ABSTRACT: In order to solve the problem of pipeline erosion—corrosion under acidic conditions with different CO2 contents of natural gas hydrate (NGH), the influence of different pH values on the erosion corrosion of X80 steel under dynamic conditions was studied. The erosion and corrosion behavior of X80 pipeline steel in multiphase flow (0.5% sand + 3% NaCl aqueous solution) at different pH was studied by rotating a cylindrical electrode apparatus combined with the electrochemical method in this paper. By analyzing the electrochemical impedance spectroscopy of the Tafel curve, the results showed that under dynamic solid—liquid two-phase conditions, with the decrease of the pH value, the self-corrosion potential of X80 steel samples was positively shifted, and the corrosion current density showed a trend of gradually increase. When the pH value is 2.0–3.0, the ac impedance spectrum of X80 steel samples shows a reactance arc in the high-frequency region and an inductive reactance arc in the low-frequency region. With the increase of the pH value, the radius of the capacitance and resistance arc of the impedance spectrogram increases gradually, and the time of the Bode spectrogram is “half peak” in the low-frequency region. As the time constant increases, the impedance decreases and the phase angle decreases; the overall polarization resistance $R_p$ on the surface increased gradually and the corrosion rate decreased significantly. Therefore, the pH value of the content of NGH with different CO2 contents has obvious influence on the solid—liquid two-phase erosion corrosion of X80 steel. With the increase of the pH value, the corrosion resistance of X80 steel increases, the corrosion rate decreases, and the corrosion strength decreases.

1. INTRODUCTION

With the rapid development of the oil and gas industry, the exploitation of gas hydrate has been paid more attention to. As a new clean energy, it has huge reserves, high energy density, and no pollution after combustion. As being processed on the seabed, natural gas hydrate (NGH) is transported to the surface of the sea through vertical pipelines, so that further treatment and utilization can be carried out. X80 pipeline steel, because of its high strength, high toughness, and excellent antistress corrosion cracking performance, is well applied in the transportation of oil and natural gas and is also widely used in the undersea transportation of NGH. Therefore, it is necessary to study the two-phase erosion corrosion of X80 pipeline steel at different pH values, so as to prevent or even avoid the occurrence of accidents during the transportation process in advance.

As a transportation pipeline, when X80 steel transports oil or natural gas, the sand-containing solid particles inside it will combine with water to form a multiphase acid corrosive medium in which gas, liquid, and solid coexist, which can easily cause corrosion in the pipeline. Studies have shown that acidity of medium solution is a key factor for pipeline corrosion.1–3 Zhang Yani et al.4 proposed that the corrosion and wear of samples in different pH NaCl solutions had orthogonal interaction with the weakest interaction when the pH value is 7 through studying the corrosion and wear behavior of P110S Tubing Steel in different pH NaCl solutions. Potentiodynamic polarization curves, electrochemical impedance spectroscopy (EIS), and three-dimensional video microscopy were used in the research of Liu et al.1 to discuss the corrosion electrochemical behavior of X80 steel in red soil-simulated solution with different pH values. Results showed that the corrosion of X80 steel in red soil-simulated solution was controlled by oxygen depolarization when the pH value was 5.5, and there was a corrosion product-binding layer on the surface of the sample. With the decrease of the pH value, corrosion of X80 steel gradually changed to electrochemical activation control. When the pH value dropped to between 4.0 and 3.0, many corrosion spots appeared on the surface of X80 steel. Wang et al.7 suggested a gradual weakened effect of corrosion phenomena and a gradual decrease with the increase of the pH value by studying the electrochemical corrosion

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behavior of X70 steel in simulated solution of Chengdu soil. Yang et al. studied the corrosion behavior of 5083 Al−Mg alloy in NaCl solution of different pH values based on SECM. The results showed that in the acid system, with the increase of the pH value, the corrosion potential was positively shifted, the pitting potential is not significantly different, the dimensional blunt current density is reduced, and there was only one capacitive arc in the EIS showing a contraction trend. In the above study, static experiments have been widely adopted in previous studies when studying the influence of the pH value on corrosion behavior of X80 steel with few research studies on multiphase flow environment under dynamic conditions.

