



Corrosion wear synergistic behavior of Hastelloy C276 alloy in artificial seawater

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Abstract: A systematic investigation was carried out to discuss the corrosion and tribocorrosion behaviors of Hastelloy C276 alloy sliding against Al_2O_3 pin in artificial seawater, using a pin-on-disk tribometer integrated with a potentiostat for electrochemical control. The results show that the great decrease of open circuit potential and three orders of magnitude increase of corrosion current density occur caused by friction. There are clearly synergistic effect between corrosion and wear, resulting in corrosion-induced-wear and wear-induced-corrosion in tribocorrosion process. The contribution of pure mechanical wear to total material loss exceeds 70% in all sliding conditions, so mechanical wear is the dominant factor during tribocorrosion. For considering synergistic effect between corrosion and wear, the contribution of wear-induced-corrosion to total material loss is not very high although corrosion rate is greatly accelerated by friction. The fraction of corrosion-induced-wear to the total material loss is high and in the range of 14.6%–20.5% under all sliding conditions.

Key words: tribocorrosion; Hastelloy C276 alloy; synergistic effect; seawater

1 Introduction

In many situations, passive alloys are subjected to combined corrosion and wear, which can be defined as tribocorrosion. These can lead to the destroy or even complete removal of passive film from contact surface, resulting in accelerated metal dissolution, which in turn causes accelerated wear loss [1–4]. Over past few years, investigation on tribocorrosion behaviors of materials subjected to corrosion and wear is a hot topic. Stainless steel and CoCrMo alloy are the most frequently used metals in tribocorrosion field [2,5].

Nickel-based alloys are widely used in chemical industry (vessels, pipes, heat-exchangers and pumps), automotive industries and marine industry (pumps, impellers, valves, and other kinds of equipments). Recently, the tribocorrosion behavior of Ni–Cr alloy was investigated by researchers. The tribocorrosion behavior of overlay welded Ni–Cr 625 alloy in sulfuric and

nitric acids under potentiostatic was investigated by ESPALLARGAS and MISCHLER [6]. The tribocorrosion behavior in sulfuric acid was similar to that of stainless steels found in other studies, with much lower wear at cathodic potential compared with passive conditions. However, wear behavior in nitric acid was highly influenced by the reduction reaction of nitric acid at the electrode–electrolyte interface, which leads to the oxidation of the alloy even at cathodic potential. BI et al [7] studied the tribocorrosion property of Ni–17.5Si–29.3Cr alloy sliding against a Si_3N_4 ball in comparison with AISI 321 stainless steel in 1 mol/L H_2SO_4 solution. The results indicated that the alloy showed excellent corrosion resistance and anti-wear ability compared with AISI 321 stainless steel. However, the fundamental mechanism that determines the wear-corrosion synergism in tribocorrosion of Ni-based alloy in chloride-containing solution has not been fully understood.

The aim of this work is to assess the tribocorrosion

behavior of Ni–Cr alloy (Hastelloy C276) in artificial seawater. The counterpart material is alumina, which is considered to act as an inert antagonist in this experiment. Ceramic materials have excellent insulation, high hardness and good wear resistance, making them suitable for investigating the tribocorrosion properties of metals as counterpart materials. Corrosion wear experiments were carried out in a developed apparatus allowing for well-controlled mechanical and electrochemical conditions.

2 Experimental

2.1 Materials and solutions

The researched metallic alloy in this experiment was Hastelloy C276 nickel-based alloy with the following chemical compositions: 16% Cr, 16% Mo, 5% Fe, 4% W, 2.5% Co, 1% Mn, 0.35% V, 0.08% Si, 0.01% C, balance Ni. The alloy was machined into a ring with an outer diameter of 54 mm and an inner diameter of 38 mm. And only upper surface of the alloy was in touch with seawater and other surfaces were covered with paint for insulation to avoid electrochemical test confusion. The Al_2O_3 pin with a flat surface at one end of a cylinder (diameter: 4.7 mm, height: 13 mm) was used as counterpart material. All samples were polished down to mirror quality using 220, 500 and 1000 grade grit paper and with 1 μm diamond suspension, respectively. Corrosive solution was artificial seawater prepared according to ASTM D1141–98 standard. The pH value was 8.2. All tests were carried out at room temperature (20–25 °C).

