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Investigation into synergistic effects of DLC/TiAlN-coated tool and water-based lubrication enhanced with hybrid MXene/Ionic Liquid additive using ironing of stainless steel and tribological testing

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KEYWORDS

DLC coating
Lubrication
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Ionic Liquid
Ironing process
Tribological performance

ABSTRACT

This study investigates new tribosystems combining Water-based Lubricants (WBLs) with double-layered DLC/TiAlN tool coating. The WBLs were formulated using a hybrid additive comprising two-dimensional MXene and Ionic Liquid (IL), added with Carboxymethyl Cellulose (CMC) surfactant. At first, ironing of 1-mm-thick SUS304 stainless steel sheet was selected to investigate the combined effects, as it replicates tribologically severe industrial contact condition occurring under 20% sheet thickness reduction. Results demonstrated an 8% reduction in forming force when using DLC/TiAlN-coated tools lubricated with MXene/IL/CMC WBL, along with significantly reduced tool wear compared to dry friction condition. To confirm this, further assessment was conducted using tribological testing at room temperature. To verify these findings, additional tribological testing was conducted at room temperature. The tests revealed a 26% reduction in friction when using CMC_MXene/IL WBL, while wear decreased by 7.3% for DLC/TiAlN-coated tools compared to uncoated tools.

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1.0 INTRODUCTION

The metal forming industry continually seeks innovative solutions to enhance the efficiency, durability, and environmental sustainability of manufacturing processes. Tool degradation poses a significant challenge that directly affects product quality. This deterioration is characterized by progressive wear on the tool surface, resulting in a reduced tool lifespan. Generally, an alternative approach involves lubricant, where traditional lubrication methods often effectiveness in reducing wear and friction as well as reducing wear and improving tool performance. However, their application raises significant environmental concerns, including pollution and challenges in disposal, which contribute to ecological degradation posing environmental and ecological risks(Nowak et al., 2019). Therefore, water-based lubricants have emerged as a promising alternative due to their eco-friendly nature and potential to minimize environmental impacts. Other alternative involves coating the tool surfaces is effective to minimize tool wear and extend tool life. To overcome these limitations, advanced coatings and lubricants have emerged as promising solutions. In this context, the combination of double-layered coatings, such as Diamond-Like Carbon (DLC) and Titanium Aluminum Nitride (TiAlN), with advanced water-based lubricants, presents a compelling avenue for improving the ironing process. While diamond-like carbon (DLC) coatings offer the advantage of reducing friction and enhancing wear resistance(Kashyap et al., 2022), their application in processes like metal cutting and ironing, which involve high normal pressures, necessitates the use of effective yet potentially dangerous lubricants to prevent the coatings from detaching.

MXenes have gained significant attention in the field of tribology due to their potential as lubricant additives. MXenes are two-dimensional materials with excellent mechanical and tribological properties. The covalent functionalization of MXenes has been explored for tribological purposes, with a focus on enhancing their performance as lubricants. Parra-Muñoz et al.(Parra-Muñoz et al., 2022) provided a critical review of the existing state-of-the-art regarding MXenes' covalent functionalization, emphasizing tribological properties and needs. Lian et al.(Lian et al., 2018) conducted a systematic study of the tribological properties of MXenes as solid lubricant coatings. In their research, they prepared a 200 nm thick Ti_3C_2 MXene coating on a copper disk using a spray coating method. The result Ti_3C_2 coatings showed coefficients of friction (COF) of 0.15 and 0.19 at normal loads of 0.5 and 1.0 N, respectively. This value is lower than the uncoated copper disc. Correspondingly, the wear rate follows the same trend. In addition to MXenes, other materials such as ionic liquids and nanoparticles have also been investigated for their potential as lubricant additives. In study Khantun et al.(Khatun & Roy, 2022) explored the optimal etching duration, ranging from 2 to 48 h, for the preparation of $Ti_3C_2T_X$ MXenes. Findings indicated that shorter etching periods did not sufficiently remove aluminum, and a 24-hour etching period was identified as the most effective for preparing $Ti_3C_2T_x$ MXenes

Ionic liquids (ILs) have emerged as promising additives in lubricants due to their unique physiochemical properties and lubricating capabilities. Research has shown that ILs can significantly reduce friction coefficients and wear rates in various tribological systems(Avilés et al., 2022; Jiménez et al., 2022; Lee-Niinioja, 2022). The molecular structure of ILs plays a crucial role in their lubrication mechanisms, with specific IL compositions demonstrating superior performance in reducing friction and enhancing durability(Jieming et al., 2023; B. Wang et al., 2022). For instance, ILs containing fluorinated segments and hydroxyl endgroups have shown exceptional thermal stability and lubricity, outperforming traditional lubricants like perfluoropolyether (PFPE) in hard disk drives(B. Wang et al., 2022). Additionally, the addition of certain ILs as lubricant additives has been found to mitigate abrasive wear mechanisms and improve overall lubricating performance, making them a viable option for next-generation lubricants in various applications(Avilés et al., 2022). Rohlmann et al.(Rohlmann et al., 2021), found ionic facilitates the formation of a protective boundary layer on metal surfaces, significantly reducing friction and improving lubrication. Its phosphate anions promote the formation of a tribofilm, which helps minimize material loss and extend the life of machine components.

