Corrosion behavior of the high strength low alloy steel joined by vertical electro-gas welding and submerged arc welding methods

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Abstract: The high strength low alloy (HSLA) steel used as a large crude oil storage tank (LCOST) was often welded by vertical electro-gas welding (VEGW) and submerged arc welding (SAW). **The VEGW specimen was produced by single-pass weld with high heat input (about 100KJ/cm), and the SAW specimen was manufactured by mutil-pass weld with low heat input (about 30KJ/cm).** The corrosion behaviors of both samples were studied by electrochemical techniques and surface methods. The research results achieved in this investigation disclosed that the welding process played an important role in obtaining satisfactory corrosion property. In comparison with the SAW joint, the VEGW joint showed uniform microstructure and coarse micro-phase. The scanning vibrating electrode technique (SVET) results demonstrated that the corrosion of the VEGW joint majorly took in the heat effect zone (HAZ) and the attack for the SAW joint mainly occurred in the fusion zone (FZ). In addition, **the scanning electron microscopy (SEM) results exhibited that the VEGW joint exhibited a light local attack and the SAW joint demonstrated a**

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heavy uniform corrosion. The electrochemical results showed that the VEGW joint showed lower corrosion rate than the SAW joint. In summary, it can be discovered that the corrosion rates of welded specimens were associated with the welding-pass and the corrosion forms of **welded specimens were related** to the heat input energy.

Key words: vertical electro-gas welding; submerged arc welding; corrosion behavior; electrochemical impedance spectra; scanning vibrating electrode technique; heat effect zone

1. Introduction

The high strength low alloy (HSLA) steel has been widely used to build a large crude oil storage tank (LCOST) [1]. During build process, The horizontal welding seam is produced by submerged arc welding(SAW) and the vertical welding seam is joined by vertical electro-gas welding (VEGW)[2]. The LCOST steel has been produced in the major steel enterprise Baosteel, WISCO, Ansteel and Shougang.

The Welded joints of the LCOST steel exhibit good mechanical property and excellent welding ability, however, the corrosion problems caused by chloride have been paid more and more attention on the LCOST by now. This is due to that the domestic crude oil comes from the ocean and makes the seawater as pressure water during exploitation. In addition, the import crude oil mostly comes from the Middle East and contains many chlorides [3]. As a result, the corrosion of crude oil becomes more and more serious, which takes serious corrosion to the LCOST and shortens its service life. Therefore great economic losses are caused by the corrosion of the LCOST. On the other hand, seawater is seemed to substitute to freshwater in

hydraulic experiment during the LCOST building, which can save lots of freshwater resources and construction costs. However, seawater can lead to serious corrosion for the LCOST by the chloride. Therefore, it is necessary to study **corrosion problems** for the LCOST in chloride solution, especially to the welding joint site for the most sensitive corrosion in the LCOST.

It was proposed that the welding would increase the corrosion activity of the HSLA steel due to metallurgical changes and residual stresses introduce a series of phase transformations taking place in the fusion zone (FZ) and heat effect zone (HAZ), In recent years, there have been many researches on the mechanical property and welding ability for the welded joint of the LCOST steel [4-6], however, **there were few researches on its corrosion behaviors** [7]. As a result, it is **necessary to study the corrosion behaviors of welded joints of the LCOST steel in 3.5% NaCl solution.**

In this work, scanning vibrating electrode technique (SVET) combined with scanning electron microscopy (SEM) observation as well as electrochemical impedance spectroscopy (EIS) measurement, was used to investigate the corrosion behaviors of welding joints of the LCOST steel in 3.5% NaCl solution.

2 Experimental procedures

2.1 Sample preparation

Test specimens were cut from the longitudinally welded LCOST steel containing welded metal (WM), HAZ and base metal (BM). The chemical compositions of the LCOST steel and filler metal were shown in Table 1.

Butt welds were made on plates of 250 mm × 200 mm × 32 mm using VEGW and SAW. The size of welding groove was shown in Fig.1. The VEGW joint was made using by single-pass welding and established suitably welding parameters (i.e., 450 A welding current, 45 V welding voltage, 12 cm/min welding speed, 100KJ/cm heat input). The SAW joint was produced by multi-pass welding and optimally welding parameters (produced using welding current of 500 A, welding voltage of 30 V, welding speed of 40 cm/min, heat input of 30 KJ/cm and argon shielding flow rate of 15 L/min welding parameters). Radiographic (RT) examined all welded joints showed that they were full penetration and acceptable bead profile with no welding defects such as porosity, undercut, cracks, or lack of fusion. Cross-section micro-images of both welding technology were shown in Fig.2, which exhibited that **both of these joints** had good welded quality. Welded samples were machined by electrochemical sample with 10 mm×10 mm ×10 mm. All sides of the specimen were embedded in an epoxy resin except the exposure surface, which was ground sequentially with 400, 600, and 800 grit emery papers. The specimen was cleaned by distilled water and degreased in acetone. Finally, a specimen was subjected for 5 min to ultrasonic washing with acetone and dried in warm flowing air prior to every experiment. EIS began after a 2 h immersion for samples to reach a steady state condition.

