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Study on Cavitation Behavior of Pipe String in Gas Storage Well under Gas-Liquid Environment

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In the long-term and continuous operation process of UGS injection-production well, the outer casing joint of the oil string is prone to failure. This causes gas to enter into the annular space between the casing and string forming the gas-liquid two-phase, leading to the issue of cavitation of the string. Therefore, herein, the string cavitation caused by the gas leakage in UGS gas well during the injection and production process is analyzed through electrochemical measurements and image characterization. The results show that the pipe string had no corrosion characteristics during the long time of injection and production owing to the action of corrosion inhibitor and passivation film of 13 Cr. During the 24 h experiment, when the leakage pressure reached 5 MPa, the pipe string was mainly layered corrosion. When the pressure increased to 10 MPa, the formation and rupture of gas bubbles on the metal surface led to the destruction of two protective films, and clear cavitation pits appeared on the metal surface. Greater the pressure, more prominent was the cavitation effect.

Keywords: Cavitation behavior; UGS gas well; Electrochemical reaction; Gas-liquid two-phase environment

1. INTRODUCTION

Different from the conventional gas wells, most of the UGS (Underground natural gas storage) well groups in China are complex deep wells with depth of more than 4000 m, facing harsh working conditions such as high temperature (>160 °C), pressure (>140 MPa), and density [1-3]. The poor well conditions bring great challenges to the integrity of UGS construction and subsequent long-term production. In the UGS well studied in this work, the well depth is calculated to be 4000 m, the temperature gradient is 1 °C / 30 m, and the pressure fluctuation range within the annulus is determined to be 5–20 MPa.

During the long-term and continuous operation process, due to the operation mode of strong

production and injection in the UGS well, the joint position of the outer casing is prone to failure under the action of external alternating tensile stress, causing the gas to enter the annular space between the casing and the string [4,5]. Meanwhile, to reduce the friction between the pipe string and casing, annulus protection fluid is mostly used for lubrication. At this time, leaked gas enters to form a gasliquid mixture. Due to the reciprocating movement of the string, the temperature and pressure in the annulus also changes under different well depth conditions, and the gas-liquid two-phase flow state in the annulus also varies repeatedly (see Fig.1). The change in flow state also signifies precipitation, dissolution, generation, and rupture of bubbles. In the reciprocating movement of the tubing string in the injection-production wells, cavitation caused by the leaking gas is one of the key factors causing the failure of tubing string [6-9]. However, till date, the cavitation problem of the string is mostly targeted at the conventional gas wells, namely depth of <2500 m, pressure of <25 MPa, and temperature of 60–80 °C, which is significantly better than the service conditions of the string of UGS well group. Therefore, the research on cavitation erosion of UGS well string group is of great importance for the vibration characteristic analysis and safety evaluation of the string after leakage.

Therefore, herein, the pipe string in service environment of UGS gas well is taken as the research object, and the evolution process of material corrosion dynamics in the cavitation process of the pipe string is established through in-situ electrochemical measurement method and microscopic corrosion image characterization.



Figure 1. Gas-liquid two-phase flow in the annulus of UGS well

2. EXPERIMENT SETTINGS

Based on high temperature-pressure reaction kettle, the three-electrode test system was adopted

to conduct the electrochemical tests (see Fig.2). Annular protection liquid was adopted as the experimental solution. The reactor was pressurized with high purity N_2 and the pressure was monitored by pressure valve in real time. The temperature was adjusted with the temperature controller, and the flow velocity was controlled by rotating the blade. In the three-electrode test system, the working electrode (WE) was 13 Cr with the size of $25 \times 25 \times 2 \text{ mm}^3$ (the elemental composition in Tab.1), the reference electrode (RE) was SCE (saturated calomel electrode), and the auxiliary electrode (AE) was Pt electrode. The reference electrode was in contact with the solution through the Luggin-Haber capillary and the distance between the tip and the X80 steel surface was less than 2 mm to ignore the IR drop due to solution [10].

Working electrode pretreatment: After removal of oil with acetone, the electrode was cleaned with the deionized water, dehydrated with anhydrous ethanol, dried, and placed in a vacuum dryer for 24 h for reserve use. Prior to the experiment, waterproof sandpaper #800-#1200 was employed to polish the surface step by step until there were no clear scratches, and then, the surface was polished.

Preparation of solution. The experimental solution was the annular protective solution with sterilization, de-oxygenation, and corrosion inhibition functions in the field. It was mainly composed of a combination of 5000 mg/L inhibitor, 3 g/L de-oxygenation agent, and 175 mg/L fungicide, with the liquid density of 1.02 g/cm^3 and pH of 7.73.

