



Pourbaix diagram for HP-13Cr stainless steel in the aggressive oilfield environment characterized by high temperature, high CO₂ partial pressure and high salinity

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ABSTRACT

The Pourbaix diagram for HP-13Cr stainless steel (SS) in the aggressive oilfield environment has been revised. Extrapolation of thermodynamics data to elevated temperatures has been performed with the revised model of Helgeson. The area of the corrosion of HP-13Cr SS and the pH value of environment have been calibrated on the Pourbaix diagram using electrochemical measurements and surface analysis. The results indicated that the quinary Pourbaix diagram (Fe-Cr-H₂O-Cl⁻-CO₂) can be obtained by the overlap of three ternary Pourbaix diagrams, i.e., Fe-Cr-H₂O, Fe-Cr-Cl⁻ and Fe-Cr-CO₂ and the operational area located in the passivity region composed of Cr₂O₃ and Cr(OH)₃(s).

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1. Introduction

The exploitation of the oil and gas in the Tarim area exhibits complex geological and chemical environment, such as super deep well (deepest to 8000 m), high temperature (maximum to 180 °C), high salinity (Cl⁻ concentration was 60000 mg/L), high CO₂ partial pressure (maximum to 4 MPa) and so on. The pipelines will suffer serious corrosion in such aggressive oilfield environment [1].

HP-13Cr SS has attracted attention due to its low-carbon, high-nickel and molybdenum contents, exhibiting higher pitting corrosion resistance and the excellent mechanical properties, and has been successfully used in the down-hole strings [2]. However, the field inspection results revealed that more than 30% of the pipelines suffered from serious corrosion or even fractured in the early stage of its working-life. Nevertheless, much of HP-13Cr SS related studies [3,4] in the past was focused on low temperature (<100 °C), low CO₂ partial pressure (<2 MPa) and low salinity (Cl⁻

concentration < 10000 mg/L). However, the published results could not be directly extrapolated to aggressive oilfield environment in the field.

Pourbaix diagrams are the thermodynamics basis for better understanding the corrosion mechanism of metals in the aggressive environments [5,6]. The diagrams provide important information on regions of immunity, corrosion/possible passivity and passivity. In most favorable cases, these diagrams allow prediction of the behavior of metals in corrosive media and design the potential and the pH value to control or mitigate the corrosion rate [7]. Summary of literature relating to Pourbaix diagrams in recent years are summarized in Table 1. As seen, most of Pourbaix diagrams focused on the Metal-H₂O systems, and the temperature ranged from 25 °C to 300 °C. Beverskog et al. illustrated the Pourbaix diagrams of Cu-H₂O system, Ni-H₂O system and Cr-H₂O system from 25 °C up to 300 °C, respectively [8–10]. Chivot et al. [11] developed the Pourbaix diagrams of Co-H₂O system from 25 °C up to 150 °C. Then, with the development of nuclear power industry, some scientists devoted to develop the Pourbaix diagrams in the nuclear power environment with the feature of higher temperature and higher pressure. Cook et al. [12–14] extended the Pourbaix diagrams for the Cr-H₂O, Ti-H₂O, Fe-H₂O and Ni-H₂O system to high-subcritical and low-supercritical conditions.

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Table 1
Summary of literature relating to Pourbaix diagrams in recent years.

Steel	Medium	Pressure (MPa)	Temperature (°C)	Ref.
Cu	H ₂ O	0.1	25–300	[8]
Ni	H ₂ O	0.1	25–300	[9]
Cr	H ₂ O	0.1	25–300	[10]
Co	H ₂ O	0.1	25–150	[11]
Cr	H ₂ O	25	300–400	[12]
Ti	H ₂ O	25	300–400	[12]
Fe	H ₂ O	25	300–400	[13]
Ni	H ₂ O	25	300–400	[14]
Fe–Cr	H ₂ O	0.1	300	[15]
Fe–Cr–Ni	H ₂ O	0.1	25–300	[16]
Ni–Ti	H ₂ O	0.1	25–300	[17]
Cu	H ₂ O–SO ₄ ^{2−}	0.1	25	[18]
Fe	H ₂ O–CO ₃ ^{2−}	0.1	25	[19]
Fe	H ₂ O–Cl [−]	0.1	25	[20]
Cr	H ₂ O–Cl [−]	0.1	25	[22]
Ti	H ₂ O–Br [−]	0.1	25	[21]

Moreover, considering the wide application of stainless steel, nickel based alloys and Ni–Ti alloys in the nuclear power industry, some researchers pay attention to the Pourbaix diagrams of multi-component alloy in super critical water environment. Cubicciotti et al. [15] showed the Pourbaix diagrams for Fe–Cr–H₂O systems in Boiling Water Reactors (BWRs). Puigdomenech et al. [16] illustrated the Pourbaix diagrams for ternary Fe–Cr–Ni alloy in pure water. Shang et al. [17] developed the Pourbaix diagrams for binary Ni–Ti alloy in pure water system.

