Hydrogen Permeation Parameters of X80 Steel and Heat-Affected Zone in High-Pressure Coal Gasification Environments (Postprint)

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Abstract

Specimens of different subzones of the heat-affected zone (HAZ) of X80 pipeline steel weld joints were prepared using welding thermal simulation technique, the hydrogen permeation behavior in X80 steel and the HAZ was investigated through hydrogen permeation experiments in a high-pressure hydrogen-containing coal-derived gas environment, and the corresponding hydrogen permeation parameters were calculated. The results revealed that, compared with X80 steel, the hydrogen diffusion coefficient in the HAZ increased to varying degrees and increased with increasing peak temperature, while the adsorbed hydrogen concentration, hydrogen solubility, and hydrogen trap density showed opposite trends. Combined with microstructural analysis, it was found that high-angle grain boundary content, dislocation density, and grain boundary planarity were the main factors affecting the hydrogen permeation parameters. The severely overheated coarse-grained zone exhibited the highest hydrogen diffusion coefficient, mainly because this zone had the highest peak temperature, austenite grains underwent severe growth, grain boundary planarity increased, coarse bainitic ferrite was generated after cooling, the high-angle grain boundary content decreased significantly, the dislocation density also decreased compared with X80 steel, and the trapping effect on hydrogen was weakened.

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Study on Hydrogen Permeation Parameters of X80 Steel and Welding Heat-Affected Zone Under High-Pressure Coal Gas Environment

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Abstract

Hydrogen gas is typically present in coal gas environments, leading to hydrogen-induced permeation in pipelines, particularly in the welding heat-affected zone (HAZ). The hydrogen permeation process is a prerequisite for subsequent hydrogen embrittlement failure. With the development of the coal gas industry, fundamental research on hydrogen permeation behavior in pipelines under coal gas conditions remains unfortunately lacking and urgently needs supplementation. In this work, X80 pipeline steel was used, and HAZ samples—including the intercritical heat-affected zone (ICHAZ), fine-grained heat-affected zone (FG-HAZ), and coarse-grained heat-affected zone (CGHAZ)—were experimentally simulated using a Gleeble 3500 simulator. Hydrogen permeation tests were then conducted on X80 pipeline steel and the HAZs in a coal gas environment. Calculated results indicated that the hydrogen diffusion coefficient increased with rising peak temperature in the HAZs, while other parameters such as subsurface hydrogen concentration, hydrogen solubility, and hydrogen trap density showed the opposite trend. The mechanism underlying differences in HAZ hydrogen permeation parameters was analyzed through optical microscopy (OM), electron backscatter diffraction (EBSD), and transmission electron microscopy (TEM). The analysis revealed that the content of large-angle grain boundaries, grain boundary straightness, and dislocation density were the primary factors. Large-angle grain boundaries and dislocations can dramatically arrest hydrogen atoms, while straight grain boundaries may act as hydrogen diffusion paths. For FGHAZ, the straight grain boundaries and low dislocation density compared with the matrix played the predominant role in the hydrogen diffusion process, resulting in an increased hydrogen diffusion coefficient relative to the steel substrate. For ICHAZ and CGHAZ, the decrease in large-angle grain boundaries and dislocation density acted as the main factor. Particularly for CGHAZ, the microstructure was mainly composed of tabular bainitic ferrite (BF) with large grain size and straight grain boundaries due to the highest peak temperature, and the content of large-angle grain boundaries decreased significantly. Consequently, CGHAZ exhibited the highest hydrogen diffusion coefficient and the lowest hydrogen trap density and hydrogen solubility.

Keywords: coal gas, X80 pipeline steel, heat affected zone (HAZ), microstructure, hydrogen permeation

Introduction

China' s energy structure is characterized by "poor oil, scarce gas, and abundant coal," with coal accounting for approximately 70% of primary energy consumption. However, most coal utilization in China involves direct combustion, resulting in low energy efficiency and severe pollution. Currently, the state is actively promoting the development of the coal gas industry, which not only enables efficient coal utilization but also alleviates pressure from natural gas shortages. As an emerging industry, a bottleneck for coal gas development is how to transport the produced coal gas to destinations. Considering the efficiency and economy of various transportation methods, pipelines are undoubtedly the preferred choice [1]. It is understood that outbound pipelines such as the "Xin-Yue-Zhe" coal gas project plan to use the same high-grade pipeline steels (X70 and X80) as conventional natural gas pipelines, with transportation pressures reaching 12 MPa.

