic contrast of SIM has been developed. Early on, we began applying the technique during *in situ* observations of intra-granular transformations.[5] One of the major problems in high-temperature observations was the influence of the light emitted from the steel specimen itself on the secondary electron detector. Since this problem occurs when the specimen is large, we made a window cover of tungsten foil and put it over the specimen to restrain the emission of light from regions other than the target area as shown in **Photo 5**. As a result, even when the emission of light from the specimen would be a problem at 1,000℃ or more, the resulting noise could be reduced to such a level that it does not impede the *in situ* observation of the microstructure change.

By making *in situ* observations of a phase transformation or any

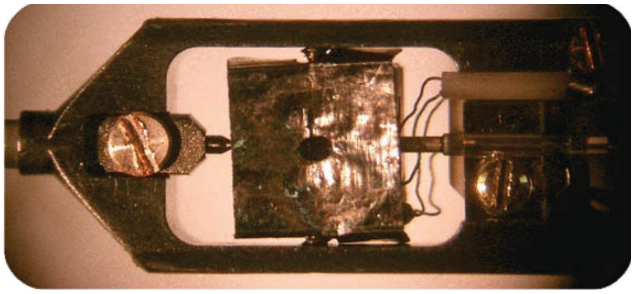

**Photo 5 Optical photograph showing top of heating stage using FIB system**

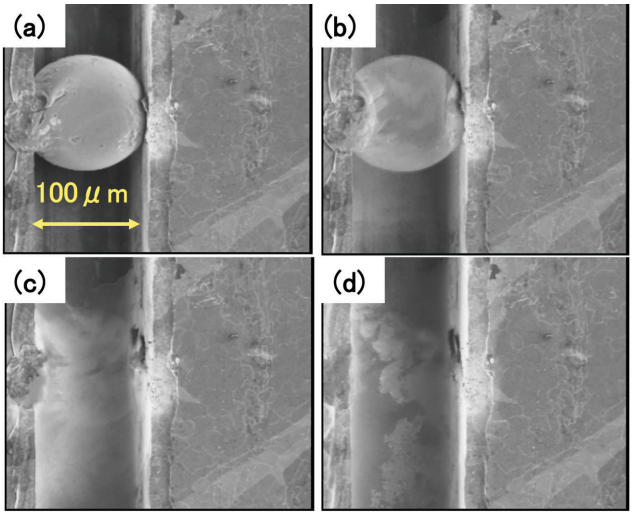


**Photo 6** ***In situ*** **observation at high temperature showing melting behavior of Cu particle under scanning ion microsope observation**

other metallurgical phenomenon that takes place at high temperature, it is possible to observe phenomena directly at that high temperature that could formerly only be conjectured after the specimen had cooled down to room temperature. The melting of metal is one of those phenomena. We prepared a particle of copper 100 $\mu$m in diameter and put it on a stainless steel base provided with a 100 $\mu$m-wide groove, as shown in **Photo 6** (a). When the specimen was directly heated by the heating stage inside the FIB equipment, the speckled pattern of the microstructure instantaneously changed as shown in Photo 6 (b) at a temperature near the melting point of copper. Then, the pattern disappeared in an instant as shown in Photo 6 (c). Since the experiment was carried out in a vacuum, we consider that the specimen sublimed inside the chamber as soon as it was melted. Photo 6 (d) shows signs of a very small amount of copper being stuck onto the groove in the stainless steel base.

The above experiment on the melting of copper has good reproducibility. Each time a particle of copper is placed properly in the groove and the right heat conduction is secured, the melting point of the copper particle is reached with nearly the same heating current of the heater. Although the melting point of copper particles in air is 1,084℃, it is uncertain how much the melting point dropped in the experiment.

## 4. Conclusion

The focused ion beam (FIB) method is an effective method of preparing specimens for transmission electron microscopy (TEM). In this paper, we show that the function of FIB in scanning ion microscopy, which utilizes secondary electrons from Ga ion irradiation, puts a new value as a very effective observation technique on the FIB system by the recent improvement in resolution and workability. In particular, the FIB serial sectioning method enables a three-dimensional image of a microstructure to be obtained by taking slice images of the target region in micron order and reconstructing those images using a computer.

Analyzing the spatial morphology of the hard M-A phase, which often becomes the origin of a steel fracture, has long been a task to make clear. In this respect, we show using the latest FIB serial sectioning technique that the three-dimensional distribution of the hard phase such as M-A has continuous distribution profiles in bulk specimens, although the M-A regions disperse among bainite lath structure by SEM observation. This finding is contrary to that obtained by conventional two-dimensional observation, and analyses of microstructures considered likely to be origins of steel fracture. Although there is still room for improvement, such as in segmentation technology, the FIB serial sectioning method is a new technology that allows for 3D imaging of regions about 100 $\mu$m in size. It is expected that combined with the EBSD method, etc., there will be dramatic progress in the technology involved in FIB in future.

Turning our attention to the high contrast of SIM images, the high-temperature, *in situ* observation technique is useful. In this paper, we demonstrate with an example that it is possible to observe the melting behavior of copper particles (up to 100 nm in size) at a temperature exceeding 1,000℃ by providing the specimen with a cover to prevent the secondary electron detector from being blinded by the light emitted from the specimen at high temperatures.

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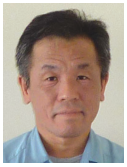

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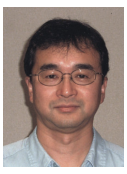

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