THE STRENGTH DETERIORATION OF STEEL ADHESIVE JOINT IN AQUEOUS ENVIRONMENT

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The strength deterioration of steel adhesive in aqueous environments was investigated. A model is proposed in which the process of progressive decrease in joint strength due to water uptake is considered as that of progressive growth of an apparent crack at the joint interface. It is found that the fracture strength after immersing the joint in water at various temperatures can be expressed as a function of single variable $D_t$.

INTRODUCTION

There are today strong demand to use of adhesive bonded joints in structures, however application of adhesive in structural bonding has been retarded due to strength drop of the joint caused by water uptake. (Brockmann(1), Adams (2), Bascom (3), Gledhill (4), Ripling (5) and Drain (6)),

In the present work, the process of decrease in bond strength of adhesive joints in aqueous environment is considered that the strength of adhesive itself does not suffer from the environmental attack but an equivalent apparent crack generates in the adhesive and grows gradually, resulting the decrease in bond strength of joint. Applying the fracture mechanics to the apparent crack, the decrease in bond strength of the joints due to water uptake is discussed.

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EXPERIMENTAL PROCEDURE

Cast sheet of the adhesive were prepared of thickness between 3.7 and 5mm and cut into 7 x 10mm rectangular plates. Also prepared is a sandwich specimen in which thin Fe foils are put into the resin as shown in Fig. 1 (b). These specimens were dried and weighed and immersed in distilled water of 30 – 50C. At various intervals a specimen was removed, dried of surface water, weighed on an balance and returned to the water.

Mild steel substrate, 7 x 10mm rectangular section and 20mm in length, was cut from hot roll plate. The surface to be bonded was degreased, grit blasted with 600 to 1000mesh abrasives, and then degreased again in an ultrasonic bath for 20min. Adhesive was applied on the treated surface and two blocks were pressed lightly together to form a butt joint with glue line thickness of about 0.02mm. Fig. 1 (c) shows the adhesive bonded specimen. The specimen was cured for 24hr at atmospheric temperature and then immersed in distilled water at 20, 30, 40 and 50C. After specified time elapsed, the specimen was removed from the water, and fractured in bending at atmospheric temperature.

EXPERIMENTAL RESULTS

Determination of Diffusion Constant

Let’s denote \( \Delta w \) the weight of water absorbed at time, \( t \), and \( w_0 \) the initial dry weight of the specimen. The ratio of \( \Delta w/w_0 \) to \( (\Delta w/w_0)_{\infty} \) is plotted against \( \sqrt{t/h} \) in Fig. 2, where \( (\Delta w/w_0)_{\infty} \) is the equilibrium value of \( \Delta w/w_0 \) and \( h \) is the thickness of the specimen. As can be seen in Fig. 2, the water uptake curves are liner at first, and the value of \( (\Delta w/w_0)/(\Delta w/w_0)_{\infty} \) are asymptotically approaching to the equilibrium value of unity. This type of diffusion is termed Fickian, and the diffusion constant, \( D \), can be calculated from the data shown in Fig. 2 using following equation, Crank (7),

\[
\frac{\Delta w/w_0}{(\Delta w/w_0)_{\infty}} = 1 - \sum_{n=0}^{\infty} \frac{8}{(2n+1)^{2}\pi^{2}} \exp \left[ -\frac{D(2n+1)^{2}\pi^{2}}{h^{2}} \right]
\]

Neglecting the higher order terms of the series, we have,

\[
D = \frac{0.0702}{(t/h)^{1/2}}
\]

where \( (t/h)^{1/2} \) is the half-time of the diffusion process. Calculated values of \( D \) are shown in Fig. 3 as a function of reciprocal of absolute temperature, which yields an activation energy of 52.2KJ/mol. And also obtained is a diffusion coefficient \( D_{0} \) of 20.4cm²/sec, and then,

\[
D = 20.4e^{-\frac{52200}{RT}}
\]
where \( R \) is the gas constant.

**Strength Deterioration in Water Environment**

The fracture strength, \( \sigma_t \), of the bonded steel specimens, after immersed in water, are plotted against the immersion time in Fig. 4. The strength increases slightly with increasing time at first, reaching the maximum value, \( \sigma_{\text{m}} \), and then decreases gradually with increasing time. This slight increase in strength observed in the initial part of the deterioration may be further progress in curving of the adhesive, and immersion time, \( t_p \), at which \( \sigma_t \) reaches its maximum strength at each temperature, is shorter the higher the temperature is. It may be plausible to consider that effective process of strength decrease takes place after \( t_p \).

Taking \( t_d = t-t_p \) and \((1-\sigma/\sigma_{\text{max}})\) on the abscissa and ordinate respectively, the data after \( t_p \) in Fig. 4 are reproduced in Fig. 5, where \( \sigma \) is the strength at time \( t_d \) and \((1-\sigma/\sigma_{\text{max}})\) is defined as the degree of strength deterioration. The average rate of deterioration, \( 1/t_d \) (subscript “T” indicate temperature) is shown in Fig. 6 as a function of temperature in the form of Arrhenius plot. The activation energy for strength deterioration process is calculated to be 56.7KJ/mol, coinciding favourably with value of 52.2KJ/mol, the activation energy for water diffusion in adhesive, \( Q_D \). It may be concluded that the deterioration of joint strength in water environment is a thermally activated process controled by diffusion, and we can write,

\[
\frac{1}{t_d} = \frac{1}{t_{do}} e^{-\frac{Q_D}{RT}} \tag{4}
\]

where \( t_{do} \) is a constant depending on \( \sigma/\sigma_{\text{max}} \). Fig. 7 shows relationship between \( 1/t_{do} \) and \( \sigma/\sigma_{\text{max}} \) and following expression is derived,

\[
\frac{1}{t_{do}} = 1.18 \times 10^5 e^{6.60(\sigma/\sigma_{\text{max}})} \tag{5}
\]

