Fracture Toughness Enhanced by Grain Boundary Shielding in Submicron-Grained Low Carbon Steel

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The enhancement of toughness at low temperatures in fine grained low carbon steel was studied, basing on a shielding theory due to dislocations and grain boundaries. Fully annealed low carbon steel was subjected to an accumulative roll bonding (ARB) process for grain refining. The grain size perpendicular to the normal direction was found to be approximately 200 nm after the ARB process. The fracture toughness of low carbon steel ARB applied was measured at 77 K by four-point bending, comparing with the fracture toughness of those without the ARB process. It was found that the value of fracture toughness at 77 K was increased by grain refining due to the ARB, indicating that the ARB process enhances toughness at low temperatures as reported in interstitial free steel and phosphorus doped interstitial free steel. It also deduces that the brittle-ductile transition (BDT) temperature shifted to a lower temperature. The enhancement of toughness and the decrease of the BDT temperature due to grain refining cannot be explained completely by the dislocation pile-up model of dislocations at grain boundaries. A simple application of the pile-up model into the shielding concept of grain boundary shielding was suggested in this paper. Therefore, another explanation is required in order to understand the mechanism of the enhancement of the BDT in ultra-fine grained materials.

In the present study, the first step for understanding the mechanism of the enhancement of toughness at low temperatures after the ARB process was attempted, basing on the crack tip shielding theory. Low carbon steel was firstly subjected to the ARB process in order to obtain a fine grained structure. Fracture toughness was measured by four-point bending tests, comparing with the results of fully annealed low carbon steel. Simulations of quasi-two-dimensional dislocation dynamics were also performed in order to discuss the effect of dislocation source spacing along a crack front on the BDT temperature. In addition to the simulation, a new concept of accommodating stress intensity at the crack tip due to grain boundaries to explain the enhancement of toughness and the decrease of the BDT temperature in fine grained materials. [doi:10.2320/matertrans.ME2007111]

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1. Introduction

Grain refining is one of the powerful methods in order to enhance mechanical property, especially strength of the materials. Severe plastic deformation, such as equal-channel angular pressing (ECAP), shot peening, high pressure torsion (HPT), accumulative roll bonding (ARB) and so on, have attracted many interests to produce materials with ultrafine grained structures. Tsuji et al. reported that interstitial free (IF) steel and phosphorous doped IF steel gained high strength without losing much toughness at low temperatures by the ARB process. High ductility is seen in the materials after the ARB process even at low temperatures such as the liquid nitrogen temperature. This indicates that the brittle-ductile transition (BDT) temperature of IF steel is decreased to a lower temperature by grain refining.

In single crystals, the basic mechanism of the BDT and enhancement of toughness is now understood by an idea of stress accommodation due to dislocations at a crack tip, so-called “dislocation shielding”. Many experimental works have been done for clarifying the theory, using silicon crystals as a model crystal. As a result, the BDT is found to be controlled by a thermal process of dislocation glide, which have been confirmed by the fact that the value of activation energy for BDT is the same as that for dislocation glide in many materials, such as semiconductors, bcc metals etc. In polycrystals, the grain size dependence of strength has been explained by the pile-up model of dislocations at grain boundaries. However, no previous work, based on the shielding theory, has attempted to explain the grain size dependence of fracture toughness and the BDT. A simple application of the pile-up model into the shielding theory implies that fracture toughness would be rather decreased and the BDT temperature would be increased as decreasing the grain size since the grain boundaries play a role as obstacles for dislocation glide, which prohibits continuous emissions of dislocations at the crack tip. Therefore, another explanation is required in order to understand the mechanism of the enhancement of the BDT in ultra-fine grained materials.

