The Effect of the Interaction between Dislocations and Hydrogen Around a Crack Tip on Hydrogen Embrittlement Under Cyclic Loading Condition*

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Abstract

For steels with lower yield stress, transgranular facet like fracture sometimes occurs under corrosive conditions. The occurrence mechanism of the transgranular facet like fracture has been analytically clarified under constant stress rate condition based on the concept of mechanical interaction between dislocations and hydrogen. In this paper, the analyses of the mechanical interaction between dislocations and hydrogen around a crack tip under cyclic loading condition were conducted to clarify the effect of stress wave forms on the feasibility of hydrogen embrittlement. This analysis showed the typical pile-up of dislocations occurs at the site of hydrogen similar as that under monotonous applied loading condition. Concerning the concentration of dislocations at the site of hydrogen atom, the dislocation pile-up intensity factor amplitude $\Delta A$ and power coefficient value of the singularity of dislocation density $n$ were defined. $\Delta A$ under fatigue condition was found to concern the feasibility of the occurrence of facet like fracture due to dislocation pile-up caused by the interaction between dislocations and hydrogen. Furthermore, the condition of Fast-Slow stress wave form was found to promote the facet like transgranular fracture as compared with that of Slow-Fast stress wave form.

Key words: Corrosion Fatigue, Dislocation Group Dynamics, Dislocation Pile-up Intensity Factor Amplitude, Hydrogen Embrittlement, Interaction between Dislocations and Hydrogen

1. Introduction

Since the hydrogen embrittlement was recognized, many researches about the hydrogen phenomenon have been continuously conducted. Nevertheless, hydrogen embrittlement for steel is still a remaining issue to be clarified in the industrial field. Concerning the embrittlement mechanism caused by hydrogen, many theories have been previously proposed such as Plannar-pressure theory, Decohesion theory, Cottrell-cloud theory and Hydrogen softening theory. These theories were proposed to describe hydrogen embrittlement mechanism corresponding to various environmental conditions and materials. However the definite fracture mechanisms of hydrogen embrittlement have not yet been clarified. Furthermore, in order to investigate the occurrence site of hydrogen embrittlement due to hydrogen concentration, analysis of hydrogen diffusion have been continuously conducted. Due to the remarkable progress of computer technology, hydrogen
diffusion behaviors have been calculated in detail based on computational science. For steels with higher yield stress, the maximum value of tensile hydrostatic stress around a crack becomes higher at the region of elastic-plastic boundary. Since the driving force of hydrogen diffusion and concentration is the gradient of hydrostatic stress, the behavior of hydrogen concentration at this region was found to become remarkable which results in intergranular cracking due to hydrogen (Delayed fracture). On the other hand, under corrosive condition, hydrogen embrittlement and anodic dissolution occur simultaneously. Therefore the dominant fracture mechanism depends on the sensitivity of each mechanisms based on the rate of hydrogen diffusion and anodic dissolution. The rate of hydrogen diffusion and anodic dissolution was numerically found to be dominated by the yield stress. On the other hand, for steels with lower yield stress, transgranular facet like fracture sometimes occurs under corrosive environmental conditions. Concerning the mechanism of transgranular cleavage fracture, on the basis of the analysis of dislocation group dynamics around a crack tip, dislocation free zone (DFZ) was noticed as a trigger point and cleavage fracture was found to occur at the end of DFZ due to inverse pile-up. It was in good agreement with experimental results of the relationship between trigger point and values of fracture toughness, which shows that facet like cleavage fracture is dominated by this mechanism. Based on this analysis mentioned above, the occurrence mechanisms of the transgranular facet like fracture was numerically clarified under the constant stress rate corrosive condition. For the materials with not so larger yield stress, since the rate of hydrogen diffusion and the dislocation velocity are sometimes competitive, the transgranular fracture was found to be sometimes caused at the site of inverse pile-up of dislocations due to its high stress concentration caused by the mechanical interaction between dislocations and hydrogen atoms.

