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Wear in Silicon Nitride Sliding Against Titanium Alloy Pairs at Different Loads Under Artificial Seawater Lubrication

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Tribological behaviors of Si_3N_4 ceramic sliding against titanium alloy at different normal loads under seawater lubricated conditions are investigated on a pin-on-disc tester. Generally, for Si_3N_4/titanium alloy pair, the friction coefficient increases from 0.43 to 0.67 with load increasing (from 10 to 30 N). Correspondingly, the wear rate is in the consistent case. When Si_3N_4 slides against titanium alloy, the wear mechanism is dominated by mechanical wear even though under seawater lubrication. With load increasing, the shear force of solid shear ($\tau_S \times A_r$) and furrow drag $\left(S \times P_e\right)\left(S \times P_e\right)$ aggravated, which should be responsible for the rising trend of friction coefficient and wear rate.

Keywords: friction, wear, seawater, Si_3N_4, titanium alloy

INTRODUCTION

It is well known that, the energy and environment crisis have been brought by the industrial development and increasing pollution. In this case, people have paid more and more attention to how to explore the ocean resources by utilizing the marine devices (Liu et al., 2013, 2015). Among these devices, some pumps, open drive system and some blades are all exposed to seawater during the service process, and subjected to corrosion, friction and wear. Especially, the tribological characteristics of the moving components have a significant influence on the smooth running and service life (Min et al., 2014; Wang et al., 2016). For example, seawater hydraulic drive is a key technology in ocean exploitation and widely applied in deep-sea submergence vehicle, underwater robot, submarine oil production system, and so on. This system requires high-performance frictional pair materials (Jia et al., 2004).

Recently, the marine mating components were generally fabricated from metal materials. For instance, titanium alloys, such as Ti6Al4V, possess and excellent corrosion resistance in seawater and are widely used in marine. But, self-mated metal pairs usually represented the severe adhesive wear, and the metal materials beared the electrochemical corrosion just because of their intrinsic (Wu et al., 2016; Ye et al., 2017). Compared with metals, the ceramic materials represented high hardness, high strength, and excellent corrosion resistance and outstanding tribological performance (Jianxin et al., 2006). So, more scientists focused on studying the tribocorrosion behavior of metals sliding against ceramics, which was considered to act as an inert antagonist in the tribocorrosion systems (Chen and Yan, 2012).

Among the ceramic materials, silicon nitride ceramics possess good overall performance (e.g., high hardness, high strength, low density, low thermal conductivity, low chemical activity, and high wear resistance), which promotes these applied as rolling bearing, machine tools, sliding bearing in water pumps. Until recently, some scholars have investigated the tribological properties of Si_3N_4-Ceramic and Si_3N_4-metal pairs in pure water environments, and the research results showed low friction and wear of silicon nitride against ceramic were obtained in water environment (Jahanmir and Fischer, 1988). The previous scholars found that, the formation of slica tribochemical film would be benefit to ultra-low friction and wear of self-mated silicon nitride ceramics (Tomizawa and Fischer, 1987). When silicon nitride ceramic slid against gray cast iron, low friction coefficient of 0.002 was obtained. And, the authors held that the research results was attributed to the tribochemical products and graphite (which was originated from gray cast iron; Gao et al., 1997). Also, other scholars further investigated the influence of sliding speed and surface roughness on the tribological properties of Si_3N_4/Si_3N_4 pairs in pure water, and found that the chemical wear ratio was related to sliding speed, but independent of roughness (Saito et al., 2001). Meanwhile, when Si_3N_4 slid against Al_2O_3, the wear mechanism was transferred from mechanical wear to tribochemical wear with the increase of sliding time (Balarini et al., 2016). As described above, for silicon nitride sliding pairs, lower friction coefficients and wear rated were obtained under water lubrication.

