Study on corrosion characteristics of Q235 steel in seawater, soil and dry-wet alternating environments focusing on Shengli oilfield

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Abstract

The coastal pipelines are the key link between the offshore and the onshore pipelines, which are located in the soil dry-wet alternating environment. Even though higher corrosion risk of metals in coastal soil dry-wet alternating environment, few papers focused on this condition, while much research about ocean and atmospheric dry-wet alternating environment was paid attention to. In this paper, a self-designed soil dry-wet alternating experimental device was used to investigate the corrosion characteristics of Q235 steel in seawater, soil and dry-wet alternating environment based on mass loss experiments, corrosion morphology analysis and electrochemical experiments. The results showed that the general corrosion rate of Q235 steel in soil and seawater environments basically remained unchanged, and in dry-wet alternating environment increased with time, while the pitting rates were 0.280 mm/a (soil), 0.420 mm/a (seawater) and 0.567 mm/a (dry-wet alternation), respectively, which was caused by the oxygen concentration cell and salt concentration cell. Meanwhile, the growth rate of pitting size in dry-wet alternating environment was much higher than that in the other two environments with different corrosion characteristics of pitting corrosion (seawater and dry-wet alternation) and a changed from general corrosion to pitting corrosion (soil). In three environments, the corrosion current density (Icorr) increased, in which on the 30th day of experiment, Icorr = 79.72 μA cm⁻² in dry-wet alternating environment, which was much higher than Icorr = 47.89 μA cm⁻² in seawater environment. In dry-wet alternating environment, the mass diffusion impedance at 10⁻² Hz and the maximum phase angle changed significantly, while the logflcorr log|Z| curve in low-middle frequency decreased significantly, and charge transfer resistance at 10⁷ Hz reduced from 10.4 Ω to 4.3 Ω, illustrating an increasing corrosion rate.

1. Introduction

In recent years, with the increase of the number, mileage and operating life of offshore oil-gas pipelines, the research on integrity management and reliability of offshore oil-gas pipelines has become the focus of attention. Compared with the onshore oil-gas pipelines, the construction, operation and maintenance costs of offshore oil-gas pipelines are higher. In the soil dry-wet alternating environment, the pipeline is more easily corroded. More importantly, once the leakage caused by the corrosion occurs, it will cause serious damage to the underwater ecosystem. The coast pipelines as the key link between the offshore and the onshore pipelines are in the soil dry-wet alternating environment [1–3], which have higher corrosion risk [1–5].

Many scholars have carried out a lot of research work on the corrosion behavior of metal materials in dry-wet alternating environment. Tsuru et al [6] used Kelvin probe and EIS method to study the influence of dry-wet alternation on metal corrosion rate and corrosion mechanism, and the results showed that the liquid film layer on metal surface was gradually thinned in the drying process, which accelerated the corrosion rate. Nishikata et al [7] studied the corrosion behavior of carbon steel in dry-wet alternating environment, and found
that the corrosion of carbon steel had three stages. (1) The increase of Cl− concentration, and the corrosion potential moved in a negative direction [8, 9]. (2) The corrosion rate increased sharply, which was caused by metal dissolution and oxygen depolarization, and the corrosion potential remained unchanged. (3) As the anodic dissolution process was inhibited, the corrosion rate decreased and the potential shifted negatively.

Much factors affected the corrosion rate of metals in dry-wet alternating environment [8–13]. For example, the dry-wet ratio had a great influence on the diffusion rate of chloride ions [8, 9]. The longer the drying time was, the more serious the metal corrosion was. Generally speaking, the more serious corrosion under dry-wet alternating was due to the decrease of the surface liquid film thickness during the dry process, and oxygen was more easily transported in the liquid film, promoting the cathode reaction process [10–13].

Different from the ocean and atmosphere environment, in the soil dry-wet alternating environment, the soil was completely immersed in seawater in wet condition with an open corrosion crevice, in which the metal in the soil can directly contact the corrosion solution with sufficient oxygen [14–17]. For dry condition, the corrosion crevice was closed and the oxygen content was lower. Moreover, part of the salt remained in the soil. Therefore, With the change of dry-wet cycles, the salt content in soil gradually increased [18]. However, much research on the corrosion characteristics of metals in ocean (splash area, tidal range area) and atmospheric dry-wet alternating environment was carried out [19], but few papers focused on the corrosion behavior of metals in coastal soil dry-wet alternating environment.

