

Research Progress on Corrosion Behavior of Oil and Gas Pipelines in Wet CO₂ and H₂S Environments

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Abstract: Corrosion of pipes is a common problem in oilfield extraction, gathering, storage and transportation. Fluid containing CO₂ and H₂S will cause serious corrosion perforation and fracture of oil and gas field pipes, which will lead to fluid leakage. It not only pollutes the surrounding environment, but also causes safety accidents. In this paper, the corrosion behavior of common pipes in the field of oil and gas field exploitation is reviewed in the presence of wet CO₂ and H₂S alone and in the coexistence environment, and the influencing factors are analyzed. The results show that CO₂ partial pressure and H₂S partial pressure play a major role in the corrosion rate of the pipe in the presence of wet phase CO₂ and H₂S alone. In addition, the effect of Cl⁻ concentration on the pipe cannot be ignored, and the corrosion rate of the pipe decreases at high Cl⁻ concentration (>5 000 mg/L). Finally, the research on the corrosion of pipes in CO₂/H₂S system is prospected, in order to provide reference for the formulation of protection measures for oil and gas fields and the development of new corrosion-resistant pipes.

Keywords: CO₂; H₂S; Interaction Effect; Corrosion Mechanism; Corrosion Factors.

1. Introduction

With the increasing demand for energy, oil and gas production increased year by year, resulting in oil and gas transportation equipment corrosion and protection has become the focus of widespread concern [1]. In view of the limitations of economy and technical feasibility, carbon steel materials are often chosen as tubing for oil and gas fields. [2].

However, in a complex environment containing multiphase fluids such as CO₂ and H₂S, pipeline systems involved in the transportation of oil and gas are often in complex environments such as high temperature, high pressure, and high acidity, resulting in frequent incidents such as pipeline damage, perforation, and oil and gas leakage [3, 4]. In particular, CO₂ induced downhole tubing corrosion poses a great threat to the integrity of oil wells, and the presence of H₂S and applied stress will accelerate these corrosion problems [5]. It is recognized that CO₂ and H₂S have almost no corrosion risk to steel in a dry environment. Only when it is dissolved in water and hydrolyzed and ionized to produce H⁺, the corrosion rate of the pipe will increase significantly [6, 7].

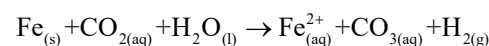
At present, a lot of scientific research has been carried out on the corrosion of oil and gas pipes casing in CO₂ and H₂S, but a relatively complete corrosion system has not yet been formed. There are still many basic theoretical and key technical problems to be further explored [8-10]. Therefore, the corrosion mechanism and its influencing factors of oil and gas pipes in the presence of CO₂ and H₂S are reviewed. The effects of water content, temperature, pressure, and Cl⁻ concentration on corrosion in the presence of CO₂, H₂S are highlighted, in order to provide some reference for the development of corrosion protection technology in oil and gas fields.

2. The Corrosion of CO₂

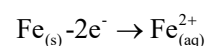
2.1. The Corrosion Mechanism of CO₂

CO₂ gas itself is not corrosive. However, when CO₂ combines with water to form carbonic acid, it will corrode the metal [11]. In the water-rich environment, the corrosion process of CO₂ on the metal undergoes multiple stages: the first is the dissolution stage. At this time, the structural framework of Fe₃C is formed inside the steel matrix, and the grains of FeCO₃ are deposited on the metal surface. Subsequently, CO₃²⁻ and HCO₃⁻ ions diffuse into the metal and react with the steel matrix to form a corrosion intermediate layer and an inner layer [12]. Under the promotion of cathodic hydrogen evolution, CO₂ accelerates the corrosion rate of metals. An electrochemical reaction occurs between the carbonic acid solution and the metal. The reaction process is as follows:

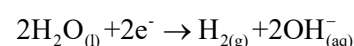
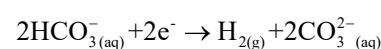
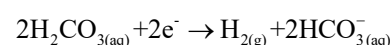
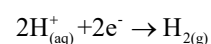
Total reaction:



Anode reaction:



Cathodic reaction:



2.1.1. Direct Reduction of Carbonic Acid

Bockris et al. [13] proposed that in the CO₂ environment, the intermediate medium that promotes the conversion of Fe→Fe²⁺ is OH⁻. When Deward et al. [14] studied the mechanism of carbon dioxide corrosion, the cathode reaction

mechanism was proposed that the undissociated carbonic acid molecules were directly reduced, which was related to the research of Ogundele et al. [15] and Linter et al. [16]. WIECKOWSKI et al. [17] observed the direct reduction of H_2CO_3 molecules and HCO_3^- ions through static cyclic voltage studies. However, Nestic et al. [18, 19] questioned some of the previous assumptions about the direct reduction mechanism. The anodic mechanism is different at different pH, and it is confirmed that H_2CO_3 is dominated by the cathodic reaction and has a limiting current at $4 < \text{pH} < 5$, which is controlled by the hydration of carbon dioxide.

2.1.2. Buffer Effect

Many scholars have introduced the 'buffer effect' [20-22] theory to confirm that even if H_2CO_3 is not used as an electroactive substance, it can fully explain the mechanism of limiting current generation. In the diffusion boundary layer, H_2CO_3 is uniformly dissociated, and the subsequent reduction of H_2CO_3 provides a reaction pathway parallel to the direct reduction of H_2CO_3 , which exists not only in CO_2 corrosion, but also in acetic acid corrosion of low carbon steel. Carbonic acid is only a 'reservoir' of hydrogen ions. Increasing the limiting current density by buffering the hydrogen ion concentration on the metal surface only affects the limiting cathode current, and has no effect on the charge transfer current [20]. Hydrogen ion reduction is the main cathode reaction [23]. In the direct reduction mechanism, H_2CO_3 is reduced by electrochemical reaction substances, while the buffer mechanism emphasizes the uniform decomposition of H_2CO_3 . The two effects are not mutually exclusive, but are different and independent processes. The buffering effect is crucial in CO_2 corrosion, and the CO_2 corrosion process may include all these cases [22].

2.2. Influencing Factors of CO_2 Corrosion

CO_2 corrosion is a complex electrochemical process, which is affected by a series of factors, including water content, pressure, Cl^- concentration, oil phase environment, pH value, flow rate, temperature and so on. The influence of pH value, flow rate, temperature and oil phase environment on CO_2 corrosion has been introduced in detail by other scholars [24].

2.2.1. Moisture Content

Water content is a key factor affecting metal corrosion. Carbon steel does not corrode in dry carbon dioxide. With the increase of relative humidity, the change of corrosion rate increases. Excessive water content will cause electrochemical corrosion, shorten the life of pipeline and cause economic loss [25]. In supercritical CO_2 and SO_2 environments, reducing water content can alleviate corrosion more than reducing sulfur dioxide content. In the supercritical CO_2 pipeline, the corrosion rate increases with the increase of water content. When the water content is greater than 50 %, the corrosion rate is accelerated and the corrosion form changes from uniform to local [26]. The water solubility increases with the increase of temperature and pressure. Under high water conditions, with the increase of temperature, pressure, CO_2 partial pressure and the content of Ca^{2+} , Cl^- , Mg^{2+} , the corrosion products of 20#steel increase, and the film is not dense enough to increase the corrosion rate [27].