Now, scholars started to focus on the friction and wear of ceramic materials under marine condition. Liu studied on the tribological performance of silicon nitride sliding against 316 stainless steel, and the friction coefficient of 0.16 was obtained in seawater and friction coefficient of 0.48 was obtained in pure water (Liu et al., 2013). Meanwhile, when Cr/GLC film slid against Si_3N_4 ceramic, the low friction coefficient of 0.06 was obtained, which attributed to the tribochemical products (Wang et al., 2016). As described above, at present some scholars have paid more attention to the tribological Si_3N_4 ceramic in seawater, but the mating materials, the sliding condition and the research results were not coincident. The systematicness and consistency of current researches were not still enough.

In this paper, the silicon nitride ceramic materials have been prepared by the method of hot-pressed sintering, and the titanium alloy was selected as the mating materials. The tribological properties of Si_3N_4/Ti6Al4V were measured by a vertical tribotester under seawater lubrication condition. The influence mechanism of normal load on the friction and wear behaviors was further discussed. This study could provide a certain guidance to develop mating components with excellent friction and wear properties under ocean environment.

EXPERIMENTAL DETAILS

Test Rig and Materials

Friction and wear tests were carried out on a vertical universal pin-on-disc device, where an upper pin slides a bottom disc in artificial seawater environment. The disc was fixed, and the pin specimen rotated at a certain velocities. The schematic diagram of the device is illustrated in **Figure 1**. A normal load was applied on the stationary disc and the friction force was continuously measured using a strain gauge.

Si_3N_4 powder, Al_2O_3 powder, and Y_2O_3 powder were used to fabricate the ceramic composites. Si_3N_4 powder (with the particle size of 0.5 μm and the purity of 90% α- Si_3N_4) was utilized as the baseline material. Al_2O_3 powder (with the particle size of 1–2 μm and the purity of 99.9%) and Y_2O_3 powder (with the particle size of 1 μm and the purity of 99.9%) were utilized as sintering additives. Meanwhile, the volume fractions of Al_2O_3 and Y_2O_3 as sintering additives were 6 and 4%. The combined powders were mixed and ball-milled in alcohol for 24 h. The specimens of silicon nitride ceramics were sintered by hot-pressing in N_2 at a temperature of 1,800°C and with a hot-pressure of 30MPa in a graphite die. **Figure 2** shows the microstructure of Si_3N_4 ceramic, composed of elongated

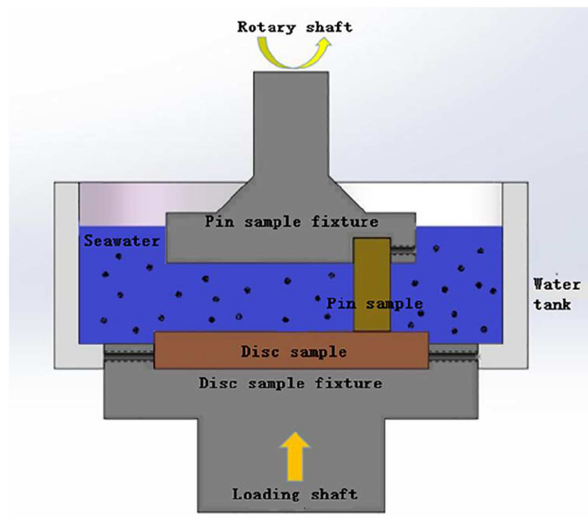

FIGURE 1 | Schematic diagram of the configuration for pin-on-disc tribometer.

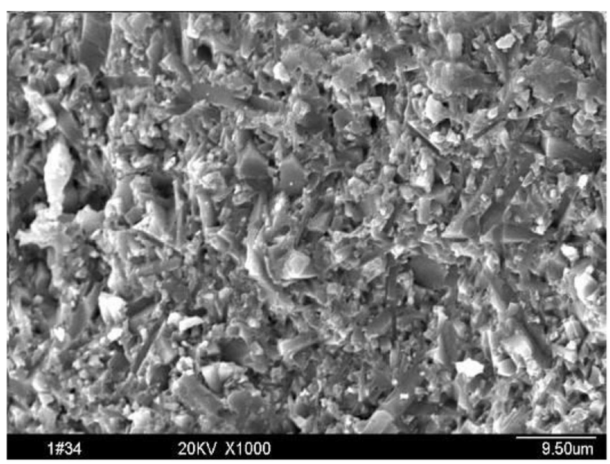

FIGURE 2 | SEM micrograph of fracture surface of Si_3N_4 ceramic.

