

MURASAWA, K., DIAB, M.R., WANG, M., NARAGINO, H., YOSHITAKE, T. and EGIZA, M. 2024. Influence of droplet-free ta-C coatings and lubrication conditions on tribological performance and mechanical characteristics of WC-Co. *Materials letters* [online], 372, article number 137058. Available from: <https://doi.org/10.1016/j.matlet.2024.137058>

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2024

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Influence of droplet-free ta-C coatings and lubrication conditions on tribological performance and mechanical characteristics of WC–Co

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ARTICLE INFO

Keywords:

ta-C coatings
Tribological performance
WC–Co substrates
Arc ion plating
Wear resistance

ABSTRACT

Cemented carbide (WC–Co) tools suffer from surface abrasion, limiting their performance. This study explores droplet-free tetrahedral amorphous carbon (ta-C) coatings deposited via arc ion plating as a solution. The coatings possess a dense, sp³-rich structure, leading to a remarkable hardness of 60 GPa compared to 37 GPa of WC–Co, and strong adhesion with a critical scratch load of 41 N. Tribological tests confirm their effectiveness. Dry sliding tests show reduced wear and lower CoF (0.123) compared to uncoated tools (0.159). Notably, water-soluble lubricants yielded the best performance (lowest CoF: 0.092, superior wear resistance), while water and mineral oil also improved performance.

1. Introduction

Cemented carbide (WC–Co) cutting tools are widely used in industrial processes but are prone to surface abrasion, a primary cause of tool failure [1]. Wear resistance is crucial for the longevity and performance of mechanical components, impacting surface quality, operational stability, and service life in both dry and lubricated conditions. To address these issues, coatings with exceptional hardness and wear resistance are essential.

Tetrahedral amorphous carbon (ta-C) coatings exhibit promising potential for enhancing the wear resistance of cutting tools. Their superior hardness and wear resistance are attributed to their high sp³ bonding content. Arc ion plating (AIP) is a deposition technique capable of producing dense ta-C films with optimal sp³ bonding [2,3], surpassing the adhesion properties of magnetron sputtering [4]. Previous studies have demonstrated the ability of AIP to achieve ta-C coatings with hardness values ranging from 19.1 to 28.6 GPa, with peak sp³ content at –100 V bias [5]. Moreover, optimization of AIP process parameters has resulted in ta-C films with exceptional hardness (56.7 GPa) and elastic modulus (721.1 GPa) [3], highlighting the technique's suitability for depositing super-hard coatings.

A significant obstacle to the widespread adoption of ta-C coatings is

the formation of macro-particles during deposition, which can adversely affect adhesion, promote oxidation, and degrade overall coating performance [6]. In tribological applications, the presence of macro-particles exacerbates issues, leading to extended running-in periods and inconsistent frictional behavior. Consequently, developing deposition processes that minimize or eliminate macro-particle formation is essential for realizing the full potential of ta-C coatings.

This study explores droplet-free ta-C coatings deposited via AIP to enhance the wear resistance of WC–Co cutting tools, focusing on mechanical and tribological evaluations under diverse lubrication conditions to optimize real-world performance.

2. Experimental methods

Tetrahedral amorphous carbon (ta-C) films were deposited on polished WC-6 %Co substrates via arc ion plating (details in [supplementary materials](#)). Surface morphology and structure (sp³ content) were analyzed using FE-SEM (JEOL JSM-IT700HR), 3D laser confocal microscopy (Olympus LEXT OLS 5000), and Raman spectroscopy (in Via, Renishaw, 532 nm). Nanoindentation (Picodentor HM500) was employed to assess the hardness and Young's modulus of the 150 nm-thick ta-C film. To minimize substrate influence, a maximum

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indentation depth of 15 nm (10 % of film thickness) was used. Film adhesion was assessed via scratch testing (Anton Paar RST3). Tribological performance of ta-C coatings was assessed using a pin-on-disk wear tester (FPR2200) under dry and lubricated (water, mineral oil, water-soluble) conditions mimicking cutting scenarios (1 N load, 600 s duration, 100 rpm, Al₂O₃ counter-body). Wear profiles were analyzed using the 3D laser confocal microscope to quantify wear resistance.