Unlike conventional natural gas, the new type of coal gas contains a certain amount of H_2, whose presence may lead to hydrogen adsorption and permeation on material surfaces, subsequently causing hydrogen embrittlement failure of pipeline steel [2,3]. Moreover, the higher the strength grade of pipeline steel, the greater its susceptibility to hydrogen embrittlement failure [4–7]. To reduce carbon emissions from conventional natural gas, the UK has conducted feasibility investigations on using existing natural gas pipelines for blended natural gas and H_2 transport, but no systematic research on hydrogen embrittlement susceptibility of pipeline steel was carried out [8,9]. To date, extensive experimental research has been conducted domestically and internationally on hydrogen permeation and hydrogen embrittlement failure of marine and buried pipeline steels caused by corrosion and improper cathodic protection, accumulating rich experience [10–14]. Regarding research on hydrogen embrittlement susceptibility of pipeline steel in high-pressure pure H_2 environments, relevant reports exist abroad [15–17]. Moro et al. [18] found that even at room temperature, X80 steel exhibits reduced elongation in 0.1 MPa H_2 environments, but the study was limited to the pipeline steel substrate. Since long-distance pipelines are typically manufactured by welding, welding thermal effects cause severe microstructural heterogeneity in the joint heat-affected zone (HAZ) [19–21], further increasing the complexity of hydrogen permeation behavior and the likelihood of hydrogen embrittlement failure in pipeline steel under coal gas environments. Hydrogen permeation is a prerequisite for hydrogen embrittlement failure of pipeline steel. Regarding research on hydrogen permeation behavior in high-grade pipeline steel under coal gas environments, no relevant reports have been found domestically or internationally to date. With the rapid development of the coal gas industry, research on hydrogen permeation behavior of high-grade pipeline steel, particularly the welding joint HAZ, in high-pressure coal gas environments has become especially necessary. This work selected X80 high-grade pipeline steel. Based on HAZ thermal cycle parameters measured from actual pipe welding operations, welding thermal simulation technology was used to prepare enlarged HAZ samples. Through hydrogen permeation experiments in high-pressure hydrogen-containing environments, combined with microstructural analysis using optical microscopy (OM), electron backscatter diffraction (EBSD), and transmission electron microscopy (TEM), this study comparatively investigated differences in hydrogen permeation behavior between pipeline steel substrate and various HAZ sub-regions in coal gas environments and their fundamental causes, providing theoretical guidance for safe design, operation, and maintenance of high-pressure coal gas pipelines.

Experimental

Materials and Sample Preparation

The experimental material was domestic X80 double-wire spiral submerged arc welded pipe, with substrate chemical composition (mass fraction, %) of: C 0.06, Si 0.27, Mn 1.81, S 0.002, P 0.011, Ni 0.30, Mo 0.31, Cu 0.28, Nb 0.07, Cr 0.02, Ti 0.015, Fe balance. High-pressure hydrogen permeation experiments employed simulated coal gas with a total pressure of 12 MPa and composition (partial pressure) of: H_2 0.24 MPa, CO_2 0.20 MPa, remainder N_2.

Samples for microstructural analysis and hydrogen permeation testing were sectioned along the pipe rolling direction with dimensions of 100 mm × 30 mm × 2.5 mm. Based on HAZ thermal cycle parameters measured from actual pipe welding operations, a Gleeble 3500 thermal simulator was used to prepare samples of the coarse-grained heat-affected zone (CGHAZ), fine-grained heat-affected zone (FGHAZ), and intercritical heat-affected zone (ICHAZ) of X80 pipeline welding joints. The welding thermal cycle curves are shown in [Figure 1: see original paper]. The specific process involved heating at 150 °C/s to peak temperatures of 1350 °C, 950 °C, and 800 °C, corresponding to CGHAZ, FGHAZ, and ICHAZ in actual welding HAZs, respectively, followed by cooling at a rate corresponding to $t_8/_5 = 15$ s. Subsequently, 24.0 mm diameter disc samples for hydrogen permeation testing were cut from the isothermal zone at the center of the thermal simulation specimens using wire electrical discharge machining.

