Corrosion behavior of the expandable tubular in formation water

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Abstract: The corrosion behavior of expandable tubular materials was investigated in simulated downhole formation water environments using a series of electrochemical techniques. The corrosion morphologies in the real downhole environment after three months of application were also observed by stereology microscopy and scanning electron microscopy (SEM). The results show that, compared with the unexpanded sample, the area of ferrite increases dramatically after a 7.09% expansion. The expanded material shows a higher corrosion current in the polarization curve and a lower corrosion resistance in the electrochemical impedance spectroscopy (EIS) plot at every studied temperature. The determined critical pitting temperatures (CPT) before and after expansion are 87.5°C and 79.2°C, respectively. SEM observations demonstrate stress corrosion cracks, and CO₂ corrosion and H₂S corrosion also occur in the downhole environment. Due to additional defects generated during the plastic deformation, the corrosion performance of the expanded tubing deteriorates.

Keywords: steel corrosion; expandable tubes; formation water; electrochemical behavior

1. Introduction

With increasing service life and more rigorous drilling conditions, exploration for oil has become more and more challenging. Fortunately, expandable tubular techniques may help this situation. These techniques have been extensively applied in expandable open-hole liner systems (OHL), expandable cased-hole liner systems (CHL), and expandable liner hangers (ELH) [1–3]. Downhole solid expandable tubular (SET) fixing equipment allows the location and operation of every kind of tool needed for drilling, completion, well repair, and oil-formation improvement. Thus, repairing the casings of damaged wells, cutting windows, side drilling, dealing with washovers, and so on can all be easily performed [4]. By using the expandable tubular techniques, the loss of hole size during setting the successive casing strings is minimized, the amounts of raw materials and cost are reduced, and the well yields are greatly improved. Therefore, these techniques are hailed as a revolutionary advance in the history of drilling engineering and technology [5–7].

However, few reports exist on the corrosion performance

of expandable tubing after being expanded in the downhole. The downhole environment is extremely corrosive and complicated, including high temperature and high pressure (HTHP), corrosive acid gases such as CO₂ and H₂S, and ultra high chloride concentration [8–11]. Field data indicate that the corrosion rate of P110 steel is as high as 10 mm/a, and the tube failed due to cracks exceeding 20 km were reported in one oil field of western China due to the downhole corrosion after only five years. For expandable tubes, the mechanical properties along with stress-strain relation and microstructure changes are closely related to their corrosion resistance in the downhole environment [12]. Thus, their corrosion performance after expansion should be given more attention to prevent well corrosion failure.

In this work, the corrosion behavior of expandable tubular materials before and after expansion was studied in formation water by electrochemical methods. The corrosion morphologies were also observed by scanning electron microscopy (SEM). The results provided the supporting information for materials research on expandable tubes and quality control for engineering applications.

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2. Experimental

2.1. Materials and solutions

The chemical composition of a seamless steel tube employed is shown in Table 1. The tube was expanded by a hydraulic drive using a cone ingot. The tube served under the oil well together with an unexpanded tube for three months. The samples were cut into the dimensions of 10 mm × 10 mm × 3 mm, embedded in an epoxy resin, leaving a working area of 1 cm², then abraded using up to 1500 emery paper, washed in distilled water, and eventually dried by cold air.

Table 1. Chemical composition of carbon steel used for the expandable tubular

wt%								
C	Si	Mn	P	S	Cr	Ni	Mo	Fe
0.45	0.26	1.41	0.013	0.005	0.24	0.044	0.092	Bal.

The test solution was simulated formation water in western China with the composition listed in Table 2. The metallograph was etched by alcohol solution containing 4vol% nitric acid. All the solutions were prepared from distilled water and analytic grade reagents. The morphologies were observed by digital microscopy VHX-2000 (Keyence, Japan) and SEM (Quanta250).

Table 2. Chemical composition of formation water in western China

mg/L								
Na ⁺	Mg ²⁺	Ca ²⁺	Ba ²⁺	Cl ⁻	HCO ₃ ⁻	SO ₄ ²⁻	pH	Total mineralization
50055	774.08	7503	20.12	92137	436.2	657.7	5.86	151600

2.2 Electrochemical testing

Electrochemical tests were carried out using a Princeton Applied-VMP3 electrochemical system. A saturated calomel electrode (SCE) and platinum sheet were used as the reference and counter electrodes, respectively. The potentiodynamic polarization measurements were performed at a scanning rate of 1 mV·s⁻¹ at temperatures of 50, 70, and 90°C. Electrochemical impedance spectroscopy (EIS) tests were performed with an applied potential amplitude of 10 mV at open circuit potential in a frequency range of 100 kHz to 10 mHz. For potentiostatic polarization measurements, the applied potential was -0.6 V vs. SCE with a growing temperature rate of 0.6°C·min⁻¹ achieved using a water bath.