3. Results and discussion

Arc ion plating successfully produced droplet-free ta-C coatings with 150 nm film thickness on WC–Co substrates, as confirmed by FE-SEM analysis (Fig. 1a and b). This approach addresses a key challenge in achieving smooth ta-C films for tribological applications. Compared to the bare substrate ($R_a = 3$ nm, $S_a = 4$ nm), the ta-C coatings exhibited slightly higher roughness values ($R_a = 7$ nm, $S_a = 7$ nm) as measured by 3D laser confocal microscopy (Fig. 1c and d). Nevertheless, these minimal surface irregularities suggest the potential for enhanced wear resistance due to reduced contact area during tribological interactions.

Raman spectroscopy (Fig. 1e) offered a non-destructive probe into the bonding configuration of carbon atoms within ta-C films. This technique exploits the characteristic vibrational modes of the material, where specific Raman peak positions correspond to distinct bonding types. Two key peaks were identified: the G-peak (~ 1550 cm⁻¹) and the D-peak (~ 1345 cm⁻¹). The G-peak signifies the presence of sp² hybridized carbon atoms arranged in a hexagonal lattice structure, as found in graphite. Conversely, the D-peak originates from defects or disorder within the carbon network, often associated with sp² clusters or dangling bonds. The I_D/I_G intensity ratio of 0.36 implies minimal structural disorder and a dominance of sp³ hybridized carbon atoms in the ta-C film [7]. This high sp³ content, estimated around 70 %, likely contributes to the exceptional hardness and wear resistance observed later in the analysis.

Nanoindentation revealed a substantial enhancement in the mechanical properties of ta-C coatings compared to the WC–Co substrate (Fig. 2a). The ta-C coatings exhibited remarkable hardness (60 ± 5 GPa) and Young's modulus (829 ± 60 GPa) compared to the substrate's 37 GPa and 770 GPa, respectively. This exceptional hardness is attributed to a dense and sp³-rich amorphous structure as confirmed by Raman analysis, resulting from highly energetic C⁺ species during deposition.

The observed plasticity index ($H/E=0.072$) suggests a good balance between elastic and plastic behavior, potentially contributing to