β-Si_3N_4. The density of Si_3N_4 ceramic was 3.31 g/cm^3, the Vickers hardness was 19.9 GPa, and the fracture toughness was 8.01 MPa·m$^{1/2}$.

The dimensions of ceramic pin specimens were 5 × 5 × 10 mm. The pin specimen surfaces are ground to a surface roughness of <0.5 µm. The dimensions of titanium alloy discs were a diameter of 44 mm and a thickness of 6 mm. The titanium alloy disc was made from Ti6Al4V alloy. The disc specimen surfaces were ground to a surface roughness of <0.8 µm. The density of disc specimen is 4.52 g/cm^3, and the hardness was HRC25.

TABLE 1 | Chemical composition of seawater.

Compositions	Contents (g/L)
NaCl	24.52
$MgCl_2$	5.23
Na_2SO_4	4.06
$CaCl_2$	1.18
KCl	0.67
$NaHCO_3$	0.21
KBr	0.10
H_3BO_3	0.03
$SrCl_2$	0.028
NaF	0.003

The worn surfaces were studies using a scanning electron microscopy (SEM) equipped with energy dispersive X-ray spectrometry (EDS) and X-ray photoelectron spectroscopy (XPS).

Procedure

The tests were conducted in artificial seawater at room temperature with normal loads of 10, 20, and 30 N and a sliding speed of 1,000 r/min (namely, 1.73 m/s). The seawater was prepared according to ASTM D1141-98, and the chemical composition of seawater was shown in **Table 1**.

The sliding distance was set to be 1,000 m. The friction coefficient was recorded automatically at the steady-state sliding. Volume wear rate of specimens were determined by using equation: $V_m = \Delta m/\rho NS$, where Δm was the weight differences before and after sliding were measured by a microbalance with a high precision (0.1 mg), ρ was the density of specimens, N was the normal load and S was the sliding distance. Three samples were run to obtain average friction coefficient and wear rate under the test condition.

RESULTS AND DISCUSSION

Tribological Behaviors of Silicon Nitride/Titanium Alloy Sliding Pairs

Figure 3 shows the friction coefficients and wear rates of Si_3N_4/titanium alloy sliding pairs at different loads. From **Figure 3A**, it can be seen that, with the increase of load, the

