Ferrite Grain Size Formed by Large Strain-High Z Deformation in a 0.15C Steel

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During large strain deformation of materials, the high angle grain boundary spacing tends to approach the order of mean thermal diffusion distances for given deformation conditions. Based on the results of microstructural and grain size analysis in low carbon steel subjected to large strain-high Z deformation, the evolved ferrite grain size was found to be controlled by the Zener-Hollomon parameter and grain boundary diffusion was found to be the controlling mechanism.

(Received August 3, 2005; Accepted September 22, 2005; Published November 15, 2005)

Keywords: large strain deformation, low carbon steel, ultrafine ferrite, Zener-Hollomon parameter, grain boundary diffusion

1. Introduction

Among the various strengthening mechanisms available, grain refinement is the only method to improve both strength and toughness simultaneously. Therefore, ultrafine grained steels with relatively simple compositions, strengthened primarily by grain refinement, show a great potential for replacing many of the existing multi-component high strength steels and have been pursued vigorously.^{1–10} Two approaches were mainly considered to obtain ultrafine ferrite grains. The first one employs heavy deformation of austenite at high temperature followed by accelerated cooling to increase the number of nucleation sites for ferrite resulting in ultrafine grained structure^{1–4} whereas the second approach uses large strain warm/cold deformation of ferrite to obtain ultrafine grain structure by recrystallization.^{5–10} Out of the above two approaches, large strain deformation in the warm working region (below A_{r3} temperature) results in finer ferrite grain size due to the lower temperatures involved. Here, we propose large strain deformation as the strain at which the high angle grain boundary (HAGB) spacing approaches thermal diffusion distance at the specified deformation temperature and time.

Some authors of this paper have previously studied^{6,7} the large strain-high Z (Zener-Hollomon parameter) deformation leading to the formation of ultrafine ferrite grains and reported that (a) there exists a critical strain for the formation of ultrafine ferrite grains, (b) their volume fraction increases with an increase in strain and (c) the size of the ultrafine ferrite grains is determined by the Z parameter. Here the Z parameter is defined as:

$$Z = \dot{\epsilon} \exp(Q_a/RT) \quad (1)$$

where $\dot{\epsilon}$ is the strain rate (s^{-1}), Q_a is the apparent activation energy for deformation ($J \cdot mol^{-1}$), R is the universal gas constant ($8.314 J \cdot mol^{-1} \cdot K$) and T is the deformation temperature (K).

An investigation^{6,7} in to the large strain-high Z deformation behavior of low carbon steel resulted in the relationship between the ferrite grain size, d (μm) and Z parameter represented by the equation:

$$d = 10^2 Z^{-0.16} \quad (2)$$

Further, the dependence of critical strain on the combined

effect of $\dot{\epsilon}$ and T on the formation of ultrafine ferrite grains has been reported.⁸

These systematic studies have in fact led to the production of ultrafine grained steel bar (18 mm square) and plate (18 mm thick) consisting of sub micrometer sized ferrite grains by large strain-high Z deformation in ferrite region.^{11,12} In spite of many studies reported on the generation of ultrafine ferrite grains, the mechanism controlling the ferrite grain size was not properly addressed. Understanding the controlling mechanism of ferrite grains size will further help in the microstructural control and optimization of processing parameters such as strain, strain rate and temperature in the production process.

During large strain warm deformation, the initial grains are compressed to pancake shape and the HAGB spacing tends to approach the order of mean thermal diffusion distance. Since the evolution of ultrafine ferrite grains takes place by thermally activated processes, the role played by the interfaces such as grain boundaries in controlling the grain size is significant. In order to clarify the controlling mechanism of ferrite grain size formed during large strain deformation, a comparison of the characteristic diffusion distance with ferrite grain size formed may lead to an insight into the role of diffusion. For this purpose, both volume diffusion and grain boundary diffusion as the controlling mechanisms of ferrite grain size are required to be considered.

Therefore, the aim of this investigation was to study the role of diffusion and the controlling mechanism of evolved ferrite grain size during large strain-high Z deformation in a low carbon steel. The experimental plan consisted of (a) isothermal constant true strain rate, large strain compression testing in the high Z region (10^{12} – $10^{16} s^{-1}$) to study the microstructural evolution and (b) detailed ferrite grain size analysis using electron backscattered diffraction (EBSD).

2. Experimental Methods

A large strain, single pass, simple compression technique designed and developed in-house is utilized for conducting the hot deformation studies using a thermo-mechanical simulator. In this anvil compression test, strain gets concentrated in the center of the specimen and the strain varies in the range of 0–4 from the surface to the center of the specimen.