In this paper, the change of coastal soil water content was simulated by a self-designed soil dry-wet alternating experimental device. Through mass loss experiments, corrosion morphology analysis and electrochemical experiments, the corrosion characteristics of Q235 steel in seawater, soil and dry-wet alternating environment were investigated.

2. Experimental settings

2.1. Environmental solution preparation
According to the distribution of oil-production plant in Shengli oilfield, a total of 84 soil (seawater) samples in seawater, soil and dry-wet alternating environment were collected, and soil properties were analyzed, as shown in figure 1. Combined with the composition and preparation of standard artificial seawater in GB/T 17 848–1999 'Test methods for electrochemical properties of sacrificial anode', the ion contents in three environments studied in this paper, as shown in table 1.
2.2. Design of dry-wet alternating device

Figure 2 showed the experimental devices for dry-wet alternating environment. The size of the experimental box was 50 × 50 × 20 cm³, in which the height of the experimental soil was 15 cm, and the height of the liquid level was 2 cm in the wet condition. Four soil water content probes were set up in the experimental soil with a height of 8 cm and a distance of 10 cm from the box wall. The Q235 sample was buried in the middle of the experimental soil. A outlet hol with a diameter of 5 cm was opened directly below the Q235 sample, and a outlet pipe and a vacuum pump were connected successively, in which the intelligent control valve was installed to discharge water through the vacuum pump.

Soil preparation. The ground original soil was immersed in deionized water for 24 h, and then the water was removed by centrifuge. Subsequently, repeat the above steps for three times and then dry for reserve. Before experiment, the pre-treated experimental soil was immersed in the solution prepared based on the table 1 for 24 h and then filtered for experiment.

Experimental process. The ratio of dry-wet alternation was set as 6 h : 6 h via the computer. The height of liquid level was monitored by the liquid level indicator and then replenished the experimental solution if insufficient. In the dry condition, the soil water content was controlled in 24 ± 1% by the intelligent control valve and the vacuum pump.

Table 1. The ion contents in seawater, soil and dry-wet alternating environments.

| Content (g g⁻¹) /% | Salt  | Mg²⁺ | Ca²⁺ | Cl⁻ | SO₄²⁻ | HCO₃⁻ | pH   |
|-------------------|-------|------|------|-----|-------|-------|------|
| Seawater          | 2.971 | 0.52 | 0.116| 1.667| 0.20  | 0.10  | 7.917|
| Soil              | 0.447 | 0.02 | 0.010| 0.278| 0.05  | 0.04  | 8.025|
| Dry-wet alternation| 0.503 | 0.03 | 0.020| 0.314| 0.06  | 0.05  | 8.000|
2.3. Corrosion rate and corrosion morphology

Experimental environment. Analytical pure reagents and deionized water were used to prepare based on the composition in table 1.

Metal sample. The metal sample was chosen as Q235 steel with a size of 50 $\times$ 25 $\times$ 3 mm$^3$ (C 0.20 wt%, Mn 1.4 wt%, Si 0.35 wt%, S 0.045 wt%, P 0.045 wt% and Fe in balance).

Sample pretreatment. The Q235 samples were cleaned with acetone, deionized water and anhydrous ethanol in sequence. The size and original weight of the each sample were measured after drying for 24 h.

Experimental design. Immersion experiments were carried out on seawater, soil and dry-wet alternating environment respectively. The total experimental time was 30 days and the experimental temperature was 25 °C. The whole experimental device was placed in a constant temperature - humidity device (GDS-3-408).

General corrosion rate. The corrosion products were cleaned by the rust-removing solution composed of 500 ml hydrochloric acid, 500 ml deionized water and 3.5 g hexamethylenetetramine. The general corrosion rate was calculated by [21]

$$v_1 = \frac{\Delta G}{S \times t \times \rho}$$

where $v_1$ was the general corrosion rate in mm/a (i.e. millimeter/year), $\Delta G$ was the mass loss in g, $S$ was the area in cm$^2$, $t$ was the experimental time in h, and $\rho$ was the density of Q235 steel.