Substituting Eq. (5) into Eq. (4), we get,

\[
\frac{1}{t_d} = 1.18 \times 10^5 e^{6.60(\sigma/\sigma_{\text{max}})} e^{-\frac{Q_D}{RT}} \tag{6}
\]

**DISCUSSION**

The butt joint specimens were immersed in water baths at \( 20 \sim 50 \degree C \) for 3days and then fractured in three point bending at atmospheric temperature. The fracture strength observed is shown in Fig. 8 as a function of temperature, and the calculated curve deduced from Eq. (6) is also shown by a solid line. The fracture strength decreases with increasing temperature because the amount of water diffused into the adhesive during the same length of time of immersion is larger...
as the temperature is higher.

Let us assume that the strength of adhesive containing water more than some critical concentration \( C \) is so low that it cannot sustain any load, so that such a part of the adhesive near surface of the bonded joint is supposed to be a crack. Then the bonded specimen, which had been immersed in water, contains an apparent crack and the length of this crack corresponds to the depth of the part of adhesive containing water more than the critical concentration \( C_r \).

The critical length of the crack, \( c_r \), is determined using following equation in the case of a small crack, that is, \( c_r \ll h \),

\[
c_r = \frac{1}{\pi} \left( \frac{K_{IC}}{\sigma_f} \right)^2
\]

(7)

where \( K_{IC} \) is fracture toughness of adhesive joint. Taking \( K_{IC} = 2.76 \text{MPa}\sqrt{\text{m}} \), (8), into Eq. (7) and using respective value of \( \sigma_f \) shown in Fig. 8, the apparent crack length is calculated and shown in Fig. 9.

It is assumed that diffusion of water into the adhesive layer is Fickian, as it is in the case of bulk adhesive. The concentration, \( C \), is expressed as following equation (7),

\[
\frac{C}{C_0} = 1 - \frac{4}{\pi} \sum_{n=0}^{\infty} \frac{(-1)^n}{(2n+1)^2} \exp \left[ -D(2n+1)\pi^2 \frac{t}{h^2} \right] \cos \left[ (2n+1)\frac{\pi x}{h} \right]
\]

(8)

where \( C_0 \) is the equilibrium concentration and \( x = h - c \), the distance from the axis of the cross section of the specimen. Taking the values of \( c \), shown in Fig. 9 and \( t = 3 \text{days} = 2.6 \times 10^4 \text{ sec} \) into Eq. (8), the water concentration at the tip of the apparent crack was determined and plotted in Fig. 9 as a function of temperature. It should be noted that almost the same values of concentration are calculated at each crack tip, regardless of the length of apparent cracks. The average value of \( C/C_0 \) is determined to be 0.42. Thus its means that the higher the temperature the deeper the apparent crack, however, the water concentration at the tip of respective crack is always constant.

The loss of joint strength caused by water diffusion, then, may be evaluated as a crack growth problem, in which the crack growth process corresponds to the penetration of water attack. From Eq. (3) and Eq. (6) we have,

\[
0.173 e^{-6.60(\sigma/\sigma_{\text{max}})} = Dt
\]

(9)

Then it should be noted that the deterioration in strength of joints caused by water uptake can be expressed as a function of a single variable \( Dt \), that is, the strength of joints, after they had been suffered from water attack can be
described by a single curve shown in Fig. 10, where \( h/\pi \) is a parameter depending on joint geometry.

Values of joint strength can be calculated from Eq. (9) and the calculated ones are shown in Fig. 5 as the solid curves, with the experimentally obtained strength values shown as individual points. The agreement between the calculated and observed strength is very good.

CONCLUSION

The deterioration in strength of steel adhesive butt joints was investigated, and the main conclusions obtained are as follows:

1. The process of the strength drop in adhesive joints due to immersion into water is a thermally activated one controlled by diffusion of water in adhesive.

2. It can be considered that the strength of adhesive containing water more than a critical concentration is too low to sustain any load, so that, the joints, which had been exposed to aqueous environment, contain an apparent crack.

3. The strength of joint, after they had been exposed to a wide range of environmental exposure conditions, can be described by a single variable of \( D_t \).

SYMBOLS USED

- \( C \) = concentration (\%)
- \( c \) = crack length (m)
- \( D \) = diffusion constant (cm²/sec)
- \( h \) = thickness of the specimen (m, mm)
- \( K_{IC} \) = fracture toughness (MPa \( \sqrt{m} \))
- \( T \) = temperature (°C, °K)
- \( t \) = time (sec)
- \( w \) = weight of specimen (gr)
- \( x \) = distance from the neutral axis (m)
- \( \sigma \) = stress (MPa)

REFERENCES

Fig. 1 Size and Dimension of Specimens. Fig. 3 Diffusion constant against reciprocal of absolute temperature.

Fig. 2 Rate of water uptake plotted against $\sqrt{t}/h$. 
Fig. 4 Three point bending strength against immersion

time in distilled water.

\[
0.173 e^{-6.60(D_{\text{td}}/D_{\text{max}})} = D_t
\]

Fig. 5 The strength deterioration in the distilled water is

plotted as a function of time \( t_d \).
Fig. 6  The rate of strength deterioration is plotted as a function of temperature.

Fig. 7  $1/t_{do}$ against $\sigma/\sigma_{max}$.

Fig. 8  Strength of joint immersed in distilled water for 3 days.

Fig. 9  Apparent crack length and water concentration at the apparent crack tip are plotted as a function of water temperature.

Fig. 10  Strength deterioration against normalized time $\sqrt{\Delta t}/(h/\pi)$. 

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