2.2 Wear measurements

Corrosion wear test setup combined with in-situ electrochemical measurements was illustrated in our previous paper. Sliding tests were carried out using an MMW–1 pin-on-disk tribometer. In-situ electrochemical measurements during sliding were fulfilled using CHI760C potentiostat. Wear track was a ring with a mean diameter of 46 mm and a width of 4.7 mm. The area of wear track was about 6.8 cm^2 . The rotation speed in this experiment was 100–400 r/min with linear velocity of 0.27–1.08 m/s and normal load was 100–200 N. After corrosion-wear tests, the metallic specimens were ultrasonically cleaned in acetone to remove debris. Gravimetric measurement before and after tribocorrosion were finished. The volume loss can be calculated as

$$V = \frac{m_0 - m_1}{\rho} \quad (1)$$

where V is the volume loss (mm^3), m_0 is the mass of material before wear (mg), m_1 is the mass of material after wear (mg), and ρ is density of the alloy. The specific wear rate, k , can be determined as

$$V = kLN \quad (2)$$

where N is the applied load and L is the total sliding distance. The morphologies of worn surfaces were examined using JEM–5600LV scanning electron microscope (SEM). All tests were repeated at least three times for reproducibility.

2.3 Electrochemical measurements

Electrochemical measurements were concluded using a CHI760C potentiostat with three-electrode test system. Metallic specimen was prepared as working electrode (WE). A saturated calomel electrode (SCE) close to working electrode served as reference electrode (RE) and platinum was used as counter electrode (CE). All potentials in this work are given with respect to the saturated calomel electrode (SCE). Several series of experiments were conducted to study corrosion-wear synergistic behaviors of the alloy: 1) Potentiodynamic test involving measuring polarization curves was initiated after a stable open potential. It was fulfilled with a potential sweep starting at -1 V and finishing at 1 V at a sweep rate of 1.67 mV/s. CHI software was used to analyze polarization data; 2) Corrosion-wear tests were carried out at open circuit potential and the evolution of open circuit potential was measured; 3) To analyze the synergistic effect between corrosion and wear, sliding wear tests polarized at -0.8 V were conducted and the transient current was measured.

3 Results and discussion

3.1 Electrochemical behavior

The evolution of open circuit potential (OCP) of Hastelloy C276 alloy under tribocorrosion is shown in Fig. 1. It can be obtained that at the start of sliding, open potential drops sharply down to more negative value due to friction. The cathodic shift phenomenon of OCP during corrosion-wear is frequently observed for

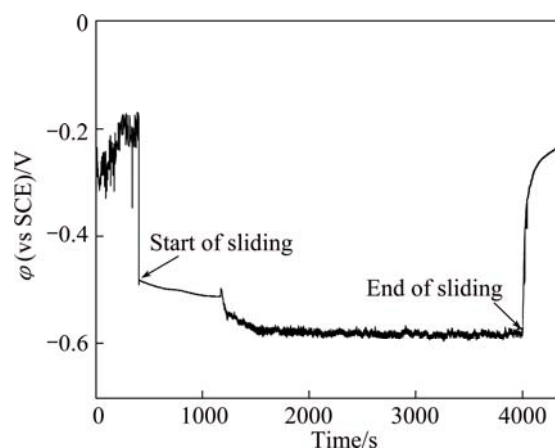


Fig. 1 Evolution of OCP of Hastelloy C276 alloy under tribocorrosion

stainless steel, TC4 and Co–Cr alloys by many researchers [8]. It can be expounded by the removal or damage of passive oxide film formed on original surface because of friction [9,10]. When friction stops, OCP starts to increase (anodic shift) abruptly and then more gradually reaches initial OCP without sliding about -0.2 V. This indicates the re-establishment of passive state in wear track.