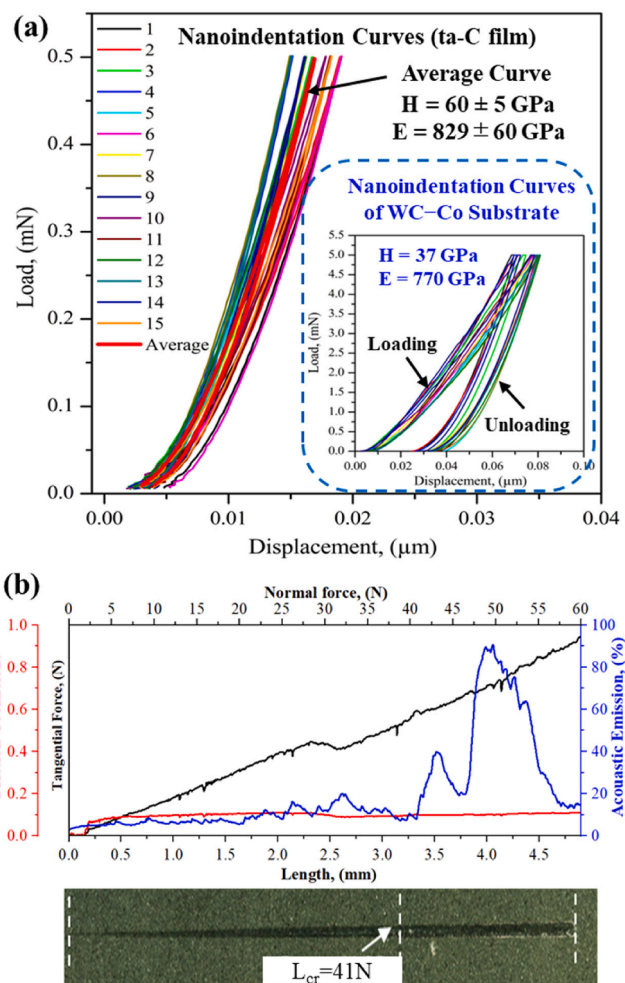


Fig. 2. (Colour online) (a) Nanoindentation results of ta-C coating and WC–Co substrate, and (b) Acoustic scratch test results with track image of the ta-C coating.

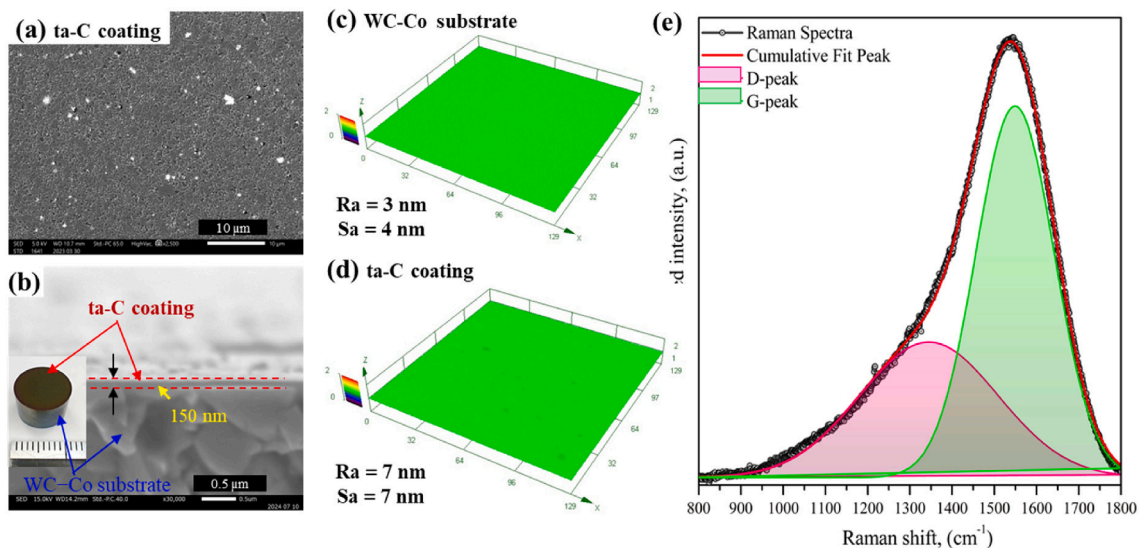


Fig. 1. (Colour online) (a) Top and (b) cross-sectional FE-SEM images of the ta-C coated WC–Co substrate, Surface topography of the (c) bare WC–Co substrate and (d) ta-C coatings, and (e) Raman spectrum of the ta-C coating.

enhanced wear resistance. Furthermore, the increased fracture toughness ($H^3/E^2 = 0.31$ GPa) compared to the substrate (0.085 GPa) implies improved resistance to crack propagation under stress. These superior mechanical properties can be attributed to the effects of plasma properties during deposition, particularly the high ion bombardment onto the bare substrate.

The adhesion strength of the ta-C film on the WC–Co substrate was evaluated using a scratch test (Fig. 2b). The critical load (L_{cr}) for film delamination was determined to be 41 N, obtained through acoustic emission and optical microscopy, indicating remarkable adhesion. The strong adhesion of the ta-C film to the WC–Co substrate is attributed to several factors. Substrate heating during deposition enhances atomic mobility, promoting better bonding between the film and substrate. Moreover, the low substrate temperature of 150 °C effectively prevents cobalt diffusion and subsequent graphitization at the interface, which could degrade adhesion.

