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Effect of Residual Hydrogen Content on Cold Crack Susceptibility in Weld Metal of High Strength Steels†

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Abstract

Recently, the lower critical stress in the Implant test of high strength steel has been approximately correlated with residual hydrogen content at temperature 100°C, which seemes to relate with HAZ cracking more closely than initial hydrogen content, independently of initial hydrogen content and preheating temperature.

In this investigation, the residual hydrogen content at 100°C was measured by means of gas chromatograph in the LB-TRC test specimen in which the susceptibility of weld metal cold cracking could be quantitatively evaluated, and the relationship between the residual hydrogen content and the lower critical stress in the LB-TRC test was investigated using HT60, HT80, HY130 and HY180.

Consequently, the experimental equations between $(\Sigma D \triangle t)_{100}$ and the residual hydrogen content of all materials used were deduced, respectively. The lower critical stress strongly depended on the residual hydrogen content at 100°C in weld metal than the total amount of residual hydrogen content at 100°C.

KEY WORDS: (Cold Cracking) (Weld Metal) (High Strength) (GTA Welding) (Weldability Test) (Hydrogen)

1. Introduction

It is well known that cold cracking occurring in HAZ or weld metal in high strength steels is attributed to diffusible hydrogen. Therefore measurement of diffusible hydrogen content in weld zone is important and usually done by JIS or IIW method in which the specimen quenched immediately after completion of welding is utilized. However, the hydrogen content in this method actually corresponds to that at high temperature.

On the other hand, it is said that cold cracking occurs at temperatures below about 100°C. Therefore a considerable amount of hydrogen is evolved from weldment during cooling down to 100°C, which is depending upon specimen size, preheating temperature, weld heat input, etc. Consequently, it is reasonably considered that cold crack susceptibility should be evaluated by the residual hydrogen content in weldment at 100°C instead of the hydrogen content at high temperature measured by JIS or IIW method. This consideration is in accord with the result that the lower critical stress in the Implant test of high strength steel approximately depends on the residual hydrogen content at 100°C independently of initial hydrogen content and preheating temperature[1].

It still remains a great problem that cold cracking within weld metal tends to occur in weldments of high and ultra high strength steels, especially in HY-type steels, of more than 80 kgf/mm² in ultimate strength level. In the previous report[2], for the purpose of investigation on the susceptibility of weld metal cold cracking of high strength steel, a new testing method named the LB-TRC (Longitudinal Bead-Tensile Restraint Cracking) test was developed by the authors.

In the LB-TRC test, it is a matter of interest to reveal whether the lower critical stress depends on the residual hydrogen content at 100°C or not. Moreover, it seems important for analysis of cracking mechanism that the effect of chemical compositions on the residual hydrogen content in weldment is revealed, since alloying elements such as Ni, Cr, Mo, Co and so on in steel increase with an increase in strength level in HY-type steels.

Therefore in this investigation the residual hydrogen content at 100°C was measured in the LB-TRC test specimen, and the relationship between the residual hydrogen content and the lower critical stress was investigated using commercial high strength steels of 60 (HT60) and 80 kgf/mm² (HT80) in ultimate strength levels and HY130 and HY180 steels.

2. Review of Study on Residual Hydrogen Content

Residual hydrogen concentration in weldment was first theoretically calculated by Fujii[3] and Chano et al.[4], solving a diffusion equation under iso-thermal condition. It was revealed that the residual hydrogen concentration depended on the term $\int Ddt$ (D: diffusivity coefficient, t: time). Factor $\Sigma D\Delta t$ (Δt: time interval) practically was substituted for the term $\int Ddt$. In Fuji's report, the critical residual hydrogen concentration at 100°C on weld bond for avoidance cracking was calculated to Tekken test utilized at various preheating temperatures. However, such analysis is practically complicated in spite of a simple diffusion model and it has great difficulties to apply it to complex geometry.

On the other hand, it was revealed by Terasaki et al.[1] that the residual hydrogen content of weldment in the Implant test could be well estimated from calculation of $\Sigma D\Delta t$ during cooling after welding and the lower critical stress in the Implant test was also related to residual hydrogen content at 100°C. However, the hydrogen content was measured by modified JIS method using mercury as a confining liquid, so that it had great difficulties to measure the residual hydrogen content at 100°C accurately. Accordingly, the relationship between $\Sigma D\Delta t$ and the residual hydrogen content was obtained by measuring the residual hydrogen content at 200°C. Then, this correlation at 200°C was applied to estimate the residual hydrogen content at 100°C.

Moreover, in the above investigation, cold crack susceptivility was related to the residual hydrogen content contained in weld zone and base metal. However, amount of hydrogen which has diffused from HAZ to base metal is assumed to be little or no influence on HAZ cracking. Especially, in the case of cold cracking within weld metal, the effect of hydrogen which has diffused from weld metal to HAZ and base metal on cracking may be much less in influence.

Recently, gas chromatograph method has been developed for measurement of hydrogen content. According to this method, it is possible to measure the hydrogen content much accurately without matters of individual error in reading and without trouble of small bubbles occurring in JIS or IIW method. Accordingly, the residual hydrogen content at 100°C can be measured easily and then the effect of chemical compositions on the residual hydrogen content measured at 100°C in the LB-TRC test specimen was investigated. Then, the lower critical stress in the LB-TRC test was correlated with the residual hydrogen content at 100°C.

