B Brittle-Ductile Transition in Low Carbon Steel Deformed by the Accumulative Roll Bonding Process

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B Brittle-ductile transition (BDT) behaviour was investigated in low carbon steel deformed by an accumulative roll-bonding (ARB) process. The temperature dependence of its fracture toughness was measured by conducting four-point bending tests at various temperatures and strain rates. The fracture toughness increased while the BDT temperature decreased in the specimens deformed by the ARB process. Arrhenius plots between the BDT temperatures and the strain rates indicated that the activation energy for the BDT did not change due to the deformation with the ARB process. It was deduced that the decrease in the BDT temperature by grain refining was not due to the increase in the dislocation mobility controlled by short-range obstacles. Molecular dynamics simulations revealed that moving dislocations were impinged against grain boundaries, creating a shielding field, and were reemitted from there with increasing strain. Grain refining led to an increase in the fracture toughness at low temperatures and a decrease in the BDT temperature. In the present paper, the roles of grain boundaries have been discussed in order to explain the enhancement in the fracture toughness of fine-grained materials at low temperatures, and the decrease in the BDT temperature.

Keywords: grain boundary shielding, crack, severe plastic deformation, accumulative roll-bonding (ARB)

1. Introduction

Solid materials are plastically deformed at high temperatures. In contrast, some materials fail without a substantial plastic deformation at low temperatures; this is referred to as brittle fracture. It is a prominent event that occasionally occurs in bcc metals as well as semi-conductors and ceramics, which is well-known as brittle-ductile transition (BDT). Since a BDT event is critical, especially in structural materials, some counter measures to decrease the BDT temperature have been suggested. One of the methods is grain refining. Tsuji et al. deformed interstitial free (IF) steel by an accumulative roll-bonding (ARB) process and measured the absorbed energy at various temperatures by conducting Charpy impact tests. They demonstrated that the IF steel gained both strength as well as ductility at low temperatures. It implies that the BDT temperature itself is decreased by the ARB process.

In single crystal materials wherein the Peierls stress is high, fracture tests provide the relation between the strain rate, \( \dot{\varepsilon} \), and the BDT temperature, \( T_{BDT} \), as follows:

\[
\dot{\varepsilon} = \varepsilon_0 \exp\left(-\frac{E_a}{kT_{BDT}}\right), \tag{1}
\]

where \( E_a \) is the activation energy, \( k \) is the Boltzmann constant, and \( \varepsilon_0 \) is a pre-factor. Many studies have confirmed that the values of \( E_a \) in eq. (1) were close to those for dislocation glide. It was deduced that the BDT behaviours in those materials are controlled by a thermal activation process related to the dislocation glide. Michot et al. pointed that the pre-factor in eq. (1) depends on the number of dislocation sources along the crack front. It indicates that the BDT temperature depends on dislocation mobility as a thermal activated term as well as dislocation source density around a crack tip as an athermal term in eq. (1).

Since dislocations have long-range stress fields, the stress field around a crack tip is required to be modified when dislocations are introduced from the crack tip. The concept of stress accommodation is termed as ‘dislocation shielding’, which has been independently suggested by Thomson and Weertman. It was experimentally confirmed by transmission electron microscopy and photoelasticity in silicon single crystals.

In polycrystalline materials, the BDT temperature strongly depends on the grain size of specimens. The BDT temperature decreases with the grain size, which could be explained by employing a dislocation pile-up model. However, the application of the shielding theory to a pile-up model deduces that the BDT temperature in fine-grained materials should increase because the dislocations cannot continue to emit from the crack tip due to the back stress of the pile-up dislocations around the grain boundaries.

In the present study, the BDT behaviours in fine-grained materials have been discussed on the basis of the shielding theory. Low carbon steel was plastically deformed by the ARB process to obtain fine-grained specimens. The temperature dependence of its apparent fracture toughness was measured at various strain rates. The activation energy obtained from the BDT temperature in roll-bonded specimens was measured and was compared with that for fully annealed specimens with a large grain size. Molecular dynamics calculations were performed in order to demonstrate the roles of grain boundaries in a BDT event. The roles of grain boundaries have been discussed in order to explain the decrease in the BDT temperatures.