Microstructural Characterization

Metallographic samples of X80 steel and HAZ sub-regions were prepared and observed using a DM2500M optical microscope (OM) to compare microstructural changes caused by welding thermal effects. For EBSD analysis, samples were prepared by electrolytic polishing in a solution of 665 mL CH_3COOH + 35 mL H_2O + 125 g CrO_3 at 15 V, with temperature controlled at (20$±$1) °C for 10 min. A SUPRA 55 scanning electron microscope (SEM) equipped with a 3D electron backscatter orientation imaging system was used to analyze samples tilted at 70°, and HKL analysis software was employed to characterize orientation distributions and large-angle grain boundary densities in X80 steel and HAZ sub-regions. Fine microstructural observations were performed using a Tecnai F30 field-emission TEM. TEM samples were prepared by twin-jet electropolishing with a 5% perchloric acid alcohol solution at 30 V.

Hydrogen Permeation Testing

The traditional Devanathan-Stachurski double electrolytic cell was combined with a high-pressure autoclave to investigate hydrogen permeation behavior in pipeline steel under high-pressure simulated coal gas environments, with the apparatus schematic shown in [Figure 2: see original paper]. The sample was sealed between the high-pressure autoclave (hydrogen charging side) and the electrolytic cell (detection side). The medium inside the autoclave was high-pressure gas containing H_2, which continuously collided with the sample surface, forming dissolved H in the sample through physical and chemical adsorption. Under the concentration gradient, dissolved H diffused through the sample interior, and H reaching the electrolytic cell side was electrochemically oxidized to H^+ while measuring the hydrogen permeation current. To ensure stable conditions in the high-pressure autoclave, gas tightness was verified before experiments. The autoclave was pressurized with 12 MPa Ar, and a pressure drop not exceeding 0.02 MPa within 24 h was required before subsequent hydrogen permeation experiments.

The detailed experimental procedure was as follows: Both circular working surfaces of the sample were ground with 600-grit waterproof abrasive paper to a final thickness of 2.0 mm, followed by degreasing with acetone. One surface served as the charging side, exposed to the high-pressure coal gas environment during testing; the other surface served as the detection side, requiring Ni plating before experiments. The Ni plating solution used Watt' s solution with a current density of 5 mA/cm^2 for 5 min. The sample (working electrode) with attached leads was sealed between the high-pressure autoclave and electrolytic cell. A Hg/HgO electrode served as the reference electrode and high-purity graphite as the auxiliary electrode. The Ni plating was passivated in 0.1 mol/L NaOH solution at a polarization potential of +300 mV (vs. Hg/HgO 98 mV) using a CHI 760D electrochemical workstation. While passivating the detection side, the autoclave was purged three times with 1 MPa Ar to minimize effects of residual O_2 on the charging side. After the detection side current decreased below 0.05 A/cm^2, the Ar in the autoclave was replaced with 0.5 MPa test gas, with this process repeated three times. Subsequently, 12 MPa simulated coal gas was introduced into the high-pressure autoclave while continuously monitoring hydrogen permeation current changes. After the hydrogen permeation current stabilized, corresponding hydrogen permeation parameters were calculated from the experimental data. During hydrogen permeation testing, the temperature inside the high-pressure autoclave was maintained at (40$±$0.5) °C.

2.1 Microstructure

[Figure 3: see original paper] shows OM images of X80 steel and the simulated welding HAZ sub-regions. As seen in Figure 3a, X80 steel consists mainly of fine acicular ferrite (AF) with a small amount of massive ferrite (MF) formed through proeutectoid reactions. Additionally, due to the controlled rolling and controlled cooling production process, grains exhibited some deformation along the rolling direction. Under welding thermal cycles, each HAZ sub-region underwent special heat treatment processes, resulting in different microstructural transformations.