On the other hand, the analysis of dislocation distribution behavior around a fatigue crack tip is important to clarify the characteristics of fatigue crack growth. Part of author previously proposed a theory of fatigue crack growth based on dislocation dynamics. After that, many fatigue crack growth theories based on dislocation dynamics have been proposed. Based on the analyses of dislocation dynamics under the cyclic load condition, temperature and load frequency dependences of fatigue crack growth rate have been theoretically derived, which is in good agreement with the experimental law of fatigue crack growth rate. Furthermore, taking account for the effects of multiple slip lines and strain hardening under cyclic loading, the theoretically proposed law was found to be quantitatively in good agreement with the experimental law. On the other hand, dynamic dislocation behaviors during unloading process and the effect of its behavior on the characteristics of fatigue crack growth rate have not yet been clarified. Especially, since the process of unloading is considered to promote the time dependent fracture, it is important to clarify this behavior under the corrosion fatigue conditions.

Therefore, in this paper, the analyses of dislocations dynamics behavior on the mechanical interaction between dislocations and hydrogen under cyclic loading condition were conducted, in order to investigate the dislocations pile-up behavior which results in transgranular facet like fracture under corrosive conditions.

2. Physical model and basic equations

Under the stress corrosion environmental conditions, dislocation emission from a crack tip occurs, which results in pre-plastic deformation and the preparation of the sites of active path where a chemical anodic dissolution is induced. Hydrogen is induced due to the chemical reaction as shown in Fig.1. The hydrogen diffuses toward the direction of the maximum hydrostatic stress field interacting with dislocations emitted from a stressed...
source around a crack tip. Previously, a corresponding physical model around a crack tip was proposed as shown in Fig.2(29). In this model, hydrogen moves toward the direction of the maximum triaxial hydrostatic stress field and concentrates at this site. Multiple slip bands are also originated from a crack tip(29).

![Chemical dissolution process around a crack tip](image1)

![Physical model of multiple slip bands originated from a crack tip](image2)

However from the viewpoint of analysis, this model is rather complex and it is difficult to conduct definite numerical analysis. Therefore, a simplification of this model was conducted to derive clear mechanical interaction between moving dislocations and locked hydrogen. The simplification by using approximated physical model of single slip line has been conducted(18),(30)-(33), and some experimental results such as, dynamic stress intensity factor(33) and fracture toughness(33) are well clarified based on this model. The approximated physical model was shown in Fig. 3(17). The physical model in Fig.3 is different from that in Fig.2, that is, for example, types of mode I and mode II, however these two dislocation models are mathematically identical. It was assured by theoretical analysis on the static dislocation groups model(34,35). In these analyses, the formula of stress intensity factor obtained by theoretical analyses based on the physical model was found to have qualitatively the same type of equations(34,35).

In this model a dislocation source, S is located at some finite distance of $x_s$ from a crack tip. In this model, the size of $x_s$ is taken as $x_s/a = 10^{-6}$, where $a$ is half of the crack length. Thus, $x_s$ amounts to order of $10^{-6}$mm in the case of the $a$ (half of the crack length) being the order of 1.0mm. In such case, $x_s$ corresponds to the order of the core cut off of the dislocation. Therefore, the source is regarded to be located at the crack tip from the viewpoint of continuum mechanics. The hydrogen was located at the site $(x_H, \rho)$, where $\rho$ is the range of Cottrell atmospheric distance between a solvent and a solute atom(36). Analyses were conducted for various value of $x_H$. On the basis of this model, the analyses of the mechanical interaction between crack tip stress field, dislocation groups and hydrogen were conducted, and the behavior of dislocation distribution was analyzed.
As the local stress distribution near a crack tip, the stress singularity near a crack tip was taken into account under a stress application as follows,

$$\tau_s(x, t) = \frac{\tau_q}{\sqrt{x} + 1},$$  \hspace{1cm} (1)

where $\tau$ is the increasing rate of stress application, $t$ is time and $x$ is the distance from the crack tip. The value of $\tau_s(x, t)$ approaches to applied stress $\tau_0$ with increase in $x$. Stress singularity of $1/\sqrt{x}$ becomes more eminent with decrease in $x$. Therefore, Eq.(1) is approximately valid as the macroscopic stress field around a crack tip. Cyclic loading conditions conducted by this analyses are shown in Table 1.