2. Experimental

Low carbon steel (0.02 mass% C) was employed, the chemical composition of which is presented in Table 1. Since
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3. Results

Figure 2 presents a TEM bright field image observed along the TD of the as roll-bonded specimen, revealing that some grains included a high density of dislocations. The grains were elongated along a direction nearly parallel to the rolling direction (RD). It was found that the grain size perpendicular to the RD was approximately 200 nm. The selected area diffraction pattern was ring-like, indicating that the boundaries between the adjacent grains had large misorientations.

Figures 3(a) and 3(b) illustrate the orientation image mappings (OIM) for the as roll-bonded specimens, indicating the normal direction (ND) and RD; these were observed along the TD at the centre of the surface by using electron backscatter diffraction (EBSD) with an accelerating voltage of 20 kV. The EBSD patterns were obtained every 0.05 μm (Hitachi H-4300SE). The grains were elongated along the RD, which has also been observed in sheets deformed by conventional rolling processes. Figure 3 roughly indicates three layers with a width of approximately 10 μm along the RD. The middle layer in the figure consists of fine grains with a width of approximately 500 nm. The top and bottom layers in the figure consist of large band-like structures, inclining towards the RD. The orientation distribution of the middle layer is relatively random as compared to that of the top and bottom layers. Figure 3(a) indicates that the ND directions of the majority of the grains were almost along (111) in the top layer. Figure 3(b) indicates that the RD directions of the grains were nearly parallel to (011) in the top and bottom layers. These figures indicate that some typical preferential orientations were developed due to the ARB process, which are usually induced by a conventional rolling process in bcc crystals.25 Figures 3(c) and 3(d) illustrate {111} and {011} pole figures obtained from the same regions in (a) and (b). They reveal the development of textures and are consistent with the results of the OIM images. The orientation scattering indicated that the trend of the texture was not very strong, and hence, the effect of those textures on the fracture event was negligible in the present study.

Figures 4(a) and 4(b) indicate the fractured surfaces of the fully annealed and the roll-bonded specimens at 77 K, respectively. Figure 4(a) exhibits river patterns, indicating a typical fracture surface in a brittle manner. Figure 4(b) shows layers with a thickness of approximately 100–200 μm parallel to the RD, corresponding to the depth of the fully annealed sheet before the ARB process. Micro-voids were observed at some places on the fractured surface, indicating that a local plastic deformation occurred during crack advance even at 77 K; the fully annealed samples demonstrated complete brittle fracture at this temperature. In order to evaluate the temperature dependence of the apparent fracture toughness, the four-point bending tests were performed.

Figures 5(a), 5(b) and 5(c) illustrate the apparent fracture toughness obtained for the roll-bonded samples as a function
of the temperature at the strain rates of $4.46 \times 10^{-4}$, $2.23 \times 10^{-3}$ and $1.12 \times 10^{-2}$ s$^{-1}$, respectively. The solid symbols indicate that the crack tip plastic zone of the fractured specimen corresponded to small-scale yielding. The open symbols indicate that the crack tip plastic zone of the ruptured specimens corresponded to large-scale yielding. The
BDT temperature was defined as one at which the highest fracture toughness was obtained although it is necessary to confirm the correlation between the BDT temperature defined in this study and that in Charpy impact tests. When fracture toughness did not definitively show the highest value, the BDT temperature was defined as the mean value of the highest temperature in fracture and the lowest temperature in flow. Figure 5 reveals that the BDT temperature increases from 114 to 135 K with the strain rate, as observed in other conventional bcc metals that were not roll-bonded.\(^{26}\) In order to evaluate the effect of grain refining, the temperature dependence of the apparent fracture toughness in the fully annealed samples was also measured.