2.2.2. Pressure

Pressure is an important factor affecting CO_2 corrosion, which interacts with temperature, CO_2 concentration and metal type [12]. Under high pressure, CO_2 is more easily dissolved in the liquid phase, thereby increasing the

concentration of CO_2 in the corrosive medium and accelerating the corrosion reaction. At a constant temperature, the pressure has a greater impact, and the local pressure difference will cause the gas-liquid interface to move and cause local corrosion or pitting [28]. In supercritical CO_2 phase, with the increase of $\text{P}_{\text{H}_2\text{S}}$ concentration, the corrosion is more serious, and the corrosion rate of aqueous phase decreases. The supercritical CO_2 pipeline has a threshold pressure (about 10 MPa), and the corrosion rate increases significantly with the increase of pressure. The solubility of H_2O in supercritical CO_2 has two trends with the change of pressure, which is divided by the phase transition point. In gaseous CO_2 , the solubility of H_2O decreases logarithmically with the increase of pressure. However, once the pressure exceeds a certain value, CO_2 enters the liquid/supercritical state, and the solubility of H_2O increases rapidly with pressure and stabilizes at a high value [29, 30]. The corrosion rate of 20# steel increases with increasing CO_2 partial pressure at high water content in oil-water mixing pipelines [27].

2.2.3. Concentration of Cl^-

Cl^- has an erosion effect on carbon steel. It is generally believed that it has little effect on uniform corrosion, and mainly affects local corrosion such as pitting corrosion [31]. Cl^- enrichment accelerates the local corrosion of passivation film and reduces its protection ability. Adequate Cl^- concentration and fluid flow can promote the initiation and propagation of pitting corrosion of carbon steel. Under the combined action of excessive Cl^- concentration and fluid flow, extremely low surface coverage will limit pitting corrosion behavior. In addition, Cl^- can change the corrosion mechanism, low concentration enhances the synergistic effect with O_2 , and high content (3000~5000 mg/L) can reduce the solubility of CO_2/O_2 and inhibit the cathodic process [32]. Cl^- has a small radius and can pass through the corrosion layer defects to the metal surface. Although it does not participate in the electrode reaction, acidification promotes the local dissolution of metal Fe and aggravates the corrosion. It also affects the stress corrosion cracking of 13Cr stainless steel. The sensitivity of stress corrosion cracking (SCC) decreases first and then increases with the concentration of Cl^- . The general corrosion rate of carbon steel is affected by Cl^- content and exposure time. The corrosion rate tends to decrease with the extension of immersion time, and the Cl^- content increases the total penetration rate of carbon steel [33]. In practical applications, considering these factors, it is very important to select the appropriate corrosion control method according to the specific application environment and metal materials. Ensure the reliability and safety of equipment and structure, reduce risks and improve overall reliability.

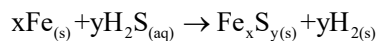
3. The Corrosion of H_2S

3.1. The Corrosion Mechanism of H_2S

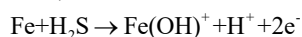
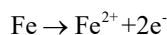
The presence of H_2S will affect the electrochemical reaction, resulting in an increase in the cathodic reaction rate, thereby accelerating the corrosion rate [34]. After H_2S is dissolved in water, it is ionized to be acidic, resulting in electrochemical corrosion of the pipe and local pitting perforation. Hydrogen atoms enriched in steel defects are constantly combined to produce hydrogen, resulting in embrittlement of steel, initiation of cracks, and stress corrosion (hydrogen embrittlement) [35]. In addition, H_2S reacts with iron to form corrosion products such as iron sulfide, which may form a film on the metal surface and affect

the subsequent corrosion process [34, 36]. When the density of the FeS product film is incomplete, the penetrating erosion ions react locally with the metal under the film, resulting in a decrease in its potential, exposing the metal to form an anode, and the surrounding FeS product film forms a cathode, thereby forming a galvanic cell and making the metal corrosion more serious [37].