### Table 1: Applied loading stress conditions

<table>
<thead>
<tr>
<th>Stress shape</th>
<th>$t_0 : t_D$</th>
<th>$t$ [s] (cycle)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Symmetry</td>
<td>(a)</td>
<td>1:1</td>
</tr>
<tr>
<td>Slow-Fast</td>
<td>(b)</td>
<td>4:1</td>
</tr>
<tr>
<td>Fast-Slow</td>
<td>(c)</td>
<td>1:4</td>
</tr>
</tbody>
</table>

The relationship between effective stress exerted on moving dislocations emitted from a stressed source and the values of dislocation velocity are written for each individual dislocation in a coplanar array of dislocation groups by the following equation.

$$V_i = \frac{M \tau_{\text{eff}/i}^m}{\tau_{0}^* \rho (x_i - x_b)^2},$$  \hspace{1cm} (2)

where $V_i$ is the velocity of $i$th dislocation, $\tau_{\text{eff}/i}$ is the effective stress exerted on each individual dislocation in terms of applied shear stress, $m$ is a material constant and $i$ is the index number of each dislocation in order of emitting from a stressed source.

$M$ is given by

$$M = \frac{V_0 (1/\tau_{0}^*)^m}{\rho (V_0 - 1 cm/s)}$$  \hspace{1cm} (3)

where $\tau_{0}^*$ is specific shear stress required to move an isolated dislocation with a velocity of 1 cm/s; and $V_0 = 1$ cm/s.

In this analysis, the effective stress exerted on the $i$th dislocation is written by Eq. (4).

$$\tau_{\text{eff}/i} = \frac{\tau_0}{\sqrt{x_i} + 1} + \left[ A \sum_{i \neq j} \frac{1}{x_i - x_j} - A \sum_{i \neq j} \frac{1}{2x_i + \sqrt{x_i^2 + x_j^2}} \right] - \frac{2A^2 \rho |x_i - x_b|^2}{\left( (x_i - x_b)^2 + \rho^2 \right)^2},$$  \hspace{1cm} (4)

where
\[ A = \begin{cases} \mu b / 2\pi(1 - \nu) & \text{For edge dislocation} \\ \mu b / 2\pi & \text{For screw dislocation} \end{cases} \]

\( \mu \) is shear modulus \((7.943 \times 10^4 \text{[MPa]})\), \( \nu \) is Poisson’s ratio \((0.35)\), \( x_i \) is the position of the \( i \)th dislocation and \( A' \) is the constant \((0.339 \times 10^{-3} \text{[MPa m^2]} \) \((36)\) which represents the magnitude of the mechanical interaction force between hydrogen and dislocations.

The first term of the right hand side of Eq. (4) is macroscopic stress field around a crack tip by applied stress. The second term is the stress of mechanical interaction between dislocations in the dislocation arrays, and the third and fourth terms are the image force of dislocations by the free boundary of crack surface \((37)\). The fifth term is the stress of mechanical interaction between dislocations and hydrogen \((36)\).

The effective stress, \( \tau_{eff,i} \), exerted on a dislocation source is written as follows,

\[ \tau_{eff,i} = \tau_0 \left( \sqrt{\frac{a}{x_i} + 1} \right) + \left[ A \sum_{j=1}^{4} \frac{1}{x_i - x_j} - \frac{A}{2x_i} - 4 \sum_{j=1}^{4} \frac{1}{\sqrt{x_i x_j}} \right] \frac{2A'p(x_i - x_j)}{[(x_i - x_j)^2 + p^2]} \]  

(5)

In this analysis the non-dimensional variables were used as follows.

\[ s_i = x_i / l \quad \text{and} \quad s_a = a / l, \]

(6)

where \( l = 0.01 \text{mm} \). When \( \tau_{eff,i} \) equals to the source activation stress, \( \tau_s \), then a new mobile dislocation is introduced at the source. Material constants for pure iron \((38)\) (edge dislocation) are used, \( m \) is material constant \((3.0)\), \( \tau_0^* \) is specific shear stress required to move an isolated dislocation \((53.9 \text{[MPa]})\) and \( \mu \) is shear modulus \((7.943 \times 10^4 \text{[MPa]})\).