Figures 6(a), 6(b) and 6(c) illustrate the apparent fracture toughness obtained for the fully annealed samples as a function of temperature at the strain rates of \(4 \times 10^6\), \(2.3 \times 10^3\), and \(1.2 \times 10^2\) s\(^{-1}\), respectively. The open symbols indicate that the specimens reached 11% strain (the test rig’s limit) during the tests without fracture; this was termed ‘ductile bending’. In the case of these specimens, the values of apparent fracture toughness shown in the figure were calculated using the stress when the load is deviated from the load-displacement curve, and thus the values of those apparent fracture toughness were the lower limits. The average value of fracture toughness at 77 K increased from 47 to 85 MPa m\(^{1/2}\) by the ARB process. The increase in the fracture toughness at 77 K implied that dislocation activity occurred in the roll-bonded specimens, which corresponded to the result that some dimples were observed on the fractured surface of the roll-bonded specimens shown in Fig. 4. However, no visible dislocation activity was observed in the fully annealed samples. The BDT temperatures in the roll-bonded specimens were lower than those in the fully annealed ones. The activation energy in eq. (1) was obtained from the results of the roll-bonded and fully annealed specimens in order to examine whether the dislocation mobility controlled by short-range obstacles had decreased.

Figure 7 illustrates the Arrhenius plots between the strain rates and the BDT temperatures. The solid squares indicate those obtained for the roll-bonded specimens. Regression lines were fitted to each set of plots, giving the activation energy, \(E_a\). The \(E_a\) values of the roll-bonded and fully annealed specimens were found to be 0.20 and 0.25 ± 0.09 eV, respectively, wherein the difference was negligible, taking the experimental accuracy into account. The error in the activation energy was due to the ambiguity in the BDT temperature observed in the case of the fully annealed samples in Fig. 6(a). The activation energy was not changed by the ARB process, implying that the decrease in the BDT temperature was not due to the decrease in the dislocation mobility controlled by short-range obstacles. Therefore, an alternative mechanism to decrease the BDT temperature is required to be considered, which has been discussed in the next section.

4. Discussion

In this section, we will discuss the mechanism behind the improvement in the fracture toughness at low temperatures, \(i.e.,\) the decrease in the BDT temperature with the grain size by the ARB process. Hodge et al.\(^{27}\) revealed a linear relation between ASTM grain-size numbers and reciprocal BDT temperatures in Fe-Ni alloys, indicating that the BDT temperature generally decreases with the grain size. We focus on the effect of grain boundaries on the decrease in the BDT temperature since the ARB process increases the volume fraction of grain boundaries in the specimen. The effect of dislocations introduced inside grains due to the ARB process is neglected in this paper, which will be discussed in a separated paper. We will not discuss the apparent fracture toughness at high temperatures since the experiment conducted in the present study was invalid in the case of fractures with large-scale yielding at the crack tip.

4.1 Dislocation pile-up against grain boundaries and fracture toughness

In the case of a grain boundary playing the role of only an obstacle against dislocation glide,\(^{23}\) the dislocations pile up against the grain boundary. This implies that grain refining should rather decrease the fracture toughness at low temper-
atures, i.e., increase the BDT temperature, which contradicts the fact that grain refining improves the fracture toughness at low temperatures, as observed in the Charpy impact tests and the present study.

Suppose \((i/C_0_1)\) dislocations had been emitted from the crack tip. Then, the critical stress intensity factor for the next dislocation to be emitted is given by the following equation in a two-dimensional approximation\(^{28}\):

\[
k_{\text{cr},j} = \frac{\mu b}{r_c} g(\theta) + \mu b \sum_{j=1}^{i-1} h \left( \frac{1}{\|r_j - r_c\|} \right),
\]

where the subscript \(i\) indicates the parameters of the \(i^{th}\) dislocation emitted from a crack tip, \(b\) and \(\mu\) represent the Burgers vector and shear modulus, respectively. \(\theta\) is the angle between the crack plane and the slip plane. \(r_c\) and \(r_j\) are the crack core size and the distance between the crack tip and \(j^{th}\) dislocation, respectively. The first term on the right-hand side contributes to that from the image force, and the second term does to that from dislocation-dislocation interaction. The dislocation-dislocation interaction term increases with the number of emitted dislocations; hence it becomes more difficult for the subsequent dislocations to be continuously emitted from the crack tip. In the presence of a grain boundary near the crack tip, some dislocations pile up against the grain boundary. Since the back stress of the pile-up dislocations inhibits the emitted dislocations from moving against the grain boundaries, it becomes difficult for the subsequent dislocations to continue emitting from the crack tip. This tendency becomes more significant when the grain size becomes smaller, i.e., materials tend to be more brittle as

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**Fig. 5** Temperature dependence of apparent fracture toughness from roll-bonded specimens. The solid symbols indicate that the crack tip plastic zone of the fractured specimen corresponded to small-scale yielding. The open symbols indicate that the crack tip plastic zone of the ruptured specimens corresponded to large-scale yielding.