Pitting rate. The maximum pit depth and corrosion morphology was obtained by Zeiss microscope, and the pitting rate was calculated by [21]

$$v_2 = \frac{h}{t}$$

where $v_2$ was the pitting rate in mm/a, $h$ was maximum pit depth in $\mu$m, $t$ was the experimental time in h.

2.4. Electrochemical experiment

A traditional three-electrode system was adopted for electrochemical tests, in which the working electrode (WE) was Q235 steel with the size of 10 $\times$ 10 $\times$ 2 mm$^3$, the reference electrode (RE) was saturated calomel electrode (SCE), and the auxiliary electrode (AE) was Pt electrode. The reference electrode was in contact with the solution through a Rukin capillary tube, and the distance between the tip and the sample surface was less than 2 mm, thus ignoring the IR drop of the solution [22, 23].

Working electrode pretreatment. The cleaned sample was welded by a copper wire on the back, and then sealed with epoxy resin, leaving a working area of 1 cm$^2$. Before electrochemical experiment, the surface was polished to be mirror-like.

Experimental design. In three experimental environment including seawater, soil and dry-wet alternating environment, the electrochemical test for a corrosion product-removing sample was carried out at different time. The workstation used for electrochemical test was PARSTAT 2273. Polarization curve and electrochemical impedance spectrum (EIS) were carried out after the whole system was stable, indicating that the variation of open-circuit potential ($E_{OCP}$) of the sample was less than $\pm$10 mV within 300 s. The scanning rate of polarization curve was set as 0.3 mV s$^{-1}$, and the scanning potential range was $\pm$250 mV versus $E_{OCP}$. The frequency range of EIS was 10$^4$–10$^{-2}$ Hz with an AC voltage of 5 mV [24]. The PowerSuite and ZSimpWin were adopted for data analysis.

3. Results and discussion

3.1. Corrosion rate

Figure 3 showed the curve of general corrosion rate of Q235 steel with time at 25 °C in seawater, soil and dry-wet alternating environment. As can be seen from the figure 3, at the initial stage of immersion (1st day), the general corrosion rate in seawater environment was the highest (0.192 mm/a), while the general corrosion rate in soil environment and dry-wet alternating environment was basically the same (0.141 mm/a and 0.147 mm/a). With the increase of immersion time, the general corrosion rate in the soil environment and seawater environment basically remained unchanged, with an average of 0.140 mm/a and 0.190 mm/a, respectively. However, the general corrosion rate in the dry-wet alternating environment increased with the increase of immersion time. On the 15th day of experiment (30 dry-wet cycles), the general corrosion rate was basically same as that in seawater environment. Subsequently, the general corrosion rate continued to increase, and the corrosion rate reached 0.225 mm/a on the 30th day (60 dry-wet cycles). Compared to the general corrosion rate in initial stage (1st day), it increased by 53.1%. Under the conditions in this paper, the relationship between the general corrosion rate ($v_1$) in dry-wet alternating environment and immersion time can be expressed as

$$v_1 = 0.1441 \times t^{0.1206} (R^2 = 0.9642)$$

[25–27].
Compared with soil environment and seawater environment, the corrosion in dry-wet alternating environment had the following two characteristics.

1. When the water in soil evaporated, indicating that environment changed from wet to dry condition, most of the salt remained in the soil. With the increase of the number of dry-wet cycles, the residual salt in soil accumulated, resulting in an increasing general corrosion rate.

2. Due to dissolved oxygen in the solution, the oxygen content in wet condition was higher than that in dry condition. Therefore, oxygen concentration alternated in the dry-wet alternating environment, leading to an increasing general corrosion rate.

Therefore, the metal corrosion was controlled by the oxygen depolarization process of the cathode in wet condition, while it was mainly controlled by the anodic process of $\text{Fe} \rightarrow \text{Fe}^{2+}$ due to the increase of salt accumulation in dry condition. The metal corrosion in dry-wet alternating environment was controlled by an alternating cathode-anode process. Therefore, the general corrosion rate continuously increased with immersion time, and then was much higher than that in seawater environment and soil environment.