2. Experimental

Low carbon steel, the chemical composition of which is shown in Table 1, was employed. Since low carbon steel exhibits rather high ductility, P was slightly doped to reduce its ductility and to demonstrate a remarkable effect of grain-refining on the fracture toughness at low temperatures. Cold-rolled 2 mm thick sheets were annealed at 1023 K for 3.6 ks in
order to complete recrystallisation. The grain size of the fully annealed sheets was found to be 0.8 mm. The sheets were cold-rolled by 50% reduction in thickness for a subsequent ARB process. The cold-rolled 25 \times 200 \times 1 \text{ mm} sheets were provided to the ARB process for three cycles. The ARB process was performed at 773 K, though the detail is shown in elsewhere.\textsuperscript{9)} The final equivalent strain was 2.93. Miniature size was performed at 773 K, though the detail is shown in elsewhere.\textsuperscript{9)} The final equivalent strain was 2.93. Miniature size of four-point bending samples were cut out from the sheet after the ARB process. Figure 1 shows a schematic drawing of the four-point bending specimen, exhibiting the normal direction (ND), rolling direction (RD) and transverse direction (TD) of the sheet. Inner and outer beam distances were 4 mm and 10 mm, respectively. A notch with a depth of 100 \mu m was introduced perpendicular to TD on the tensile surface of each specimen. The same size bending samples were also cut out from a sheet without the ARB process for comparison. The strain rate at the tensile surface was set to be 4.5 \times 10^{-4} \text{ s}^{-1}. Four point bending tests were carried out at 77 K to measure fracture toughness with six and seven specimens in fully annealed and ARBed samples, respectively.

3. Results

Figure 2 shows a TEM bright field image of a sample just after the ARB process, observed from TD. Some grains contain dislocations of high density. Grains are elongated to the direction parallel to RD. It was found that the grain size perpendicular to RD is approximately 200 nm, showing that ultra-fined grains actually developed in low carbon steel by the ARB process.

Figure 3 shows load-displacement curves obtained at 77 K at a strain rate of 4.5 \times 10^{-4} \text{ s}^{-1}. Curve (a) from an as annealed sample indicates a curve of typical brittle fracture, where rupture took place without any applicable plastic deformation. Curve (b) from an ARBed sample shows slight plastic deformation just before it broke. It corresponds to the result shown by Tsuji \textit{et al.}\textsuperscript{9)} that the absorption energy of ARBed materials measured by Charpy impact tests at 77 K is higher than that for large grained.

Figure 4(a) shows a SEM image from the as annealed sample cleaved at 77 K, indicating a typical transgranular fracture surface where a crack extended from the beneath of the notch (from left to right in the figure). This corresponds to Curve (a) in Fig. 3, where the specimen fractured without any applicable plastic deformation. Figure 4(b) shows a SEM image from the ARBed sample fractured at 77 K. Six layers of about 100–200 \mu m width are seen parallel to RD due to ARB process. Figure 4(c) shows an enlarged image of the area surrounded by a rectangle of (c) in Fig. 4(b), indicating a brittle fracture surface. Grains are elongated along RD due to the ARB process as seen in Fig. 2. Almost all the area of the fracture surface indicate the brittle fracture surface of this kind. Figure 4(d) shows an enlarged image of the area surrounded by a rectangle of (d) in Fig. 4(b), exhibiting dimples on the fracture surface. This indicates that local plastic deformation took place during crack advance, which corresponds to Curve (b) in Fig. 3. Fracture toughness, $K_f$, in a four-point bending test is given by Ref. 20):

$$K_f = \sigma_f \sqrt{\pi c f(\xi)}$$

with $\xi = c/h$,

$$f(\xi) = 1.112 - 1.40\xi + 7.33\xi^2 - 13.08\xi^3 + 14.0\xi^4,$$  \hspace{1cm} (1)

where $\sigma_f$ is the fracture stress, $c$ is the crack length and $h$ is the specimen height. The fracture toughness of as annealed and ARBed samples was measured at 77 K at a strain rate of 4.5 \times 10^{-4} \text{ s}^{-1} on a tensile surface of the beam.
Table 2 shows the values of fracture toughness of as annealed and ARBed samples fractured at 77 K as shown in Fig. 3. The measured values were statistically treated to show the average values with the standard deviations in the table. The values of the fracture toughness of ARBed and fully annealed samples are found to be 38.5 ± 2.2 MPa m¹/² and 22.5 ± 2.9 MPa m¹/², respectively. The error ranges are much smaller than the difference in the two average values, indicating that the difference in fracture toughness is not due to the experimental error. The fracture toughness of low carbon steel was increased by the ARB process, that is, the toughness of the material was enhanced by such grain refinement. This is in good agreement with the result of Charpy impact tests obtained by Tsuji.9) Two kinds of mechanisms which enhance the ductility of fine grained materials at low temperatures will be discussed in the next session.