### 3. Results of analyses

#### 3.1 Dynamic behaviors of dislocation distribution for the case without hydrogen

In order to analyze the dynamic behavior of dislocation distribution under the cyclic loading condition, analysis without the interaction between dislocations and a hydrogen atom was conducted under symmetrical stress wave form condition, as shown in (a) of Table1. The illustration of dislocation distribution behavior was shown in Fig.4, which corresponds to the time sequential distribution of dislocations emitted from a stressed source around a crack tip. Dislocations emitted from a stressed source move on the slip line apart from the crack tip due to applied cyclic shearing stress, and the dislocation free zone (DFZ) was caused \((19)\). This behavior was shown by the analytical results of time sequential characteristics of dislocation distribution for each cycle as shown in Fig.5. A new dislocation was emitted from a stressed source around a crack tip when the applied stress \( \tau \) takes some specified value at each cycle. The relationship between the ratio of the applied stress balanced by \( \tau_{net} \) at the time of dislocation emission \((\tau_{emission} - \tau_{min})\) to the applied stress amplitude, \((\tau_{peak} - \tau_{min})\) and the number of emitted dislocations were shown in Fig.6. At the first load cycle, new dislocations were emitted when the applied stress took over 77% of the value of the stress amplitude \((\tau_{peak} - \tau_{min})\). Furthermore, the stress of dislocation emission increases with increase in the number of applied stress cycles. After the first load cycle, a new dislocation was emitted when the applied stress takes about the 90% of the value of stress amplitude. Due to the AFM observation of the crack growth, a new slip deformation was experimentally shown to occur only when the applied stress takes a higher value during loading process under fatigue condition \((39)\). Therefore, the analytical result obtained in this analysis was in good agreement with the experimental result \((39)\). This behavior was considered to be caused by the back stress due to the emitted dislocations in the slip line. Dislocations emitted at the first cycle remain around a crack tip, and the back stress against dislocation emission by these dislocations is exerted on the dislocation source.
3.2 The effect of stress wave form on the dynamic behaviors of dislocation distribution for the case with the mechanical interaction between dislocations and hydrogen

Taking account for the effect of the mechanical interaction between hydrogen atom induced by corrosive reaction and dislocations on the hydrogen embrittlement, the analyses were conducted under various cyclic applied stress wave form conditions. Time sequential characteristics of dislocation distribution under Slow-Fast(a) and First-Slow(b) conditions at the 4th applied stress cycle were shown in Figs.7 respectively. These results showed, the typical back flow of trailing dislocations toward the crack tip occurs under the Slow-Fast stress wave condition as shown in Fig.7 (a). This behavior was caused by back stress due to
preceding dislocations around a hydrogen atom during loading process which overcome the applied load, and it disappears when applied load approaches to the maximum value and preceding dislocations are released from the lock by hydrogen. On the other hand under the Fast-Slow stress wave condition, the back flow of dislocations does not occur which is different from that under the Slow-Fast stress wave condition as shown in Fig.7 (b). This will be due to the fact that under the Fast-Slow stress wave condition, stress increases rapidly to the maximum value which overcome the back flow stress of preceding dislocations. Therefore, dislocations emitted from a stressed source gradually approach to the lock part by hydrogen.

Fig. 7 Time sequential behavior of dislocation distribution around a crack tip with locked hydrogen under cyclic loading conditions: (a) Slow-Fast stress wave condition; (b) Fast-Slow stress wave condition

Comparison of dislocation density toward the site of hydrogen under the Slow-Fast stress wave form condition with that under the Fast-Slow stress wave form condition were shown in Fig.8. The characteristic of the singularity of dislocations pile-up against the hydrogen was shown in Fig.9. These results show that the relationship between the distribution of dislocation density $f(x)$ and the distance from the hydrogen were given by Eqs.(7) and (8) respectively.

$$f(x) = 8.8 \times \frac{l}{x^{0.52}} \quad \text{(Fast-Slow stress wave condition)} , \quad (7)$$

$$f(x) = 7.5 \times \frac{l}{x^{0.55}} \quad \text{(Slow-Fast stress wave condition)} . \quad (8)$$
According to Eqs. (7) and (8), \( f(x) \) was generalized as follows.

\[
f(x) = n A \cdot x^n.
\]

(9)

By calculating the Eq. (10) using the Eq. (9), dislocation pile-up intensity factor \( A \) is calculated. Where, \( A \) is a parameter which represents the intensity of dislocation pile-up, named dislocation pile-up intensity factor.

\[
\lim_{x \to 0} x^n f(x) = A.
\]