Figure 4 showed the curve of pitting corrosion rate of Q235 steel with time at 25 °C in seawater, soil and dry-wet alternating environment. In different environments, the pitting corrosion rates all showed a slight increase trend. During the experiment period, the average pitting corrosion rates were $0.280 \text{ mm a}^{-1}$ (soil environment), $0.420 \text{ mm a}^{-1}$ (seawater environment), and $0.567 \text{ mm a}^{-1}$ (dry-wet alternating environment), respectively. The reason that the pitting corrosion rate in dry-wet alternating environment was highest was the combined result of the oxygen-and salt-concentration differential cell.

1. With the evaporation of water in soil, the diffusion channel was formed in the soil. Due to the crevice structure inside the soil, it was difficult for air to fill in time. As a result, the oxygen concentration near the metal was lower than the external environment, forming an oxygen concentration differential cell with the external cathode and the metal as the anode.
Due to the inhomogeneity of the soil structure, the water content of the metal surface may be variable at different locations. When the water evaporated, the residual salt content of the metal was also different. In the continuous dry-wet alternating process, the difference of salt content at different positions on the metal surface was becoming larger, so as to form a salt concentration differential cell, which was anode at the high salt-concentration position and cathode at the low salt-concentration position [8, 17, 29].

Figure 5. The curve of open circuit potential ($E_{OCP}$) of Q235 steel with time at 25°C in seawater, soil and dry-wet alternating environment.
3.2. Electrochemical tests

Figure 5 showed the curves of open circuit potential ($E_{OCP}$) of Q235 steel with time at 25 °C in seawater, soil and dry-wet alternating environment. At the initial stage, the open circuit potential ($E_{OCP}$) was $-0.687$ V (seawater environment), $-0.425$ V (soil environment) and $-0.404$ V (dry-wet alternating environment), respectively, indicating that the Q235 steel in seawater environment had the largest corrosion tendency [35]. Furthermore, the $E_{OCP}$ in different environments showed a decreasing trend with time, indicating that the longer the experimental time was, the greater the corrosion tendency of Q235 steel was. At 30th day, the $E_{OCP}$ of Q235 steel in dry-wet alternating environment ($-0.527$ V) was negative to that in the soil environment ($-0.454$ V), but positive to that in the seawater environment ($-0.707$ V). The results indicated that the dry-wet alternation could significantly increase the corrosion tendency of Q235 steel and then aggravate the corrosion behavior [35, 36].

Figure 6 and figure 7 showed the polarization curves including corrosion potential ($E_{corr}$), corrosion current density ($I_{corr}$) and Tafel-slope ratio ($r = \beta_a/\beta_c$) of Q235 steel with time at 25 °C in seawater, soil and dry-wet alternating environment. In different environments, the polarization curves of Q235 steel showed activation corrosion characteristics. In seawater environment, the polarization curves of Q235 steel basically coincided with each other at different experimental times, and the $E_{corr}$ was basically unchanged ($-0.725$ V), while the $I_{corr}$ increased, indicating that the corrosion rate of Q235 steel increased with time. The corrosion kinetics of Q235 steel in soil environment and dry-wet alternating environment was characterized by negative shift of $E_{corr}$ and increase of $I_{corr}$ [37]. Compared with the Q235 steel in seawater environment, the $E_{corr}$ of Q235 steel was more positive and the corrosion rate was smaller in soil environment. However, though the $I_{corr}$ (17.84 $\mu$A cm$^{-2}$) of Q235 steel in dry-wet alternating environment was much lower than that in seawater environment (32.63 $\mu$A cm$^{-2}$) at the initial stage, the $I_{corr}$ increased rapidly with time, and reached 79.72 $\mu$A cm$^{-2}$ at the 30th day of experiment, which was much larger than that in seawater environment (47.89 $\mu$A cm$^{-2}$).

The ratio of Tafel slope ($r = \beta_a/\beta_c$) showed that $r$ was greater than 1 in different environments, indicating that the corrosion behavior of Q235 steel was controlled by the anode process. Meanwhile, in seawater environment, $r$ was basically unchanged with an average of 1.732, while in soil environment and dry-wet alternating

![Figure 6. The polarization curves of Q235 steel with time at 25 °C in seawater, soil and dry-wet alternating environment.](image-url)
environment, the ratio \((r)\) fluctuated relatively greatly, with an average of 2.546 and 5.831, respectively. In other words, the process of anodic polarization control weakened successively in dry-wet alternating environment, soil environment and seawater environment [37, 38].