Experimental conditions: As shown in Fig.1, the annular space was 893 m deep, calculated temperature was about 70 °C, and the calculated annular pressure was 5–20 MPa. Under the studied experimental conditions, the gas-liquid ratio of the mixed fluid was described by the pressure value in the reactor, and the cavitation process of the fluid was described by setting the stirring speed of 10 m/s.



Figure 2. Experimental apparatus: high temperature - pressure reactor

Experimental setup. The workstation used for the electrochemical test was PARSTAT 2273. The scanning rate of polarization curve was set as 0.3 mV/s, and the scanning potential range was ± 250 mVvs.OCP. The electrochemical impedance measurement range was 10^2-10^{-5} Hz, and the

disturbance voltage was ± 10 mV. When the fluctuation of the open-electrode potential was less than 5 mV in 300 s [11], the whole system was considered to be in the stable state, and the polarization curve and electrochemical impedance test could be performed.

Data processing. Electrochemical data processing was performed using "PowerSuite" software and "ZSimpWin" software.

Three groups of test pieces were set for each working condition, of which two groups were used for electrochemical test and the third group was used for corrosion morphological observation. After the specimens were removed and cleaned, the surface morphology was observed under scanning electron microscope (Nova Nano SEM450).

 Table 1. 13Cr element composition (Mass fraction, %)

С	Si	Mn	Cr	Mo	S	Р	Cu	Ni	Ν	V
0.03	0.20	0.35	13.00	1.00	0.003	0.015	0.25	4.00	0.02	0.02

3. RESULTS AND DISCUSSION

3.1 Analysis of corrosion kinetics process

3.1.1 At the beginning of the experiments

Fig.3 shows the change of open-circuit potential (E_{OCP}) and polarization curve of 13Cr under different pressure conditions at the beginning of the experiments of stabilization. With the increase in pressure in the annulus, the open-circuit potential displayed a trend of positive deviation, which indicates that the corrosion tendency of the 13Cr increased. At the same time, when the pressure increased from 5 MPa to 10 MPa, the open-circuit potential demonstrated a large positive deviation, up to 140 mV, suggesting that the reaction process on the metal surface underwent a fundamental change [12].

From the point of the field test results, when the pressure in the annulus reached 5 MPa, prominent gas leak in the annulus had occurred. Under the condition of 0–5 MPa pressure, two polarization curves were synchronized, at which corrosion potential (E_{corr}), corrosion current density (I_{corr}), and cathode / anode Tafel constant (β_a and β_c) were generally equal. Moreover, there was clear passivation range in the anodic polarization curves, indicating that the solution could be approximately regarded as the single liquid phase with little dissolved gas. Under these circumstances, the corrosion was inhibited by the corrosion inhibitor and passivation film of Cr, and the impact of dissolved gas could be neglected [13-15]. With the increase of gas pressure in the annulus, the gas solubility in the protective solution was also increased. When the pressure was 10 MPa, the whole polarization curve moved towards the right and upwards, the corrosion rate increased, and the corrosion potential was positively shifted. However, there was still a clear passivation interval in the anodic polarization curve. However, with the further increase in pressure to 15 MPa and 20 MPa, the anode passivation area completely disappeared and the metal displayed activated corrosion, at which the corrosion rate may

increase by 6-7 times [16].

As shown in the Tab.1, both corrosion potential and corrosion current density increased with the increase in pressure. At the same time, when the pressure was greater than 5 MPa, both of them increased rapidly, indicating that the corrosion process on the metal surface had changed significantly. From the system as a whole, in the gas-liquid two phase environment, firstly, annulus protection fluid formed a protective film on the metal surface. Secondly, 13 Cr is a stainless steel and can form a layer of passivation film to slow down the corrosion of tubing string [17,18]. However, with the rise in pressure and under the action of flow rate, formation and rupture of gas bubble on the metal surface directly led to the destruction of two layers of protective film [19-21]. Therefore, when the pressure increased to 15 MPa, the metal activation corrosion morphology was displayed.