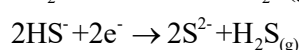
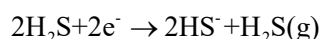
Total reaction:



Anode reaction:



Cathodic reaction:



3.2. Influencing Factors of H₂S Corrosion

3.2.1. Pressure

Pressure has an important influence on H₂S corrosion, which has a great relationship with temperature. First of all, high pressure makes H₂S more easily dissolved in the liquid phase. The increase of H₂S partial pressure will lead to the increase of the solubility of H₂S in water and the corresponding increase of mass concentration, which will lead to the increase of the concentration of H⁺ in the solution and the decrease of the pH value of the solution, which will aggravate the corrosion of H₂S to oil and gas field pipes. Secondly, high pressure will affect the corrosion products, which may form a denser or looser product layer, and may also cause product phase transition, affecting the protection performance of the metal. Furthermore, the pressure change produces a local pressure difference, which causes the gas-liquid interface to move, causing local corrosion or pitting. The concentration of corrosive medium in the high-pressure area is high, and the flow restriction in the low-pressure area is easy to form a local corrosion environment. The pressure interacts with temperature, H₂S concentration, metal type and other factors, and the corrosion rate increases significantly under high temperature and high pressure. The corrosion behavior of different metals is different under specific pressure and H₂S concentration.

3.2.2. Temperature

In general, raising the temperature can accelerate the corrosion rate of H₂S on the pipe. The reason is that the change of temperature will affect the formation and state of the corrosion product film, thus changing the protectiveness of the corrosion product film and affecting the corrosion rate. Wikjard et al. [38] found that in the solution with pH= 3.3, H₂S partial pressure of 2MPa, and temperature of 80~180 °C, the phase transition of the marjinoite occurs, and the phase transition sequence is: marjinoite→ cubic FeS phase pyrite→ pyrrhotite→ pyrite. According to the ion selectivity of corrosion products, Sun et al. [38] found that when the temperature of P110 oil well pipe was 60°C and 80°C respectively, cationic selective pyrite could inhibit the diffusion of corrosive ions to the substrate, and the corrosion rate was low and the corrosion was uniform. When the temperature is 100°C and 205°C respectively, the anionic selective pyrrhotite will cause the accumulation of corrosion scale and Cl⁻ between the substrate, the corrosion rate is high and localized corrosion will occur. Huang et al. [39] found that under the environment of total pressure of 1.5MPa,

L245NS pipeline steel is extremely serious corrosion at 30~90°C, and the maximum corrosion rate appears at 60°C, reaching 0.5149mm/a, when it continues to heat up to 120°C, it becomes severe corrosion.

3.2.3. Cl⁻ concentration

The corrosive medium of H₂S generally contains Cl⁻, and the presence of Cl⁻ also accelerates the corrosion of H₂S on oil and gas pipelines. The product film formed by H₂S corrosion will be destroyed by Cl⁻, preventing the passivation of the pipe surface, which will aggravate the corrosion of H₂S to the pipe. Shannon et al. [40] found that Cl⁻ weakened the interaction between metal and corrosion products, and had an effect on the formation of adhesive sulfides. Therefore, the corrosion product film was more likely to fall off under the action of Cl⁻, which accelerated the corrosion of oil well pipes. However, when the concentration of Cl⁻ is high to a certain extent, the corrosion rate of the metal will decrease. The reason is that Cl⁻ has a high adsorption performance and will replace H₂S and HS⁻ to adsorb on the metal surface, thereby reducing the corrosion rate of the metal. Zhang et al. [41] found that with the increase of Cl⁻ content, the corrosion potential shifted negatively, the activation and dissolution of the metal anode accelerated, the localized corrosion of the substrate strengthened, and the corrosion rate increased first and then decreased.

4. The Corrosion of H₂S/CO₂

With the rapid development of the petrochemical industry, the status of crude oil as a core energy source has become increasingly prominent. However, the corrosion of carbon steel in oil and gas pipelines in some reservoirs with high CO₂ and H₂S content in Sichuan, Shaanxi, Gansu and Ningxia provinces is still a key bottleneck restricting the sustainable development of the industry [42]. H₂S/CO₂ corrosion not only causes serious damage to oil production, storage and transportation equipment, but also poses a potential threat to the safe production of oil and natural gas in China. Although the corrosion of carbon steel in H₂S/CO₂ environment has been systematically studied, some progress has been made. However, the effective corrosion prevention and control technology in this environment is still insufficient and needs to be further improved. Therefore, strengthening the research on CO₂ and H₂S corrosion is one of the important directions for the current and future technological progress of the oil and gas industry.