The above research studies mostly adopt static experiments to study the influence of the pH value on the corrosion behavior of pipe steel, but there are few relevant research studies on X80 steel in the multiphase flow environment under dynamic conditions, and the change span of the pH value is not large, especially the pH value of CO2 NGH with different contents on the seabed. Based on X80 pipeline steel as the research object, this paper on NaCl solution as a corrosive medium, by adopting the method of electrochemical corrosion tests and morphology observation method to study the pH value of different concentrations of CO2 gas hydrate on the corrosion behavior of X80 pipeline steel scouring effect, for the effective control of submarine pipelines of NGH scouring corrosion harm, guarantee the safe operation of pipelines.

2. BACKGROUND

NGH is an ice-like solid compound that is formed (or can be formed) naturally from small gas molecules such as CO2 and H2S and water molecules under certain pressure and temperature conditions. The composition of NGH in the seabed and tundra is very complex. As shown in Figure 1, hydrates in the tundra often contain CH4, C2H6, C3H8, CO2, and n-C4H10, among which the content of C3H6 and C3H8 is as high as 21%. However, it was also found in the hydrate samples in Wu Li area of Qinghai that the content of CO2 is generally more than 90% and the content of CH4 is only less than 40%. In NGHs in the East China Sea, in addition to methane hydrates, CO2 hydrates are no longer rare, and the content of CO2 is as high as 84%. As shown in Figure 2, as the concentration of CO2 increases, the pH value in the solution will become lower and lower, even lower than 3.84. At this time, the solution will be extremely acidic. Therefore, the higher the concentration of CO2 in NGH, the more acidic it will be. In the transportation of NGH, especially in the transportation of NGH or CO2 gas hydrate with high CO2 content, solid−liquid two-phase flow will be involved in the pipeline, and the acid corrosion of CO2 will be very serious. Therefore, in this paper, mixed solutions with different pH values were used to simulate NGH with different CO2 content for experiments, so as to study the influence of acidity of NGH with different CO2 content on erosion−corrosion of X80 steel.

3. DISCUSSION AND ANALYSIS OF EXPERIMENTAL RESULTS

3.1. Potentiodynamic Polarization Curve. Figure 3 shows the polarization curves of X80 steel in sand−salt solution.

![Figure 1. Distribution characteristics of NGH in China.](image1)

![Figure 2. Relation curve between CO2 concentration and pH.](image2)

![Figure 3. Potentiodynamic polarization curves of X80 pipeline steel at different pH values.](image3)
solution at different pH values, and Table 1 shows the corresponding fitting results. As can be seen from Figure 3, the polarization diffusion phenomenon exists in the cathodic polarization curve when the pH value is 4.0, 5.0 and 6.5, revealing that the scour corrosion of X80 steel is controlled by oxygen depolarization under this condition. As the acidity decreases to the pH value 2.0 and 3.0, the polarization diffusion phenomenon on the cathodic polarization curve disappears in activation and dissolution state where the scour corrosion of X80 steel is controlled by electrochemical activation. The results indicate a low concentration of hydrogen ions in the solution when the pH value is between 3.0 and 6.5, where oxygen reduction reaction is the main chemical reaction of the cathodic reaction of X80 steel erosion−corrosion. When the pH value decreases, the concentration of hydrogen ions in the solution increases, the O₂ reduction is inhibited, the effect of oxygen depolarization in the cathodic reaction is weakened, and the scour corrosion reaction is transformed into activation control. As shown in Table 1, with the decrease of the pH value, the self-corrosion potential is positively shifted, and the corrosion current density presents a trend of gradual increase. The results indicate that the decrease of the pH value aggravates the erosion−corrosion of X80 steel.