Hereafter, the residual hydrogen content at 100°C which is represented in ml per 100 g of fused metal and $\Sigma D\Delta t$ at 100°C are designated $(H_R)_{100}$ and $(\Sigma D\Delta t)_{100}$, respectively.

3. Materials Used and Experimental Procedures

3.1 Materials used

Base metals used were HT60, HT80, HY130 and HY180 steels, which were weldable heat-treated high strength steels whose nominal ultimate strengths were 60, 80, 100 and 140 kgf/mm², respectively. Among these, HY130 and HY180 steels had exceptionally high toughness in low temperature range.

Filler wires for automatical GTA welding were F60, F80, F(130) and F(180) which were well matched with base metals in strength levels, respectively. F60 and F80 were commercial filler wires and the others were experimental ones. The chemical compositions of these base metals and filler wires are shown in Table 1.

3.2 The LB-TRC test

The LB-TRC test has been developed to evaluate the susceptibility of weld metal cold cracking. Testing method has been described in detail elsewhere[2]. Briefly

Table 1   Chemical compositions of base metal and filler wire used

Material		Thickness (mm)	Diameter (mm)	Chemical composition (wt%)									
				C	Si	Mn	P	S	Cu	Ni	Cr	Mo	Co
Base metal	HT60	35	—	0.13	0.34	1.33	0.019	0.008	—	0.02	0.02	0.21	—
	HT80	25	—	0.13	0.27	0.86	0.011	0.004	0.25	1.08	0.50	0.43	—
	HY130	35	—	0.08	0.21	0.78	0.005	0.001	0.03	5.20	0.48	0.53	—
	HY180	22	—	0.11	0.13	0.17	0.006	0.003	—	10.40	1.88	1.01	8.05
Filler wire	F60	—	1.6	0.07	0.51	1.80	0.008	<0.005	0.01	0.03	0.02	0.31	—
	F80	—	1.6	0.09	0.52	1.57	0.007	<0.005	0.04	2.60	0.05	0.52	—
	F(130)	—	1.6	0.10	0.36	1.72	0.003	<0.005	0.01	2.46	1.19	0.67	—
	F(180)	—	1.6	0.11	0.16	0.09	0.004	0.005	—	10.0	2.10	1.10	8.20

describing, it essentially consists of two plates with U-groove butted together without gap to provide a slit across which a test bead is laid as shown in **Fig. 1**, and then a constant tensile load is applied parrallel to the weld line at about 150°C of weld metal after the completion of welding with or without preheating.

In this study, test welds were deposited about 40 mm in bead length with an automatic GTA welding with commercially pure argon and argon-hydrogen ($Ar + H_2$) mixed shielding gas and constant wire feed system utilizing 300A, 14V, 120 mm/min. Hydrogen gas content was changed to 0.35, 1.0 and 2.0% in volume. Total gas flow rate was 20 l/min. These mixing gases were precisely obtained through gas mixing chamber. The preheating temperature of specimen was 75 and 125°C. Then, the lower critical stress of weld metal (σ_{cr}) were evaluated.

3.3 Measurement of residual hydrogen content in the LB-TRC test specimen

Essentially, $(H_R)_{100}$ in the LB-TRC test specimen should be measured using the LB-TRC test specimen itself. However, the length of that specimen as shown in Fig. 1 is too long to be inserted itself into the vessel of gas chromatograph apparatus for collecting hydrogen.

Then, the small-sized specimen which had the same groove geometry of the LB-TRC test specimen was adopted for convenience for measuring $(H_R)_{100}$, as shown in **Fig. 2**. The hydrogen which evolved from side and bottom surface during cooling in this specimen was
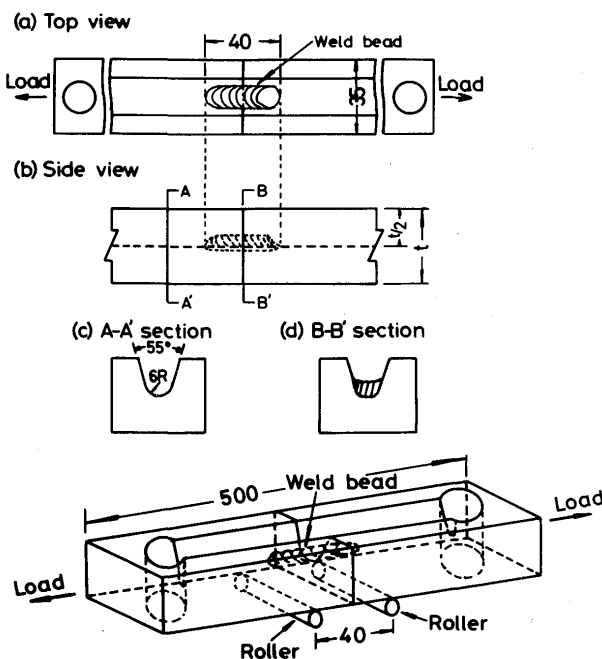
judged to be negligible from estimation of a simple calculation.