3. Results and discussion

3.1. Metallographic characterization

Fig. 1 presents the section ring images before and after

expansion, indicating the average sizes of $\phi 114.3$ mm × 7.02 mm and $\phi 122.4$ mm × 6.13 mm, respectively. The tubing was expanded by 7.09%. Based on the calculated measured sites as shown Fig. 1(c), the original wall unevenness was 3.2% and rose to 8.7% after expansion. This wall unevenness may be explained in two possible ways: (1) the unevenness was inherited and enlarged on the basis of the initial slight dimensional deviation during expansion; (2) the force direction of the expansion ingot and the central axis of the tube could not always remain in a straight line, resulting in a few migrations along the circumference. To diminish this effect, the lubrication and rotation of the ingot during expansion were advised. However, the tube still met the standard of the American Petroleum Institute (API), which is a maximum expansion of 12.5% [13].

The metallographs of the two samples are shown in Fig. 2. Before expansion, they consist of ferrite (F) and pearlite (P) with a relatively uniform distribution (as shown in Figs. 2(a) and (c)). After expansion in Fig. 2(b), the pearlite lamella changes little, while the ferrite significantly increases

Fig. 1. Expandable tubular images: (a) before expansion; (b) after expansion; (c) thickness measured sites.

Fig. 2. Metallographic images observed by stereological microscopy and SEM before ((a) and (c)) and after expansion ((b) and (d)).

with a large phase area filled. Thus, it can be concluded that ferrite was the primary contributor to the steel plastic deformation. Both the experimental and simulative results suggested that pearlite was responsible for the toughness of carbon steel, while its deformability was determined by ferrite [14–16]. One could imagine that this microstructural transformation would alter the steel's corrosion performance due to the change in the potential difference between two

grains, which was the driving force of electrochemical corrosion [17].

3.2. Electrochemical measurements

The potentiodynamic polarization curves of the carbon steel before and after expansion in formation water at different temperatures are shown in Fig. 3. Generally, the corrosion potential declines and the corrosion current increases

Fig. 3. Potentiodynamic polarization results of the carbon steel before and after expansion in formation water at different temperatures: (a) 50°C; (b) 70°C; (c) 90°C.

with increasing temperature for both the samples. In addition, the anodic polarization curves are not obviously different, indicating that the mechanism of anodic electrochemical reaction does not change. At the cathodic part, the limiting diffusion current of oxygen is evident at 50°C, but is weakened at 70°C and 90°C; this implies that the dominant cathodic reaction changes from oxygen reduction to hydrogen evolution when the temperature increases. This phenomenon is attributed to the transfer step and the activity of H^+ , which are favored at higher temperatures in the presence of mass chlorides.

By comparison, the corrosion current of the expanded sample is higher than that of the unexpanded sample at every tested temperature, and their differences increase with temperature, especially at high anodic potentials. These results demonstrated that the corrosion resistance of the expandable tube declined, and that this decline was more pronounced at higher temperatures.

Fig. 4. EIS results of the carbon steel before and after expansion in formation water at different temperatures: (a) 50°C; (b) 70°C; (c) 90°C.

The equivalent circuit in Fig. 5 was used to fit the EIS data, where R_s is the solution layer resistance, Q_c the capacitance of the corrosion product layer, R_{ct} the charge transfer resistance, and W the Warburg impedance. The fitting results are listed in Table 3. It can be seen that Q_c increases with temperature, indicating that the product layer is less homoge-

neous and more defective [19]. In contrast, R_{ct} decreases

Fig. 5. Equivalent circuit used for modeling the EIS results.

Table 3. Parameters of fitting results from the EIS equivalent circuit

Sample	Temperature / °C	$R_s / (\Omega \cdot \text{cm}^2)$	$Q_c / (\Omega^{-1} \cdot \text{s}^n \cdot \text{cm}^{-2})$	$R_{ct} / (\Omega \cdot \text{cm}^2)$	n	$Y_w / (\Omega^{-1} \cdot \text{s}^{1/2} \cdot \text{cm}^{-2})$
Before expansion	50	2.45	0.00049	1153.1	0.7946	0.04998
	70	1.97	0.00097	602.7	0.7623	0.01292
	90	1.82	0.00112	437.7	0.7460	0.01626
After expansion	50	2.42	0.00097	754.8	0.8043	0.03902
	70	1.84	0.00115	489.3	0.7650	0.05972
	90	1.70	0.00219	338.7	0.7778	0.01627

Note: n stands for the capacitance exponent of the corrosion product layer, and Y_w the diffusion impedance.

with temperature. At every temperature, Q_c is higher and R_{ct} is lower after expansion. These EIS results are in good agreement with the polarization curves.