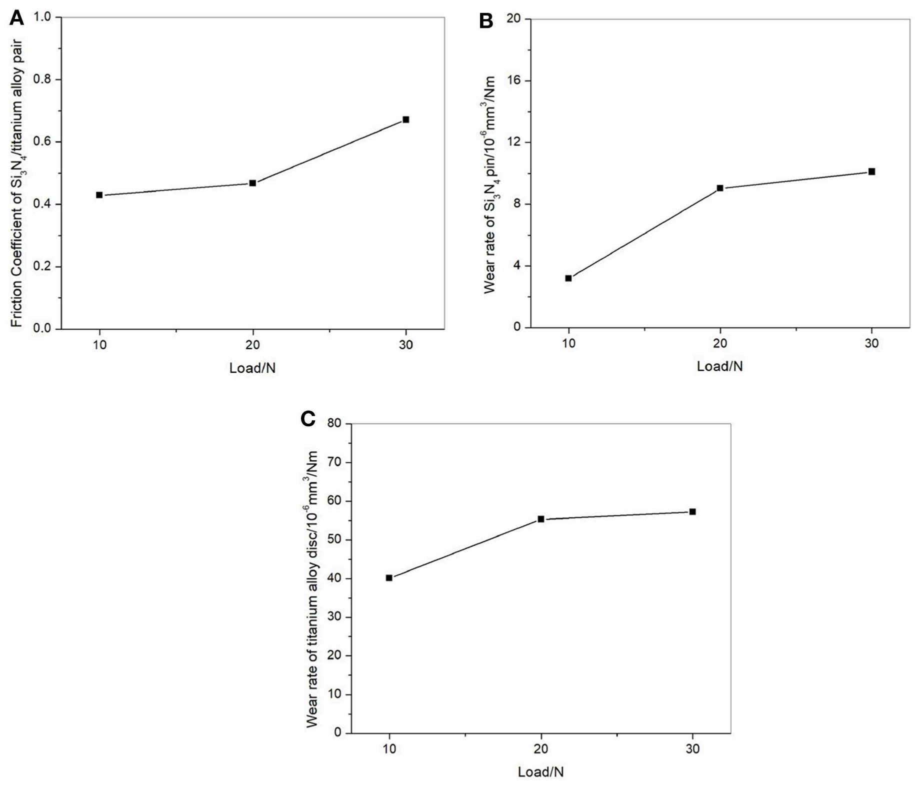

FIGURE 3 | Titanium alloy in sliding contact against Si_3N_4 in seawater, **(A)** Friction coefficient, **(B)** wear rate of pin specimen, **(C)** wear rate of disc specimen.

friction coefficient increased from 0.43 for 10 N to 0.67 for 30 N. Similarly, it can be found from **Figure 3B** that the wear rate of Si_3N_4 pin also increased with the increase of load, and the wear rate are all at the magnitude of 10^{-6} mm^3/Nm. Compared with the wear rates of pin, the wear rates of titanium alloy are higher and all at the magnitude of 10^{-5} mm^3/Nm. Obviously, the increase of load deteriorated to both of the friction and wear properties of Si_3N_4/titanium alloy in seawater.

Figure 4 shows the worn surfaces of titanium alloy disc sliding against Si_3N_4 pin at the different loads. From the figures, it can be seen that, at a low load of 10 N, wear surface was relatively smooth and contained a few micro grooves filled with a little spalling particles. When the load reached to 20 N, plastic deformation and more significant furrow was discovered from the worn surface of titanium alloy. And, at the load of 30 N, the worn surface of titanium alloy presented the severe plastic deformation and furrow with greater depth and width. While, it is interesting that no significant tribochemical film could be observed. Maybe, under this test condition, the tribochemical products were difficult to form a continuous surface film. XPS results of titanium alloy against Si_3N_4 pin at a load of 10 N were shown in **Figure 5**. The XPS results confirm the formation of

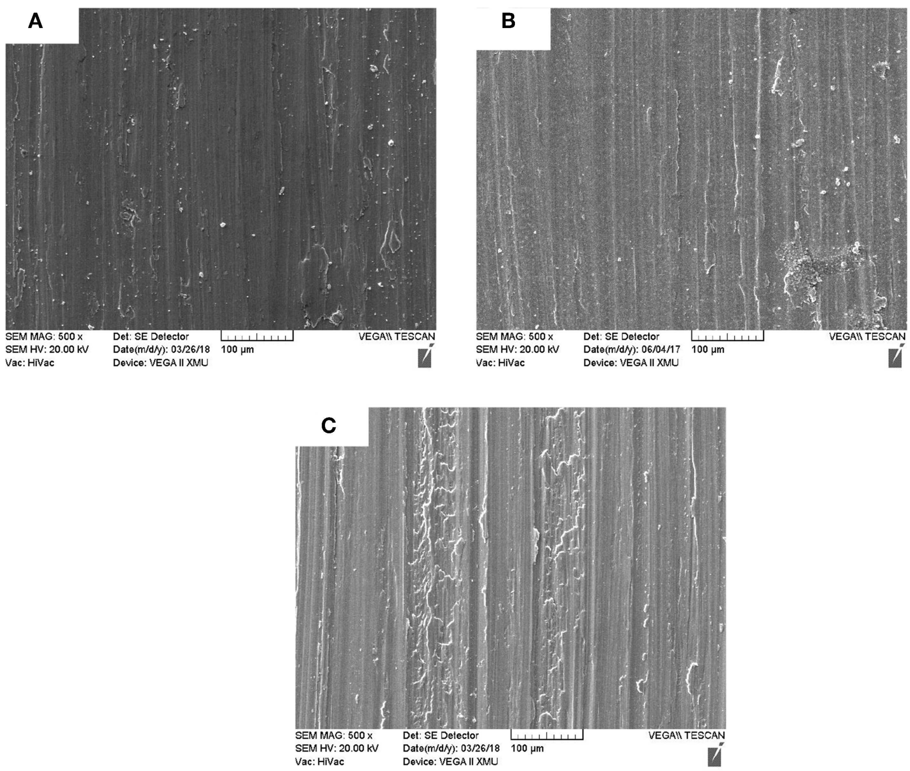

FIGURE 4 | SEM images of titanium alloy disc against Si_3N_4 pins at the loads of **(A)** 10 N, **(B)** 20 N, and **(C)** 30 N in seawater.