4. Discussion

4.1 Accommodation of stress intensity at a crack tip due to dislocations

When an external stress is applied on a specimen with a crack, the stress, σij, near a crack tip in the absence of dislocations is expressed by polar coordinates (r, θ):

\[
\sigma_{ij} = \frac{K_1}{\sqrt{2\pi r}} f_j(\theta),
\]

where \( K_1 \) is the stress intensity factor given as \( K_1 = \sigma_a/(2\pi r)^{1/2}. \) (\( \sigma_a \): applied stress, \( a \): crack length, \( S \): specimen shape factor), \( f_j(\theta) \) is function of \( \theta \) and \( r \) is the distance from the crack tip. When dislocations are emitted from the crack tip and stay around there, the intensified stress, \( \sigma_j^D \), is modified by the superposition of the stress field due to the dislocations. The stress near the crack tip due to the dislocations, \( \sigma_j^D \), is given by:

\[
\sigma_j^D = \frac{k_D}{\sqrt{2\pi r}} f_j(\theta),
\]

where \( k_D \) is the stress intensity factor caused by dislocations, given by the following equation:

\[
k_D = \sum_i \left( \frac{3\mu b_i}{(1 - \nu)(2\pi r)} \cos \frac{\theta}{2} \sin \theta \right),
\]

where \( \nu \) is shear modulus, \( b \) is the scalar of the Burgers vector. When an applied stress and dislocations are present, the total stress near the crack, \( \sigma_j^T \), is derived from eqs. (1) and (2),

\[
\sigma_j^T = \sigma_j + \sigma_j^D = (K_1 + k_D)f_j(\theta)/\sqrt{2\pi r},
\]

with \( k_1 = K_1 + k_D. \)

Here \( k_1 \) is termed “local stress intensity factor”.

The crack extension force in the plane strain condition under a simple mode I stress field is given by:

\[
R_D = \left( \frac{3\mu b_i}{(1 - \nu)(2\pi r)} \cos \frac{\theta}{2} \sin \theta \right)
\]

with \( R_D \) being the crack extension force.
\[ F_c = \frac{k_1^2}{E} (1 - \nu^2), \]  
(7)

where \( E \) is the Young’s modulus.

Opposing this elastic force is a resistive force due to the surface energy. The criterion for the crack extension is given with the surface energy, \( \gamma \): \(^{(25)}\)

\[ F_c = \frac{k_1^2}{E} (1 - \nu^2) = 2\gamma. \]  
(8)

Replacing \( k_1 \) in eq. (6) with (8) gives the applied stress intensity factor for crack extension,

\[ K_{IC} = \sqrt{\frac{2E\gamma}{1 - \nu^2}} - k_0. \]  
(9)

When dislocations are introduced by an external tensile load, the sign of \( k_0 \) term due to dislocations is negative to restrain the concentration of tensile stress around the crack tip, which was confirmed by direct TEM observations of dislocations around a crack tip. \(^{(26,27)}\) This means that the stress intensity due to the dislocations generated around the crack is compressive which is against the external applied stress. Therefore, the applied stress intensity factor increases, i.e., the fracture toughness increases. This idea is called “shielding effect” due to dislocations.