Friction and wear characteristics of the ta-C coating were evaluated using pin-on-disk tests against a ceramic Al_2O_3 disk in open air without/with external lubricating. Al_2O_3 (Fig. 3a-onset), known for its high hardness and abrasion resistance, presents a significant tribological challenge. Fig. 3a presents the variation of the friction coefficient of ta-C film with sliding time under dry and lubricant sliding. Fig. 3b summarizes the steady-state CoF and wear volume of ta-C film compared with bare substrate.

The tribological performance of ta-C coatings was evaluated using a pin-on-disk test against a challenging alumina (Al_2O_3) counter body, simulating real-world harsh friction and wear environments encountered in various applications [8]. Compared to the bare WC–Co substrate (CoF=0.159), the ta-C coating exhibited a lower CoF (0.123) under dry sliding conditions. This reduction can be attributed to two key mechanisms. Firstly, wear-induced graphitization likely occurred at contact asperities due to localized temperature rise during friction. This graphitization process creates a low-shear transfer layer between the ta-C coating and the Al_2O_3 counter body, potentially reducing friction by minimizing interfacial shear strength. Secondly, the superior plastic resistance index (H/E) of the ta-C coating ($H/E=0.072$) compared to the substrate ($H/E=0.048$) suggests a better balance between elastic and plastic behaviour. This property can contribute to improved wear resistance by allowing the coating to absorb and dissipate contact stresses more effectively.

The lubrication conditions significantly impacted the tribological behaviour of the ta-C coatings. Water, introduced as a coolant, slightly reduced the coefficient of friction ($\mu = 0.116$) compared to dry sliding. This decrease is likely due to the ability of water to absorb frictional heat

and maintain lower contact temperatures, thereby reducing graphitization and interfacial adhesion. Mineral oil, with its lubricating properties, further minimized the friction coefficient ($\mu = 0.097$) and wear rate. The oil likely formed a lubricating film at the contact interface, reducing shear forces and minimizing material transfer between the ta-C coating and the counter body.

The most favorable tribological performance was achieved with water-soluble lubricants ($\mu = 0.092$), which effectively manage frictional heat and provide a lubricating film similar to mineral oil, leading to the lowest friction coefficient for ta-C coatings (Fig. 3c–g). The exceptional wear resistance of ta-C coatings under lubricated conditions is attributed to several key factors: high hardness ($H=60$ GPa) enhances resistance to deformation and wear, an ultra-smooth, droplet-free surface ($R_a = 7$ nm) reduces contact area, high sp^3 content ($\sim 70\%$) ensures mechanical strength, and strong film-substrate adhesion ($L_{cr} = 41$ N) maintains coating integrity. These properties significantly improve tribological properties of WC–Co tool.

4. Conclusion

This study demonstrates the effectiveness of droplet-free ta-C coatings, deposited via arc ion plating, in significantly enhancing the wear resistance of WC–Co cutting tools. The resulting coatings exhibited exceptional properties, including a high sp^3 content of approximately 70 %, a hardness of 60 ± 5 GPa, and strong adhesion to the substrate ($L_{cr} \approx 41$ N). Compared to uncoated tools, ta-C coated tools displayed a 22 % reduction in coefficient of friction under dry conditions and a remarkable 31 % reduction when using water-soluble lubricants. This translates to a coefficient of friction of just 0.092, accompanied by minimal wear. These findings underscore the potential of ta-C coatings to substantially improve the durability and performance of WC–Co cutting tools in demanding industrial applications.

