

See discussions, stats, and author profiles for this publication at: <https://www.researchgate.net/publication/233058680>

Tribological properties of SiC coatings deposited by r.f. magnetron sputtering as a function of substrate temperature

Article in *Philosophical Magazine Letters* · July 2011

DOI: 10.1080/09500839.2011.583287

CITATIONS

7

READS

80

6 authors, including:



Sonali Dash

Chandigarh University

218 PUBLICATIONS 2,960 CITATIONS

[SEE PROFILE](#)



Srinath Rajagopalan

Vel Tech - Technical University

428 PUBLICATIONS 25,799 CITATIONS

[SEE PROFILE](#)



Ashok Bahuguna

Indira Gandhi Centre for Atomic Research

6 PUBLICATIONS 101 CITATIONS

[SEE PROFILE](#)

Some of the authors of this publication are also working on these related projects:



Self-cleaning coating for solar panel applications [View project](#)



Studies on Hampi Musical Pillars [View project](#)

This article was downloaded by: [Indira Centre for Atomic Research]

On: 22 August 2011, At: 20:29

Publisher: Taylor & Francis

Informa Ltd Registered in England and Wales Registered Number: 1072954 Registered office: Mortimer House, 37-41 Mortimer Street, London W1T 3JH, UK



Philosophical Magazine Letters

Publication details, including instructions for authors and subscription information:

<http://www.tandfonline.com/loi/tphl20>

Tribological properties of SiC coatings deposited by r.f. magnetron sputtering as a function of substrate temperature

N. Kumar ^a, S. Dash ^a, S. Rajagopalan ^a, Ashok Bahuguna ^a, A.K. Tyagi ^a & Baldev Raj ^a

^a Surface and Nanoscience Division, Material Science Group, Indira Gandhi Centre for Atomic Research, Kalpakkam 603 102, Tamil Nadu, India

Available online: 03 May 2011

To cite this article: N. Kumar, S. Dash, S. Rajagopalan, Ashok Bahuguna, A.K. Tyagi & Baldev Raj (2011): Tribological properties of SiC coatings deposited by r.f. magnetron sputtering as a function of substrate temperature, *Philosophical Magazine Letters*, 91:7, 465-472

To link to this article: <http://dx.doi.org/10.1080/09500839.2011.583287>

PLEASE SCROLL DOWN FOR ARTICLE

Full terms and conditions of use: <http://www.tandfonline.com/page/terms-and-conditions>

This article may be used for research, teaching and private study purposes. Any substantial or systematic reproduction, re-distribution, re-selling, loan, sub-licensing, systematic supply or distribution in any form to anyone is expressly forbidden.

The publisher does not give any warranty express or implied or make any representation that the contents will be complete or accurate or up to date. The accuracy of any instructions, formulae and drug doses should be independently verified with primary sources. The publisher shall not be liable for any loss, actions, claims, proceedings, demand or costs or damages whatsoever or howsoever caused arising directly or indirectly in connection with or arising out of the use of this material.

Tribological properties of SiC coatings deposited by r.f. magnetron sputtering as a function of substrate temperature

N. Kumar*, S. Dash, S. Rajagopalan, Ashok Bahuguna,
A.K. Tyagi and Baldev Raj

Surface and Nanoscience Division, Material Science Group, Indira Gandhi Centre for Atomic Research, Kalpakkam 603 102, Tamil Nadu, India

(Received 30 September 2010; final version received 18 April 2011)

Silicon carbide coatings have been deposited by r.f. magnetron sputtering on AISI 304LN stainless steel at four different substrate temperatures. At the lowest substrate temperature of 300°C, the film was amorphous whereas at a substrate temperature of 600°C a crystalline SiC phase was obtained. However, at substrate temperature of 800°C, the films were amorphous again. The tribological properties of the coatings were also found to change with substrate temperature. The coefficient of friction was found to be low for the crystalline films. Such a low value of coefficient of friction is indicative of superior wear resistance. A higher coating thickness enhanced the tribological properties while any surface roughness of the substrate and coating was found to degrade these properties. Micro-Raman spectra of the films varied with substrate temperature. Some phases were found to disappear and new phases were observed in the wear track with progressive sliding.