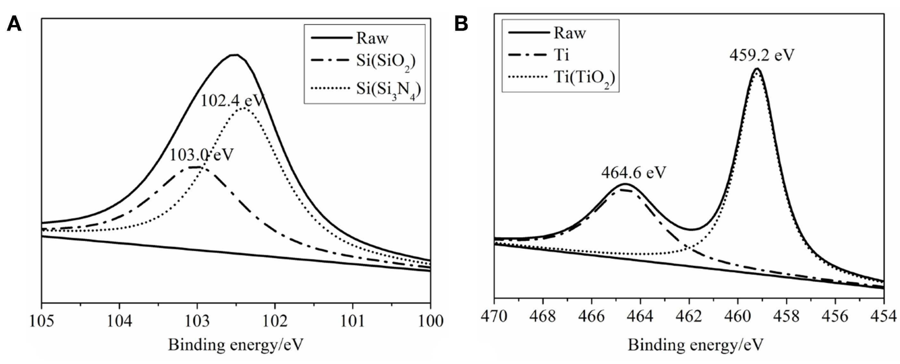

FIGURE 5 | XPS results of the worn surface of the titanium alloy disc against the silicon nitride pin at a load of 10 N: **(A)** Si_{2p} and **(B)** Ti_{2p}.

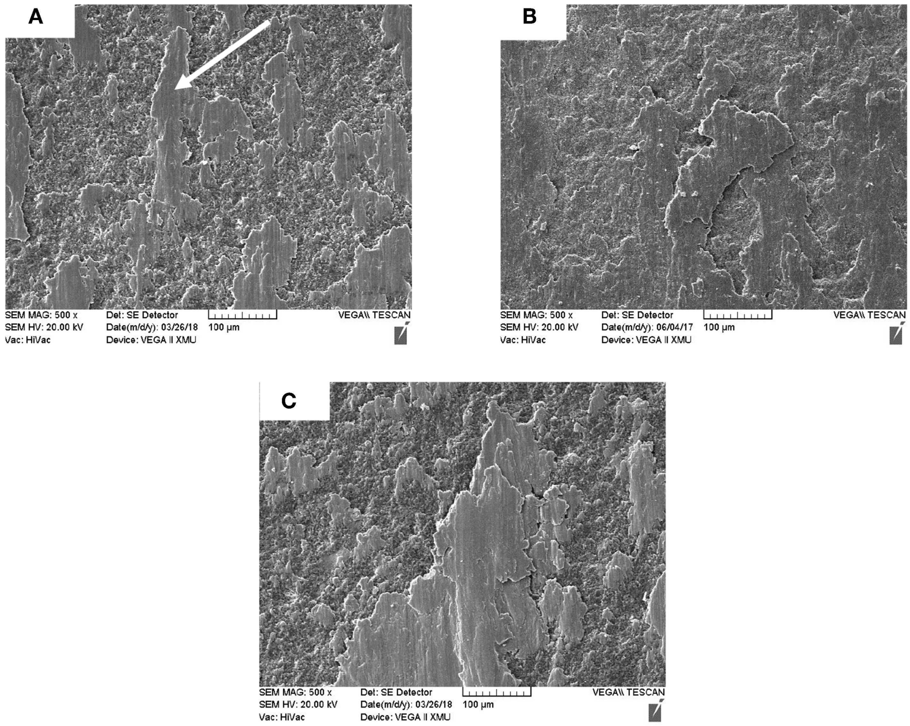

FIGURE 6 | SEM images of Si_3N_4 pins against titanium alloy disc at the loads of **(A)** 10 N, **(B)** 20 N, and **(C)** 30 N in seawater.