4.2 Model for two dimensional dislocation dynamics simulation

Hirsch and Roberts \(^{(28)}\) summarised the BDT temperature of single crystal silicon from several groups and plotted \( \ln(\text{strain rate}) \), \( \ln(\dot{\epsilon}) \), against reciprocal BDT temperature, \( 1/T_{\text{BDT}} \), showing that each line from the collected data exhibits the same angle of slopes in the \( \ln(\dot{\epsilon}) - 1/T_{\text{BDT}} \) diagram but shows different interceptions of the lines with the axis of \( \ln(\dot{\epsilon}) \). This indicates that the activation energy for the BDT is constant over the whole results used, being independent from the shape or measure of the experiment, however, the BDT temperature depends on the way of the experiment. Hirsch pointed out from the result that the BDT temperature depends on the number of dislocation sources and spacing along a crack front. Here, the effect of the dislocation source spacing is evaluated, using a following model of dislocation glide from a crack tip.

Figure 5 shows a schematic drawing of a quasi-two-dimensional model for dislocation dynamics simulation employed in this study, which is suggested by Roberts et al. \(^{(29)}\) A crack was loaded in a simple opening mode with a constant rate \( dK/dt = 0.01 \text{ MPa-m}^{-1/2} \text{ s}^{-1} \). The other components of modes II and III were set to be zero. Dislocation sources, marked as \( \odot \) in Fig. 5, locate along a crack front at the distance of \( d_c \) ahead and parallel to the crack front as shown Fig. 5. Dislocations emitted from each source and bow out on a slip plane with an angle of 45° with respect to the crack plane, forming concentric loops. This geometry of the crack and slip plane corresponds to a system of (010)[100] crack and (110) slip plane. The basic idea of this model is that dislocations need to travel a certain distance to shield the crack tip efficiently. When a dislocation loop travels a distance of \( d_{\text{crit}} \), a segment of dislocation loop perpendicular to the crack front meets another loop segment coming from the next source. Since they have the same Burgers vector but opposite dislocation line vector, they react and vanish. Then, only a loop segment parallel to the crack front remains at the distance of \( 2d_{\text{crit}} \) from the crack front. In the present study, the value of shielding at the position \( Z_{\text{crit}} \) on the crack front in Fig. 5 is calculated.

The criterion for the first dislocation to emit was defined with a total force on the first dislocation, \( f_d^1 \), exceeds zero:

\[ f_d^1 > 0 \quad \text{with} \quad f_d^1 = f_a + f_m, \]  
(10)

where \( f_a \) and \( f_m \) are the force due to applied stress intensity and mirror force, respectively. Once the first dislocation emitted, dislocation-dislocation interaction force, \( f_{dd} \), is also taken into account for the emission criterion, i.e., \( f_{dd} \) was added in the right hand term in eq. (10).

Dislocations generated at the crack tip glide away from the crack tip on a slip plane with an angle of 45° with respect to the crack plane as shown in Fig. 5, where dislocations do not cross slip in this model. The velocity of the \( \text{th} \) dislocation in iron is described by an Arrhenius type empirical law of the form:\(^{(30)}\)

\[ v^i = v_0 \left( \frac{t_d^i}{t_0} \right)^m \exp \left( - \frac{Q}{kT} \right), \]  
with \( t_d^i = \frac{f_d^i}{b} \),  
(11)

where \( v_0, t_0 \) and \( m \) are constants, \( k \) is the Boltzmann constant, \( T \) is the absolute temperature and \( Q \) is the activation energy for dislocation glide. The values of the constant parameters used in Equation (11) are obtained from the result of the direct measurement of dislocation velocity in pure iron at 198 K in Ref. 30). The activation energy for dislocation glide used in this study was obtained by Burner et al. \(^{(31)}\) as an activation enthalpy for a double kink nucleation along a screw dislocation in iron. The value is the same with that for BDT. Those parameters are listed in Table 3. The calculation was terminated when the local stress intensity factor, \( k_i \), exceeds 0.8 MPam\(^{1/2} \) which is calculated from a value of surface energy and Yong’s modulus of iron. \(^{(32)}\)