Keywords: magnetron sputtering; SiC films; phases; tribology; friction

1. Introduction

Silicon carbide coatings are technologically advanced materials characterised by high hardness, wear resistance and low friction. In addition, their chemical inertness, high thermal stability and dielectric properties are useful for applications involving extreme sliding conditions [1]. High hardness and high toughness provide abrasion and fracture resistance, aspects that further improve the tribological properties of SiC coatings [2]. They can be deposited by a variety of techniques such as laser-assisted deposition [3], dynamic ion mixing [4], plasma-enhanced chemical vapour deposition [5] and magnetron sputtering [1,6]. Magnetron sputtering is an efficient technique on account of its simplicity, high deposition rates and wide acceptability to industry. Amorphous SiC mostly forms at low deposition temperatures, which has a hardness comparable to that of crystalline SiC [7,8]. Furthermore, because of its excellent mechanical properties, a-SiC has been also used for a variety of high-temperature and machining applications, such as MEMS coatings [9], dielectric copper diffusion barriers [10], sliding friction [11,12] and wear

*Corresponding author. Email: niranjan@igcar.gov.in

resistance [13]. It has been shown that the mechanical properties of a-SiC coatings can be increased by different approaches including the control of composition [14].

In this study, changes in the structure of the coatings deposited by r.f. magnetron sputtering are investigated as a function of substrate temperature. The role of substrate temperature on the tribological properties of the coatings is studied and Raman spectroscopy is used to correlate the tribological properties with evolution of various phases occurring on the surface of coatings and in wear scars.

2. Experimental techniques

AISI 304 LN steel was used as a substrate in which to deposit the SiC. The films were sputtered from a silicon carbide target using an r.f. magnetron sputtering system. A discharge at a pressure of 3×10^{-3} mbar in high-purity argon gas with an r.f. power of 100 W was maintained during deposition. The substrate temperatures used were 27°C, 300°C, 600°C and 800°C. The crystal structure of the coatings was examined by GIXRD with an incidence angle of 1°. The linear reciprocating mode of a ball-on-disk CSM tribometer was used to carry out tribological tests of the coatings. A spherical steel ball (100Cr6 SS) of diameter 6 mm was used as a sliding body to measure the coefficient of friction with the normal load and sliding speeds kept constant at 1 N and 2 cm/s, respectively. Tests were performed under ambient conditions (dry and unlubricated) with a relative humidity of 62%. The film thickness and surface roughness were measured using stylus profilometer (Dektak 6M). The adhesive strength of the coatings was measured using a micro-scratch tester (Revetest, CSM instruments, Switzerland). A spherical diamond indenter with a radius of curvature of 200 μm was used to carry out progressive load scratch tests. Micro-Raman measurements were performed using a spectrometer (Horiba Jobin Yvon) equipped with a microscope to record the phase evolution on the surface and the wear tracks. For these measurements, laser radiation with a wavelength 514.5 nm was used as an excitation source at a power of 5 mW.

3. Results and discussion

The surface roughness of the coatings was found to be dependent on the roughness of the substrate. At a high substrate roughness of 82 nm the coating roughness was found to be 15 nm and with decreasing substrate roughness the coating roughness was also found to decrease as shown in Figure 1. At a deposition temperature of 800°C, the surface of the coating was found to have an rms roughness of 21 nm; this high value may arise from a combination of substrate roughness and roughness induced by the high deposition temperature. Higher deposition temperature causes larger grain sizes to occur, which in turn gives a rougher surface. The thickness of the coating was found to be 1.6 μm at a deposition temperature of 300°C. This value is found to decrease with deposition temperature. It was only 0.6 μm at a deposition temperature of 800°C. The effect of coating thickness may also influence the roughness parameters. With increase in thickness of the coatings, the effect of substrate roughness becomes reduced.

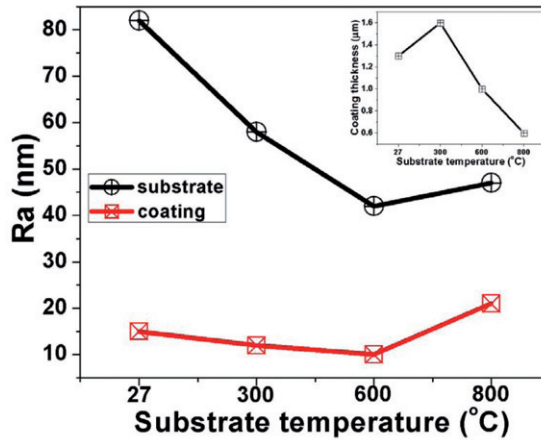


Figure 1. Substrates and coating surface roughness versus film thickness.