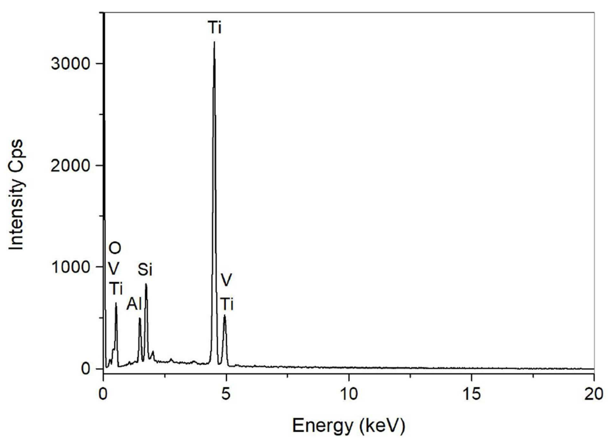

FIGURE 7 | EDS results of transferred layer on the Si_3N_4 pin against titanium alloy disc at a load of 10 N.

tribochemical products of SiO_2 and TiO_2. So, it can be concluded that some tribochemical products formed during the friction process, but a continuous and significant tribofilm did not form on the wear surfaces.

Figure 6 shows the worn surfaces of corresponding Si_3N_4 pin. As shown in **Figure 6**, metal transferred film and weaken crack can be observed on the worn surfaces of Si_3N_4 pin against titanium alloy disc at the load of 10 N (see **Figure 6A**); with the increase of load, besides transferred film, plastic deformation and furrows gradually are observed in **Figures 6B,C**. **Figure 7** shows EDS results of the transferred layer on the worn surface of Si_3N_4 pin against titanium alloy disc at a load of 10 N. From this figure, it can be seen that the transferred layer area is mainly composed of Ti, V, Al, Si and O and so on. The results indeed confirm the formation of transferred layer.

It was well known that, during friction process of ceramic/metal sliding pairs, the hard micro-bulges would plow the metal surface, and so that then some metal transferred layer would form on the ceramic surface (Hua et al., 2008; Chen et al., 2010, 2019). The metal transferred layer repeatedly formed and peeled off, which contributed to the wear of ceramic and metal. In this study, it can be clearly seen that obvious transferred layer formed on the silicon nitride ceramic surface (as shown in **Figure 6**). Simultaneously, even though a continuous tribofilm cannot be obviously observed, the results indicated that some tribochemical products formed on the wear surfaces (as shown in **Figure 5**). So, the wear surface morphology can be schematically shown in **Figure 8**. From the figure, it can be seen that the ceramic pin surface morphology was composed of transferred layer, ceramic surface, and some tribochemical products.

In our study, under seawater lubrication, the adsorbed water film, tribochemical products, and transferred layer would inhibit the direct contact of surface micro-bulges at a certain degree. Meanwhile, the direct contact of some micro-bulges would result in adhesion, abrasive wear and wear debris. From **Figures 4**, **6**, it can be observed that, for Si_3N_4/titanium alloy pair sliding in seawater, at a lower load, metal transferred film on the ceramic pin and smoother metal disc surface could be observed (see **Figures 4A**, **6A**). At a higher load, more significant transferred film on the ceramic pin and rough metal disc surface appear (see **Figures 4C**, **6C**). The contact mode can be shown in **Figure 9**. Obviously, combined with the friction and wear results, the direct contact of surface micro-bulges would aggravate with the increase of load, and the source of friction force would alter.

According to classical adhesion theory (Bhushan, 1996, 2006), the average friction force(F)at the interface between ceramic and metal can be originated from F_s is the shear force at the direct contact solid surface, F_p is the furrow drag, and F_lF_l is the shear force at the liquid surface. Meanwhile, F_s can be calculated according to the equationτ_s. Where, A_r is the sum of real solid contact area for load, and τ_s is critical shear stress at the direct contact interface between two bodies. Similarly, the shear force at the liquid surface can be calculated as $F_l = A_l \times \tau_l$. Meanwhile, the furrow drag can be calculated as $F_p = S \times P_e$. Where, S is furrow area, and P_e is furrow drag per unit area.