4.3 Calculated BDT temperatures

Figure 6(a) and (b) show calculated \( K_a \), \( k_1 \) and \( k_0 \) at 80 K plotted against time with \( d_{\text{crit}} \) of 5 nm and 50 nm, respectively. \( K_a \), applied stress intensity factor, increases linearly.
with increasing time with a slope of 0.01 MPa·m$^{-1/2}$·s$^{-1}$. Since shielding dislocations emitted from the crack tip, the sign of $k_D$ denotes negative value. $k_D$ drops every time when each segment of dislocation loops reaches $2d_{crit}$ from the crack tip, when the dislocation starts to shield the crack tip. $k_I$ increases with the same increment with that of $K_a$ until the first dislocation reaches $2d_{crit}$ from the crack tip, then $k_D$ drops from $K_a$ due to shielding by dislocations. $k_I$ also oscillates due to the oscillation of $K_a$. $K_a$ is obtained from the value of $K_a$ when $k_I$ exceeds 0.8 MPa·m$^{-1/2}$, which is calculated from the surface energy of iron in eq. (9). Figure 6(b) shows a graph in such a case that $2d_{crit}$ is set to be 50 nm. When a dislocation source spacing increases, $k_I$ reaches 0.8 MPa·m$^{-1/2}$ faster than in case of $2d_{crit} = 5$ nm, hence $K_I$ for $2d_{crit} = 50$ nm is smaller than that for $2d_{crit} = 5$ nm at 80 K.

Figure 7 shows calculated fracture toughness against temperature. Solid square, circle and triangle indicate results for $2d_{crit}$ of 5 nm, 50 nm and 300 nm, respectively. The values of fracture toughness are much lower than those experimentally obtained in the present study. This is due to a usage of the simplified model in the simulation, where only a single source and slip plane are activated. In this study, the BDT temperature is defined as a temperature at which fracture toughness increases from the value derived from Griffith level (0.8 MPa·m$^{-1/2}$). Dashed line in Fig. 7). It is to be noted here that the BDT curve shifts to the right with increasing the value of $2d_{crit}$. It indicates the BDT temperature decreases with decreasing the dislocation source spacing. This result of simulation assumes that the enhancement of toughness of the material subjected to ARB is due to decreasing the source spacing along the crack front. In this simulation, the effect of grain boundaries ahead of the crack tip is not taken into account. The effect will be considered in the next sub-session.

### 4.4 Grain boundary shielding

As pointed out previously in the present paper, the classical pile-up model cannot fully explain the enhancement of toughness of materials at low temperatures due to reducing the grain size. Since the volume fraction of grain boundaries is relatively higher than that of conventional grained materials, it is deduced that not only dislocations emitted at the crack tip but also grain boundaries accommodate the stress intensity at the crack tip, which can be termed “grain boundary shielding”.

Figure 8 shows a simplified concept of the grain boundary shielding, in which a crack is loaded under mode I tensile stress. Grain boundaries are shown by dashed lines. When the applied load is increased, the shear stress (dashed arrows in Fig. 8(a)) on a slip plane ahead of the crack tip is also increased, and then a dislocation is emitted from the crack tip. An edge dislocation, for instance, is emitted as shown in the figure. The dislocation has such a stress field that shown by solid arrows in Fig. 8(b), shielding the crack tip. It is the concept of “shielding effect” due to dislocations discussed in the previous sub-session. When the dislocation approaches a grain boundary, the dislocation is absorbed at the grain boundary (Fig. 8(c)). It is to be noted here that even though the dislocation is absorbed at the grain boundary, the stress field due to the dislocation still remains around the grain boundary. Actually, Shimokawa et al. calculated interactions between edge dislocations and a tilt grain boundary by quasi-continuum simulations, in which edge dislocations generated from a crack tip and glide away to a tilt grain boundary under a shear stress. They indicate that the dislocations are absorbed at the grain boundary, remaining their stress around there. The result of the simulation deduces the existence of the stress which still accommodates stress intensity at the crack tip even though the dislocation is absorbed at the grain boundary. When the stress field in the next grain becomes high enough, another dislocation emit at the grain boundary in the next grain and glide away to the
A new concept of grain boundary shielding was also suggested to explain the enhancement of toughness in ultra-fine grained materials.

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