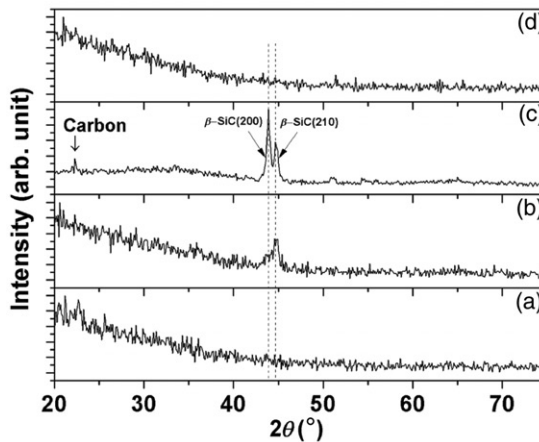


Figure 2. XRD of SiC coatings at substrate temperatures of (a) 27°C, (b) 300°C, (c) 600°C and (d) 800°C.

X-ray diffraction analysis of samples confirms that the deposition products are β -SiC and free carbon, as illustrated in Figures 2a and b. There are three peaks in the spectrum assigned as disordered sp^3 -carbon peaks (111) at 22.4° and β -SiC with preferred orientation of (200) and (210) diffraction line 43.9° and 44.7°, respectively. At low (27°C) and high (800°C) deposition temperatures in Figures 2a and d respectively, these peaks are found to be absent owing to the formation of amorphous compounds of SiC. At lower deposition temperatures, the adatom mobility and the energy of the carbon atoms become low which restricts the formation of crystalline SiC. At high deposition temperature, the formation kinetics and kinetic energy of the various species of adatoms are expected to be higher,

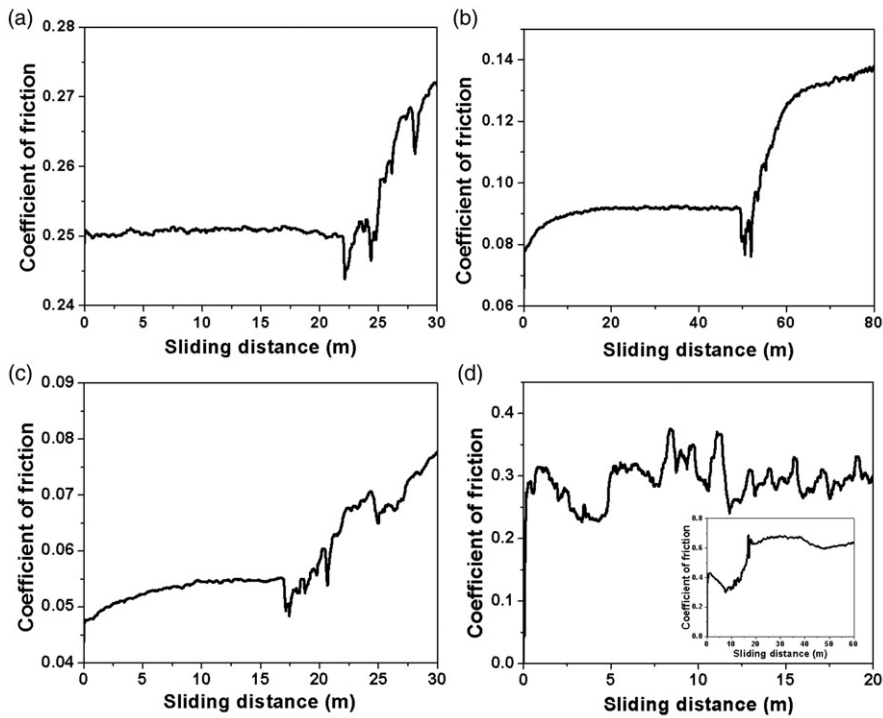


Figure 3. Coefficient of friction of coatings deposited at substrate temperatures of (a) 27°C, (b) 300°C, (c) 600°C, and (d) 800°C.

leading to fractionation which also disrupts the formation of an ordered lattice structure of SiC on account of stoichiometric defects in the coatings.