For the ICHAZ farther from the heat source, welding thermal effects were relatively minor. As shown in Figure 3b, the peak temperature of 800 °C only caused partial austenitization of the base material. During heating, non-austenitized ferrite grains underwent some coarsening, while austenitized ferrite transformed primarily into fine granular bainite (GB) upon cooling, distributed randomly with partially coarsened ferrite. This region retained the rolling texture characteristics of the base material, with noticeably decreased microstructural uniformity.

As the HAZ peak temperature increased further, as shown in Figure 3c, complete austenitization occurred in the base material at 950 °C, the rolling texture disappeared, and grain boundaries began to straighten. Due to the low austenitization temperature and the inhibiting effect of carbide-forming elements such as Nb and Ti, austenite grain growth was significantly restricted. During cooling, a microstructure dominated by polygonal ferrite (PF) and MF formed, with some GB appearing due to the fast cooling rate. When the HAZ peak temperature reached 1350 °C, exceeding the temperature for rapid austenite grain growth (generally 1100 °C) [22], the inhibiting effect of Nb, Ti and other elements weakened, and severe austenite grain growth occurred with increasingly straight grain boundaries. During cooling, a small amount of MF formed through proeutectoid reactions, subsequently transforming primarily into coarse tabular bainitic ferrite (BF), as shown in Figure 3d.

2.2 Hydrogen Permeation Behavior of X80 Steel and HAZ Sub-regions

[Figure 4: see original paper] shows the hydrogen permeation current density-time relationship curves for X80 steel and HAZ sub-regions in the 12 MPa coal gas environment, where time zero represents when coal gas was introduced into the autoclave. It can be observed that both X80 steel and HAZ sub-regions require a certain period—the breakthrough time—before hydrogen permeation current density increases continuously with time and finally reaches a steady-state diffusion process. The steady-state current densities $i\infty$ for X80 steel and HAZ sub-regions are listed in . X80 steel exhibits the lowest $i\infty$, while for the HAZ, $i\infty$ increases continuously with rising peak temperature. Since $i\infty$ is proportional to hydrogen diffusion flux, the relative magnitude of hydrogen diffusion flux in X80 steel and HAZ sub-regions is also reflected by their respective $i\infty$ values.

2.3 Accurate Calculation Method for Hydrogen Diffusion Coefficient

Before calculating the hydrogen diffusion coefficient, an appropriate calculation method must first be selected. Currently, primary methods for calculating hydrogen diffusion coefficients include the breakthrough time method, time-lag method, Laplace method, and Fourier method [23].

The formulas for calculating hydrogen diffusion coefficients using the breakthrough time method and time-lag method are respectively [23]:

$D = L^2/(6t\_b)$ (1)
$D = L^2/(15t\_L)$ (2)

where D is the hydrogen diffusion coefficient, L is sample thickness, t_b is breakthrough time, and t_L is lag time corresponding to when hydrogen permeation current reaches 63% of steady-state current. As shown in Equations (1) and (2), for samples of a given thickness, the accuracy of diffusion coefficients calculated using these two methods is closely related to the starting point of hydrogen permeation current monitoring. Since both the test gas purging process and subsequent pressurization cause hydrogen adsorption and permeation, precisely determining the start time for hydrogen permeation current monitoring represents a challenge in gas-phase hydrogen permeation experiments. Consequently, these two calculation methods are not suitable for accurate determination of hydrogen diffusion coefficients in gas-phase hydrogen permeation test systems.