The values of $(H_R)_{100}$ using this specimen were measured as follows;

Test bead on the small-sized specimen was laid by means of the same conditions in the LB-TRC test. As soon as the specimen cooled down to 100°C at the butted location after the completion of welding, the specimen was inserted into the vessel for collecting hydrogen after quenching into ice water. After extraction for 48 hrs'. duration at 45°C, hydrogen content was measured by means of gas chromatograph, of which the measurement accuracy was 0.01 ml. Moreover, the cooling rates of specimen after welding were changed using 3 methods (Method 1, 2, 3), as shown in **Fig. 3**. Besides in Method 4 the specimen was quenched rapidly after completion of welding at about 650°C, in order to measure the total hydrogen content originally involved. Hydrogen content was represented in ml per 100 g of fused metal under the condition of the normal state (0°C, 1 atm). Each measurement was repeated for 4 to 8 times under the same condition and then hydrogen content was represented as the average of those results.
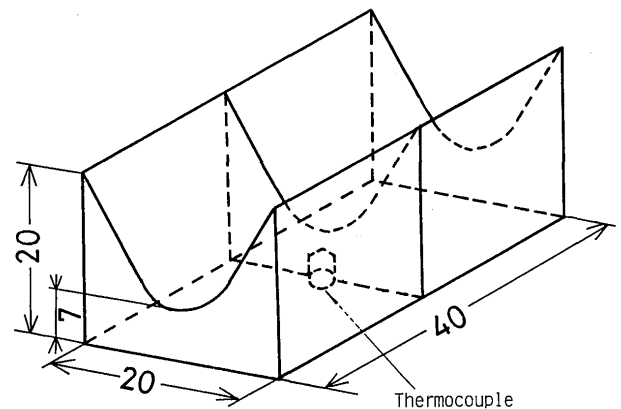

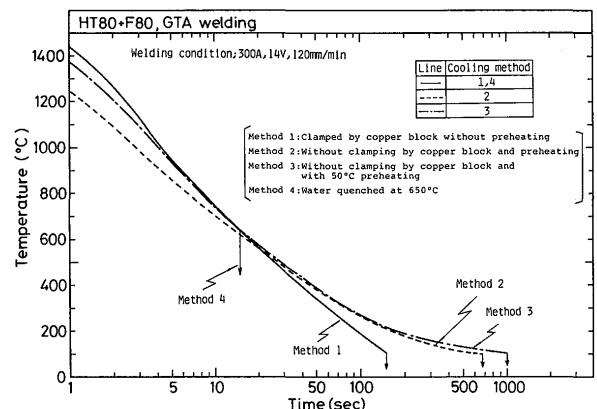**Fig. 2** Small-sized specimen for measurement of residual hydrogen content

Fig. 3 Welding thermal cycles of various cooling methods

Fig. 1 LB-TRC test specimen and testing method

On the other hand, in order to estimate the availability of the above method, $(H_R)_{100}$ in the LB-TRC test specimen of standard size for HT80 steel was measured and compared with that in the small-sized specimen, using the same technique with the exception that the standard specimen was cut off to small size to put into the collector vessel, immersing in liquid nitrogen to prevent evolution of hydrogen. The measurement was done under two methods in which specimens were quenched immediately after completion of welding and quenched after cooling down to 100°C at butted location.

In **Fig. 4** the data of the comparative method are shown in different mark with Ar + 2%H_2 shielding gas. The value of $(H_R)_{100}$ in small-sized specimen shows a good accordance to that in the LB-TRC test specimen at the same $(\Sigma D\Delta t)_{100}$. This result indicates that $(H_R)_{100}$ in small-sized specimen is regarded as that in LB-TRC test specimen itself. Then, $(H_R)_{100}$ in all materials used were measured using this small-sized specimen.

3.4   Calculation of values for $\Sigma D\Delta t$

The values of $(\Sigma D\Delta t)_{100}$ in small-sized specimen were calculated from cooling curves as shown in Fig. 3. These cooling curves were divided into the steps of the temperature ranges which were each 100°C from 1500 to 800°C because of short holding time and were each 25°C from 800 to 100°C, and then Δt were time intervals corresponding to these temperature ranges. Moreover, the value of the mean diffusivity coefficients of hydrogen for each small temperature range was adopted as D.

Diffusivity coefficient, D, essentially depends on materials. However, D of materials used in this work were unknown, so that D in pure iron was selected here for convenience regardless of materials. It was given by

$$D_1 = 1.51 \times 10^{-2} \exp(-11970/RT) \quad \cdots (1.\text{a})$$
$$(773 \leq T)$$

$$D_2 = 1.4 \times 10^{-3} \exp(-3200/RT) \quad \cdots (1.\text{b})$$
$$(473 \leq T < 773)$$

$$D_3 = 0.12 \exp(-7820/RT) \quad \cdots (1.\text{c})$$
$$(373 \leq T < 473)$$

D : diffusivity coefficient (cm^2/sec)

T : absolute temperature (°K)

R : gas constant (1.986 cal/mol°K)

Equation (1. a) was obtained by Sykes et al.[5], and Eq. (1. b) and Eq. (1. c) by Johnson and Hill[6]. Transformation from austenite was assumed to occur at 500°C for all materials used.

