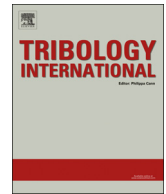




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Friction and wear properties of CrSiCN coatings with low carbon content as sliding against SiC and steel balls in water

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ARTICLE INFO

Article history:

Received 22 May 2015

Received in revised form

10 August 2015

Accepted 15 August 2015

Available online 24 August 2015

Keywords:

CrSiCN coatings

Mechanical properties

Friction properties

Water lubrication

ABSTRACT

CrSiCN coatings were deposited on Si wafer and 316L stainless steels by using unbalanced magnetron sputtering system via adjusting the trimethylsilane (TMS) flows, and their composition, microstructure and nano-hardness were characterized by X-ray diffraction (XRD), X-ray photoelectrons spectroscopy (XPS), scanning electron microscopy (SEM) and nano-indenter, respectively. The friction behaviors sliding against SiC and SUS440C balls in water were investigated. The results showed that the CrSiCN coatings presented nanocomposite microstructure of Cr(C, N) nanocrystallites embedded into amorphous matrix of Si–C–N. The coatings showed maximum hardness (19.1 GPa) deposited at the TMS flow of 10 sccm. Compared with different mating balls, the CrSiCN/SiC tribopairs exhibited better friction and wear properties than that of the CrSiCN/SUS440C steel tribopairs in water.

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1. Introduction

Due to their favorable corrosion resistance, superior wear resistance and good thermal stability chromium nitride (CrN) coatings have been successfully employed into cutting tool and mold fields [1–4]. However, the high friction coefficient of 0.7 has been obtained as the CrN coatings slid against steel balls in air [5]. In addition, when the CrN coatings slid against Si₃N₄ balls at different loads (0.5–1.5 N) in water, the friction coefficient fluctuated in a range of 0.33–0.55 [6]. The high friction coefficient could not satisfy the application requirements under extreme conditions. Furthermore, unsatisfactory hardness of CrN coatings stands in their way of wide application, especially in tribology, which considers hardness as a crucial impact. Currently, many research works have been done by introducing metal (Ti, Al, W) [7–9] or nonmetal (Si, C, B) elements [10–17] to improve the mechanical and tribological properties of CrN coatings. As seen in Table 1, the C element was proposed to be doped in the CrN coatings, and then

the CrCN showed low friction coefficient due to the self-lubrication of amorphous carbon [11,12]. For example, the CrCN coating with the carbon content of 89.2 at% exhibited low friction coefficient ($\mu=0.18$) as sliding against Si₃N₄ balls in air, but its hardness reduced to 1800 HV owing to a large amount of amorphous carbon [12]. It is known that the hardness was one of the most important material properties affecting their resistance to wear [18,19], so the decrease of hardness resulted in an increase of wear rate [12,20,21]. Wang et al. [12] have reported the CrCN coating with low carbon content (15.35 at%) slid against Si₃N₄ ceramic balls in water, the friction coefficient was below 0.2 and the hardness increased from 18.5 GPa to 22.5 GPa as compared with CrN coating. However, some Refs. [18,19] pointed out the hardness of CrCN coatings increased, which was caused by their high residual stress. Actually, higher residual stress leads to poor adhesion to substrate, and deteriorates the tribology of CrCN coatings. As compared with CrCN coating, the CrSiN coating usually showed high hardness (≥ 24 GPa) due to solid solution strengthening and the formation of nano-composite structure [22–24], better corrosion and oxidation resistance [25,26], and improved adhesion strength and tribological properties [15,23]. For instance, Lin et al. [23] have reported the hardness of CrSiN coatings increased from 26 GPa to 31–38 GPa when the Si content

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Table 1
References related to tribological properties of CrN-based coatings.

References	Tribopairs	Hardness (GPa)	Normal load (N)	Speed or frequency	Ball-on-disk type	Lubricant	C,Si content (at%)	Friction coefficient	Coatings' specific wear rate (mm ³ /Nm)
Fuentes et al. [11]	CrN/100Cr6 CrCN/ 100Cr6	12.5 13.02	2	10 cm/s	Reciprocating	In air	0 C ₂ H ₂ /[N ₂ +Ar]= 0.2–0.48	0.8 0.35–0.6	5.6 × 10 ⁻⁵ 3.6 × 10 ⁻⁵
Wang et al. [12]	CrN/Si ₃ N ₄ CrCN/Si ₃ N ₄	18.5 22.5	2	0.1 m/s	Turning	Distilled water	0 15.35	0.25 0.197	< 2.27 × 10 ⁻⁶ 2.27 × 10 ⁻⁶
Yamamoto et al. [13,14]	CrN/CrN CrSiN/ CrSiN	14–15 17	2	0.5 m/s	Turning	Distilled water	0 < 45 > 60	0.2 0.2–0.3 < 0.02	10 ⁻⁷ 10 ⁻⁷ 10 ⁻⁸
Geng et al. [15]	CrN/SUS CrSiN/SUS	14–15 17	20	0.37–1 m/s	Turning	Distilled water	0 80	0.14–0.07 0.06	No data No data
	CrN/WC CrSiN/WC	11.8 6.8–22	5	2.5 Hz	Reciprocating	Water/in air Water/in air	0 2.5–18.4	0.44/0.62 0.35–0.48/ 0.36–0.65	10 ⁻⁷ < 10 ⁻⁷
Jeon et al. [16]	CrN(CrCN)/steel	22	1	0.2 m/s	Turning	In air	–	0.62	No data
	CrSiN/steel	35					9.2	0.46	No data
	CrSiCN/Steel	43					9.2/15.6	0.42/0.32	No data
Cai et al. [17]	CrN/WC–Co	19.9	2	20 cm/s	Turning	In air	0	0.81	1.89 × 10 ⁻⁶
	CrSiCN/ WC–Co	21.6/(13.6)					1.3/(3.4)	0.71/(0.67)	1.06 × 10 ⁻⁶ /8.0 × 10 ⁻⁷