This will facilitate the observation of regions deformed to various strain levels in a single specimen.⁸⁾

A commercial JIS SM490 grade hot rolled steel plate with a chemical composition of (composition in mass% C-0.16, Si-0.41, Mn-1.43, P-0.014, S-0.004, Fe-Balance) was used in the present study. Test specimens having dimensions of 15 mm length $\times$ 13 mm width $\times$ 12 mm thickness were cut from the steel plate. The specimens were machined such that the compression axis was perpendicular to the rolling direction. The initial microstructure consisted of a lamellar structure in which ferrite and pearlite bands are formed alternatively at substantially equal intervals as shown in Fig. 1(a). The initial nominal size (d_0) of ferrite grains was 15 μm (ASTM grain size). The average intervals of pearlite bands was 27 μm . Using the thermo-mechanical treatment simulator capable of controlling the specimen temperature, strain and strain rate under high Z deformation conditions, a specimen was heated to a temperature in the range of 773 to 923 K which is lower than the A_{e1} point (about 993 K). Specimens were heated at 5 Ks⁻¹ to their specified temperature by direct resistance and compressed in a single stroke after soaking for 60 s. The compressive deformation was carried out in the time periods of 1.38 and 138 s so as to impose apparent nominal strain rates of 1 and 0.01 s⁻¹ respectively. Immediately after the deformation, the specimens were *in-situ* water-quenched. The schematic of the experimental methodology and sectioning plan of the specimen for observation is presented in Fig. 1(b). Measurement of grain boundary misorientation was carried out using OIMTM software (TexSEM Lab Inc. USA) by EBSD in a Schottky type (Philips XL30 SFEG) scanning electron microscope (SEM) operated at 15 kV. For this purpose, the cross section of the deformed specimens was electrolytically polished using 100 mL HClO₄ + 900 mL CH₃COOH solution to obtain a smooth and flat surface.

The measurement of ferrite grain size was carried out from the boundary maps obtained through EBSD patterns and includes all grains surrounded by boundaries with misorientation larger than 15°. However, grains with boundaries partially surrounded by misorientation (θ) $5^\circ \leq \theta \leq 15^\circ$ were also selected separately since many of the new high angle grain boundaries ($\theta > 15^\circ$) were found to be surrounded by a segment with low angle boundary ($5^\circ \leq \theta \leq 15^\circ$).⁶⁾ During the grain size measurement, only grains with aspect ratio less than 3 were considered to avoid deformed/work hardened grains being included. The measurement of the ferrite grain size was carried out from the boundary maps by tracing out the individual grains manually. Grain size (d) was defined as the square root of the average grain area measured, which is equal to the ASTM grain size.

3. Results and Discussion

3.1 Microstructural evolution at large strains and ferrite grain size measurement

Figures 2(a)–(f) shows a set of the boundary maps of the deformed specimens at a strain of 4 at temperatures in the range of 773–923 K and at strain rates of 0.01 and 1 s⁻¹ respectively. In the present study, grain boundaries are distinguished by three colors according to the misorientation,

Fig. 1 (a) Optical photomicrograph of the initial microstructure of the steel used. (b) Schematic of the experimental methodology and sectioning plan for microstructural observation and grain size determination.

θ . The red line represents the high angle grain boundaries of $\theta \geq 15^\circ$. The dark blue and the light blue lines represent low angle grain boundaries of $5^\circ \leq \theta < 15^\circ$ and $1.5^\circ \leq \theta < 5^\circ$, respectively. The presence of a black point in the pattern indicates invalid data where the image quality (IQ) or the confidence index (CI) is too low (IQ < 50 and CI < 0.1).

In order to understand the mechanism of formation of new ultrafine grains, a comparison of the theoretical and observed ferrite pancake thickness (HAGB spacing) was carried out. The increase in the HAGB area due to the shape change introduced by the imposed strain depends both on the amount of strain as well as the nature of deformation.^{13,14)} The relationship between the original grain size (d_0), the HAGB

Fig. 2 Boundary maps of the specimen deformed at (a) 773 K, 1 s^{-1} ; (b) 773 K, 0.01 s^{-1} ; (c) 823 K, 1 s^{-1} and (d) 823 K, 0.01 s^{-1} (e) 923 K, 1 s^{-1} and (f) 923 K, 0.01 s^{-1} at a strain of 4. The markings in red on the right hand side of the micrographs represent the calculated HAGB spacing as per eq. (3).