Generally, OCP measurement enables to gather electrochemical information in wear track and can easily evaluate the electrochemical behavior of metal surface. However, this electrochemical technique is powerless for assessing corrosion kinetics. Potentiodynamic polarization method was used to evaluate corrosion kinetics during tribocorrosion in this experiment [9].

The polarization curves of Hastelloy C276 alloy under corrosion-only and sliding conditions are shown in Fig. 2. Passive phenomenon and very low passive current density can be seen under corrosion-only, which is comparable with commonly stainless steel and titanium alloys, suggesting strong passivation ability [8,11]. The polarization curves obtained during tribocorrosion reflect corrosion behavior and repassivating ability when passive film is mechanically damaged or removed by friction. Since contact area of friction couples is constant in this experiment, the calculation of current density is based on the area of wear track. The polarization curve exhibits significant current oscillations, which can be attributed to instability of corrosion-wear system and simultaneous interaction of damage and recovery of passive film. Moreover, contact positions of friction couples change continuously, which also contributes to oscillations of curves [12,13]. The corrosion potential in tribocorrosion shifts to lower value, more active potentials by about 0.3 V compared with that under corrosion-only condition, which is consistent with result of OCP measurement. Apparent passive region is also observed, which means that Hastelloy C276 alloy

exhibits passivation performance even in tribocorrosion. Importantly, corrosion current density during sliding is increased by three orders of magnitude compared with that under corrosion-only condition. It can be concluded that corrosion rate is significantly increased by friction in this experiment. The degradation of passive film causes high dissolution of metal, resulting in wear-induced-corrosion.

The corrosion current I measured in polarization curves during tribocorrosion can be considered the sum of two parts of currents I_a and I_p :

$$I = I_a + I_p \quad (3)$$

where I_a is the current produced from wear track, and I_p is current produced from unworn area. For the unworn area, I_p cannot exceed current value measured at the same potential in corrosion-only curves. By comparing the values of I_p and I_a around corrosion potential, it can be concluded that I is almost equal to I_a . The measured corrosion current and its evolution with potential during tribocorrosion, are characteristic of the electrochemical behavior in wear track [9].

Table 1 shows corrosion potential (ϕ_{corr}) and the corresponding corrosion current density (J_{corr}). The J_{corr} was determined using Tafel extrapolation method. The current density increases obviously with the increase of normal load and rotation speed, which is much larger than that under corrosion-only condition. High normal load and rotation speed result in low open circuit potential and high corrosion current density. Rotation speed is linked to the time interval of two successive contact events during sliding. Increasing contact frequency causes less time available for repassivation between contact intervals. Therefore, the amount of active area in wear track is large at high rotation speed, which results in low corrosion potential and high corrosion current density. The increase of corrosion current density with increasing normal load can be explained by the severely damaged passive film, severe plastic deformation and high densities of defects [9,14].

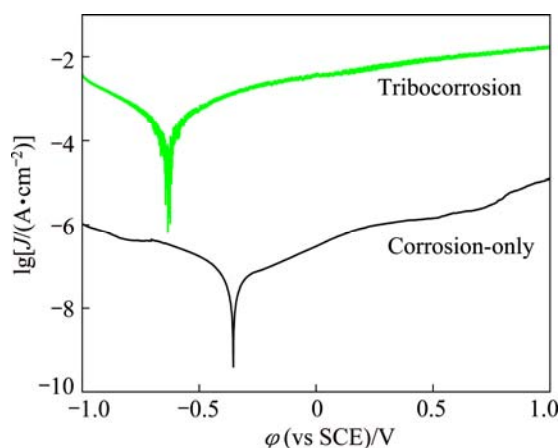


Fig. 2 Polarization curves of Hastelloy C276 alloy under corrosion-only and sliding conditions

