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Investigation using 4D-CT of massive-like transformation from the δ to γ phase during and after δ-solidification in carbon steels

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Abstract. For a long time, the γ phase in metallic alloys has seemed to be produced through a peritectic reaction between the δ and liquid phases. However, direct observations have shown that a massive-like transformation, in which the δ phase transforms into the γ phase in the solid state, is dominant during or after solidification of the δ phase in carbon steels. To characterize such massive-like transformation, we use time-resolved tomography (4D-CT) to demonstrate the volume change during cooling from the melt and the crystallographic orientation relationship between the δ and γ phases. The volume changes from solidification and from the massive-like transformation from the δ to the γ phase were −3% and −0.5%, respectively. The transformation from the δ to the γ phase finished quickly, as demonstrated by the volume change. Fine γ grains were produced even in a single δ grain through the massive-like transformation. Also, the refined γ grains showed a wide crystallographic distribution.

1. Introduction

Recently, time-resolved in-situ tomography (4D-CT) has been used to observe the solidification of metallic alloys [1,2,3,4,5,6]. One key issue in 4D-CT is achieving sufficiently high time resolution in order to observe microstructure evolution. The solidification of Al–Cu alloys has been observed threedimensionally by 4D-CT [1-5]. Solid grains isolated by a liquid phase, change in the solid fraction during cooling, shrinkage due to solidification, and the solid/liquid interfacial area (f_{ss}>0.7) have been observed or measured in Al–4mass%Cu alloys [1]. Segregation of Y₂O₃ particles during solidification of Al–Mg–Si alloys (Al-6082) with 0.54 vol.% of added Y₂O₃ particles (~500 nm) has also been demonstrated [2]. Dendrite growth calculated by a phase field model has been compared with the dendritic structure in Al–15mass% Cu alloys, obtained by 4D-CT [3]. The dendrite morphology, even early in solidification, was reconstructed by a 4D-CT technique using the interlaced view sampling (TIMBIR) method [4], which improved the time resolution. In addition to solidification, semisolid deformation in Al–15mass%Cu alloys has been observed by 4D-CT [5]. This study found that grains isolated by the remaining liquid phase experienced transgranular liquidation cracking even at low stress (1–40 MPa) [5]. This finding gives new insight into controlling grain structure during solidification. Solidification of Ni–14mass%Hf, Fe–11mass%Hf, and Co–18mass%Hf alloys have also been observed [7]. Adding the Hf
increased the contrast resolution between the solid and liquid phases, enabling solidification to be observed at high temperatures. Thus, 4D-CT techniques are expected to be useful in studying various metallic alloys.

An earlier study attempted to use 4D-CT to observe solidification in metallic alloys with low contrast resolution, such as Fe–C–Mn–Si alloys [8]. Although these reconstructed images had rather poor quality due to the low contrast resolution and low number of projection images, a filter method using a phase field model significantly improved the image quality. The development of the primary and secondary arms was observed, and the average curvature of the solid/liquid interface was obtained as a function of solid fraction. Characterizing dendritic growth, as done in Ref. [8], will be useful for developing physical models and numerical simulations.

Using 4D-CT with monochromatized X-rays has another advantage: X-ray diffraction spots can be measured in synchronization with rotation by using a 2D detector, and sequential analysis of the diffraction data can be used to obtain the distribution of crystallographic orientations over time. For example, it can find the relationship of crystallographic orientation between a mother phase and a product phase in a phase transformation. Additionally, the volume fractions of constituent phases can be evaluated by integrating the diffraction intensities, and the volume change from thermal expansion and phase transformation can be found simply by counting voxels. Thus, this technique is attractive for studying phase transformation in situ.

For a long time, the γ phase has appeared to be produced through a peritectic reaction between the δ and liquid phases in carbon steel with a C content lower than 0.5 mass%. However, X-ray transmission imaging [9,10,11,12] has shown that a dominant massive-like transformation, in which the δ phase transforms into the γ phase in the solid state. As shown in Fig. 1, there is little nucleation of the γ phase, and the δ phase was undercooled below the peritectic reaction. Thus, the δ phase transformed into the γ phase in the solid state. Laser-scanning confocal microscopy has also shown that the δ phase massively transforms into the γ phase [13]. Moreover, the massive transformation was selected in Fe–Cr–Ni alloys [14]. Even so, the massive-like transformations in Fe–C and Fe–Cr–Ni alloys are not yet fully understood. Thus, to further understand massive-like transformations, we should measure the volume change during cooling and analyze the crystallographic orientation relationship between the δ and γ phases by using 4D-CT.