The coefficient of friction is found to vary with deposition temperature as shown in Figure 3. In each figure, there are regimes of low and high friction as shown in Figures 3a–c. Low-friction regimes are caused by sliding over the SiC coatings. A clear transition stems from failure of the SiC layer, yielding high value of coefficient of friction. This value is found to be unstable after a transition towards the higher value, which is attributed to third body interaction with worn out SiC particles from the deformed steel substrate. At low deposition temperature (300°C), where the crystalline phase of SiC is practically absent (Figure 2a), the value of the coefficient of friction is found to be 0.25 in the lower regime. After ~22 m of wear distance, the coating is found to break down and the coefficient of friction marginally rises to 0.27. In this condition, the wear rate is found to be $1.6 \times 10^{-7} \text{ mm}^3/\text{Nm}$. Coatings deposited at high substrate temperatures (300°C and 600°C) have a significantly lower coefficient of friction as seen from Figures 3b and c where the crystalline phase of SiC is detected from GIXRD. These steady values before the frictional transition were found to be lower by 0.054 and 0.092 at the deposition temperatures of 300°C and 600°C, respectively. A reduced wear rate of $2.8 \times 10^{-8} \text{ mm}^3/\text{Nm}$ is found in coatings deposited at a substrate temperature of 600°C. The wear rate rises to $4.7 \times 10^{-8} \text{ mm}^3/\text{Nm}$ for coatings deposited at 300°C.

This value drops to $3.8 \times 10^{-7} \text{ mm}^3/\text{Nm}$ for coatings deposited at 800°C . The coating deposited at 800°C has minimum wear resistance with a high value of coefficient of friction and an unstable friction curve. The higher wear rate is concomitant with a high value of friction coefficient as shown in Figure 3d. The wear rate of the AISI 304LN substrate obtained under similar test condition is found to be $2.7 \times 10^{-6} \text{ mm}^3/\text{Nm}$. These experimental findings indicate that a low friction coefficient is associated with improved wear resistance. In addition, a low coefficient of friction is associated with low cohesive and easy shearability of the contacting surfaces. Such materials display a low deformation of wear track. High wear resistance impedes sliding after longer sliding distances. The transition between the friction regimes is determined by the thickness of the coatings. With increase in thickness, a steady value of the coefficient of friction persists over longer sliding distances. It is seen from Figure 1 that the thickness of the coating is high for the sample deposited at 300°C and for which a shift towards higher coefficient of friction is found after a longer wear distance ($\sim 49 \text{ m}$). As seen from Figure 3c and for a coating thickness of $\sim 1 \mu\text{m}$, the transition in the value of the coefficient of friction occurs rather early i.e. after a short sliding distance of $\sim 17 \text{ m}$. This significant difference in the coefficient of friction could also arise from either coating or substrate surface roughness. It is also noted that the coefficient of friction decreases with decrease in substrate and coating surface roughness. As shown in Figure 3a, when the substrate and coating roughness are found to be high, a higher value of friction results. A few sliding passes can smoothen the roughness and eliminate its influence on the coefficient of friction. However, at a further sliding distance, the substrate surface, which is otherwise shielded by the SiC coating, is exposed and becomes a contributing factor. The roughness of the coating surface can be destroyed during early passes of tribo-sliding and thus no longer have an effect on the coefficient of friction. Figure 3d does not depict a low-friction regime. The altogether absence of a low-friction regime arises from the amorphous nature of the coating on the one hand, and on the other, from a built in higher surface roughness of $\sim 21 \text{ nm}$ in this $\sim 0.6 \mu\text{m}$ coating thickness which is also low. After a few sliding passes, coating failure is observed which increases the value of the coefficient of friction. Three-body interactions also destabilise this value as shown in the figure. In the inset of Figure 3d the evolution of the coefficient of friction of the steel substrate is shown, which is significantly higher compared to that of the SiC coating. Even, after the failure of the SiC coating, the coefficient of friction is significantly lower compared to steel. This may be explained by the formation of a tribo-layer which prohibits direct tribo-contact with the substrate during sliding [15]. The adhesive strength between coating and substrate was measured using progressive load scratch tests. Coating deposited at 300°C shows the highest adhesive strength of 18 N which decreases for the 300°C , 600°C and 800°C deposited films to 16 N , 14 N and 8 N , respectively. There is no direct correlation of the friction coefficient to the adhesive strength of the coating with the substrate. The adhesive strength between the coating and substrate is related to interfacial properties whereas friction is a surface property of the material. Adhesion of the sliding surfaces is known to be an important factor for the dissipation of frictional forces. Larger adhesion causes higher shear strength, which is attributed to larger friction. Accordingly, the frictional force is known to be a combination of effects from adhesion and deformation. The adhesive force