(10)

The effective factor on fatigue life is not the maximum stress \( \tau_{\text{max}} \) but the stress amplitude \( \Delta \tau = (\tau_{\text{max}} - \tau_{\text{min}}) \). Therefore instead of \( A \), \( \Delta A \) is considered to be an effective factor on dislocation concentration under cyclic loading, where \( \Delta A \) is defined as the difference between \( A \) and \( A_{\text{min}} \), that is \( A - A_{\text{min}} \) named the dislocation pile-up intensity factor amplitude. The time sequential characteristic of \( n \) and \( \Delta A \) was shown in Figs. 10 and 11 respectively. The values of \( n \) and \( \Delta A \) change during the process of stress loading and unloading respectively. Typical difference of the power coefficient value of the singularity of dislocation density \( n \) was not observed between Slow-Fast and Fast-Slow stress wave forms as shown in Fig.10. However, a typical difference of the dislocation pile-up intensity factor amplitude \( \Delta A \) was observed between these conditions as shown in Fig.11. Therefore, in order to clarify the effect of stress wave forms on inverse pile-up of dislocations which result in transgranular facet like fracture, the following parameter was proposed.

\[
TR = \frac{I_{\Delta A}}{I_A}
\]

(11)

The average estimated values of \( \Delta A \) are plotted against \( TR \) as shown in Fig.12. The difference of \( \Delta A \) between Slow-Fast and Fast-Slow stress wave form conditions was observed even under the same applied load amplitude and load frequency. Note that, typical pile-up of dislocations occurred under the condition of Fast-Slow stress wave form. This result corresponds to the possibility of occurrence of facet like fracture due to dislocation pile-up under Fast-Slow stress wave form condition. Actually, typical effect of applied stress wave form on corrosion fatigue crack growth rate (CFCGR) was observed for pure titanium\(^{40} \). The relationship between \( TR \) and CFCGR was shown in Fig.13\(^{40} \). For this material, the transgranular cleavage fracture is dominant which will be caused by typical pile-up of dislocations. This result is in good agreement with the corresponding analytical results.
Furthermore, the analyses of the number of dislocations emitted from a stressed source around a crack tip were conducted under various yield stress and stress wave form conditions. This behavior closely concerns the number of active path for anodic dissolution. The relationship between the number of dislocations emitted from a stressed source around a crack tip and the number of applied load cycles was shown in Fig.14 under various yield stress and stress wave form conditions. Furthermore, the number of dislocations emitted from a stressed source at specified load cycle is controlled by that of the first cycle, which is the increasing ratio of the number of dislocations to that of the first cycle, $\eta = n/n_1$. The relationship between $\eta$ and the dislocation free zone (DFZ) were shown in Fig.15, where $n_1$ is the number of dislocations emitted from a stressed source around a crack tip at the first applied load cycle. These results showed typical effect of applied stress wave form on the
number of dislocations emitted from a stressed source around a crack tip did not exist for pure iron with lower yield stress (ductile materials). However, the effect of applied stress wave form on the number of dislocation emission becomes remarkable for materials with higher yield stress (material with higher $\tau_0^*$) as shown in Fig.15. $\eta$ is plotted against TR in order to show the effect of applied stress wave form more clearly as shown in Fig.16, where $t_0$ is the applied load rising time, $t_D$ is the applied load descending time. These results show the effect of stress wave form on anodic dissolution was not so remarkable for materials with lower value of $\tau_0^*$ (ductile materials), however this effect becomes remarkable with increase in $\tau_0^*$ (brittle materials). According to the corrosion fatigue crack growth tests for 2.25Cr-1Mo steel, the effect of applied stress wave form on crack growth rate was observed(38) as shown in Fig.17. This result is in good agreement with the analytical result.

![Fig. 14 The number of dislocations emitted from a stressed source around a crack tip under various yield stress and stress wave form conditions](image1)

![Fig. 15 Relationship between the ratio of the number of dislocations emission from a stressed source of specified load cycles to that of the first cycle and the dislocation free zone under various yield stresses and stress wave form conditions](image2)

![Fig. 16 Effect of stress wave form on the number of dislocations emitted from a crack tip for various yield stress materials](image3)
4. Considerations

On the basis of these results, the following behaviors were considered to occur.