Figure 8 showed the EIS curves including Nyquist diagrams and Bode plots of Q235 steel with time at 25 °C in seawater, soil and dry-wet alternating environment, figure 9 was the equivalent circuit model, and figure 10 presented the curves of fitting parameters of EIS curves of Q235 steel with time.

In the seawater environment, Nyquist diagrams were characterized by capacitive-inductive reactance arc. Considering the corrosion characteristics of Q235 steel in seawater environment, \(R_s(Q(R_p(L(C_{dl}R_{ct}))))\) was selected as the equivalent circuit, in which \(R_s\) was solution resistance, \(Q\) was capacitance, \(R_p\) was polarization resistance, \(L\) was inductance, \(R_0\) was equivalent resistance of inductance, \(C_{dl}\) was double-layer capacitance, and \(R_{ct}\) was charge transfer resistance. However, in the land soil environment and the alternate soil dry-wet environment, Nyquist diagrams showed complete capacitive arc resistance characteristics in soil environment and dry-wet alternating environment, so the equivalent circuit was chosen as \(R_s(Q(R_p(C_{dl}R_{ct}))))\).

In seawater environment, the corrosion rate of Q235 steel was larger than that in the others. Therefore, a large number of corrosion products accumulated on metal surface, and then the corrosion product layer was formed to protect Q235 steel. However, because of the porous characteristics of corrosion product layer, the chloride ions in solution can penetrate the corrosion product layer to the metal matrix, and then promote metal activation corrosion to damage corrosion product layer. Therefore, the synergistic effect between the formation and destruction of corrosion product layer led to the appearance of the inductive reactance characteristics. At the same time, it can be seen from the Bode plots that the maximum phase angle and frequency changed little with \(f_{\text{max}} = 2.6-13.7\, \text{Hz}\) and \(\varphi_{\text{max}} = 69-72^\circ\) in \(\log f-\log |Z|\) curve. Meanwhile, in \(\log f-\log |Z|\) curve, the charge transfer impedance in \(10^3\, \text{Hz}\) basically remained unchanged \((|Z| = 5.1-7.1\, \Omega)\), indicating that the electrochemical reaction process of Q235 steel stayed the same with time, while the mass diffusion impedance reduced from 1951 \(\Omega\) to 213 \(\Omega\), meaning that corrosion product layer was damaged by Cl\(^-\) to accelerate the diffusion process.

Figure 7. The changes of fitting parameters of polarization curves including corrosion potential \((E_{\text{corr}})\), corrosion current density \((I_{\text{corr}})\) and Tafel-slope ratio \((r = \beta_a/\beta_c)\) of Q235 steel with time at 25 °C in seawater, soil and dry-wet alternating environment.
In soil environment and dry-wet alternating environment, the metal corrosion process was mainly controlled by oxygen depolarization. For another aspect, due to the crevice structure existing in the soil, the metal corrosion process was also controlled by oxygen concentration cell. Therefore, in soil environment, the polarization resistance \( R_p \) and the charge transfer resistance \( R_{ct} \) of Q235 steel decreased, showing a more severe corrosion process. Meanwhile, mass diffusion impedance at \( 10^{-2} \) Hz \( (|Z| = 1018–1964 \Omega) \) and the maximum phase angle \( (f_{\text{max}} = 13–41 \text{ Hz}\) and \( \phi_{\text{max}} = 57–61^\circ) \) changed little in Bode plots. However, in dry-wet alternating environment, mass diffusion impedance at \( 10^{-2} \) Hz \( (|Z| = 250–2578 \Omega) \) and the maximum phase angle \( (f_{\text{max}} = 7.9–41.8 \text{ Hz}\) and \( \phi_{\text{max}} = 39–65^\circ) \) changed much more than that in soil environment. Impedance curve at low frequency was occurred in the sharply down, and the charge transfer impedance at \( 10^5 \) Hz reduced from 10.4 \( \Omega \) to 4.3 \( \Omega \) in log-log \( |Z| \) curve, illustrating that the corrosion rate greatly accelerated with the increase of dry-wet cycles [39].