In this study, even some tribochemical products formed on the wear surface, but the tribochemical products might not efficiently protect and lubricate the wear surfaces. In such a setting, with the lubrication of seawater, the friction force should be calculated as follows:

$$F = A_r \times \tau_s + S \times P_e + A_l \times \tau_l$$

Combined with the friction and wear behaviors, it can be concluded that, the shear stress of solid and furrow drag should play a leading role. So, the equation can be transformed to $F \approx A_r \times \tau_s + S \times P_e$.

Some predecessors have indicated the existence of water or surface film had significant influence to the adhesion theory (Skopp et al., 1995; Deng and Cao, 2007). But, this lubrication effect was not observed in this study. In this case, if the surface contact state is elastoplastic contact, the friction coefficients of the pairs can be expressed as:

$$\mu \approx KN^{\frac{1}{3}} \times \tau_s + S \times P_e/N$$

According to this equation and experimental phenomenon, with the increase of load, the direct contact area between the micro-bulges of ceramic and metal surface aggravated, and the adhesive wear and furrow drag became more serious. Even under the lubrication and cooling of seawater, the friction coefficients and wear rates increased with the increase of load.

CONCLUSIONS

1. The normal load influences the tribological behaviors of Si_3N_4 sliding pairs under seawater considerably. The friction coefficient of Si_3N_4/titanium alloy pairs increases with the increase of normal load at a range from 0.43 to 0.67.
2. For Si_3N_4/titanium alloy pairs, with the increase of load, the shear stress between direct solid contact and furrow drag aggravated. Even under the lubrication and cooling of seawater, the friction coefficients, and wear rates increased with the increase of load.
3. The tribochemical products did not effectively protect and lubricated the wear surfaces. The wear mechanism of Si_3N_4

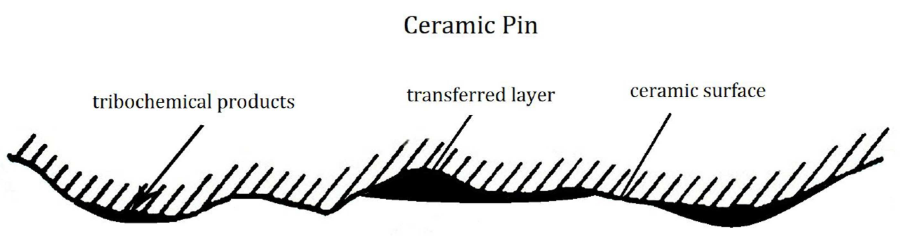

FIGURE 8 | Schematic diagram of ceramic pin surface.

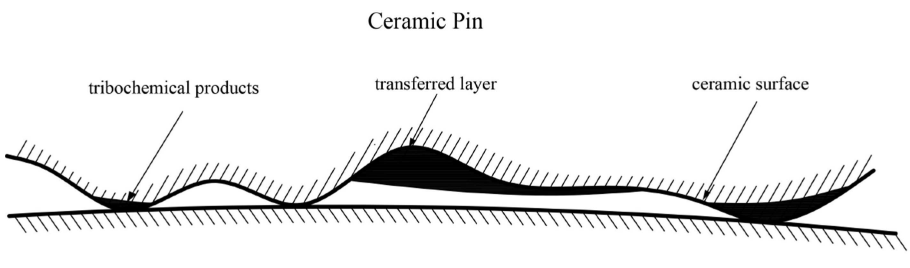

FIGURE 9 | Schematic diagram of contact model of Si_3N_4/titanium alloy pairs.

slides against titanium alloy is dominated by mechanical wear even though under seawater lubrication.

DATA AVAILABILITY

The raw data supporting the conclusions of this manuscript will be made available by the authors, without undue reservation, to any qualified researcher.

AUTHOR CONTRIBUTIONS

JZ and SM: conceptualization. WC and MZ: methodology. WC: formal analysis. JZ and SM: investigation. HL: resources. ZW: data curation. JZ and WC: writing–original draft preparation. JZ and SM: writing–review, editing, and supervision. SM and WC: project funding acquisition.

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