CRediT authorship contribution statement

Koki Murasawa: Writing – original draft, Resources, Methodology. **Mohamed Ragab Diab:** Writing – original draft, Methodology, Formal analysis, Validation, Visualization. **Mei Wang:** Methodology, Validation, Visualization. **Hiroshi Naragino:** Methodology, Investigation. **Tsuyoshi Yoshitake:** Writing – review & editing, Supervision, Funding acquisition. **Mohamed Egiza:** Writing – review & editing, Writing – original draft, Supervision, Conceptualization, Project administration.

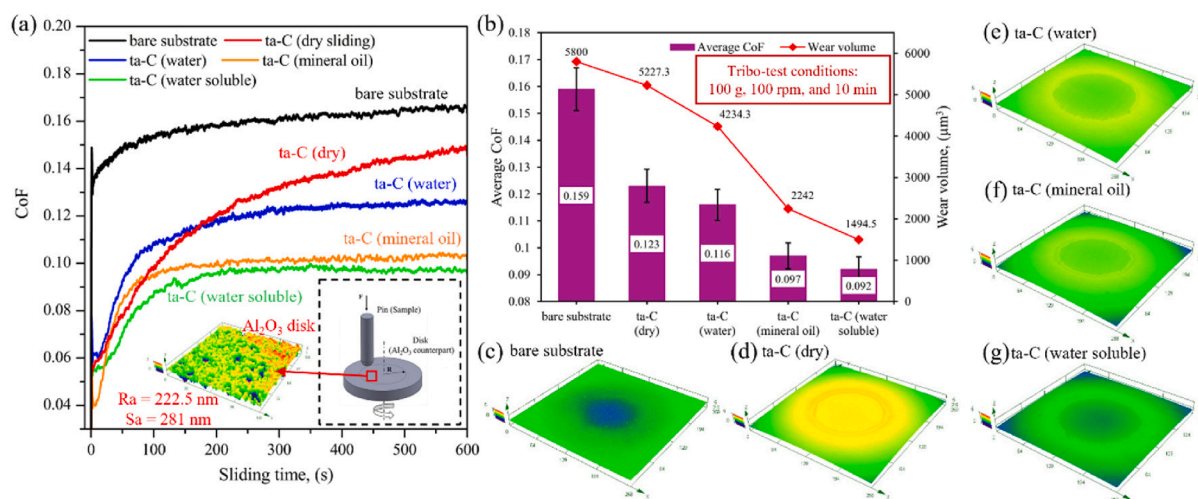


Fig. 3. (Colour online) (a) Friction coefficient of ta-C coating against Al_2O_3 disk. (b) Average CoF and wear volume of ta-C coating under dry and lubricated conditions. (c–g) Wear morphology of ta-C pin-samples after dry and wet sliding tests.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The authors do not have permission to share data.

Acknowledgment

This work was supported by Advanced Machining Technology and Development Association, JST A-STEP Stage II (AS2915051S).

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.matlet.2024.137058>.