Table 1 Corrosion potential (ϕ_{corr}) and current density (J_{corr}) for Hastelloy C276 alloy

Condition	$J_{\text{corr}}/(\mu\text{A}\cdot\text{cm}^{-2})$	$\phi_{\text{corr}}/\text{V}$
Corrosion only	0.68	-0.355
100 N, 200 r/min	703	-0.639
150 N, 200 r/min	789	-0.681
200 N, 200 r/min	842	-0.713
100 N, 100 r/min	664	-0.617
100 N, 300 r/min	778	-0.664
100 N, 400 r/min	824	-0.692

3.2 Wear behavior

Corrosion rate is significantly enhanced by friction and synergistic effect between wear and corrosion results in accelerated material loss. In order to determine the contribution of the synergy to total material loss, wear test was conducted at a cathodic potential of -0.8 V, which is lower than the corrosion potentials measured under both tribocorrosion and corrosion-only conditions. Under this electrochemical condition, there is no electrochemical corrosion and material loss is only caused by mechanical wear. Similar test procedure was employed by other investigators to evaluate the mechanical wear component for many alloys [15,16]. Figure 3 shows the evolution of current density at -0.8 V under 100 N and 200 r/min. Clearly, the measured current is cathodic before, during and after sliding, confirming that no corrosion occurs.

Table 2 shows the wear loss, mean friction coefficient and specific wear rate (k) of Hastelloy C276 alloy under both OCP and cathodic potential conditions. It can be seen that wear loss is much greater at OCP than that at cathodic potential of -0.8 V. At OCP, both

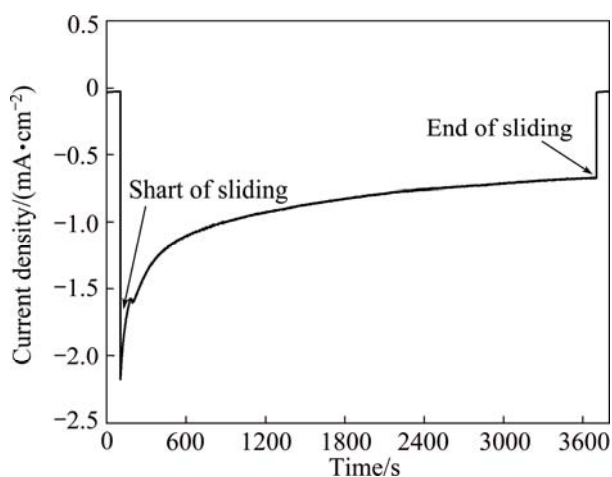


Fig. 3 Evolution of current density for Hastelloy C276 alloy at -0.8 V

corrosion and wear take place, resulting in accelerated wear loss. Clearly, corrosion has a significant effect on wear of Hastelloy C276 alloy. It is obvious that wear loss increases with increasing normal load and rotation speed. The specific wear rate is an effective parameter when sliding condition is different. Specific wear rate is in the range of 10^{-4} mm³/(N·m) for Hastelloy C276 alloy. The wear rate decreases clearly with the increase of rotation speed. The average friction coefficient of Hastelloy C276 alloy is in the range of 0.45–0.52 and increases slightly with increase of normal load.

Metal wear debris is transferred on the pin surface as shown in Fig. 4(a). The wear loss of Al₂O₃ pin is large and the surface is much roughened, as shown in Fig. 4(b). The roughened Al₂O₃ surface would abrade the metallic surface and lead to the transfer of opposing metallic debris, resulting in the severe wear. IWABUCHI et al [8,17] reported the same serious damage of Al₂O₃, which may be caused by cold welding. Such a rough surface of Al₂O₃ pin was also observed for the combination with Ni-alloy in this work.