spacing (TH_α) parallel to compression axis and the true strain (ε) is given by

$$TH_\alpha = d_0 \exp(-\varepsilon) \quad (3)$$

It should be noted that Equation (3) is valid only if there are no other microstructural reconstitution mechanisms operating at the specified deformation conditions. In Fig. 2, the markings in red on the right hand side of the micrographs represent the HAGB spacing at a given strain ($\varepsilon = 4$) as per eq. (3). It may be noted that the magnification used for obtaining the boundary maps shown in Fig. 2 is not same for all and is selected based on the grain size of the specimen. A close observation of Figs. 2(a) and (b) along with the calculated HAGB spacing shown on the right hand side reveals that the calculated HAGB spacing closely matches with the observed spacing, indicating the microstructure to be essentially work hardened without any major reconstitution.

On the other hand, comparison of Fig. 2(e) with that of Fig. 2(f), which were deformed at 923 K at 1 s^{-1} and 0.01 s^{-1} respectively, indicates a large variation between the calculated and observed HAGB spacing. This suggests a large-scale migration of grain boundaries during deformation. Further, a comparison of Figs. 2(e) with (f) indicates that significant migration of the grain boundaries has taken place when deformation time has increased from 1.38 s ($\dot{\varepsilon} = 1 \text{ s}^{-1}$) to 138 s ($\dot{\varepsilon} = 0.01 \text{ s}^{-1}$). In view of the large variation between the calculated and observed HAGB spacing, the process of new grain formation is attributed to dynamic recrystallization (DRX).

Intermediate behavior may be noted by comparing the Figs. 2(c) and (d) where both pancake grains and equiaxed grains are seen and a relatively close correlation between the calculated and observed HAGB spacing is noticed. In other words, strain hardening along with dynamic recovery or

(a). Work hardening with or without Dynamic Recovery

(b). Dynamic Recrystallization (DRX)

Fig. 3 Schematic illustration of classification of microstructural evolution during large strain warm deformation of 0.15 carbon steel.

recrystallization of the microstructure has taken place.

From the above observations, it may be noted that the large strain-high Z deformation reveals occurrence of dynamic recovery characterized by compressed and elongated type microstructure, whereas the low Z deformation shows the prevalence of DRX as evidenced by clear equiaxed grains. In fact, Glover and Sellars¹⁵⁾ noted a change in the restorative mechanism from dynamic recovery at high Z values to that of DRX at low Z values during high temperature deformation of α -Fe. This observation was further examined and verified by Maki¹⁶⁾ and Tsuji¹⁷⁾ in ferrite for the occurrence of DRX. Based on the above observations, the microstructural manifestations during large strain warm deformation of low carbon steel are summarized in Fig. 3 with respect to the formation mechanism of new ultrafine grains surrounded by HAGB's.

Figure 4 shows the average ferrite grain size of the newly evolved grains surrounded by whole boundaries having misorientation larger than 15° (closed symbols) and by grains partially surrounded with boundaries having misorientation between 5° and 15° (open symbols) under various deformation conditions by EBSD analysis. It may be noted from the figure that the average ferrite grain size does not depend on strain. The average size of newly evolved grains surrounded by boundaries with misorientation larger than 5° or 15° is almost same. The volume fraction and average misorientation of the new ultrafine ferrite grains increases gradually with strain while the grain size itself does not change with strain. The average ferrite grain sizes were in the range of 0.26 to 1.1 μm .⁶⁾

3.2 Controlling mechanism of ferrite grain size

During large strain deformation, the HAGB spacing decreases and there is a possibility of approaching the order of mean thermal diffusion distances (given by the character-

istic diffusion distance, $\sqrt{Dt}$ where D is the appropriate diffusion coefficient based on the operating controlling mechanism of diffusion).