It has been quantitatively demonstrated that temperature is an important criterion for describing the onset of pitting or localized corrosion [20]. Here, the potentiostatic polarization method was used to determine and compare the critical pitting temperatures (CPT) of the two samples, and the results are shown in Fig. 6. At a polarized potential of -0.6 V vs. SCE, the unexpanded sample shows a plateau in the cathode current from 20°C to 30°C , while the plateau for the expanded sample occurs in the anode current at 26°C . Subsequently, they both increase gradually with temperature until a sudden current increase is observed. The abrupt rises in current density occur at 87.5°C and 79.2°C in the unexpanded and expanded samples, respectively. The CPTs obtained herein, however, are not exactly the same as the “pitting” temperature described for passive metals [21–22] because the currents far exceed $100 \mu\text{A}\cdot\text{cm}^{-2}$. These temperatures are actually the critical temperatures when severe localized corrosion occurs in the downhole environment.

The above results showed that the corrosion performance of the tube deteriorated after expansion. During the deformation process, lattice distortion occurred and the grains were stretched, slipped, and thinned, resulting in substructure,

inhomogeneous deformation, and residual stress [23–25]. With increasing dislocation density and dislocation interaction, the corrosion activity was enhanced, leading to poor corrosion resistance.

Fig. 6. Potentiostatic polarization results for the carbon steel before and after expansion in formation water at an applied potential of -0.6 V vs. SCE.

3.3. Corrosion morphologies

Fig. 7 shows the typical corrosion morphologies as determined by stereology microscopy after three months of use in the downhole environment. Both uniform corrosion and localized corrosion are observed with taupe corrosion

Fig. 7. Typical corrosion morphologies (before (a) and after (b) expansion) and calculated corrosion area (before (c) and after (d) expansion) of the carbon steel used for three months in the downhole environment.

products covering the surface. The total corrosion area was obtained using the built-in area measuring software; the corrosion area ratio was only 17.1% for the unexpanded sample, while it increased to 49.3% after expansion. The number of corrosion area units between 0–50 μm^2 also immensely increased after expansion.

The typical localized corrosion topography and depth

were also observed. As shown in Fig. 8, the single pit of the unexpanded sample is relatively small, with a diameter of 125 μm and a depth of 15 μm . After expansion, however, the pits are about two times larger and become coterminous with a rugged topography. Both Figs. 7 and 8 indicated that the steel was much more seriously corroded after expansion, which was consistent with the electrochemical measurements.

Fig. 8. 3D stereological microscopy image (before (a) and after (b) expansion) and corresponding 2D sectional drawing (before (c) and after (d) expansion) of the carbon steel used for three months in the downhole environment.

Fig. 9 shows the morphology and corrosion scales of the expanded steel used for three months in the real downhole environment. Some cracks are observed in Fig. 9(a), indicating that stress corrosion cracking (SCC) occurs. Thus, not only the corrosion performance deteriorates, but the stress corrosion cracking sensitivity is also enhanced after expansion due to the decrease in toughness after deformation. The corrosion scales are compact and exhibited good adherence to the matrix, although the interfaces are not smooth. The energy dispersive spectroscopy (EDS) map indicated O-rich products inside the circle, implying that the main corrosion products were Fe_2O_3 and FeOOH . C and S were identified at the margin of products, which could be from FeCO_3 [26–28] and FeS [29–31]. Therefore, CO_2 corrosion and H_2S corrosion also occurred in the downhole environment as the following equations.

At the beginning of the downhole corrosion process, the corrosive medium in the formation water could preferentially corrode the steel, generating iron oxides and protecting the corroded surface. CO_2 corrosion and H_2S corrosion, on the contrary, as the relatively chronic and slower processes, only occurred around the edge of the protected surface where some active sites still existed. Thus, FeCO_3 and FeS were primarily found at the margin of the corrosion products.

4. Conclusions

The corrosion behavior of the expandable tubular was investigated in formation water to simulate the downhole corrosion environment by a series of electrochemical techniques at different temperatures. The corrosion morphology was also characterized by SEM.

(1) After a 7.09% expansion, the wall unevenness increases and the ferrite phase is greatly stretched, while the pearlite has no obvious change.

Fig. 9. SEM morphologies and EDS maps of the expanded steel used for three months in the downhole environment: (a) stress corrosion cracks; (b) surface corrosion products; (c)–(f) corresponding EDS maps of Fig. 9(b).

(2) The corrosion current is higher after expansion, especially at high temperature. The impedance value and CPT decrease after expansion, indicating deterioration in corrosion performance.

(3) Both the corrosion area and localized corrosion increase remarkably after plastic deformation. Stress corrosion cracks and the corrosion products of FeCO_3 and FeS are detected after three months of use in the downhole environment.

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