The Laplace method calculates hydrogen diffusion coefficients using the formula [24]:

$i\_t = A \cdot \exp(-\pi^2 Dt/L^2)$ (3)

where i_t is hydrogen permeation current density at time t, and A is a constant. By plotting ln(i_t) vs. t^{-1} and fitting, the slope can be obtained to calculate D. For more intuitive analysis of the Laplace method' s applicability, Equation (3) can be further transformed. Assuming t_1 and t_2 are two time monitoring points during the hydrogen permeation transient process, substituting into Equation (3) yields:

$\ln(i\_t_1) = \ln(A) - \pi^2 D t_1/L^2$ (4)
$\ln(i\_t_2) = \ln(A) - \pi^2 D t_2/L^2$ (5)

Subtracting and simplifying Equations (4) and (5) gives:

$\ln(i\_t_1/i\_t_2) = -\pi^2 D(t_1 - t_2)/L^2$ (6)

Analysis reveals that the accuracy of diffusion coefficients calculated using the Laplace method is also closely related to hydrogen permeation current monitoring time and is therefore unsuitable for gas-phase hydrogen permeation test systems.

The Fourier method calculates hydrogen diffusion coefficients using the expression [24]:

$\ln(1 - i\_t/i\infty) = B - \pi^2 Dt/L^2$ (7)

where B is a constant, and D can be calculated from the slope of the $\ln(1 - i\_t/i\infty)$ vs. t curve. Similarly assuming t_1 and t_2 as two time monitoring points during the transient process, substituting into Equation (7) gives:

$\ln(1 - i\_t_1/i\infty) = B - \pi^2 D t_1/L^2$ (8)
$\ln(1 - i\_t_2/i\infty) = B - \pi^2 D t_2/L^2$ (9)

Subtracting and simplifying Equations (8) and (9) yields:

$\ln[(1 - i\_t_1/i\infty)/(1 - i\_t_2/i\infty)] = -\pi^2 D(t_1 - t_2)/L^2$ (10)

Equation (10) shows that diffusion coefficient calculation accuracy is affected by L, $i\infty$, and the current values at times t_1 and t_2, as well as the difference between t_2 and t_1. Among these parameters, sample thickness L and $i\infty$ can be measured accurately. For the transient hydrogen permeation process, when two specific current values are selected, their magnitudes and corresponding time differences can be precisely obtained without being affected by the starting time of hydrogen permeation current monitoring. Consequently, the Fourier method can accurately determine hydrogen diffusion coefficients in materials.

2.4 Hydrogen Permeation Parameters

The Fourier method was used to fit the transient hydrogen permeation curves of X80 steel and HAZ sub-regions, with results shown in [Figure 5: see original paper]. Based on the fitted curve slopes, hydrogen diffusion coefficients D for X80 steel and HAZ sub-regions were calculated and are listed in .

After hydrogen permeation reaches steady state, the diffusion flux becomes time-independent and hydrogen concentration at each location in the sample no longer changes with time. At this point, Fick' s first law can be used to calculate the sub-surface hydrogen concentration C_0 [25]:

$C_0 = (i\infty \cdot L)/(F \cdot D)$ (11)

where F is Faraday' s constant (F = 96485 C/mol). Substituting the L, i∞, and D values for X80 steel and HAZ sub-regions into Equation (11) yields the corresponding adsorbed hydrogen concentrations C_0, which are listed in .

According to Sieverts' law, the sub-surface hydrogen concentration C_0 is proportional to the square root of the equilibrium hydrogen pressure p, with the ratio representing hydrogen solubility S in the material, expressed as [26]:

$S = C_0/\sqrt{p}$ (12)

Substituting the adsorbed hydrogen concentrations and hydrogen partial pressure in the coal gas into the above equation yields the hydrogen solubility S values in X80 steel and HAZ sub-regions, shown in .

Due to hydrogen traps in steel, the experimentally obtained D is always lower than the ideal lattice diffusion coefficient (D_L). This delay in hydrogen diffusion is strongly related to hydrogen trap density [27]. Therefore, determining hydrogen trap density in steel is essential for analyzing hydrogen permeation behavior. Let N_T be the hydrogen trap density in the material; the relationship between apparent diffusion coefficient D and lattice diffusion coefficient D_L can be expressed as [27]:

D = D_L/(1 + N_T) (13)

Transforming this yields the calculation formula for hydrogen trap density N_T:

N_T = (D_L/D) - 1 (14)

where D_L is taken as 1.28×10^{-4} cm^2/s [28]. The calculated N_T values for X80 steel and HAZ sub-regions are listed in .