2. Experiments
We studied a specimen of Fe–0.18C–0.6Mn–0.3Si (mass%) carbon steel, hereafter referred to as 0.18C steel. The specimen was 0.8 mm in diameter and 2–3 mm in height. Its height was sufficiently small to observe the entire specimen when evaluating specimen volume. The specimen was cooled from a melt at a cooling rate of 0.33 K/s in a vacuum (approximately < 1 Pa). The temperature gradient at the specimen was less than 1 K/mm.

Figure 1. Example of the massive-like transformation observed in 0.45mass%C steel at a cooling rate of 0.83 K/min [9,10].
To measure the volume change and observe diffraction spots during cooling from the melt, we performed 4D-CT experiments at beamline BL20XU of the SPring-8 synchrotron radiation facility (Hyogo, Japan), as shown in Fig. 2. The size of the X-ray beam was adjusted using the slits, and the intensity was adjusted using the absorber, if needed. The specimen, inserted into a sintered $\text{Al}_2\text{O}_3$ pipe with inner and outer diameters of 0.8 mm and 2 mm, respectively, was placed in a graphite furnace. Transmission images were obtained by a beam monitor with a format of $1024 \times 1024$ pixels (pixel size: 6.5 μm × 6.5 μm). X-ray diffraction spots were observed with a flat-panel sensor with a format of $1008 \times 682$ pixels (pixel size: 100 μm × 100 μm, frame rate: 30 fps). Transmission images at a frame rate of 100 fps and diffraction images at a frame rate of 30 fps were continuously obtained by using monochromatized X-rays with an energy of 37.7 keV. The sample was rotated at 0.25 rps. Convolution back-projection (CBP) image reconstruction was performed using 200 transmission images over a 180° rotation every 4 s. Normal vectors of the $(110)_\delta$, $(200)_\delta$, $(211)_\delta$, and $(111)_\gamma$ planes in the specimen were calculated from the positions of X-ray diffraction spots on the flat-panel sensor and the rotation angle. The resolution of crystallographic orientation was 3° because 120 frames over 360° were obtained by the flat panel sensor. Because the flat panel had a small solid angle, the beam size was relatively large (4 mm horizontal and 3 mm vertical), and the diffraction spots changed rapidly after the massive-like transformation, we could not obtain a crystallographic orientation map of the specimen. The specimen volume was evaluated by counting specimen voxels (solid and liquid phases). Porosity detected in the specimen was excluded for this evaluation.

![Figure 2. Setup of 4D-CT for observing transmission images and X-ray diffraction spots.](image)

![Figure 3. Reconstructed 3D image after solidification and the massive-like transformation. Concave shape at the top was caused by solidification shrinkage.](image)
3. Results and Discussion

3.1. Volume change

Figure 3 shows a reconstructed 3D image of the 0.18C steel (solid and liquid phases) after the massive-like transformation following the ferrite solidification. The concave shape at the top was caused by solidification shrinkage. It was rather easy to track the boundary between specimen and vacuum because of the large difference in X-ray absorption coefficient. In addition, the reconstructed 3D images showed porosities with a diameter of more than 20 μm. The specimen volume was simply evaluated by counting specimen voxels (solid and liquid phases). The accuracy of the volume measurement depended on a threshold value to extract specimen voxels from the 3D images. However, the volume normalized by the volume of the δ phase immediately before the massive-like transformation has a relatively high sensitivity to volume change.

Figure 4 shows the change in volume during cooling from the melt at 0.33 K/s for the 0.18C steel. The volume of the liquid phase gradually decreased due to thermal shrinkage. The volume expansion coefficient estimated from the slope was $125 \times 10^{-6}$ K$^{-1}$. Thus, the linear expansion coefficient, $\alpha$, was estimated to be $42 \times 10^{-6}$ K$^{-1}$. Jimbo and Cramb reviewed the reported values of liquid density ($\rho$) of Fe [15], giving the temperature dependence of density as shown in Eq. (1).

$$\rho = \rho_m + \Lambda (T - T_m)$$  \hspace{1cm} (1)

Here, $\rho_m$ is the density at the melting point, $\Lambda$ is the coefficient, $T$ is the temperature, and $T_m$ is the melting point. The reported density at the melting point ($\rho_m$) ranges from $6.97 \times 10^3$ kg/m$^3$ to $7.28 \times 10^3$ kg/m$^3$. In addition, the reported coefficient, $\Lambda$, ranges from $-0.8 \times 10^{-3}$ kg/m$^3$K to $-1.6 \times 10^{-3}$ kg/m$^3$K. The linear expansion coefficient calculated from the reported values ranges from $38 \times 10^{-6}$ K$^{-1}$ to $74 \times 10^{-6}$ K$^{-1}$. The measured value in the present study lies within the range of reported values. Thus, the present method has at least the same accuracy as the methods used in the previous works. Because the reported values were very scattered, further work was required to evaluate the accuracy of 4D-CT.