The value of $(\Sigma D\Delta t)_{100}$ at butted location in the LB-TRC test specimen was calculated from the above technique.
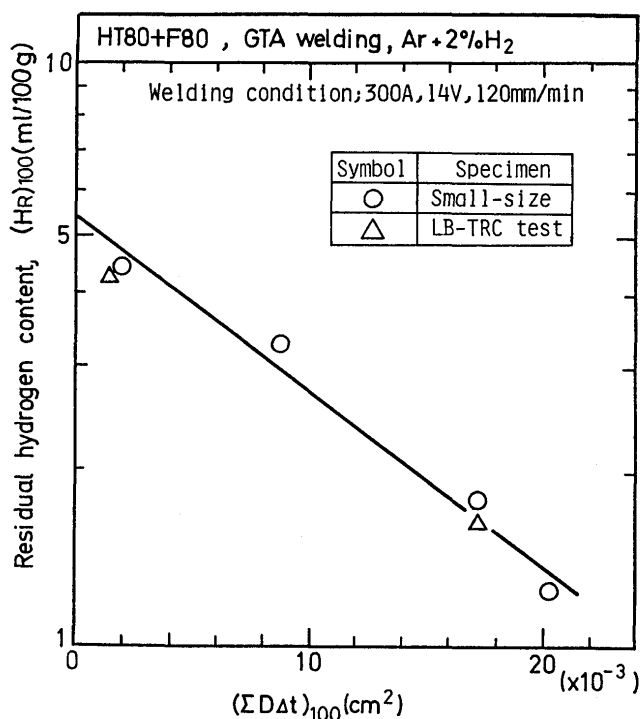

Fig. 4   Comparison of residual hydrogen content at 100°C in the LB-TRC test specimen and small-sized specimen
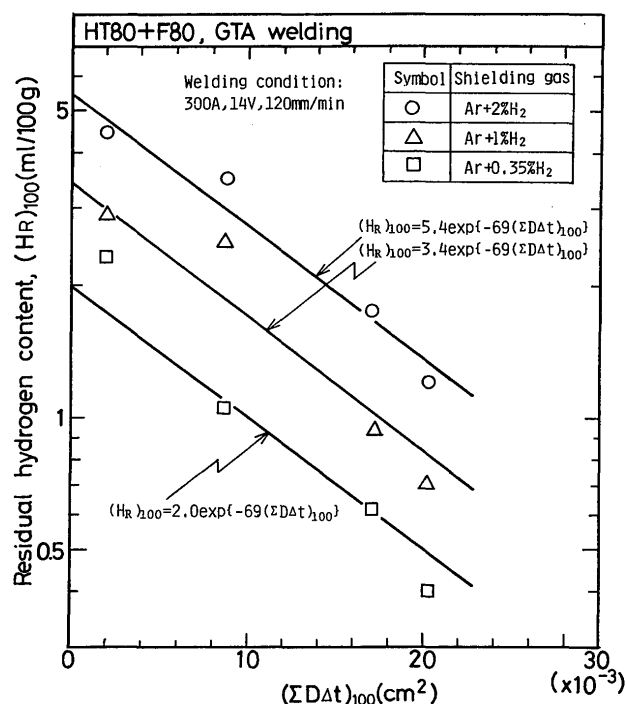

Fig. 5   Relationship between $(\Sigma D\Delta t)_{100}$ and the residual hydrogen content at 100°C with various initial hydrogen content in HT80 weldment

4. Result and Discussion

4.1 Residual hydrogen content in the LB-TRC test

Figure 5 shows the relationship between $(\Sigma D\Delta t)_{100}$ and $(H_R)_{100}$ in HT80 weldment with $Ar + 0.35\%H_2$, $Ar + 1\%H_2$ and $Ar + 2\%H_2$ shielding gases. By the regression analysis of the data, $(H_R)_{100}$ is expressed as a function of $(\Sigma D\Delta t)_{100}$ independently of initial hydrogen content given by

$$(H_R)_{100} = H_0 \exp \{ -69 (\Sigma D\Delta t)_{100} \} \ldots \ldots (2)$$

where H_0 is initial hydrogen content which corresponds to the residual hydrogen content at zero of $(\Sigma D\Delta t)_{100}$. The values of H_0 are obtained by extrapolating from each line in Fig. 5.

Figure 6 shows the relationship between $(\Sigma D\Delta t)_{100}$ and the dimensionless parameter of the residual hydrogen content, $(H_R)_{100}/H_0$, in all materials used. These relationships are expressed as the following equations.

For HT60

$$(H_R)_{100}/H_0 = \exp \{ -83 (\Sigma D\Delta t)_{100} \} \ldots (3.a)$$

For HT80

$$(H_R)_{100}/H_0 = \exp \{ -69 (\Sigma D\Delta t)_{100} \} \ldots (3.b)$$

For HY130

$$(H_R)_{100}/H_0 = \exp \{ -46 (\Sigma D\Delta t)_{100} \} \ldots (3.c)$$

For HY180

$$(H_R)_{100}/H_0 = \exp \{ -41 (\Sigma D\Delta t)_{100} \} \ldots (3.d)$$

These equations show that the coefficient of $(\Sigma D\Delta t)_{100}$ increases with an increase in strength of the material. This means that hydrogen in weldment tends to evolve easily with a decrease in strength of material and also alloying element. If the true D for each material is experimentally decided in future, these four relationship will be considered to be coincided as one simple relation.

4.2 Relationship between residual hydrogen content and lower critical stress of weld metal

The values of $(H_R)_{100}$ in the LB-TRC test for all materials used were calculated from Eq. (3) using $(\Sigma D\Delta t)_{100}$ measured from cooling curve. Then, the relationship between the calculated $(H_R)_{100}$ and the lower critical stress (σ_{cr}) in the LB-TRC test was investigated. These relationships in all materials used are shown in **Fig. 7 (a), (b), (c)** and **(d)**. The lower critical stress decreases with an increase in $(H_R)_{100}$ and increases with an increase in preheating temperature at the same $(H_R)_{100}$ in all materials.