Figure 8. The EIS curves including Nyquist diagrams and Bode plots of Q235 steel with time at 25 °C in seawater, soil and dry-wet alternating environment.
3.3. Corrosion morphology

Figure 11 showed the corrosion morphology of Q235 steel with time at 25 °C in soil, seawater and dry-wet alternating environment by Zeiss 3D microscope. It can be seen that at the initial stage of the experiment (1st day), obvious pitting pits appeared on the surface of Q235 steel in seawater and dry-wet alternating environments, in which the pit size in seawater environment was maximum, followed by that in dry-wet alternating environment and soil environment [40]. As the experiment was processed through, the number of pits was increasing in all three environments. On 10th day, the obvious local corrosion area on Q235 steel...
occurred in seawater and dry-wet alternating environment, while the smallest general corrosion rate \( v_1 \) in figure 3 were observed on 15th day in the soil environment. Furthermore, the corrosion of Q235 steel was aggravated in different environments with time [41]. Compared with the Q235 steel in seawater environment, the pitting corrosion characteristics of Q235 steel were more obvious in soil environment and dry-wet alternating environment. In the experiment period of 20–30 days, the pit size and depth of Q235 steel in soil environment gradually increased, and the corrosion in local areas became obvious. However, in dry-wet alternating environment, the surface of Q235 steel was divided into corroded area and non-corroded area while the pitting corrosion intensified, which was mainly caused by the salt concentration battery during the dry-wet cycle.

4. Conclusion

In this paper, the corrosion characteristics of Q235 steel in seawater, soil and dry-wet alternating environment were investigated based on mass loss experiments, corrosion morphology analysis and electrochemical experiments. The following conclusions were drawn.

1. During immersion experiments, the general corrosion rate of Q235 steel in soil and seawater environments basically remained unchanged, with an average value of 0.140 mm/a and 0.190 mm/a, respectively. However, the general corrosion rate of Q235 steel in dry-wet alternating environment increased with the increase of experimental time, described by \( v_1 = 0.1441 t^{0.1206} \).

2. The pitting rates of Q235 steel were 0.280 mm/a (soil), 0.420 mm/a (seawater) and 0.567 mm/a (dry-wet alternation), respectively. Combined with the results of corrosion morphology, the highest pitting rate in dry-wet alternating environment was caused by the oxygen concentration cell and salt concentration cell.

3. In seawater environment, the corrosion potential \( E_{\text{corr}} \) of Q235 steel was basically unchanged \((-0.725 \text{ V})\), and the corrosion current density \( I_{\text{corr}} \) increased. The corrosion behaviors of Q235 steel in soil and dry-wet alternating environments were characterized by negative shift of corrosion potential \( E_{\text{corr}} \) and increase of corrosion current density \( I_{\text{corr}} \). On the 30th day of experiment, \( I_{\text{corr}} = 79.72 \mu \text{A cm}^{-2} \) in dry-wet alternating environment, which was much higher than \( I_{\text{corr}} = 47.89 \mu \text{A cm}^{-2} \) in seawater environment.

4. In dry-wet alternating environment, the mass diffusion impedance at \( 10^{-2} \text{ Hz} \) \( (|Z| = 250–2578 \Omega) \) and the maximum phase angle \( (f_{\text{max}} = 7.9–41.8 \text{ Hz}, \varphi_{\text{max}} = 39–65^\circ) \) changed significantly greater than that in soil environment. The \( \log f - \log |Z| \) curve in low-middle frequency decreased significantly, and charge transfer resistance at \( 10^5 \text{ Hz} \) reduced from 10.4 \( \Omega \) to 4.3 \( \Omega \), illustrating an increasing corrosion rate.

5. In corrosion morphology, the pitting corrosion of Q235 steel was the main corrosion in seawater and dry-wet alternating environment, while in soil environment, with the increase of experiment time, the corrosion

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**Figure 11.** The corrosion morphology of Q235 steel with time at 25 °C in (a) soil, (b) seawater and (c) dry-wet alternating environment by Zeiss 3D microscope.
behavior of Q235 steel gradually changed from general corrosion to pitting corrosion. Meanwhile, the growth rate of pitting size in dry-wet alternating environment was much higher than that in the other two environments.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

Author’s Contributions

All authors contributed equally to this work.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

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