Thus, the D, C_0, S, and N_T values for X80 steel and HAZ sub-regions were obtained. Under welding thermal effects, HAZ microstructure underwent significant changes, consequently affecting hydrogen permeation parameters. Specifically, D exhibited an increasing trend with rising peak temperature, while C_0, S, and N_T showed the opposite trend.

2.5 Mechanism of Microstructural Influence on Hydrogen Permeation Parameter Differences

The welding thermal cycle process significantly affects material microstructure, grain size, and dislocation density. Both dislocations and grain boundaries act as hydrogen traps that can effectively retard hydrogen diffusion in steel [29]. To further analyze the fundamental reasons for microstructural changes in the joint HAZ affecting hydrogen permeation parameters, EBSD analysis and TEM observation of X80 steel and HAZ sub-regions were performed.

[Figure 6: see original paper] shows the bcc phase orientation maps for X80 steel and HAZ sub-regions (with grain boundaries >15° considered large-angle grain boundaries), accompanied by inverse pole figure (IPF) legends, where regions of the same color represent similar orientations. [Figure 7: see original paper] shows the grain boundary misorientation angle distribution curves for X80 steel and HAZ, also using 15° to distinguish between low- and high-angle grain boundaries. [Figure 8: see original paper] shows TEM images of X80 steel and HAZ sub-regions.

As seen in Figure 6a, X80 steel has a high density of large-angle grain boundaries, though local aggregation of low-angle grain boundaries exists. Grain boundaries act as hydrogen traps and significantly influence hydrogen diffusion. Teus et al. [30] found that hydrogen diffusion at grain boundaries requires higher activation energy than in the crystal lattice. Jothi et al. [31] used numerical simulation to determine that grain boundary angles of 15°–45° can effectively retard hydrogen diffusion in Ni. Smoluchowski [32] proposed that at low grain boundary angles, lattice diffusion controls hydrogen diffusion, while at higher angles (>15°), hydrogen diffusion along grain boundaries becomes the dominant factor. Statistical calculations based on the grain boundary angle distribution curve in Figure 7 show that large-angle grain boundaries account for 66% in X80 steel, while low-angle grain boundaries account for only 34%. Therefore, the small grain size and high content of large-angle grain boundaries constitute an important reason for the low hydrogen diffusion coefficient in X80 steel. The green region in Figure 6a occupies approximately 45% of the analyzed area, belonging to the {101} crystal plane family—the close-packed planes of the bcc structure—which also act as barriers to hydrogen diffusion to some extent. Figure 8a shows a TEM image of X80 steel with typical acicular ferrite structure and extremely high dislocation density. Dislocations, as hydrogen traps, further enhance the trap effect hindering hydrogen diffusion in X80 steel. In summary, the high content of large-angle grain boundaries, dislocation density, and presence of close-packed planes can all effectively retard hydrogen diffusion in X80 steel, acting as hydrogen traps, thereby resulting in relatively high hydrogen solubility and low diffusion coefficient.

Under welding thermal effects, the HAZ experienced special heat treatment processes. For ICHAZ, incomplete austenitization occurred, and the room-temperature microstructure consisted mainly of coarsened ferrite and granular bainite transformed from undercooled austenite. The green region in Figure 6b represents ferrite that coarsened during the welding thermal cycle, whose preferred orientation did not disappear and continued to act as close-packed planes hindering hydrogen diffusion. However, grain coarsening in this region reduced the final large-angle grain boundary content to 60% (Figure 7), weakening the hydrogen trap effect of large-angle grain boundaries. Additionally, TEM observation of ICHAZ in Figure 8b shows that dislocation density in this region decreased compared with X80 steel, correspondingly reducing the dislocation hydrogen trap effect. Combined, these factors caused the diffusion coefficient of ICHAZ to increase relative to X80 steel, while hydrogen trap density and hydrogen solubility decreased.