Therefore the reason why σ_{cr} does not depend on $(H_R)_{100}$ regardless of preheating is considered as follows;

One-dimensional schematic illustration of hydrogen distribution is qualitatively shown in **Fig. 8** whose abscissa represents inward distance from bead surface to base metal, where $X < l_0$ shows weld bead and $X > l_0$ shows HAZ and base metal, and ordinate represents hydrogen concentration. Consider two cases of different initial hydrogen concentration in weld metal represented in $C_0 1$ and $C_0 2$. Moreover, it is assumed that preheating is applied in weld metal of $C_0 2$ hydrogen concentration and is not applied in that of $C_0 1$. Therefore after different lapse of time two hydrogen distributions in specimens at the same $100°C$ change to Curve 1 (from $C_0 1$) and Curve 2 (from $C_0 2$), respectively, of which areas under these curves are the same. Of course specimen of Curve 2 has longer lapse of time and larger $(\Sigma D\Delta t)_{100}$ than that of Curve 1. Although the residual hydrogen content $(H_R)_{100}$ is the same in these curves, it is expected that hydrogen content contributed to cause cracking in weld metal is much more in Curve 1 than in Curve 2. This is considered as one of reasons for the results in Fig. 7. Thus, it seems necessary to consider the shape of hydrogen distribution of $(H_R)_{100}$ in the LB-TRC test, because cold cracking occurred in weld metal in this test. The residual hydrogen content in weld metal $((H_{RW})_{100})$ is adopted as the parameter in which the distribution of $(H_R)_{100}$ is considered. The value of $(H_{RW})_{100}$ is defined
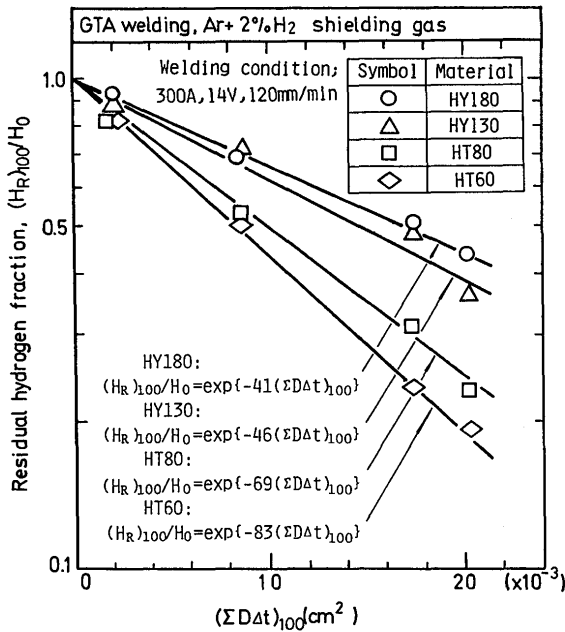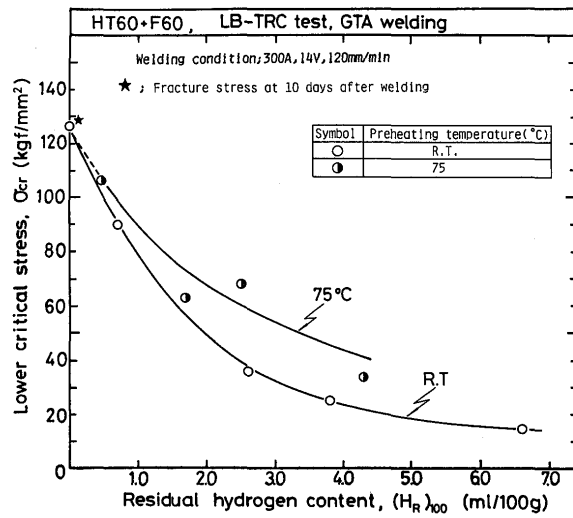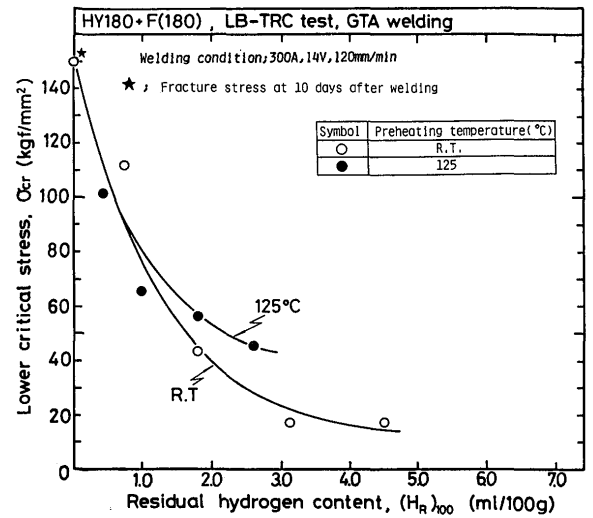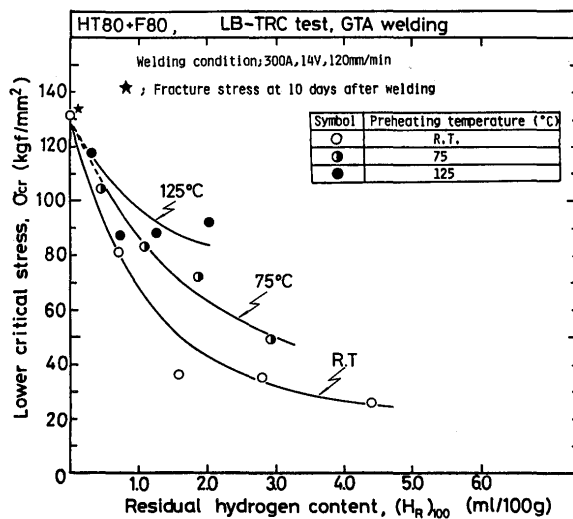