3.3 Synergistic effect

In addition to the commonly observed phenomena related to the cathodic shift of OCP, the increase of corrosion current and the accelerated wear loss, several interesting phenomena deserve further discussion in the present experiment. These include correlation between materials loss and wear mechanism as well as the synergistic effect between corrosion and wear.

Corrosion current density in Table 1 shows the existence of accelerated corrosion by wear. The results in Table 2 clearly show the existence of accelerated wear loss by corrosion. There is obvious synergistic effect between corrosion and wear, which leads to corrosion-induced wear and wear-induced corrosion. Efforts have been made by researchers to quantify such synergistic effect using the following approach [8,18–20]. The volume loss may be explained by defining the following terms:

Table 2 Wear loss, mean friction coefficient and specific wear rate (k) of Hastelloy C276 alloy in seawater

Condition	Open circuit potential			Polarization at -0.8 V		
	Wear loss/ mm ³	Mean friction coefficient	k / (mm ³ ·N ⁻¹ ·m ⁻¹)	Wear loss/ mm ³	Mean friction coefficient	k / (mm ³ ·N ⁻¹ ·m ⁻¹)
100 N, 100 r/min	37.4	0.458	3.85×10^{-4}	28.6	0.467	2.94×10^{-4}
100 N, 200 r/min	67.7	0.471	3.48×10^{-4}	51.3	0.468	2.64×10^{-4}
100 N, 300 r/min	89.5	0.468	3.11×10^{-4}	72.2	0.479	2.48×10^{-4}
100 N, 400 r/min	116.3	0.474	2.99×10^{-4}	96.3	0.481	2.48×10^{-4}
150 N, 200 r/min	93.2	0.493	3.19×10^{-4}	74.1	0.503	2.54×10^{-4}
200 N, 200 r/min	124.3	0.502	3.20×10^{-4}	102.5	0.513	2.64×10^{-4}

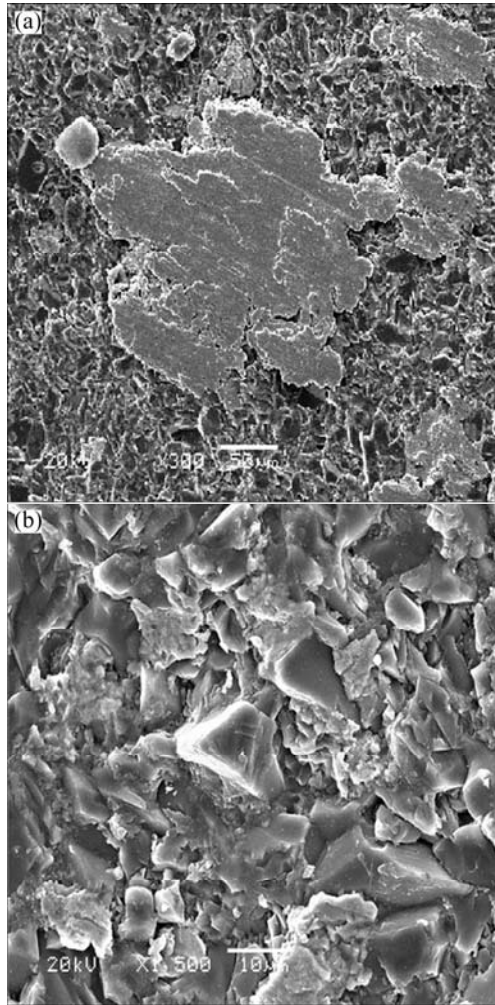


Fig. 4 SEM images of wear track on Al_2O_3 pin after friction test: (a) Transfer metal debris; (b) Roughened surface

$$K_{wc} = K_w + K_c \quad (4)$$

where K_{wc} is the total volume loss owing to corrosion-wear, K_w is the material loss owing to wear, and K_c is the material loss owing to corrosion. Here, K_w can be divided into K_{w0} and ΔK_w :