2.2 Experimental device description

Both electrochemical measurements were performed in a standard three-electrochemical cell, with a saturated calomel electrode (SCE) as the reference electrode, a platinum sheet as the auxiliary electrode (AE), and the samples as the

working electrode (WE). Polarization curve and EIS were carried out in a VMP3 electrochemical corrosion testing apparatus at room temperature in 3.5% NaCl solution. For the potentiodynamic curve, the sweeping potential was from -0.25 V to 0.25 V with a scanning rate of 0.1667 mV/s. EIS measurements were performed at the open circuit potential with AC amplitude of 5 mV, **and the applied frequencies** ranged from 0.01 Hz to 10 KHz. At least three tests were conducted under each condition to confirm the validity of the experiment measurements.

The SVET measurements were conducted through a PAR370 Scanning Electrochemical Workstation. The detailed setup was described as previous works [8]. A video camera was used for imaging and controlling the distance between the Pt-Ir micro-probe and the work electrode surface, which was set at 100 μm . The vibrating amplitude of the micro-electrode was 25 μm and the vibrating frequency was 400 Hz in the direction to the surface. The potential of the microelectrode was proportional to its position in the vibrating plane. The difference of the potentials when the microelectrode was located at the vibrating peak and valley, respectively, ΔE , was measured by an electrometer incorporated in M370. The solution resistance between the vibrating peak and valley, R is determined by $R = d/k$, where d is the vibrating amplitude of the microelectrode ($35\mu\text{m}$) and k is the solution conductivity. The SVET current I was then obtained by $I = \Delta E/R$ polishing with diamond paste, and etched by a mixture of 5% nitric acid and ethanol.

Micro-hardness test was conducted through a nano-indentation measuring device (TI900 Tribo-Indenter, Hysteron). In a force-controlled mode, the indenter tip

(Berovich type triangular pyramid) was loaded with a peak force of 3000 μN at a rate of 50 $\mu\text{N/s}$.

Scanning electrode microscopy (SEM) was utilized to investigate the corrosion morphologies of removed corrosion scales for the SAW joint and the VEGW joint after 144 h of immersion in 3.5% NaCl solution.

3 Result and discussion

3.1 Microstructure of SAW and VEGW welds

Fig. 3 shows the micrographs for the VEGW samples of WM, HAZ and BM, respectively. Various micro-phases are observed in different zones. The BM is character to bainite and acicular ferrite. The WM is featured with allotriomorphic ferrite, acicular ferrite and tempered bainite. The HAZ contains incomplete heat affected zone (ICHAZ), fine grain heat affected zone (FGHAZ) and coarse grain heat affected zone (CGHAZ), as shown in Fig3b(I), Fig3b(II) and Fig3b(III), respectively. The CGHAZ adjacent to the fusion line could be resulted from nearly complete austenite dissolution on heating and subsequent ferrite grain growth. The microstructure of CGHAZ is polygonal ferrite and pearlite. The micro-phase of FGHAZ is fine polygonal ferrite, pearlite and acicular ferrite, while the micro-phase of ICAZ is featured with acicular ferrite and granular bainite.

Fig. 4 shows optical micrographs of the cross section of the different welding regions (BM, HAZ and WM) of the SAW joint. The different zone in the HAZ is demonstrated in Fig. 4b, which contains ICAZ(Fig4b(I)), FGHAZ(Fig4b(II)) and CGHAZ (Fig4b(III)), respectively. The microstructure of CGHAZ consists of stripped

ferrite with tempered bainite. The micro-phase of FGHAZ is the mixture phase of pearlite and nubble ferrite. The micro-phase of ICHAZ is featured with acicular ferrite and granular bainite. The WM microstructure consists of pearlite and polygonal ferrite. Compare with the VEGA joint, the SAW joint shows finer micro-phase in welding zone, which is relate to the high heat input by the VEGW technology.