Fig. 6 Relationship between $(\Sigma D\Delta t)_{100}$ and the ratio of the residual hydrogen content at $100°C$ to the initial hydrogen content in all materials used (HT60, HT80, HY130, HY180)
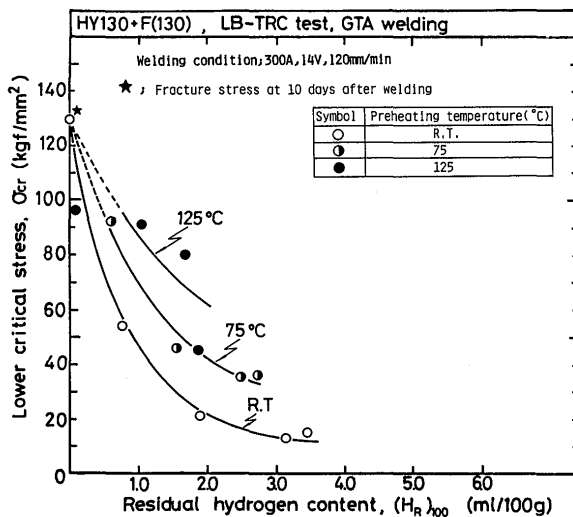
(a) HT60 + F60

(b) HT80 + F80

(c) HY130 + F(130)

(d) HY180 + F(180)

Fig. 7 Effect of the residual hydrogen content at 100°C on the lower critical stress in the LB-TRC test
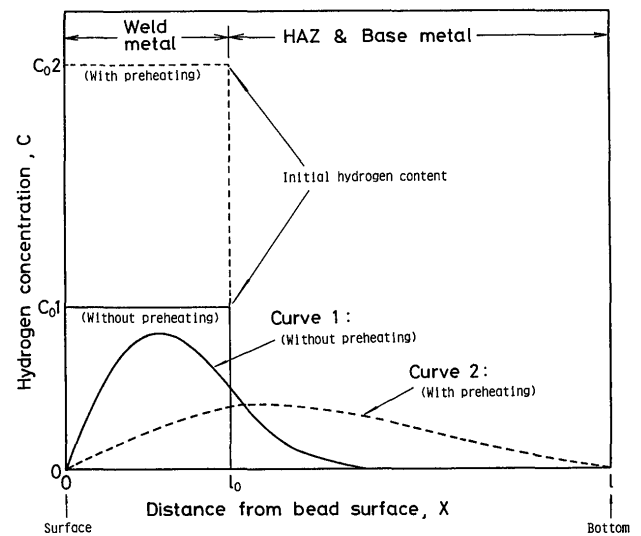

Fig. 8 Qualitative hydrogen distributions at 100°C in thickness direction under the same residual hydrogen content at 100°C in cases of preheating and no preheating

by

$$(H_{RW})_{100} = R_W \times (H_R)_{100}$$

$$(R_W = \int_0^{l_o} Cdx \Big/ \int_0^l Cdx) \quad \ldots \ldots \quad (4)$$

where R_W is calculated using analytical solutions of one-dimensional diffusion equation obtained by Fuji et al.[3] Calculation of R_W is explained in Appendix.
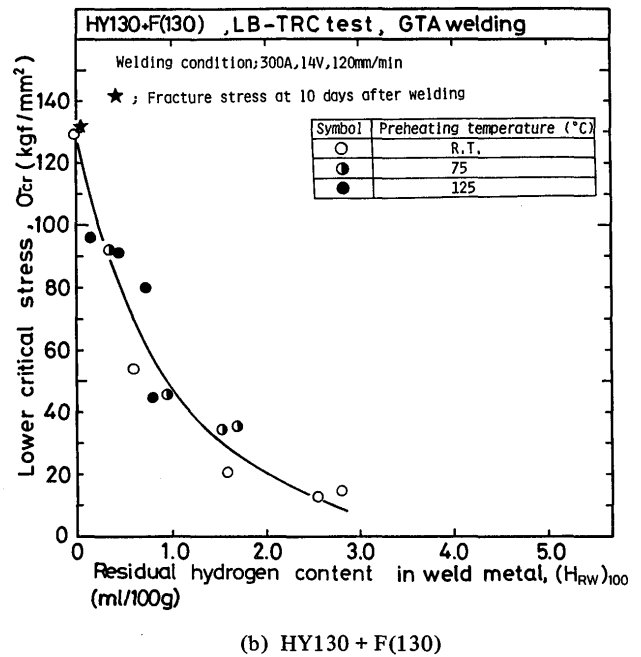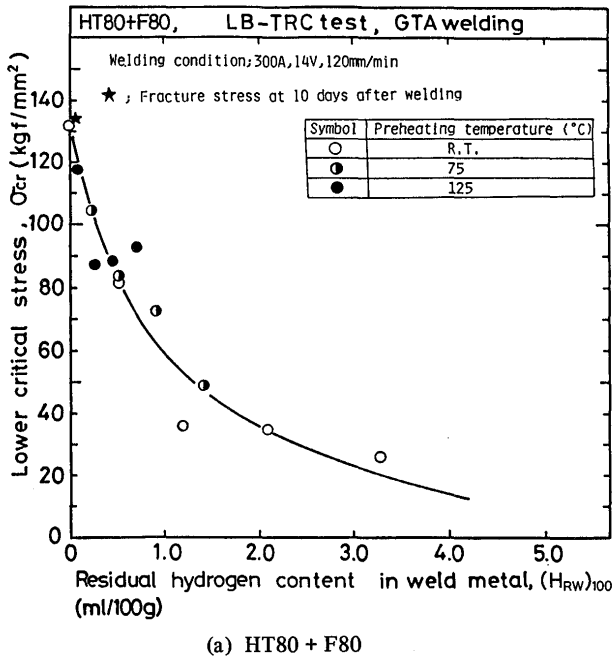
(a) HT80 + F80