$$K_w = K_{w0} + \Delta K_w \quad (5)$$

where K_{w0} is the pure wear loss in the absence of corrosion, and ΔK_w is the volume loss owing to the effect

of corrosion on wear. And K_c can be divided into K_{c0} and ΔK_c :

$$K_c = K_{c0} + \Delta K_c \quad (6)$$

where K_{c0} is volume loss owing to corrosion in the absence of wear, and ΔK_c is volume loss owing to effect of wear on corrosion, or the enhancement of corrosion induced by wear. Moreover, the material loss due to synergistic effect between corrosion and wear (ΔK) can be expressed as

$$\Delta K = \Delta K_c + \Delta K_w \quad (7)$$

The material loss caused by corrosion K_c is given by Faraday equation as

$$K_c = Q / (ZF) \quad (8)$$

$$K_c = Mit / (ZF) \quad (9)$$

where Q is the charge passed, Z is the number of electrons involved in corrosion process, F is Faraday constant (96500 C/mol), M is the atomic mass of alloy and can be calculated by considering the mass fraction of components of the alloy, I is corrosion current, and t is corrosion time. Volume loss induced by wear in the absence of corrosion, K_{w0} , can be estimated by measuring volume change in cathodic condition at -0.8 V. As corrosion current cannot be directly measured at OCP, current density value is taken as corrosion current density calculating from polarization curve by standard Tafel procedure (Table 1).

The results of various contributions to material loss of Hastelloy C276 alloy in tribocorrosion are given in Table 3. The contribution of K_{c0} to material loss is very small and can be negligible as evidenced from Table 3. It can be found that ratio of synergistic effect (ΔK) is in the range of 17.2%–22.4%, indicating that synergistic effect between corrosion and wear is significant. The ratio of $\Delta K / K_{wc}$ decreases slightly with increasing normal load and rotation speed, which means that synergistic effect is significant especially at low normal load and rotation speed. In addition, the contribution of pure wear to total material loss exceeds 70%, which means that pure wear is the dominant factor during corrosion–wear process.

Table 3 Contribution of corrosion and wear to material loss under different corrosion-wear conditions

Condition	K_{wc}/mm^3	K_{w0}/mm^3	$\Delta K/\text{mm}^3$	$\Delta K_c/\text{mm}^3$	$\Delta K_w/\text{mm}^3$	$(\Delta K/K_{wc})/\%$	$(\Delta K_w/K_{wc})/\%$	$(\Delta K_c/K_{wc})/\%$
100 N, 100 r/min	37.4	28.6	8.8	2.5	6.3	23.5	16.8	6.7
100 N, 300 r/min	89.5	72.2	17.3	2.9	14.4	19.3	16.1	3.2
100 N, 400 r/min	116.3	96.3	20.0	3.0	17.0	17.2	14.6	2.6
100 N, 200 r/min	67.7	51.3	16.5	2.6	13.9	24.4	20.5	3.9
150 N, 200 r/min	93.2	74.1	19.1	2.9	16.2	20.5	17.4	3.1
200 N, 200 r/min	124.3	102.5	21.8	3.1	18.7	17.5	15.0	2.5

Figure 5 shows the fractions of material loss components K_{wo} , ΔK_w and ΔK_c . It should be emphasized that although electrochemical dissolution is significantly accelerated and corrosion rate is increased by hundreds of times due to friction, the contribution of wear-induced-corrosion to total material loss ($\Delta K_c/K_{wc}$) is not very high. Actually, the maximum value is 6.7% under 100 N and 100 r/min and the minimum value is only 2.5% under 200 N and 200 r/min. The ratio $\Delta K_c/K_{wc}$ decreases obviously with the increase of normal load and rotation speed. The ratio of the corrosion-induced-wear to total material loss ($\Delta K_w/K_{wc}$) is high, which is in the range of 14.6%–20.5% under all experiment conditions. Corrosion-induced-wear obviously increases the material loss. It should be emphasized that contribution of wear-induced-corrosion and corrosion-induced-wear to synergism in tribocorrosion of several passive alloys has been studied. JIANG et al [18] found that contact frequency affects the synergistic effect between wear and corrosion. At high frequency, contact interval is sufficiently short and repassivation of the wear track is prevented, which makes the wear track always in active state, leading to a large contribution of wear-induced-corrosion to total material loss. While at low frequency, great contact interval time gives wear damaging through surface and subsurface crack initiation and propagation enough time to take place, leading to a large contribution of corrosion-induced-wear to synergism. The results obtained in this experiment show that under all experiment conditions, pure mechanical wear is dominant; for synergism between corrosion and wear, corrosion-wear-induced is dominant.