varied in the scope of 6.7–10.2 at%. When the CrSiN coatings slid against WC–Co balls in air, the friction coefficient decreased from 0.55 to 0.4–0.43, and the wear rate of CrSiN coatings dropped to the lowest values of 5–6 × 10⁻⁷ mm³/Nm. Geng et al. [15] also reported the scratch critical load (L_c) of CrSiN coatings increased from 19 N to 20–30 N. When they slid against WC balls in air, the friction coefficient and wear rates of CrSiN gradually decreased to 0.36 and 0.95 × 10⁻⁷ mm³/Nm at the Si/(Cr+Si) ratio of 12.7 at% in Table 1. Thus, the CrSiN coatings possess good wear resistance due to their high hardness. To further improve the mechanical and tribological properties of CrN or CrSiN coating, the simultaneous addition of Si and C elements into CrN coatings has been proposed. Refs. [17,27] pointed out the CrSiCN coatings have high corrosion resistance, high crack resistance and good tribological properties. Li et al. [28] have reported the CrSiCN coating showed the lower friction coefficient and higher adhesion strength than TiSiCN coating. Jeon et al. [16] also reported the micro-hardness of CrSiCN coatings was about 43 GPa owing to the formation of nc-Cr (C, N)/a-Si₃N₄ (SiC) composite, and the average friction coefficient largely depended on the contents of carbon and silicon in the coatings as sliding against steel balls in air. But for the friction and wear properties for CrSiCN coating in water environment, there are no relative works up to now.

In this paper, the CrSiCN coatings were deposited on Si wafers and 316 L stainless steels using unbalanced magnetron sputtering, and their compositions were controlled by adjusting TMS flows. The microstructure, mechanical properties and friction behavior of CrSiCN coatings sliding against SiC and SUS440C balls in water were investigated.

2. Experimental details

2.1. Deposition of CrSiCN coatings

CrSiCN coatings were deposited on Si (100) wafers and 316L stainless steel substrates simultaneously using closed-field unbalanced magnetron sputtering system (UDP-650, Teer Coatings Limited, UK). The purity of Cr is 99.9%. 316L stainless steel disks ($\Phi 30 \times 4$ mm²) were polished by a metallographic polishing

machine (UNIPOL-820) to make their surface roughness $R_a=30$ nm. Then they were cleaned ultrasonically in deionized water and ethanol for 20 min. After the substrates were installed on substrate holder, the chamber was evacuated to 4×10^{-4} Pa. Prior deposition, the substrates were sputter cleaned with Ar plasma generated by glow discharge at a bias voltage of –450 V for 30 min. Pure Ar working gas was introduced into the chamber via a MKS mass flow meter in a constant flow mode (30 sccm). A pure Cr interlayer ($\sim 0.4 \mu\text{m}$) was first deposited onto substrates for 15 min, and then the deposition of CrSiCN coatings was started for 80 min. There was no external heater to heat the substrate. However, the substrates were biased to attract the ions, and then the ion bombardment during coating deposition could cause the temperature rise of the substrates. A floating thermocouple was placed near the rotating substrate holder. The substrate temperature due to ion bombardment was approximately 150 °C. During deposition, the work gas pressure was maintained at 0.23 Pa, the rotation speed of substrate holder was 10 rpm and the bias voltage applied to substrate holder was –80 V. The target current for each Cr target was 4 A. The partial pressure of nitrogen reactive gas was controlled by optical emission monitor (OEM) with rapid feedback. This feedback process can be done by observation of the spectral line emission of the sputtering metal target and thus the flow of nitrogen was adjusted dynamically by a fast-response piezoelectric valve. In this experiment, the OEM setting for the nitrides was 50%. For the addition of Si element in the coatings, the TMS flow was controlled by a MKS mass flow meter with a flow rate of 10, 20, 30 sccm during sputtering. According to the TMS flow amount, the CrSiCN coatings in the next section were denoted as CrSiCN-10, CrSiCN-20, CrSiCN-30, respectively.

2.2. Microstructure, mechanical and tribological characterization of CrSiCN coatings

The coatings' surface and cross-section micrographs were observed using a field emission scanning electron microscope (SEM) (JEOL-JSM-7001F), and their crystal phase and bonding structure were characterized using a X-ray diffractometer (XRD, Ultima IV, Japan) with Cu K α radiation source ($\lambda=0.1542$ nm) at a scan rate of 5° per minute from 20° to 80° and X-ray photoelectron

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