(b) HY130 + F(130)

Fig. 9 Relationship between the residual hydrogen content in weld metal at 100°C and the lower critical stress in the LB-TRC test
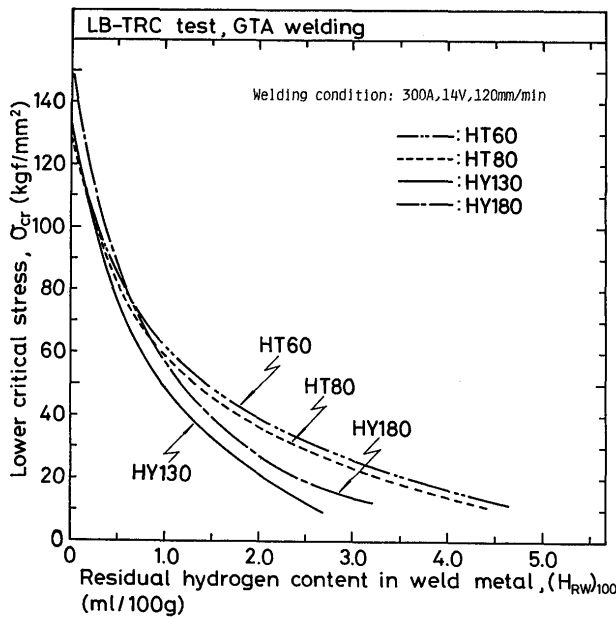

Fig. 10 Relationships between the residual hydrogen content in weld metal at 100°C and the lower critical stress in all materials used (HT60, HT80, HY130, HY180)

Figure 9 (a) and (b) show the relationship between $(H_{RW})_{100}$ and σ_{cr} in HT80 and HY130 steels. These results indicate that regardless of preheating temperature σ_{cr} is closely related to $(H_{RW})_{100}$ and better than to total residual hydrogen $(H_R)_{100}$. Moreover the same relations are seen in the other steels. Consequently, it is

shown that the lower critical stress in the LB-TRC test is correlated well with the residual hydrogen content within weld metal.

The relationships between $(H_{RW})_{100}$ and σ_{cr} in all materials used are collectively summarized in **Fig. 10**. From Fig. 10, the lower critical stress, σ_{cr}, decreases in order of HT60, HT80 and HY130 under the same $(H_{RW})_{100}$ and this tendency coincides with the reverse of strength levels which has been widely known. However, σ_{cr} in HY180 steel is a little higher than that in HY130 steel, the reason of which is not revealed at this moment.

5. Conclusions

The residual hydrogen content at 100°C $((H_R)_{100})$ in the LB-TRC test for high strength steels of HT60, HT80, HY130 and HY180 was measured using gas chromatograph method. Moreover, the relationship between $(H_R)_{100}$ and the lower critical stress (σ_{cr}) in the LB-TRC test, and the relationship between the residual hydrogen content in weld metal $(H_{RW})_{100}$ and σ_{cr} were investigated. Main conclusions are summarized as follows:

(1) The values of $(H_R)_{100}$ in all materials used are related to $(\Sigma D\Delta t)_{100}$ given by

For HT60

$$(H_R)_{100}/H_0 = \exp\{-83\,(\Sigma D\Delta t)_{100}\}$$

For HT80

$$(H_R)_{100}/H_0 = \exp\{-69\,(\Sigma D\Delta t)_{100}\}$$

For HY130

$$(H_R)_{100}/H_0 = \exp\{-46\,(\Sigma D\Delta t)_{100}\}$$

For HY180

$$(H_R)_{100}/H_0 = \exp\{-41\,(\Sigma D\Delta t)_{100}\}$$

where H_0 is initial hydrogen content and D is diffusivity coefficient given by Sykes et al. and Johnson and Hill. It is considered that the difference in coefficient of $(\Sigma D\Delta t)_{100}$ is caused by the difference in diffusivity coefficient of material.

(2) The lower critical stress, σ_{cr}, is not related to $(H_R)_{100}$, because σ_{cr} increases with an increase in preheating temperature at the same $(H_R)_{100}$.

(3) Hydrogen content which has diffused from weld metal to HAZ and base metal is considered to have little effect on weld metal cold cracking, so that $(H_{RW})_{100}$ is calculated from $(H_R)_{100}$ using some assumptions. Consequently, σ_{cr} of weld metal of various steels in the LB-TRC test approximately depends on $(H_{RW})_{100}$ regardless of preheating temperature. From these results it is clearly defined that weld metal cracking strongly depends on $(H_R)_{100}$ rather than $(H_R)_{100}$.

Acknowledgement

References

1) T. Terasaki et al. : Trans. JWS, Vol. 10 (1979), No. 1, pp. 53-57.