For FGHAZ, the higher peak temperature caused complete austenitization of

X80 steel. After cooling to room temperature, the formed polygonal ferrite, massive ferrite, and granular bainite exhibited good isotropy, the crystallographic texture disappeared (Figure 6c), and the barrier effect of close-packed planes on hydrogen diffusion diminished. Furthermore, compared with X80 steel, the proportion of large-angle grain boundaries in FGHAZ increased to 81% (Figure 7). However, enhanced carbon migration and diffusion capacity at high temperatures caused austenite grain boundaries to become straight, making hydrogen diffusion paths along grain boundaries more direct and weakening the grain boundary hydrogen trap effect. Figure 8c shows that dislocation density in FGHAZ ferrite decreased significantly compared with X80 steel, reducing the dislocation hydrogen trap effect. In summary, the disappearance of preferred orientation, straightening of grain boundaries, and reduction in dislocation density caused FGHAZ diffusion coefficient to increase compared with X80 steel, while hydrogen trap density and hydrogen solubility decreased.

As the HAZ peak temperature continued to increase, in CGHAZ, the bainitic ferrite grains transformed from undercooled austenite exhibited obvious preferred orientation (Figure 6d). The hydrogen permeation test surface consisted mainly of close-packed planes from the bcc structure {101} crystal plane family, occupying approximately 70% of the analyzed area. Due to increased high-temperature dwell time, carbonitrides dissolved, second-phase particle pinning effects weakened, and diffusion of carbon and alloying elements at high temperatures was significantly enhanced, further increasing grain boundary straightness. Severe austenite grain growth occurred, with average grain size of approximately 40 m, and large-angle grain boundary content decreased significantly to only 46% (Figure 7). Figure 8d shows the fine structure of CGHAZ, revealing that coarse bainitic ferrite substructures are lath-shaped, with low-angle grain boundaries between lath interfaces and high-angle grain boundaries between lath packet interfaces. Compared with other HAZ regions, CGHAZ has slightly higher dislocation density but still lower than X80 steel, meaning the dislocation hydrogen trap effect in CGHAZ is smaller than in X80 steel. In summary, although close-packed planes and dislocations can somewhat retard hydrogen diffusion, the increased grain size and drastic reduction in large-angle grain boundary content played a more important role in weakening the hindrance to hydrogen diffusion. Consequently, this region has the highest diffusion coefficient and the lowest hydrogen trap density and hydrogen solubility. Yazdipour et al. [33] heat-treated X70 steel under different conditions to obtain samples with various grain sizes, and through experimental research combined with numerical simulation, confirmed that the hydrogen diffusion coefficient reaches a maximum when X70 steel grain size is approximately 46 m, consistent with our research results.

Conclusions

1. The original microstructure of X80 steel consists of fine acicular ferrite with high densities of dislocations and large-angle grain boundaries, along with some preferred orientation. Hydrogen diffusion planes include mostly

close-packed planes from the {101} crystal plane family, which can effectively retard hydrogen diffusion in X80 steel and act as hydrogen traps. Therefore, X80 steel has relatively high hydrogen trap density and hydrogen solubility, along with a low diffusion coefficient.

2. For ICHAZ and CGHAZ, the increased grain size, reduced large-angle grain boundary content, and decreased dislocation density lead to weakened hydrogen trap effects, which constitute the main reason for increased hydrogen diffusion coefficients relative to X80 steel. Particularly for CGHAZ, severe austenite grain growth occurred at high temperature, forming coarse bainitic ferrite upon cooling with average grain size reaching 40 m. Large-angle grain boundary content decreased to only 46%, weakening the grain boundary hydrogen trap effect and resulting in reduced hydrogen trap density and hydrogen solubility compared with X80 steel.

3. For FGHAZ, although the peak temperature was not high enough to cause severe austenite grain growth and the room-temperature microstructure consisted mainly of polygonal ferrite and massive ferrite with minor granular bainite, and despite this region having the finest grain size and highest large-angle grain boundary content, the disappearance of preferred orientation, reduced dislocation density, and increased grain boundary straightness weakened the hydrogen trap effect. Consequently, FGHAZ exhibited increased hydrogen diffusion coefficient and decreased hydrogen trap density and hydrogen solubility compared with X80 steel.

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Note: Figure translations are in progress. See original paper for figures.