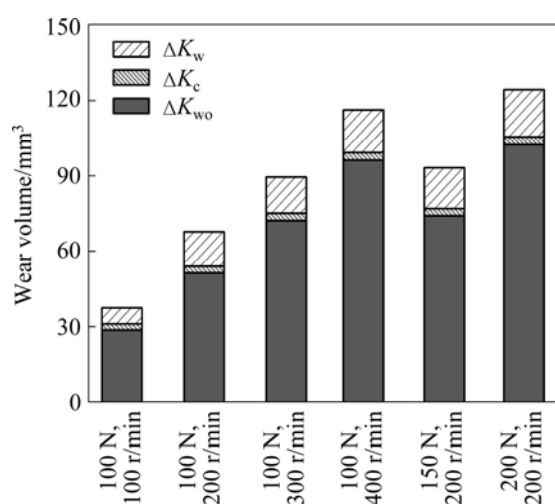


Fig. 5 Fractions of material loss factors under tribocorrosion of Hastelloy C276 alloy

Figure 6 shows the typical micrographs taken from wear tracks under OCP and cathodic potential conditions at 100 N and 200 r/min. It can be seen that worn surfaces at cathodic potential are characterized by mainly severe

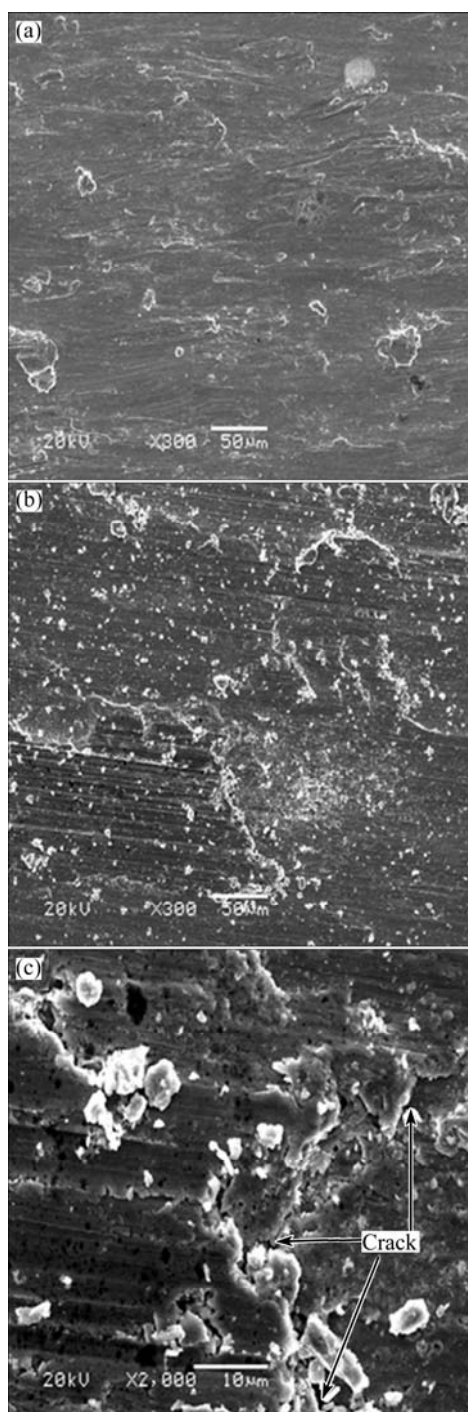


Fig. 6 Typical micrographs taken from wear tracks for Hastelloy C276 alloy at 100 N and 200 r/min: (a) Cathodic potential; (b) Contact area at OCP; (c) Crack at OCP