2) F. Mtsuda et al.: Trans. JWRI, Vol. 8 (1979), No. 1, pp. 113-119

3) T. Fujii: Abstracts JWS Spring Meeting, No. 22 (1978), April, pp. 50-53 (in Japanese)

4) F. R. Coe, Z. Chano: Res. Report Weld. Inst., 1973, November

5) C. Sykes, et al.: J. Iron Steel Inst., 1947, No. 6, pp. 155-180

6) E.W. Johnson, M.L. Hill: Trans. Met. Soc. of AIME, Vol. 218 (1960), No. 12, pp. 1104-1111

Appendix   Calculation of R_W

Figure A-1 shows the relationship between τ (= $\Sigma D\Delta t/l_0^2$) and R_W. When the calculation is practiced using Fig. A-1, D in $\Sigma D\Delta t/l_0^2$ of abscissa should be used the value of each material. However, as already described in 3.4, D in each material is unknown, so that $(\Sigma D\Delta t)_{100}$ in $\Sigma D\Delta t/l_0^2$ is rectified as follows;

In equation (3), the coefficient of each $(\Sigma D\Delta t)_{100}$ depends on the material. However, if $(\Sigma D\Delta t)_{100}$ is calculated using true D in each material, this coefficient essentially must be constant independently of material. Now, HT60 contains the fewest alloying element of all materials used, so D in HT60 seems to be the closest to D
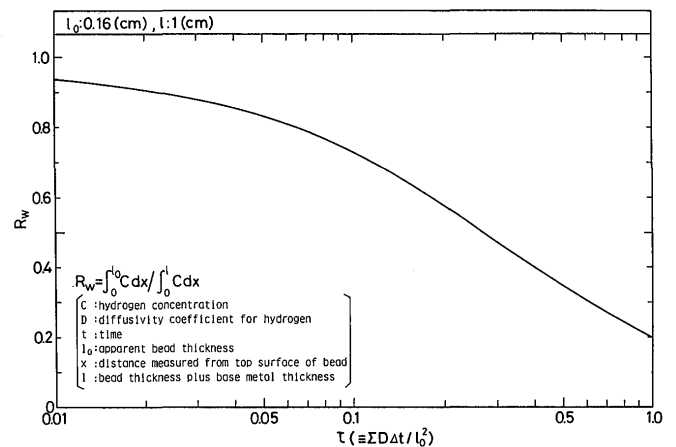

Fig. A-1   Change of the calculated ratio of the residual hydrogen content in weld metal at 100°C to that containing in the whole specimen, R_W, as a function of time factor τ
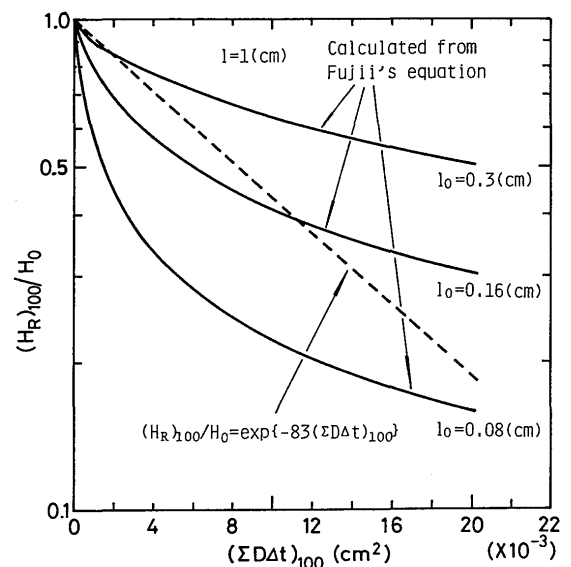

Fig. A-2   Comparison of theoretical curves by Fujii's equation and experimental one

in pure iron. Therefore $(\Sigma D \Delta t)_{100}$ in each material was conveniently rectified using each coefficient in equation (3) on the basis of $(\Sigma D \Delta t)_{100}$ in HT60. For example, in HY130

$$(\Sigma D \Delta t)_{100} \text{ in HY130} = 46/83 \{(\Sigma D \Delta t)_{100} \text{ in HT60}\} \cdots \text{(A-1)}$$

Accordingly, $(\Sigma D \Delta t)_{100}$ in HT80 and HY180 are also obtained by multiplying $(\Sigma D \Delta t)_{100}$ in HT60 by 69/83 and by 41/83, respectively.

Moreover, it is also important to select the apparent bead thickness (l_0) on calculating R_W. Then, l_0 is selected by comparing the curves of equation (3. a) in HT60 with that of theoretical equation by Fujii. **Figure A-2** shows the relationship between $(\Sigma D \Delta t)_{100}$ and $(H_R)_{100}/H_0$ of the theoretical results at various l_0 and the experimental result in HT60. The theoretical curve changes as a function of l_0. Now actual bead thickness is about 0.3 (cm), however, the theoretical curve at l_0 of 0.3 (cm) doesn't correspond to the experimental curve. Then the theoretical curve at l_0 of 0.16 (cm) approximately matched the experimental curve in the range of $(\Sigma D \Delta t)_{100}$ from 0 to 0.02 (cm^2) by the regression analysis. Therefore, l_0 of 0.16 (cm) was used in order to calculate $(\Sigma D \Delta t)_{100}/l_0{}^2$.

Consequently, R_W is calculated from Fig. A-1 using rectified $(\Sigma D \Delta t)_{100}$ in each material and l_0 of 0.16 (cm).