**Fig. 6** Temperature dependence of apparent fracture toughness from fully annealed specimens. Open symbols indicate that the specimens reached 11% strain (the test rig’s limit) during the tests without fracture; these are classified as “ductile bending”. The arrows indicate that the values of the actual fracture toughness are higher than those plotted.
the grain size decreases. This contradicts the fact that grain refining decreases the BDT temperature. Therefore, other functions of the grain boundary are required to be discussed. In the next subsection, two functions of the grain boundary have been discussed: its role as a dislocation sink and source, and its role in accommodating stress due to the impinged dislocations via grain boundary shielding. Firstly, the role of the grain boundary as a dislocation sink and source is discussed.

### 4.2 The role of grain boundaries as sinks and sources

Figure 8 indicates the configurations of atoms around a crack under the application of a tensile stress perpendicular to the crack plane, as calculated by using molecular dynamics (MD). Σ5 symmetric tilt boundaries were set to be lying parallel to the crack plane. The embedded atom method for aluminum developed by Mishin et al.\(^{(29)}\) was used. Although the crystal structure was different from that of low carbon steel, the MD simulations revealed the role of the grain boundary the essence of which is considered to be independent of the crystal structures. Periodic conditions were applied at each boundary of the calculation box. The number of total atoms was set to be 731640. A tensile stress perpendicular to the crack plane was applied at a strain rate of \(8 \times 10^8 \text{s}^{-1}\) at 100 K. The details regarding the calculation condition are presented elsewhere.\(^{(30)}\) Figures 8(a), 8(b), 8(c) and (d) present images after the application of a tensile strain normal to the crack plane, \(\varepsilon_{zz}\), of 0.016, 0.021, 0.032 and 0.038, indicating that the dislocations were emitted from the crack tip with increasing \(\varepsilon_{zz}\). The first dislocation emitted from the crack tip that glided against the grain boundary and then was impinged against the grain boundary is shown in (b) and (c). Figure 8(e) illustrates an enlarged image of (c), indicating the dislocations reemitted from the grain boundary (the dashed arrows indicate the direction of the dislocation glide). Figure 8(f) presents an enlarged image of the dashed square in (d), indicating that the dislocation reemissions continued as \(\varepsilon_{zz}\) increased. Six dislocations were emitted on reaching \(\varepsilon_{zz} = 0.038\).

Thus, the MD calculations indicated that grain boundaries act not only as barriers for dislocation motion but also
of dislocations impinged against the grain boundary, as shown in (b), (c) and (d). The normal stress on the crack plane, \(\sigma_{zz}\), derived from \(\tau_{x'x'}\) was found to be compressive, indicating the formation of a shielding field at the crack tip. It is termed as ‘grain boundary shielding’.\(^{31}\) which is the essential mechanism required to understand the mechanism behind the decrease in the BDT temperature and enhancement in the fracture toughness of fine-grained materials at low temperatures.

5. Conclusion

The BDT behaviour was investigated in low carbon steel deformed by an ARB process. The BDT temperature was decreased by the ARB process. Arrhenius plots between the BDT temperatures and the strain rates indicated that the activation energy was not changed by the ARB process. This indicated that the decrease in the BDT temperature by grain refining was not due to the increase in the dislocation mobility for short-range obstacles. MD simulations revealed that dislocations emitted from a crack were impinged against grain boundaries and were reemitted as the strain increased as well as create a shielding field. It is therefore concluded that the decrease in the BDT temperature by grain-refining is due to not the increase in dislocation mobility controlled by short range obstacles but the increase in the number of dislocation sources.

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