If the operating controlling mechanism is volume diffusion, then D is given by:

$$D_v = D_v^0 \exp\left(\frac{-Q_v}{RT}\right) \quad (4)$$

where D_v^0 is the frequency factor ($5.0 \times 10^{-1} \text{ cm}^2 \cdot \text{s}^{-1}$), Q_v is the activation energy for volume diffusion ($240 \text{ kJ} \cdot \text{mol}^{-1}$), R is the universal gas constant and T is the absolute temperature.¹⁸⁾

If the operating controlling mechanism is grain boundary diffusion, then D is given by:

$$D_{gb} \delta = 5.4 \times 10^{-8} \exp\left(\frac{-Q_{gb}}{RT}\right) \quad (5)$$

where the pre-exponential constant $5.4 \times 10^{-8} \text{ (cm}^3 \cdot \text{s}^{-1}\text{)}$ is the product of grain boundary diffusion constant (D_{gb}^0) and grain boundary thickness (δ), Q_{gb} is the activation energy for grain boundary diffusion ($155 \text{ kJ} \cdot \text{mol}^{-1}$), and R and T have their usual meaning.¹⁸⁾ Assuming δ to be 1 nm, one can compute the value of D_{gb} at any temperature.¹⁹⁾

The characteristic diffusion distance or distance into the volume and along the grain boundaries in a polycrystalline material are given by Ref. 20):

$$L_v = \sqrt{D_v t} \quad (6)$$

$$L_b = \sqrt{D_{gb} t} \quad (7)$$

Figures 5(a) and (b) shows the comparison between the measured ferrite grain size (d) and the diffusion distance $\sqrt{D_v t}$, considering volume diffusion data [Fig. 5(a)] and diffusion distance $\sqrt{D_{gb} t}$, using the grain boundary diffusion data [Fig. 5(b)]. The ferrite grain size data for the specimens

Fig. 4 Average ferrite grain size of the newly evolved grains surrounded by whole boundaries having misorientation larger than 15° (closed symbols) and by grains partially surrounded with boundaries having misorientation between 5° and 15° (open symbols) under various deformation temperatures and strain rates of (a) 0.01 s^{-1} and (b) 1 s^{-1} with designation.⁶⁾

deformed at 600°C at 0.01 and 1 s^{-1} was also included in this graph. While calculating the characteristic diffusion distance as per eqs. (6) and (7) reported in Fig. 5, the adiabatic temperature rise during deformation was also taken into consideration. The comparison of ferrite grain size with the characteristic diffusion distance assuming grain boundary diffusion [Fig. 5(b)] is found to be in far better agreement than that using the volume diffusion data [Fig. 5(a)]. From eq. (3), the calculated TH_α at a strain of 4 (for initial grain size of $15 \mu\text{m}$) becomes $0.28 \mu\text{m}$ which is in the comparable range with $\sqrt{D_{\text{gb}}t}$ indicating that grain boundary diffusion plays an important role in determining the ferrite grain size. On the other hand, it is not comparable with $\sqrt{D_v t}$, which is completely out of range with TH_α . This implies that grain boundary diffusion, but not volume diffusion plays an important role in determining the ferrite grain size. Therefore, it may be considered that the deformed ferrite grain structure is fully recrystallized when HAGB spacing (TH_α) is smaller than the characteristic diffusion distance by grain boundary diffusion ($\sqrt{D_{\text{gb}}t}$); otherwise, atoms diffuse only to a short distance leaving a mixture of elongated and newly generated grains.

Fig. 5 (a) Comparison of the observed ferrite grain size ($\epsilon = 4$) and the characteristic diffusion distance during large strain deformation of 0.15C steel assuming the prevalence of volume diffusion. (b) Comparison of the observed ferrite grain size ($\epsilon = 4$) and the characteristic diffusion distance during large strain deformation of 0.15C steel assuming the prevalence of grain boundary diffusion.

To further understand the strong dependence of ferrite grain size, d on Z parameter given by eq. (2), the relationship between $\sqrt{D_t t}$ and Z is considered. Multiplying eqs. (1) and (4) and assuming that the activation energy for deformation is equal to the activation energy for volume diffusion, i.e. $Q_a = Q_v$, we obtain

$$D_v Z = D_v^0 \dot{\epsilon} \quad (8)$$

The characteristic diffusion distance by volume diffusion is given by,

$$\sqrt{D_v t} = \sqrt{\frac{D_v^0 \dot{\epsilon}}{Z}} t = \sqrt{\left(\frac{D_v^0 t}{\exp(Q_v/RT)} \right)} \quad (9)$$

On the other hand, if the activation energy for deformation is equal to the activation energy for grain boundary diffusion, *i.e.* $Q_a = Q_{gb}$, we obtain the characteristic diffusion distance ($\sqrt{D_{gb} t}$) as:

$$\sqrt{D_{gb} t} = \sqrt{\frac{D_{gb}^0 \dot{\epsilon}}{Z \delta}} t = \sqrt{\left(\frac{D_{gb}^0 t}{\exp(Q_{gb}/RT) \delta} \right)} \quad (10)$$