References

- [1] Z. Tang, X. Huang, C. Wang, B. Liao, D. Lin, J. Yao, L. Zheng, Micro-drilling characteristics of sequential glass fiber-reinforced resin-based/copper foil multi-layer sheets, *Int. J. Adv. Manuf. Technol.* 107 (1) (2020) 399–410, <https://doi.org/10.1007/s00170-020-05038-x>.
- [2] B. Scheffel, O. Zywitzki, T. Kopte, Diamond-like films of tetrahedral amorphous carbon deposited by anodic arc evaporation of graphite, *Surf. Coat. Technol.* 477 (2024) 130305, <https://doi.org/10.1016/j.surfcoat.2023.130305>.
- [3] M. Wang, L. Zhang, G. Lin, Improved mechanical and tribological properties of diamond-like carbon films by adjusting pulsed substrate bias, *Diam. Relat. Mater.* 130 (2022), <https://doi.org/10.1016/j.diamond.2022.109402>.
- [4] R. Lan, Z. Ma, C. Wang, G. Lu, Y. Yuan, C. Shi, Microstructural and tribological characterization of DLC coating by in-situ duplex plasma nitriding and arc ion plating, *Diam. Relat. Mater.* 98 (2019) 107473, <https://doi.org/10.1016/j.diamond.2019.107473>.
- [5] Y.S. Zou, Y.F. Wu, H. Yang, K. Cang, G.H. Song, Z.X. Li, K. Zhou, The microstructure, mechanical and friction properties of protective diamond like carbon films on magnesium alloy, *Appl. Surf. Sci.* 258 (4) (2011) 1624–1629, <https://doi.org/10.1016/j.apsusc.2011.10.031>.
- [6] Y.-J. Jang, J.-I. Kim, W.-Y. Lee, J. Kim, Friction properties of thick tetrahedral amorphous carbon coating with different surface defects under dry contact conditions, *Appl. Surf. Sci.* 550 (2021) 149332, <https://doi.org/10.1016/j.apsusc.2021.149332>.
- [7] A.C. Ferrari, Determination of bonding in diamond-like carbon by Raman spectroscopy, *Diam. Relat. Mater.* 11 (3) (2002) 1053–1061, [https://doi.org/10.1016/S0925-9635\(01\)00730-0](https://doi.org/10.1016/S0925-9635(01)00730-0).
- [8] K. Wegener, F. Kuster, S. Weikert, L. Weiss, J. Stirnimann, Success story cutting, *Procedia CIRP* 46 (2016) 512–524, <https://doi.org/10.1016/j.procir.2016.04.110>.

Influence of Droplet-Free ta-C Coatings and Lubrication Conditions on Tribological Performance and Mechanical Characteristics of WC–Co

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Deposition of ta-C Films by arc ion plating method:

Tetrahedral amorphous carbon (ta-C) films were deposited on cemented carbide (WC–Co) substrates using cathodic arc ion plating (Fig. 1). This high-energy technique utilizes a vacuum arc to generate highly ionized carbon vapor from a target (99.99% pure carbon). The negatively biased substrate attracts these positive carbon ions, leading to film growth. K-type WC–Co substrates ($\phi 10 \times 5.5$ mm) with a polished surface roughness (R_a) of $0.003 \mu\text{m}$ were employed. Prior to deposition, the substrates underwent ultrasonic cleaning in a sequence of acetone, methanol, and finally, deionized water. The films were deposited under vacuum (≈ 1 Pa) onto a heated substrate (≈ 150 °C) maintained at a negative bias voltage (≈ 300 V). The resulting ta-C films had a thickness of approximately 150 nm and a deposition rate of $0.3 \mu\text{m/hr}$.

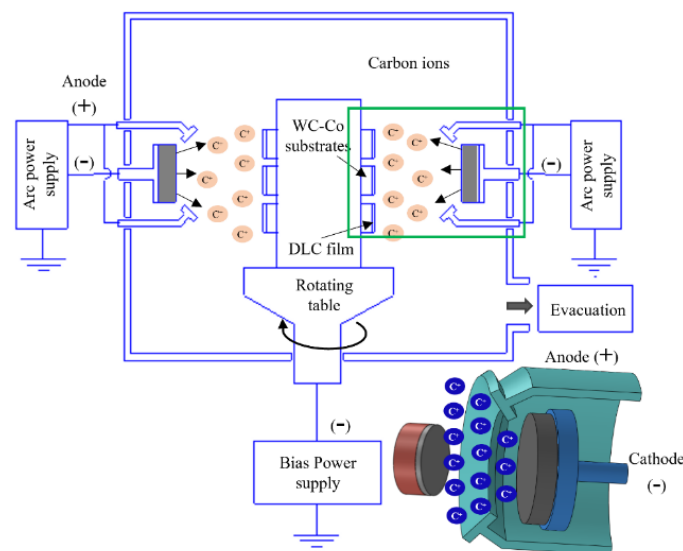


Fig. 1 (Colour online) Schematic diagram of cathodic arc ion plating used for ta-C deposition.