Typical microhardness profiles of welding samples are shown in Fig. 5. It can be shown that **the hardness values of the VEGW joint are symmetric distribution as a centre of the FZ**. The micro-hardness of the BM is about 230 HV. WM and HAZ show little lower micro-hardness values, 190 HV for WM and 165~185 HV for HAZ. It can be concluded that the VEGW joint shows relatively the uniform fluctuations of micro-hardness in welding zone (WZ). Generally, the micro-hardness profiles of the SAW joint reveal that the micro-hardness values gradually decrease along the welding direction, which is related to mutil-pass welding technology [9]. The micro-harness values of the SAW joint are 205~235 HV for WM and 195~245 HV for HAZ. It can be confirmed that the SAW joint exhibits more obvious micro-hardness fluctuate in welding zone than the VEGW joint, which is due to the mutil-pass weld for the SAW joint that makes the welding zone suffering many times metallurgical change and making worse microstructure uniformity. Thus, **it is indicate that the microstructure homogeneity of VEGW is better than that of SAW**.

3.2 Corroded surface characterization

Fig.6 shows corrosion morphologies of **welding samples** in 3.5% NaCl solution after 144 hours. It can be shown in Fig. 6(a) that the VEGW joint exhibits obvious

local corrosion in the welding zone and light corrosion in whole welding joint, which is related to the microstructure characteristics of welding samples. **For the VEGW joint, it can be observed many coarse micro-phases in welding zone in Fig. 3, and it can be revealed symmetrical micro-hardness fluctuation in welding joint in Fig.5.** Thus, the VEGW joint exhibits a light local corrosion in 3.5% NaCl solution. The SAW joint presents a heavy corrosion uniform attack in Fig 6(b), which is related to the homogeneity of the microstructure. It can be shown in Fig 5 that the SAW joint shows obvious hardness fluctuation in welded zone, which reveals the inhomogeneous microstructure of the SAW joint. This leads to heavy corrosion for the SAW joint in 3.5% NaCl solution.

3.3 SVET measurement

SVET is a new technique, which can measure the local corrosion potential under untouched sample surface. The electrochemical reaction took place on the active surface of the welded specimen in 3.5% NaCl solution. The ion current could flow during reaction process and lead to the small change of corrosion potential. SVET can measure this small change of the corrosion potential. Fig 7 shows the SVET maps to measure on the welding specimen after 1 h of immersion in 3.5%NaCl solution. Fig 7(a) reveals that, for the VEGW joint, corrosion currents of BM show lower than those of HAZ and WM, thus BM exhibits a local cathode zone and forms local form on the sample surface (in Fig 6(a)). The distribution of the corrosion currents shows a relatively uniform distribution in BM. There is a maximum dissolution current density up to 3.85×10^{-4} A/mm² in the HAZ adjacent to the BM and the smallest current

density of about 1.9×10^{-4} A/mm² in the BM. As shown in Fig 7(b), it can be seen that the SAW joint exhibits worse homogeneous distribution and higher corrosion current in the WM and the HAZ. As a result, the SAW joint exhibits heavy corrosion in Fig.6(b). A maximum dissolution current density is up to 4.5×10^{-4} A/mm² in the WM adjacent to the HAZ, and the minimum current density of about 2.1×10^{-4} A/mm² in the BM. Compared with the VEGW joint, It can be indicate that the SAW joint show higher corrosion current than the VEGW joint in 3.5%NaCl solution, which is fit to the SEM results.

3.4 Polarization curve

Fig 8 shows the potentiostatic polarization for the WM, HAZ and BM of welding samples after 1 h in 3.5%NaCl solution. It can be shown **that both of these welding samples** show similar corrosion behaviors. Anodic current densities continuously increase with an increase in corrosion potentials, which indicates that all samples display an active dissolution behavior in 3.5%NaCl solution.

Table 2 shows corrosion potentials (E_{corr}), polarization resistances (R_p), Tafel slopes (b_a and b_c , anodic and cathodic, respectively) and corrosion currents (I_{corr}) simulated by the polarization curve results. As shown in Table 2 for the VEGA joint, It can be observed that HAZ has a more negative potential (-530 mV_{SCE}) than WM(-513.29 mV_{SCE}) and BM (-470 mV_{SCE}), which means that its HAZ shows an obvious electrochemical reaction activity. At a meantime, the corrosion current density of BM is $16.80 \mu\text{A cm}^{-2}$, which is close to WM ($18.77 \mu\text{A cm}^{-2}$), and lower than that of HAZ ($21.39 \mu\text{A cm}^{-2}$). It can be shown that there is a high dissolution current density at