4.1. The Corrosion Mechanism of H₂S/CO₂

There is a clear competitive and synergistic relationship between CO₂ and H₂S in the corrosion process [43]. In the process of CO₂ corrosion, H₂S can not only accelerate the corrosion rate through the cathode reaction, but also delay the corrosion rate through the continuous generation of FeS. Yun et al. [44] showed that the presence of H₂S would change the corrosion mechanism of CO₂, making the formation of corrosion layer more complex. The dominant factor depends largely on the partial pressure in the mixed environment of CO₂/H₂S. In the CO₂/H₂S coexistence system, when the CO₂/H₂S partial pressure ratio is less than 20, the corrosion process is mainly dominated by H₂S. At this time, the FeS product film is dense and has good adhesion, which has a good protective effect on the pipe wall. As the partial pressure continues to increase, the corrosion rate decreases [45]. When the CO₂/H₂S partial pressure ratio rises to 20~500, and the

greater the H₂S partial pressure, the faster the corrosion rate. When the CO₂/H₂S partial pressure ratio is greater than 500, the corrosion effect of CO₂ is dominant, and the corrosion products are generally Fe_x(CO₃)_y and iron oxide with good stability, and the corrosion product film is complete and dense.

4.2. Influencing Factors of H₂S Corrosion

The partial pressure ratio of CO₂ to H₂S has a significant effect on the corrosion behavior of carbon steel. On the one hand, the increase of gas partial pressure makes the water in the air quickly convert into liquid, which accelerates the corrosion and fracture of the pipeline. On the other hand, with the increase of pressure, the gas solubility also changes, which reduces the pH value of the solution. If the pH value decreases, it will promote the dissolution of the rust film on the pipeline and accelerate the erosion of the pipeline. In addition, the partial pressure ratio of CO₂ to H₂S affects the morphology of corrosion products (loose or dense), which is the key factor determining the corrosion rate. The loose product film structure will promote the local galvanic corrosion of carbon steel, thus accelerating the corrosion. The dense product film can effectively inhibit the further corrosion of carbon steel [46].

In addition to the partial pressure of CO₂ and H₂S, factors such as temperature, water content, flow rate, medium composition (such as Cl⁻ content) and material properties also have a significant impact on the corrosion process. For example, high temperature will aggravate the corrosion reaction, and the increase of flow rate may lead to the aggravation of erosion corrosion and local corrosion. In addition, the corrosion resistance of metals with different materials in CO₂/H₂S environment is also significantly different.

5. Conclusion

(1) When CO₂ gas is dissolved in water, the electrochemical corrosion process will be significantly accelerated. The rate of this corrosion phenomenon is regulated by multiple environmental parameters, including water content, partial pressure, and Cl⁻. The partial pressure is the most critical factor. Most of the final products of the corrosion reaction are mainly ferrous carbonate and iron oxides.

(2) H₂S is ionized to be acidic after dissolved in water, resulting in electrochemical corrosion of the pipe and local pitting perforation. Pressure, temperature, and Cl⁻ concentration all have an effect on the corrosion of H₂S. Among them, the partial pressure of H₂S will act synergistically with pH and temperature to exacerbate the corrosion of the pipe.

(3) The coexistence of CO₂ and H₂S corrosion is dominant in the actual working conditions, and the partial pressure ratio has a significant effect on the corrosion process. However, due to the complexity of the coexistence system of CO₂ and H₂S, the competition and synergy mechanism of the two in the corrosion process is not clear. Therefore, it is an important research direction in the field of corrosion science in the future to strengthen the in-depth study of the corrosion behavior of CO₂/H₂S coexistence system and construct a comprehensive and accurate corrosion prediction model.

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