deformation regions interrupted by fine furrows parallel to the sliding direction, as shown in Fig. 6(a). Small cracks and thin flakes are also found on the worn surface. Under OCP condition, many wear debris are observed in Figs. 6(b) and (c), exhibiting many small corrosion pits inside the wear track at OCP. The corrosion current density is high inside the wear track during tribocorrosion, resulting in the formation of corrosion pits. And there are no apparent corrosion pits under

cathodic potential condition.

Wear can highly accelerate corrosion current density due to the following reasons. 1) Passive film is damaged or removed by wear. Consequently, bare surfaces expose to electrolyte and suffer from large metal dissolution. 2) Wear causes plastic deformation in wear track and increases the densities of point defects, cracks and dislocations, which make active surface and high corrosion rate. The presence of corrosion pits shown in Fig. 6(c) is attributed to the local corrosion in active area with high electrochemical activity [2].

Corrosion-induced-wear can be understood through the failure mechanism of passive film. According to the model proposed by JIANG et al [18], wear in tribocorrosion can be treated as a low cycle fatigue process involving crack initiation and propagation. Figure 6(c) shows the micro cracks and fatigue cracks in wear track. The tip of crack has high-density of point defects and dislocations, which have high activity and prone to dissolution. Corrosion can promote the crack initiation and propagation on the worn surface. In addition, corrosion pits can serve as a site for crack nucleation and a site of stress concentration, which can also promote crack initiation and propagation.

4 Conclusions

1) The cathodic shift of open circuit potential of Hastelloy C276 alloy in artificial seawater under sliding contact with alumina pin due to friction is confirmed and current density is increased by three orders of magnitude as compared with that without sliding.

2) There is clear synergistic effect between corrosion and wear, which leads to corrosion-induced-wear and wear-induced-corrosion during tribocorrosion. Wear loss of Al_2O_3 pin is also large when it slides against Hastelloy C276 alloy and the surface is roughened.

3) The contribution of pure wear to total material loss exceeds 70% and pure wear is the dominant factor in corrosion–wear process. For considering synergism between corrosion and wear, the contribution of wear-induced-corrosion to total material loss is not very high although corrosion rate is greatly accelerated by friction. The fraction of corrosion-induced-wear is high and in the range of 14.6%–20.5% for all experiments.

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Hastelloy C276 合金在模拟海水环境中的 腐蚀磨损协同作用

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摘 要: 利用带有电化学测试体系的销-盘腐蚀磨损试验机系统研究 Hastelloy C276 合金在模拟海水中与 Al_2O_3 陶瓷对磨时的腐蚀和腐蚀磨损行为。结果表明, 摩擦作用不仅使 Hastelloy C276 合金的开路电位大幅下降, 而且将合金的腐蚀电流密度提高了 3 个数量级。Hastelloy C276 合金的磨蚀与磨损之间存在明显的协同作用, 磨损促进了腐蚀, 反过来腐蚀也加速了磨损, 在本实验摩擦条件下纯机械磨损在金属总磨损量中的占比均超过 70%, 这说明机械磨损作用是合金腐蚀磨损失效的主导因素。虽然摩擦使得 Hastelloy C276 合金的腐蚀速度得到极大提高, 但是磨损对腐蚀的促进量在金属总磨损量中的占比并不大, 而腐蚀对磨损的促进量在总磨损量中的占比较高, 在 14.6%~20.5% 范围内变化。

关键词: 腐蚀磨损; Hastelloy C276 合金; 协同作用; 海水

(Edited by Wei-ping CHEN)