3.2. Electrochemical Impedance Spectroscopy. EIS test results under different acidity levels are shown in Figure 4. As can be seen from Figure 4a,b, when the pH value is 2.0−3.0, the erosion−corrosion behavior of X80 steel in sandy salt solution is basically similar. EIS test results reveal that capacitive arc resistance is in the high-frequency region and inductive arc resistance in the low-frequency region. With the increase of the pH value, the high frequency of the ac

Table 1. Fitting Results of Polarization Curves for X80 Pipeline Steel at Different pH Values

| PH   | Ecorr/VAg/AgCl | Icorr/A·cm⁻² | βa/mV·dec⁻¹ | βc/mV·dec⁻¹ |
|------|----------------|--------------|-------------|-------------|
| 2    | −0.471         | 7.532 × 10⁻³ | 5.505       | 5.292       |
| 3    | −0.558         | 4.283 × 10⁻³ | 5.64        | 3.238       |
| 4    | −0.611         | 6.907 × 10⁻⁴ | 8.728       | 1.259       |
| 5    | −0.634         | 6.453 × 10⁻⁴ | 10.603      | 1.65        |
| 6.5  | −0.665         | 5.862 × 10⁻⁴ | 9.840       | 1.278       |

Figure 4. EIS of X80 pipeline steel under different pH values. (a) pH = 2, 3. (b) pH = 4, 5, 6.5. (c) Impedance diagrams. (d) Phase angle diagram.
impedance spectrum shows a little incomplete capacitive reactance arc, while the low-frequency reactance arc disappears, and the incomplete capacitive reactance arc appears in the low-frequency region; when the pH value ranges from 4.0 and 6.5, EIS is composed of three incomplete capacitive reactance arcs. Obviously when the pH value increases, the capacitive reactance arc radius of the ac impedance spectrum increases gradually. The Bode spectrum shows “half peak” in the low-frequency region, and the time constant increases, the impedance decreases, and the phase angle also decreases. This shows that the corrosion resistance of X80 steel increases and the corrosion strength decreases with the increase of the pH value. It is generally believed that the capacitive reactance arc represents the electrochemical reaction and the inductive reactance arc represents the interface property: the generation of the inductive arc may be caused by the adsorption process or pitting nucleation, or the two processes may exist at the same time. When the pH value increased from 2.0 to 6.5, the low-frequency area to form a new capacitive reactance arc may be due to the adsorption electrode surface corrosion products and change of the electrode surface state, forming a new corrosion resistance and slowing down the corrosion. Microcapacitive reactance arcs appeared in the high-frequency region, probably because Cl$^{-}$ is up to 3.5% in the medium, and the corrosion products are continuously subjected to mechanical erosion by particles under the action of Cl$^{-}$, so pitting pits are easy to form on the surface of the sample. When the pH value is 2.0–3.0, the low-frequency arc appeared at low frequency. Visibly, when pH is low, the process on the surface of the electrode includes not only the overall average polarization but also the process of local activation polarization, mainly by the adsorption process of H$^{+}$ ions attached to the surface of the sample. The integrity of the surface oxidation film was destroyed by dissolution, and uneven corrosion was found when the metal is exposed to the acidic solution.

In order to further analyze the influence of the pH value on the scour corrosion behavior of X80 steel, equivalent circuits as shown in Figure 5 were used for fitting. Where $R_{0}$ represents the solution resistance; $Q_{dl}$ represents the double-layer capacitance of the reaction interface; $R_{p}$ is the overall polarization resistance of the sample. Because the corrosion product exists on the electrode surface when the pH value is 4.0–6.5, the equivalent components $Q_{dl}$ and $R_{p}$ of the corrosion product layer are introduced into the equivalent circuit diagram. $Q_{dl}$ represents the capacitance element of the corrosion product layer, and $R_{p}$ represents the resistance of the corrosion product layer. When the pH value is 2.0–3.0, there are no corrosion products on the electrode surface, and the pitting nucleation leads to low-frequency inductive reactance arc, so only $R_{0}$ and $Q_{dl}$ need to be introduced into the equivalent circuit to reflect the relaxation behavior of pitting nucleation. That is, $R_{0}$ represents the polarization resistance at the pitting nucleus, and $Q_{dl}$ represents the change of film thickness at the pitting nucleus. In addition, because of the dispersion effect of double-layer capacitance, constant phase angle element $Q$ is used to replace capacitance $C_{dl}$ in the equivalent circuit, $Q = Y_{0} - 1/(j\omega n)$, where $Y_{0}$ is the admittance constant and $n$ is the dispersion index. The fitting results are shown in Table 2.