![Figure 4](image_url)  \hspace{1cm} Figure 4 Volume change of 0.18mass%C steel during cooling from the melt. The volume was normalized by the volume immediately before the massive-like transformation.

Because solidification occurred in the undercooled melt, solidification completed within 20 s even at the low cooling rate. The volume change due to solidification was $\sim$3%. After solidification, the volume of the δ phase decreased, and the linear expansion coefficient was estimated to be $39 \times 10^{-6}$ K$^{-1}$. The massive-like transformation occurred at 780 s. The volume shrinkage due to the massive-like
transformation was 0.5%. This shrinkage completed within the time resolution of 4D-CT (4 s). This fast volume change is consistent with reported in-situ observations with X-ray transmission imaging [9-12]. The observed shrinkage of 0.5% will help in understanding the strain and stress induced by the massive-like transformation. In addition, the linear expansion coefficient of the γ phase after the massive-like transformation was estimated to be $48 \times 10^{-6}$ K$^{-1}$.

3.2. Crystallographic orientation

Figure 5 shows an example of X-ray diffraction pattern of the γ phase after the massive-like transfixion, obtained by the flat panel sensor. The intensity of X-ray diffraction was integrated over 360-degree rotation.

![Figure 5](image)

Figure 5 X-diffraction of γ phase after the massive-like transfixion, obtained by the flat panel sensor. The intensity of X-ray diffraction was integrated over 360-degree rotation.

![Figure 6](image)

Figure 6 Distribution of crystallographic orientation of (a) δ phase at 2s before the massive-like transformation, (b) γ phase at 2 s after the massive-like transformation and (c) γ phase at 325 s after the massive-like transformation.

3.2. Crystallographic orientation

Figure 5 shows an example of X-ray diffraction pattern of the γ phase after the massive-like transformation. In this figure, 120 diffraction images over a 360° rotation were superimposed to show the diffraction rings. In the analysis, the positions of diffraction spots were measured on every diffraction image. The normal vectors of diffraction planes were geometrically calculated from the positions and the rotation angle of the specimen. Thus, we obtained a distribution of crystallographic orientations with an angle resolution of 3°, as shown Fig. 6. Each point on the stereographic projection corresponds to a normal vector of the diffraction plane. Normal directions outside the green circle in Fig. 6(a) were not measured because of the flat panel sensor had a limited solid angle. As shown in Fig. 6(a), the crystallographic orientation of the δ phase before the massive-like transformation showed that dendrites of the δ phase with a single orientation grew in the entire specimen. Namely, the massive-like transformation occurred in a single crystal of the δ phase.

Many diffraction spots of the γ phase appeared after the massive-like transformation (Fig. 5), indicating that it produced multiple γ grains. Figure 6(b) shows the distribution of the closest packed
plane (111) at 2 s after the massive-like transformation. The color denotes the intensity of the diffraction spots. Ref. [9], using a white X-ray beam with a size of 50 μm × 50 μm, suggested that the closest packed plane (110) of the δ phase and the closest packed plane (111) of the γ phase tended to be adjoined. However, many (111) spots of γ grains appeared in the present study, so it was rather difficult to identify the crystallographic orientation relationship between the δ and γ phases in the specimen with a diameter of 0.8 mm and a height of 2–3 mm. In the present study, we found that the massive-like transformation produced fine γ grains with a wide crystallographic distribution, even though there was a certain crystallographic orientation relationship. Producing refined γ grains will be important to understanding microstructure evolution during and after solidification in carbon steel.

4. Summary
Using 4D-CT, we measured the change in volume during cooling from the melt. The distribution of crystallographic orientation was also measured for the δ and γ phases.

- The volume changes due to solidification and the massive-like transformation from the δ to the γ phase were 3% and 0.5%, respectively. This quantitative measurement will be useful for understanding the strain and stress induced by the massive-like transformation.
- The linear expansion coefficient of liquid was estimated to be $40 \times 10^{-6}$ K$^{-1}$. The linear expansion coefficients of the δ and γ phases were estimated to be $39 \times 10^{-6}$ K$^{-1}$ and $48 \times 10^{-6}$ K$^{-1}$, respectively.
- 4D-CT will be a useful tool for measuring volume changes from solidification phenomena.
- Fine γ grains were produced even in a single δ grain through the massive-like transformation.
- The refined γ grains showed a wide distribution of crystallographic orientations.
- The massive-like transformation was characterized by a volume change of 0.5% and the production of fine γ grains.

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