HAZ, which is in accordance with the SEVT result. For the SAW joint, WM shows the most negative corrosion potential ($-789.1\text{mV}_{\text{SCE}}$) than HAZ ($-604.6\text{ mV}_{\text{SCE}}$) and BM ($-472.2\text{ mV}_{\text{SCE}}$), which imply that its WM is prone to attack in 3.5%NaCl solution. Furthermore, the corrosion current of WM is $23.07\mu\text{A cm}^{-2}$, which is obviously more than those of HAZ ($19.9\mu\text{A cm}^{-2}$) and BM ($17.10\mu\text{A cm}^{-2}$). The results show the WM of SAW joint exhibits higher corrosion rate than the others in 3.5%NaCl solution.

The corrosion behaviors of both joints are listed in Table 2. It can be discovered that the SAW joint exhibits more positive corrosion potentials and higher corrosion currents than the VEGW joint, which means the SAW joint manifests lower corrosion tendency and higher corrosion rate in 3.5% NaCl solution, which agrees with the SEM result.

3.5 EIS measurement

The key aim of the EIS experiment is to provide insight into the characteristics and kinetics of the electrochemical process occurring at the interface of Fe/solution in 3.5% NaCl solution. The working electrodes had been immersed in 3.5% NaCl solution until the formation of a stable state. Fig. 9 illustrates the typical Nyquist diagrams of welding samples obtained at different immersion time. all samples exhibit similar corrosion behaviors during corrosion process, and the Nyquist plots show increasing capacitance arcs with the increasing time in Figs. 9(a) and 9(b), indicating an activation-like behavior. With an increase in the immersion time, EIS shows two capacitance arcs in Figs. 9(c) and 9(d), and a new capacitance arc is related to the

formation of the local corrosion on sample surface.

According to **EIS results** in Fig. 9, the equivalent circuits are illustrated in Fig. 10. Fig. 10a is fitted to the impedance spectra with one-time constant. Fig. 10b is simulated to the impedance spectra with two time constants. In these models, R_s is the solution resistance, CPE_{pit} and R_{pit} correspond to the local corrosion capacitance and resistance, respectively. R_{ct} is the reaction resistance. CPE_{dl} is the double-layer capacitance. A constant phase element representing a shift from an ideal capacitor is used instead of the capacitance itself, for simplicity. The impedance of a constant phase element is defined as $Z_{CPE} = [Q(j\omega)^n]^{-1}$ [10~12], where Q is a proportional factor, ω is the frequency and $-1 \leq n \leq 1$. The value of n seems to be associated with the non-uniform distribution of current as a result of the roughness and surface defect.

It is well recognized that the polarization resistance (R_p) is inversely proportional to the corrosion rate. The R_p value is equal to the R_{ct} value during the first 144 h and the sum of the R_{ct} and R_{pit} value after 144 h of immersion. The R_p values obtained by analyzing impedance spectra are presented in Table 3. The R_p values increase with the increasing corrosion time within 144 h of immersion, which is relates to the deposition of corrosion scales. After 144 h, the R_p values exhibit decreasing trend, which is relate to the formation of local corrosion on sample surface. **During corrosion process, the VEGW joint presents higher R_p values than the SAW joint in 3.5% NaCl solution**, which complies with the PC results. The reason for this is associated with the welding technology. As shown in Fig 5, The SAW joint by multi-pass welding shows inferior homogeneity. As a result, the SAW joint shows

high corrosion rate in 3.5% NaCl solution.

4 conclusions

This study has performed an experimental investigation into the corrosion behaviors of welding specimens of the LCOST steel by the VEGW and the SAW in 3.5% NaCl solution. The experimental findings support the following major conclusions:

(1) The VEGW joint presents coarser micro-phase and more uniform microstructure in welding zone than the SAW joint.

(2) The SVET results reveal that the VEGW joint shows small corrosion current the welding zone and the main corrosion in HAZ, while the SAW joint exhibits high corrosion current in the welding zone and the key attack in FZ.

(3) SEM results demonstrate that the VEGW joint shows slight local corrosion and the SAW joint exhibits a heavy uniform attack.

(4) Electrochemical results show that the VEGW joint shows smaller corrosion rate than the SAW joint.

(5) EIS show that the welding samples show two corrosion processes in 3.5% NaCl solution. EIS shows one increasing capacitance arc in the first 144 hours due to the deposition of corrosion products, and exhibits two capacitance arcs after 144 hours owing to the formation of the local corrosion on sample surface.