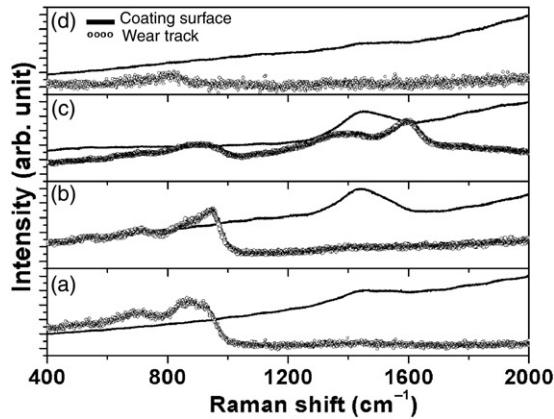


Figure 4. Micro-Raman of surface and wear scars of coatings deposited at substrate temperatures of (a) 27°C, (b) 300°C, (c) 600°C, and (d) 800°C.

between the contacts depends on the many factors, such as the chemical structure, the atomic configuration, the potential energy and the roughness of the surface. The surface energy is known to be less for crystalline compared to amorphous phases. To establish the composition and phase-dependent wear resistance properties of such a coating, micro-Raman spectral investigations on the coating surface and the tribo-track were carried out. These are shown in Figure 4.

The Raman lines of silicon and carbon are strongly dependent on the structural properties of the coatings, in particular, both the line position and the FWHM of the transverse optical phonon mode are strongly dependent on the local order prevailing in the silicon network. On the coating surface, the disordered form of sp^3 -bonded amorphous carbon exhibits a line centred at 1437 cm^{-1} as shown in Figures 4a–d. This peak occurs with high intensity and low broadening as shown in Figure 4b and c. This disorder accompanying sp^3 -bonding is rather low [16]. The spectral line is found to be feeble as shown in Figure 4d. Raman analysis did not reveal the existence of sp^2 -bonding which is normally encountered in amorphised carbon networks. Vibrational modes representing the formation of crystalline a SiC phase is also not detected at the coating surface. It is known that stoichiometric SiC does not have a Raman active vibrational mode at 514.5 nm [16,17]. After tribological tests, micro-Raman spectra from the wear track shows significant differences compared to the spectra obtained from the coating surface. Some new phases appear and some existing phases disappear in the wear tracks. Peaks corresponding to the formation of amorphous carbon are found to be absent in the wear track as seen from Figures 4a, b and d. The sliding brings about wearing out of this surface layer. In Figure 4c, it is seen that vibrational bands appear at 1368 cm^{-1} and 1595 cm^{-1} corresponding to evolution of ‘disordered’ D- and ‘graphitic’ G- bands of carbon networking [18]. A new phase indicated by the line at 532 cm^{-1} corresponds to the formation of amorphous silicon [8]. Peaks centred at 708 cm^{-1} , 896 cm^{-1} and 944 cm^{-1} correspond to the formation of a SiC phase as shown in Figures 4a, b and c. The weak feature at 804 cm^{-1} is assigned to the formation of SiC

as presented in Figure 4d. These spectra reflect the phonon density of states, which changes in the wear track during tribo-chemical reaction [16,19]. These spectral lines correspond to SiC and free silicon in wear tracks. Formation of SiC with sp^3 -phases as shown in Figure 4c bring about significant decline in coefficient of friction. This is illustrated in Figure 3c.