Concerning the concentration of dislocations at the site of hydrogen atom which related to facet like transgranular fracture for ductile materials under corrosive conditions, dislocation pile-up intensity factor amplitude $\Delta A$ and power coefficient value of the singularity of dislocation density $n$ were defined. The effect of stress wave form on $\Delta A$ was found to promote facet like fracture. Average value of $\Delta A$ under Fast-Slow stress wave condition was found to be larger than that under Slow-Fast stress wave condition, which is in good agreement with experimental results (40). This behavior will become dominant under the condition of intermediate values of yield stress between ductile materials with lower yield stress which is less than 400MPa and brittle materials with higher yield stress which is more than 1000MPa (12) and the facet like fracture was found to appear under corrosive conditions (12,16).

On the other hand, typical effect of applied stress wave form on the number of dislocations emitted from a stressed source around a crack tip which concerns the number of active path for anodic dissolution does not exist for materials with lower yield stress (ductile materials). However, the effect of applied stress wave form on the number of dislocation emission becomes remarkable for materials with higher yield stress. These results show the effect of stress wave form on anodic dissolution was not so remarkable for materials with lower value of $\tau_0$ (ductile materials), however this effect becomes remarkable with increase in the resistant stress of dislocation motion, $\tau_0^*$ which corresponds to the yield stress. It is promoted under Slow-Fast stress wave condition which is in good agreement with experimental results (41).

Therefore, as is shown in previous paper (17,20,42), the mechanisms of anodic dissolution, facet like fracture and hydrogen embrittlement related to hydrogen concentration were found to appear due to the competitive relationships between the rate of anodic dissolution, the interaction of dislocations and hydrogen, and hydrogen embrittlement related to hydrogen diffusion and concentration dominated by the competitive relationship between the rates of dislocation motion and hydrogen diffusion.

5. Conclusions

The analyses of the effect of mechanical interaction between dislocations and hydrogen around a crack tip on hydrogen embrittlement under cyclic loading condition were conducted, and the following results were obtained.

1) The dynamic behaviors of dislocations emitted from a stressed source around a crack tip during the loading and unloading process were clarified under cyclic loading condition. These results showed after the first load cycle, a new dislocation was
emitted from a stressed source when the applied stress takes about the 90% value of the stress amplitude ($\tau_{\text{peak}} - \tau_{\text{min}}$). Furthermore, the stress of dislocation emission gradually increases with increase in the number of applied stress cycles which corresponds to the experimental observation.

2) Typical pile-up of dislocations occurs at the site of hydrogen similar as the results under monotonous applied loading condition. On the other hand, the back flow of trailing dislocation toward the crack tip occurs under Slow-Fast stress wave condition which is different from that under Fast-Slow stress wave form conditions.

3) Concerning the concentration of dislocations at the site of hydrogen atom which related to facet like transgranular fracture for ductile materials under corrosive conditions, dislocation pile-up intensity factor amplitude $\Delta A$ and power coefficient value of the singularity of dislocation density $n$ were defined. The effect of stress wave form on $\Delta A$ was found to promote facet like fracture. Average value of $\Delta A$ under Fast-Slow stress wave condition was found to be larger than that under Slow-Fast stress wave condition, which is in good agreement with experimental results.

4) Typical effect of applied stress wave form on the number of dislocations emitted from a stressed source around a crack tip which concerns the number of active path for anodic dissolution does not exist for materials with lower yield stress (ductile materials). However, the effect of applied stress wave form on the number of dislocation emission becomes remarkable for materials with higher yield stress. These results show the effect of stress wave form on anodic dissolution was not so remarkable for materials with lower value of $\tau_0$ (ductile materials), however this effect becomes remarkable with increase in the resistant stress of dislocation motion, $\tau_0$ which corresponds to the yield stress. It is promoted under Slow-Fast stress wave condition which is in good agreement with experimental results.

5) For ductile materials with intermediate yield stress under corrosive condition, facet like fracture is found to occur due to the typical pile-up of dislocations. It will be promoted under Slow-Fast stress wave condition, which is in good agreement with experimental result. On the other hand under Slow-Fast stress wave form condition, anodic dissolution is found to become remarkable due to the dislocation emission which related to the formation of active path for anodic dissolution.

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