Figure 3. Open-circuit potential curves and polarization curves of 13Cr in annular protective solution under different leaked-gas pressure of 0-20 MPa with the flow velocity of 2 m/s at the beginning of the experiments

Table 1. The fitting results of polarization curves of 13Cr in annular protective solution under different leaked-gas pressure of 0-20 MPa with the flow velocity of 2 m/s at the beginning of the experiments

Pressure /(MPa)	0	5	10	15	20
$E_{\rm corr}/({\rm mVvs.SCE})$	-980	-972	-821	-825	-787
$I_{\rm corr}/(\mu A/m^2)$	0.11	0.12	0.20	0.27	17.29
$\beta_{\rm a}/({\rm mV/dec})$	114	123	311	382	542
$\beta_{\rm c}/({\rm mV/dec})$	1047	912	567	454	118
$r=eta_{a}/eta_{c}$	0.11	0.14	0.55	0.84	4.61

Fig.4 displays the electrochemical impedance curves of 13Cr under different pressure conditions at the beginning of the experiments. Under pure liquid phase condition, Nyquist diagrams displayed clear bi-capacitation resistance, signifying the protective film of the annulus and passivation film of 13Cr. Therefore, the fitting circuit was selected as $R_s(QR_p)(Q_{dl}R_{ct})$. With the increase in pressure, under the action of flow velocity, Nyquist diagrams displayed clear Warburg diffusion characteristics, signifying that the formation - rupture process of gas double at the metal surface damaged the protective films [22,23]. Therefore, the fitting circuit was selected as $R_s(QR_p)(Q_{dl}(R_{ct}W)$: R_s as solution resistance, Q as the system capacity, R_p as the polarization resistance, Q_{dl} as passivation membrane capacitance matrix, R_{ct} as the charge transfer resistance, and W as Warburg diffusion impedance. The fitting results are shown in Tab.2, where the fitting errors were all of the order of 10⁻⁴, displaying a high fitting accuracy [24].

With the gas leakage, the pressure in the annulus increased. However, when the pressure increased from 0 to 5 MPa, the high frequency characteristic of 13Cr did not change, while the mass diffusion process occurred at low frequency, indicating that the outer corrosion inhibitor layer was affected at this time [25]. With further increase in pressure, the capacitor-reactance radius of both the high frequency and low frequency decreased, indicating that the damage of the gas bubble to the double layer was intensified. As shown in the Bode plots, when the pressure reached 10 MPa, the characteristic of intermediate frequency resistance had completely disappeared. When the pressure reached 15–20 MPa, the bilayer membrane had less capacitor resistance. In the Nyquist diagrams, there was a large Warburg diffusion tail, while in the Bode plots, the impedance dropped rapidly and maximum phase angle occurred at the high frequencies. These results indicate that the bilayer membrane structure was completely destroyed and the metal structure was in the process of activated corrosion [26,27]. This result is the same as that of the polarization curve analysis.

The change rules of the relevant fitting results with pressure showed that the two capacitances namely Q and Q_{dl} increased with the rise in pressure, while the resistances of R_p and R_{ct} decreased. As per the change law of charge transfer resistance (R_{ct}), when the pressure was greater than 10 MPa, the charge transfer resistance basically remained unchanged, suggesting that the surface passivation layer was completely destroyed and the metal had entered the activated corrosion stage, i.e., cavitation occurred.





Figure 4. EIS curves of 13Cr in annular protective solution under different leaked-gas pressure of 0-20 MPa with the flow velocity of 2 m/s at the beginning of the experiments

Table 2. Fitting results of EIS curves of 13Cr in annular protective solution under different leaked-gaspressure of 0-20 MPa with the flow velocity of 2 m/s at the beginning of the experiments

Pressure /(MPa)	0	5	10	15	20
$R_{\rm s}/(\Omega\cdot{\rm cm}^2)$	1.84	1.89	2.04	3.68	2.66
$Q/(\mathrm{F/cm^2})$	1.88×10 ⁻⁶	1.88×10 ⁻⁶	1.66×10 ⁻⁶	1.71×10 ⁻⁶	2.73×10 ⁻⁴
n	0.49	0.70	0.75	0.51	0.70
$R_{\rm p}/(\Omega \cdot {\rm cm}^2)$	1.55×10^{5}	1.31×10^{5}	7.93×10 ⁴	3.33×10 ³	1.88×10^{2}
$Q_{ m dl}$ /(F/cm ²)	2.63×10 ⁻⁸	8.74×10 ⁻⁷	9.67×10 ⁻⁷	1.40×10 ⁻⁵	9.55×10 ⁻⁵
п	0.68	0.65	0.65	0.60	0.43
$R_{\rm ct}$ /($\Omega \cdot {\rm cm}^2$)	1.11×10^{5}	9.40×10^4	1.73×10^{3}	2.32×10^{3}	2.01×10^{2}
$W/(\Omega \cdot \mathrm{cm}^2)$	-	2.99×10 ⁻⁵	1.67×10 ⁻⁴	1.01×10 ⁻⁴	1.03×10 ⁻³
Error	1.78×10 ⁻⁴	3.26×10 ⁻⁴	4.19×10 ⁻⁴	0.93×10 ⁻⁴	7.73×10 ⁻⁴