The circuit diagram (a) and (b) of Figure 5 were used to fit the ac impedance spectrum (a) and (b) of Figure 4, respectively. The fitting results are shown in Table 2. In view of the limitations of the conventional electrochemical testing methods in studying local corrosion, only the overall corrosion of the electrode is considered here, and then, only the relationship between variables and $R_{p}$ is studied. Figure 6 is obtained by plotting the pH value and $R_{p}$ data in Table 2.

As shown in Figure 6, the effect of pH under sand-bearing conditions can be summarized as follows. When the pH is 2 to 6.5, the overall polarization resistance $R_{p}$ of the sample surface gradually increases with a significant decrease of the corrosion rate. Meanwhile, when the pH is 3, $R_{p}$ changes most strikingly, which is similar to the test results of Liu et al. When the pH value is low, the concentration of H$^{+}$ is relatively high. Under the action of mechanical scouring, the protective effect of the corrosion product film is no longer obvious, and the corrosion rate significantly increased, so $R_{p}$ is relatively high. When the pH value is large, the bonding layer of the corrosion product formed on the surface of the working electrode will play a certain role in inhibiting corrosion.

3.3. Observation of Corrosion Morphology. Figure 7 shows the corrosion morphology of X80 steel after erosion corrosion in sand–salt solutions with different pH values. Figure 7a,b are corrosion morphologies of X80 steel after erosion in slurry with the pH values of 2 and 3, respectively. There are dense hole-shaped erosion pits distributed on the surface of the sample, and pitting corrosion appears on the sample because pitting corrosion pits will occur on the surface of the material because of the involvement of the reducing agent H$^{+}$ in the solution, while the solid particles in the solution will impact the exposed metal, which aggravates the damage to the surface of the sample. Figure 7c–e shows corrosion morphology diagrams of X80 steel after erosion corrosion in slurries with the pH values of 4, 5, and 6.5, respectively. Compared with Figure 7a,b, the number of corrosion pits in Figure 7c–e decreases and the depth became shallow, and the surface of the sample after erosion–corrosion as a whole relatively leveled off, indicating that the strength of erosion corrosion of X80 steel decreased. This is because the H$^{+}$ concentration decreased and the concentration of OH$^{-}$ in high pH solution is opposite bigger, leading to the surface of the electrode being prone to generate protective Fe(OH)$_{3}$, which will prevent the diffusion of corrosive ions to the surface of the anode and hinder the dissolution of iron. It can be seen that with the increase of the pH value, pitting pits on the
Table 2. Electrochemical Impedance Fitting Results of X80 Pipeline Steel at Different pH Values

| pH  | $R_p$/Ω·cm² | $Q_u$ | $n$ | $R_e$/Ω·cm² | $L_i$/H·cm⁻² | $C/F$·cm⁻² | $R_e$/Ω·cm² |
|-----|-------------|-------|-----|-------------|-------------|------------|-------------|
| 2.0 | 2.794       | 0.01572 | 0.8  | 1.084       | 4.423       | 4.865      | 1.132       |
| 3.0 | 2.89        | 0.004198 | 0.8083 | 13.38     | 4.37        | 8.855      | 1.186       |
| 4.0 | 1.98        | 0.00685  | 0.9125 | 38.2       | 29.47       | 4.183 × 10⁻⁶ | 1.24        |
| 5.0 | 1.805       | 0.006542 | 0.9024 | 53.33      | 3.205 × 10⁸ | 3.568 × 10⁻⁶ | 0.4401      |
| 6.5 | 2.045       | 3.627 × 10⁻⁶ | 1     | 97.85      | 18.96       | 10.84      | 97.85       |

Figure 6. Relationship between the pH value and $R_p$ value of salt resistance in sand containing salt solution.