4. Conclusions

Silicon carbide coatings were deposited by r.f. magnetron sputtering on AISI 304LN steel at four different substrate temperatures. A clear transition from amorphous silicon carbide to a crystalline phase of this compound occurred on changing the substrate temperature from 300°C and 600°C, while at a higher substrate temperature of 800°C, the coating was again amorphous. This is attributed to the accumulation of defects and elemental deficiencies in the lattice caused by the segregation of free silicon and carbon. The tribological properties of these coatings are influenced by the relative proportions of the phases comprising the coating. The coefficient of friction and wear rate were found to be least in the case of the crystalline phase of SiC with the formation of sp^3 -bonding. These were found to be 0.052 and $2.8 \times 10^{-8} \text{ mm}^3/\text{Nm}$, respectively for the coating deposited at a substrate temperature 600°C. Higher coating thicknesses caused a decrease in the friction while surface roughness of both substrate and coating increased this value. Micro-Raman spectra of the coatings varied with deposition temperature with the formation of a tribo-layering on the wear track. This phase was found to reduce the friction. The evolution of amorphous carbon in the wear track also reduces the friction.

Acknowledgements

The authors thank Dr T.R. Ravindran, CMPD/IGCAR, for the micro-Raman spectroscopic measurements, Ms S. Kalavathi, CMPD/IGCAR, for carrying out the XRD, and Ms G. Vijayakumari, SND/IGCAR for specimen preparation. They are also grateful to Dr C.S. Sundar, Director, MSG/IGCAR, for sustained encouragement. Dr N. Kumar especially thanks Prof E.A. Davis, Editor of *Philosophical Magazine Letters*, for his valuable guidance.

References

- [1] M. Lattemann, E. Nold, S. Ulrich, H. Leiste and H. Holleck, *Surf. Coat. Technol.* 174–175 (2003) p.365.
- [2] P. Andersson and A. Blomberg, *Wear* 174 (1994) p.1.
- [3] H. Sung, G. Erkens, J. Funken, A. Voss, O. Lemmer and E.W. Kreutz, *Surf. Coat. Technol.* 54–55 (1992) p.541.
- [4] J.P. Rivike, M. Zaytouni and J. Delafond, *Surf. Coat. Technol.* 67 (1994) p.43.
- [5] H. Yoshihara, H. Mori and M. Kiuchi, *Thin Solid Films* 76 (1981) p.1.
- [6] L. Guzman, S. Tuccio, A. Miotello, N. Laidani, L. Calliari and D.C. Kothari, *Surf. Coat. Technol.* 66 (1994) p.458.
- [7] A.K. Costa and S.S. Camargo, *Surf. Coat. Technol.* 163–164 (2003) p.176.

- [8] A.K. Costa, S.S. Camargo Jr., C.A. Achete and R. Carius, *Thin Solid Films* 377–378 (2000) p.243.
- [9] N. Ledermann, J. Baborowski, P. Mural, N. Xantopoulos and J.-M. Tellenbach, *Surf. Coat. Technol.* 125 (2000) p.246.
- [10] M.J. Loboda, J.A. Seifferly, C.M. Grove and R.F. Schneider, *Mater. Res. Soc. Symp. Proc.* 447 (1997) p.145.
- [11] B. Goranchev, K. Reichelt, J. Chevallier and P. Hornsho, *Thin Solid Films* 139 (1986) p.275.
- [12] Y. Awad, M.A. El Khakani, D. Brassard, R. Smirani, N. Camire, M. Lessard, C. Aktik, M. Scarlete and J. Mouine, *Thin Solid Films* 518 (2010) p.2738.
- [13] M. Le Contellec, J. Richard and A. Guiyarc'h, *Thin Solid Films* 58 (1979) p.407.
- [14] Y. Awad, M.A. El Khakani, C. Aktik, J. Mouine, N. Camire, M. Lessard, M. Scarlete, H.A. Al-Abadleh and R. Smirani, *Surf. Coat. Technol.* 204 (2009) p.539.
- [15] S. Wilson and A.T. Alpas, *Wear* 245 (2000) p.223.
- [16] F. Neri, S. Trusso, C. Vasi, F. Barreca and P. Valisa, *Thin Solid Films* 332 (1998) p.290.
- [17] M. Lattemann, S. Ulrich, H. Holleck, M. Stuber and H. Leiste, *Diamond Related Mater.* 11 (2002) p.1248.
- [18] A.C. Ferrari and J. Robertson, *Phys. Rev. B* 61 (2000) p.14095.
- [19] V.I. Ivashchenko, P.E.A. Turchi and V.I. Shevchenko, *Phys. Rev. B* 75 (2007) p.085209.