The relationship between the Z parameter and $\sqrt{Dt}$ is plotted in Fig. 6. It may be noted from Fig. 6 that the variation of $\sqrt{D_v t}$ and $\sqrt{D_{gb} t}$ with Z parameter can be represented by straight lines ($\sqrt{D_{gb} t}$, represented by dotted line and $\sqrt{D_v t}$, represented by solid line). In other words the dotted line represents the grain boundary diffusion and solid line represents the volume diffusion as the controlling mechanism for ferrite grain size. On investigation, it was found that, for volume diffusion case, $\sqrt{D_v t}$ becomes equal to $1.41Z^{-0.5}$ whereas for grain boundary diffusion case, $\sqrt{D_{gb} t}$ is $1.47Z^{-0.5}$ at a fixed strain of 4 resulting in near overlapping of the two lines (represented by solid and dotted lines).

In order to understand the variation of ferrite grain size with Z parameter to get further insight into the controlling mechanism, the grain size data is plotted in Fig. 6. For this

purpose, both volume diffusion and grain boundary diffusion were considered separately and $Q_{gb} = 155 \text{ kJ} \cdot \text{mol}^{-1}$ and $Q_v = 240 \text{ kJ} \cdot \text{mol}^{-1}$ were used in the computation of Z. It

Fig. 6 Variation of the observed ferrite grain size in tested specimens as a function of Zener-Hollomon parameter along with the computed diffusion distance assuming the prevalence of grain boundary diffusion ($\sqrt{D_{gb} t}$, represented by dotted line) and volume diffusion ($\sqrt{D_v t}$, represented by solid line). Ferrite grain size data is separately plotted assuming grain boundary diffusion ($Q = 155 \text{ kJ} \cdot \text{mol}^{-1}$) as well as volume diffusion ($Q = 240 \text{ kJ} \cdot \text{mol}^{-1}$).

Fig. 7 Schematic of (a) Turnbull model for grain boundary migration and (b) the development of microstructure as a function of strain during low, medium and large strain warm deformation of 0.15 carbon steel.

may be noted that the nominal strain rates (0.01 and 1 s^{-1}) at which the tests were conducted have been corrected to actual strain rates (0.03 and 3 s^{-1}) to incorporate the effect of strain distribution within the specimen. From Fig. 6, it is interesting to note that the relationship between the ferrite grain size and Z parameter assuming grain boundary diffusion (data in grey) matches very well with the dotted line relating the characteristic diffusion distance with Z parameter. On the other hand, the ferrite grain size data plotted assuming the prevalence of volume diffusion (data in black) matches poorly with the solid line relating the characteristic diffusion distance with Z parameter.

In order to further understand the reason for the ferrite grain size to follow grain boundary diffusion rather than volume diffusion, the mechanism of grain boundary migration is considered here. Figure 7(a) shows the schematic for grain boundary migration during large strain warm deformation and Fig. 7(b) shows the schematic for the development of microstructure as a function of strain during low, medium and large strain warm deformation of 0.15 carbon steel. Fig. 7(a) is drawn on the basis of the observation of Turnbull²¹⁾ that the activation energy for grain boundary migration is equal to the activation energy for grain boundary diffusion. Also it was noted²¹⁾ that the grain boundary migration distance is comparable to grain boundary diffusion distance whereas, it is many orders of magnitude larger than volume diffusion. Therefore, the grain boundary migration distance (x) is comparable to the grain boundary diffusion distance ($L_b = \sqrt{D_{gb}t}$) as shown in Fig. 7 and not to the volume diffusion distance ($L_v = \sqrt{D_vt}$). In the present study, new ultra fine ferrite grains are formed by the migration of grain boundaries and their size is closely related to the grain boundary diffusion distance. Therefore, the mechanism by which the ferrite grain size is controlled by Z parameter is attributed to be grain boundary diffusion. It should be noted that, carbon exists in the form of spheroidized cementite particles (in specimens deformed to large strains) which may cause pinning effect. However, its effect is insignificant here since the volume fraction of cementite is small and needs further study to examine its effect at higher volume fractions.

4. Conclusions

The role of diffusion as the controlling mechanism of

ferrite grain size was studied during large strain-high Z deformation of 0.15 carbon steel in the present study. It was found that characteristic diffusion distance by grain boundary diffusion matches very well with ferrite grain size formed. Therefore grain boundary diffusion controls the ferrite grain size. The ferrite grain size dependency on Z parameter can be attributed to grain boundary diffusion during large strain deformation.

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