The influence of pH values of NGH with different CO₂ content on the two-phase erosion corrosion of X80 steel pipeline under dynamic conditions was studied by a rotating cylindrical electrode experimental device. The electrochemical measurement and corrosion morphology observation methods were used to obtain the following conclusions:

(1) When the pH value ranges from 4.0 to 6.5, the polarization diffusion phenomenon exists in the cathodic polarization curve and the erosion corrosion of X80 steel was dominated by oxygen depolarization. When the pH value is 2.0–3.0, the polarization diffusion phenomenon on the cathodic polarization curve disappears and is shown to be in the state of activation solution, where the scour corrosion of X80 steel is controlled by electrochemical activation. With the decrease of the pH value, the corrosion current density presents a trend of gradual increase.

(2) When the pH value is between 2.0 and 3.0, the ac impedance spectrum shows a small incomplete capacitive reactance arc, while the low-frequency reactance arc disappears, and the incomplete capacitive reactance arc appears in the low-frequency region; when the pH value ranges from 4.0 to 6.5, EIS consists of three incomplete capacitive reactance arcs. With the increase of the pH value, the arc radius of capacitive reactance of the ac impedance spectrum increases gradually. The Bode spectrum shows “half peak” in the low-frequency region, with the time constant increasing, impedance decreasing, and phase angle decreasing; when the pH value is between 2 and 6.5, the overall polarization resistance $R_p$ on the sample surface increases gradually and the corrosion rate decreases significantly. Therefore, the corrosion resistance of X80 steel increases, and the corrosion strength weakens as the pH value increases.

5. EXPERIMENTAL SECTION

5.1. Methods. In order to study the influence of pH of corrosion medium on erosion—corrosion behavior, the corrosion medium was prepared with five different pH values of deionized water, with pH values of 2.0, 3.0, 4.0, 5.0, and 6.5, respectively, NaCl concentration of 3.5% (mass concentration), sediment concentration of 2% (mass fraction), medium flow rate of 2 m/s (converted to speed of 2546 t/min), experimental temperature of room temperature (20 °C), and scour erosion time of 12 h.

The working electrode material is X80 steel, with its chemical composition shown in Table 3. The sample is featured by an outer diameter of 15 mm, an inner diameter of 9.53 mm, a height of 6.39 mm, and a working area of 3.01 × 10⁻⁴ m². The sample was polished to a specification of 1000 # by SiC sandpaper step by step, then degreased with acetone, washed with distilled water, and finally air-dried with cold air to record the weight of the sample before the experiment.

5.2. Electrochemical Measurement. The electrochemical measurement was carried out on a CHI920D electrochemical workstation in Chenhua, Shanghai. Using a three-electrode system, working electricity is the X80 pipeline steel sample, reference electricity is Ag/AgCl, auxiliary electricity is platinum sheet.

5.2.1. Potentiodynamic Polarization. The scanning rate of the potentiodynamic polarization curve test is 1 mV/s, and the scanning range is ±500 mV of self-corrosion potential. The corresponding corrosion current density and polarization slope of anode and cathode were analyzed by Powersuite.

5.2.2. Electrochemical Impedance Spectroscopy. At the self-corrosion potential, the measured frequency range is 0.01 Hz to 10 MHz.
Hz to 100 kHz and the disturbance voltage amplitude is 10 mV. ZSimpWin software was used to fit the electrochemical impedance data, and the parameters of each element were obtained.

5.3. Analysis and Microstructural Characterization. After the erosion corrosion, the sample was washed with hydrochloric acid and hexamethylenetetramine. After air-drying, a Smartzoom 5 digital microscope was used to observe and analyze the corrosion morphology of the sample surface.

![Image](https://example.com/image.png)

**Figure 7.** Erosion and corrosion morphology of X80 pipeline steel at different pH values. (a) pH = 2. (b) pH = 3. (c) pH = 4. (d) pH = 5. (e) pH = 6.5.

| C   | Si  | Mn  | P   | Cu  | Cr  | Ni  | Mo  | Nb  | Ti  | Al  | Fe  |
|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| 0.036 | 0.200 | 1.771 | 0.012 | 0.220 | 0.223 | 0.278 | 0.184 | 0.110 | 0.019 | 0.021 | allowance |

**Table 3. Chemical Composition and Content of X80 Pipeline Steel (Mass Fraction/%)**

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**Notes**

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