3.1.2 Measurement after 24h

Fig.6 shows the open-circuit potential and polarization curves of 13Cr under different pressure conditions after experiments for 24 h. The fitting results are shown in Tab.3. Compared with the stage at the beginning of the experiments, the open-circuit potentials of 13Cr were all positively offset under different pressure conditions, indicating that with the progress in corrosion process, the corrosion tendency of 13Cr increased [28]. Under the action of flow rate, the double-layer film on the metal surface was in the process of continuous destruction and formation under the action of erosion. At this time, the polarization corrosion process was in the activation corrosion process, and prominent anodic passivation interval completely disappeared. These results suggest that the double-layer protective film structure on the metal surface was damaged by the leaked gas under long-term experimental conditions. At the same time, with the rise in pressure, the corrosion potential and corrosion current

density increased significantly, suggesting that with more severe damage of bubbles to the metal surface, the corrosion control process changed from anode control to cathode control [29].



Figure 6. The open-circuit potential curves and polarization curves of 13Cr in annular protective solution under different leaked-gas pressure of 0-20 MPa with the flow velocity of 2 m/s after experiment 24 h

Table 3. The fitting results of polarization curves of 13Cr in annular protective solution under differentleaked-gas pressure of 0-20 MPa with the flow velocity of 2 m/s after experiment 24 h

Pressure /(MPa)	0	5	10	15	20
$E_{\rm corr}/({ m mVvs.SCE})$	-784	-780	-737	-704	-679
$I_{\rm corr}/(\mu A/m^2)$	1.20	1.29	7.97	8.43	12.46
$\beta_{\rm a}/({\rm mV/dec})$	141	249	89	80	56
$\beta_{\rm c}/({\rm mV/dec})$	120	91	115	90	109
$r=\beta_{a}/\beta_{c}$	1.17	2.74	0.77	0.89	0.52

Fig.7 displays the electrochemical impedance curves of 13Cr under different pressure conditions after experiments for 24 h. After long time of experiment, under the effect of flow velocity, double membrane structure on the surface was in the destruction and generation process. Therefore, under the condition of pure liquid, Warburg apparent diffusion impedance characteristics occurred in its Nyqisut diagram. Hence, the fitting circuit was selected as $R_s(QR_p)(Q_{dl}(R_{ct}W): R_s$ as the solution resistance, Q as the system capacity, R_p as the polarization resistance, Q_{dl} as the passivation membrane capacitance matrix, R_{ct} as the charge transfer resistance, and W as the Warburg diffusion impedance. As the pressure increased, the metal substrate was completely exposed to the corrosion environment at this time, and the polarization curve displayed that the metal was in the activated corrosion state at this time. Therefore, under the pressure of 10–20 MPa, there were two time constants in the Bode plots. Hence, the fitting circuit was selected as $R_s(Q(R_p(C_{dl}R_{ct})))$, where C_{dl} is the capacitance of the double electric layer. The fitting results are shown in Tab.4, where the fitting errors were all of the order of 10⁻⁴, demonstrating a high fitting accuracy.



Figure 7. EIS curves of 13Cr in annular protective solution under different leaked-gas pressure of 0-20 MPa with the flow velocity of 2 m/s after experiment 24 h

As per the electrochemical impedance curves, under the conditions of pure liquid (0–5 MPa), due to the flow action, the double membrane diffusion occurred, while the radius of capacitive reactance arc decreased due to the increased pressure, presenting an increased capacitance $(Q=7.27\times10^{-7}\rightarrow1.00\times10^{-6} \text{ F/cm}^2)$ and decreased resistance $(R_p=2.15\times10^4\rightarrow1.17\times10^4 \ \Omega\cdot\text{cm}^2)$. Therefore, there were two time constants in the log*f*- φ curves at highest frequency (10⁵ Hz) and lowest frequency (10⁻² Hz), named as the mass diffusion process and charge transfer process, respectively. With the rise in pressure, the Nyqusit curves of 10–20 MPa displayed the characteristics of double capacitive reactance and the Warburg diffusion characteristics disappeared. This is because when the pressure reached 10 MPa, the corrosive environment changed from single liquid phase to the gas-liquid two-phase. Under long-term experiment, the double-layer film on the metal surface was destroyed, and the metal matrix was completely exposed to the corrosive environment, displaying the state of activated corrosion. At the same time, with the increase in pressure, the cavitation effect on the metal surface became stronger, leading to a gradual decrease in the double layer capacitance on the metal surface ($C_{dl}=2.51\times10^{-7}\rightarrow1.60\times10^{-8}$ F/cm²), while the charge transfer resistance remained basically unchanged ($R_{ct}=3.39\times10^{6} \ \Omega \cdot cm^{2}$).