(6) It can be discovered that the corrosion rate of the welding joint is associated with the welding pass, and the corrosion form of the welding sample is related to the welding heat input.

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Fig 1 Schematic illustration of welded groove (a) VEGW joint (b) SAW joint

Fig 2 Cross-section macro-images of both the welding technology (a) VEGW joint (b) SAW joint

Fig 3 Optical micrographs of different zones for welded joints using Vertical Electro-Gas Welding (a) base metal (b) heat effected zone (c) welded metal

Fig 4 Optical micrographs of different zones for welded joints using transverse Submerged-Arc Welding (a) base metal (b) heat effected zone (c) welded metal

Fig 5 Distribution character of the hardness for both samples (a) VEGW joint (b) SAW joint

Fig 6 SEM micrograph of samples after 144 hours immersion (a) VEGW joint (b) SAW joint

Fig 7 SVET measurements of all weld joints in 3.5%NaCl solution (a) VEGW joint (b) SAW joint

Fig 8 Polarization curves measured the sample in 3.5% NaCl solution (a) different zone of the VEGW joint (b) different zone of the SAW joint (c) whole welded joint

Fig.9 EIS of the welding joint of the VEGW and SAW in 3.5% NaCl (a)4 hours (b) 8 hours (c)144hours (d) 1224hours

Fig 10 Equivalent circuits for EIS obtained in 3.5% NaCl solution (a) former stage (b) latter stage

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(a) base metal (b) heat effected zone (c) welded metal

Fig 4 Optical micrographs of different zones for welded joints using transverse SAW

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(a)

(b)

Table 1 Composition of the HSLA steel

Table 2 Fitted results to polarization curves for the sample in 3.5% NaCl solution

Table 3 Parameters of fitted results with equivalent circuits for EIS at different times

Table 1 Composition of the HSLA steel and (wt%)

Material	C	Mn	Si	Al	Mo	Cr	Cu	Nb	Ni	S	P
Base metal	0.090	1.44	0.185	0.029	0.1	0.03	0.01	0.014	0.21	0.0015	0.0043
SAW wire	0.090	1.32	0.1	0.012	0.8	0.02	0.15	0.01	0.35	0.001	0.004
VEGW power	0.100	1.26	0.6	0.023	0.9	0.26	0.12	0.01	0.28	0.001	0.003

Table 2 Fitted results to polarization curves for the sample in 3.5% NaCl solution

Sample		E_{corr} (mV vs. SCE)	I_{corr} ($\mu\text{A cm}^{-2}$)	β_c (mv.dec ⁻¹)	β_a (mv.dec ⁻¹)
VEGW	BM	-470.67	16.80	463.3	242.6
	HAZ	-530.00	21.39	545.2	256.2
	WM	-513.29	18.77	476.7	246.2
	weldment	-0.841	22	155	214
SAW	BM	-472.20	17.10	472.2	235.9
	HAZ	575.68	19.9	604.6	258.6
	WM	-547.16	23.07	789.1	227
	weldment	-0.786	36.7	150	235

Table 3 Parameters of fitted results with equivalent circuits for EIS at different times

Corrosion		R_s	Q_{dl}	n_{dl}	R_{ct}	Q_{pit}	N_{pit}	R_{pit}	R_p
time/ h		$/(\Omega \text{ cm}^2)$	$/(\Omega^{-1} \text{ cm}^{-2} \text{ s}^n)$		$/(\Omega \text{ cm}^2)$	$/(\Omega^{-1} \text{ cm}^{-2} \text{ s}^n)$		$/(\Omega \text{ cm}^2)$	$/(\Omega \text{ cm}^2)$
4	VEGW	3.942	6.26×10^{-3}	0.78	378.41				378.41
	SAW	5.336	5.52×10^{-3}	0.78	331.22				331.22
8	VEGW	3.95	6.94×10^{-3}	0.76	590.91				590.91
	SAW	5.37	5.18×10^{-3}	0.79	373.12				373.12
144	VEGW	4.16	5.55×10^{-3}	0.67	255.91	7.15×10^{-4}	0.701	4.36	260.27
	SAW	3.42	7.76×10^{-3}	0.71	122.49	5.78×10^{-4}	1	1.63	124.12
1224	VEGW	3.31	2.84×10^{-2}	0.89	225.68	1.78×10^{-2}	0.64	3.41	229.09
	SAW	3.56	1.29×10^{-2}	0.75	131.42	1.07×10^{-2}	0.68	3.99	135.41