At the same time, the effect of aggressive ion, such as SO₄^{2−}, CO₃^{2−} and Cl[−] on the Pourbaix diagram were also discussed on the Pourbaix diagram by many scientists. For example, Marcus et al. [18] built the Pourbaix diagram of Cu–H₂O–SO₄^{2−} system. Refait et al. [19,20] revised the Pourbaix diagram of Fe–H₂O–CO₃^{2−} system and Fe–H₂O–Cl[−] system. Garcia-Anton et al. [21,22] showed the Pourbaix diagram of Ti–H₂O–Br[−] system and Cr–H₂O–Cl[−] system.

As for the Pourbaix diagram of HP-13Cr SS in the aggressive

oilfield environment, the effects of high temperature, high CO₂ partial pressure and high salinity containing Cl[−] are still unclear, and the quinary Pourbaix diagram of Fe–Cr–H₂O–Cl[−]–CO₂ has not been founded. Moreover, the location of pH value of the formation water and the potential of HP-13Cr SS exposed to the aggressive environment has not been identified on the Pourbaix diagram. Thus, it is hard to tell whether corrosion occurs in the immunity, corrosion, passive and passivity/possible regions, which is unbenefited to better understand the corrosion mechanism of HP-13Cr SS in aggressive environment.

Moreover, the Pourbaix diagrams usually calculated at two concentration levels, 10^{−6} and 10^{−8} mol/L. The former is the conventional corrosion limit stipulated by Pourbaix, and the latter is intended to be used in high purity waters, indicating the materials have lower corrosion sensitivity. The corrosion sensitivity of HP-13Cr SS in aggressive environment was unclear. Therefore, in the present work, the Pourbaix diagram for HP-13Cr SS in the aggressive environment was developed. The applicability of two dissolved species (10^{−6} mol/L and 10^{−8} mol/L) was discussed.

The temperature and pressure in this manuscript was studied in couple to simulate the aggressive oilfield environment of the different depths in the well. We selected four depths and the data was 1500 m, 3000 m, 6500 m and 8000 m, which correspond to 95 °C/2.8 MPa, 120 °C/3.2 MPa, 150 °C/3.6 MPa and 180 °C/3.8 MPa, respectively. Furthermore, by means of the electrochemical tests and surface analysis, the potential of HP-13Cr SS and the pH value of formation water under different for temperature and pressures in such environment had been calibrated on the Pourbaix diagram.

2. Background

2.1. Composition of the Pourbaix diagram

The chemical species could be formed for the HP-13Cr SS in the aggressive oilfield environment as listed in Table 2. Moreover, based on the variety of the chemical species, the Pourbaix diagram

Table 2
Thermodynamic data of the species considered.

Species	ΔG^0 (KJ mol ^{−1})	ΔS^0 (J mol ^{−1})	$\Delta Cp^0(T)$ (J mol ^{−1} K ^{−1})			Ref.
			a	b × 10 ^{−3}	c × 10 ^{−5}	
H ₂ O	−273.19	70.08	75.44	0	0	[31]
H ₂	0	130.60	27.28	3.26	0.50	[31]
H ⁺	0	0	0	0	0	[31]
O ₂	0	205.15	29.98	4.14	−1.67	[31]
FeCr ₂ O ₄	−1339.40	152.26	157.02	29.29	−17.14	[31]
Fe	0	27.28	14.11	29.70	1.79	[31]
Fe ²⁺	−78.90	−182.18	−188	0	0	[31]
Fe ₂ O ₃	−740.98	89.96	98.28	77.82	−14.85	[31]
Fe(OH) ₂ ⁺	−438.06	−142	0	0	0	[20,40]
FeOH ⁺	−270.88	120	0	0	0	[20,40]
Fe ₄ (OH) ₈ Cl	−2139.90	−264.42	252	0	0	[20,40]
FeOOH	−408.60	49.85	98.43	0	0	[20,40]
Fe(OH) ₂	−486.89	−80.00	97.13	−2.87	0	[20,40]
Fe(OH) ₂ 2FeOOH	−346.06	−7.46	97.92	0	0	[20,40]
FeCO ₃	−666.70	92.91	82.14	0	0	[19,40]
Cr	0	23.54	26.90	−3.78	−2.78	[31]
Cr ₂ O ₃	−1049.96	82.37	146.82	−3.86	−5.23	[31]
Cr ²⁺	−166.00	−126.48	−314.51	0	0	[31]
HCrO ₄ [−]	−773.62	89.96	−282.31	0	0	[31]
CrOH ²⁺	−431.80	−151	160	0	0	[10]
Cr(OH) ₃ (aq)	−834.13	−38	480	0	0	[10]
Cr(OH) ₂ ⁺	−633.19	−92	340	0	0	[10]
CrO ₄ ^{2−}	−727.75	50.21	−251	0	0	[10]
H ₂ CO ₃	−623.16	187.45	0	0	0	[40]
HCO ₃ [−]	−586.85	98.42	0	0	0	[40]
CO ₃ ^{2−}	−527.90	−50.00	0	0	0	[40]

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