Pressure /(MPa)	0	5	10	15	20
$R_{\rm s}/(\Omega\cdot{\rm cm}^2)$	1.24	1.52	3.40	3.21	1.45
$Q/(\mathrm{F/cm^2})$	7.27×10 ⁻⁷	1.00×10 ⁻⁶	2.50×10 ⁻⁶	4.11×10 ⁻⁶	8.06×10 ⁻⁶
n	0.49	0.50	0.50	0.46	0.44
$R_{\rm p}/(\Omega \cdot { m cm}^2)$	2.15×10^{4}	1.17×10^{4}	1.43×10 ⁴	1.41×10^{4}	4.81×10 ⁴
$Q_{ m dl}$ /(F/cm ²)	5.75×10 ⁻⁵	1.33×10 ⁻⁴	-	-	-
n	0.63	0.65	-	-	-
$R_{\rm ct}$ /($\Omega \cdot {\rm cm}^2$)	1.41×10^{4}	3.48×10^4	-	-	-
$W/(\Omega \cdot \mathrm{cm}^2)$	1.39×10 ⁻⁵	1.96×10 ⁻⁵	-	-	-
$C_{\rm dl}$ /(F/cm ²)	-	-	2.51×10 ⁻⁷	1.25×10 ⁻⁷	1.60×10 ⁻⁸
$R_{\rm ct}$ /($\Omega \cdot {\rm cm}^2$)	-	-	3.57×10^{6}	3.28×10^{6}	3.31×10 ⁶
Error	6.17×10 ⁻⁴	4.83×10 ⁻⁴	3.66×10 ⁻⁴	1.42×10^{-4}	1.29×10 ⁻⁴

Table 4. Fitting results of EIS curves of 13Cr in annular protective solution under different leaked-gaspressure of 0-20 MPa with the flow velocity of 2 m/s after experiment 24 h

3.2 Analysis of cavitation morphology

When the experimental time was 24 h, the sample was taken out and the surface corrosion products were removed using standard derusting liquid [30,31]. As shown in Fig.8, the local characteristics of metal surface under different gas pressures were analyzed via SEM. As shown in Fig.9, through binarization treatment, the distribution characteristics of the corrosion pits were observed on the whole metal specimen surface.

When there was no gas leakage, i.e., the gas pressure in the corrosive environment was 0, the entire metal surface was jointly protected by the corrosion inhibitor and passivation film, and the metal surface had no prominent corrosion characteristics, as shown in Fig.8(1). When the gas pressure leakage reached 5 MPa, although the internal corrosion environment could still be approximated as pure liquid phase, the erosion aggravated the damage to the double-layer membrane structure and the metal surface presented the characteristics of layered corrosion [32]. When the gas pressure increased to 10 MPa, clear cavitation characteristics appeared on the metal substrate. Higher the pressure, more were the cavitation pits, indicating that the corrosive environment changed into the gas-liquid two-phase and led to cavitation. As shown in the binarization treatment of the overall metal corrosion pit distribution, the number of cavitation pits increased significantly with the rise in pressure, indicating that cavitation was the main reason for the corrosion failure of the string due to gas leakage in the

injection and production process of gas wells.



Figure 8. Analysis of local cavitation characteristics of 13Cr surface in annular protective solution under different leaked-gas pressure of 0-20 MPa with the flow velocity of 2 m/s after experiment 24 h



Figure 9. Development of cavitation characteristics on 13Cr surfaces in annular protective solution under different leaked-gas pressure of 10-20 MPa with the flow velocity of 2 m/s after experiment 24 h

4. CONCLUSION

Herein, the string cavitation caused by gas leakage in injection and production process of UGS gas well was analyzed via electrochemical test and image characterization. The main conclusions are as follows:

(1) When no gas leakage occurred, the string was protected by the corrosion inhibitor and passivation film, and the matrix was well protected under the erosion conditions for a long time. No prominent corrosion characteristics appeared on the surface of the matrix. When clear gas leak occurred and the pressure in the environment increased to 5 MPa, the electrochemical behavior of the metal did not change substantially at the beginning of the experiments and 24 h of experiment, indicating that the corrosion environment at this time was still a single liquid phase. Under the pressure of 5 MPa, the erosion effect increased, and the metal surface was primarily subject to layered corrosion.

(2) When the pressure in the annulus increased to 10 MPa owing to gas leakage, the formation and rupture of gas bubbles on the metal surface led to the destruction of the double-layer protective film, and prominent cavitation pits appeared on the metal surface. Greater the pressure, more prominent was the cavitation effect.

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