The effectiveness of coating is often linked to the number of layers it contains; in addition, a greater number of layers can also improve other characteristics, such as hardness (Caliskan et al., 2017; Shuai et al., 2020). Double-layer coatings are widely used in research and industrial applications to improve material performance by combining the benefits of two different layers. The main advantage of using two-layer coatings is the ability to increase wear resistance, where the base layer provides hardness and durability, while the top layer reduces friction and prevents surface damage (Liu et al., 2022). This combination also improves lubricity, as a well-designed top layer, such as diamond-like carbon (DLC), can significantly reduce friction, improving the tribological behavior of the coated surface. In addition, two-layer coatings offer increased heat and oxidation resistance, making them suitable for high-temperature environments. The base layer, such as titanium aluminum nitride (TiAlN), provides excellent heat resistance and oxidation protection, allowing the coating to maintain its integrity under extreme conditions (Sebbe et al., 2024).

This research investigates the synergistic effects of a DLC/TiAlN double-layered coating system, and a novel water-based lubricant enhanced with $Ti_3C_2T_x$ MXene multilayer nanoflakes and the ionic liquid tributyl(methyl)phosphonium dimethyl phosphate. The DLC/TiAlN coating is renowned for its exceptional hardness, wear resistance, and thermal stability, making it an ideal candidate for protecting forming tools under extreme conditions. Meanwhile, the incorporation of MXene nanoflakes and ionic liquids into the lubricant aims to reduce friction, improve thermal conductivity, and enhance the lubricant's load-bearing capacity. MXenes, a class of two-dimensional materials, exhibit unique mechanical and tribological properties, while ionic liquids offer excellent thermal stability and boundary lubrication characteristics.

By integrating these advanced materials, this study aims to optimize the ironing process, reduce tool wear, and improve the surface quality of formed components. Furthermore, the use of water-based lubricants aligns with the growing demand for environmentally friendly manufacturing practices. This research not only contributes to the understanding of the tribological interactions between advanced coatings and lubricants but also provides practical insights for the development of sustainable and efficient metal forming technologies. Through experimental analysis and performance evaluation, this work seeks to establish a foundation for the application of DLC/TiAlN coatings and MXene-enhanced lubricants in industrial metal forming processes.

2.0 EXPERIMENTAL PROCEDURE

2.1 Material

Double Layered Coating:

The tool surfaces were coated with single and double hard coatings using the High-Power Impulse Magnetron Sputtering (HiPIMS) technique, through the support from CemeCON Scandinavia A/S. The DLC/TiAlN was deposited on tool surfaces, with the DLC coating serving as the top layer. The average thickness of the test coatings 4 ± 0.4 µm. Figure 1 shows the cross-sectional view of DLC/TiAlN.

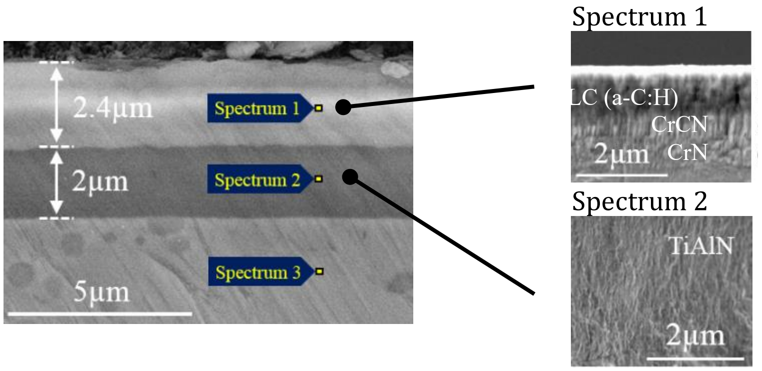


Figure 1: A cross-sectional view of the tool surface with deposited DLC/TiAlN coating (Note: Spectrum 1 – DLC, Spectrum 2 – TiAlN, and Spectrum 3 – Cutting tool surface)

Water-Based Lubricants:

Commercialized Ti_3C_2Tx MXene multilayer Nanoflake with Purity 99.9 % (Nanoshell, India) and ionic liquid tributyl(methyl)phosphonium dimethyl phosphate with >95 % (Tokyo Chemical Inc, Japan) were used as additive in this research. The preparation of WBLs involves the use of 0.5 % additives, distilled water as oil based, and surfactants to increase the stability of the particle dispersion. 0.25% Ti_3C_2Tx Mxene Multilayer Nanoflake (Mxene) and 0.25% Tributyl(methyl)phosphonium Dimethyl Phosphate (IL) as additive are first dispersed in distilled water using a magnetic stirrer. Afterwards, 0.5% surfactant Carboxyl Methyl Cellulose (CMC) was added to the mixture to increase the viscosity and ensure stable dispersion, with continuous stirring at 1000 rpm for 30 minutes at 50°C. The mixture was then processed in a homogenizer for 5 minutes at 8000 rpm, repeated twice. Figure 2 illustrates the production process of water-based lubricants.

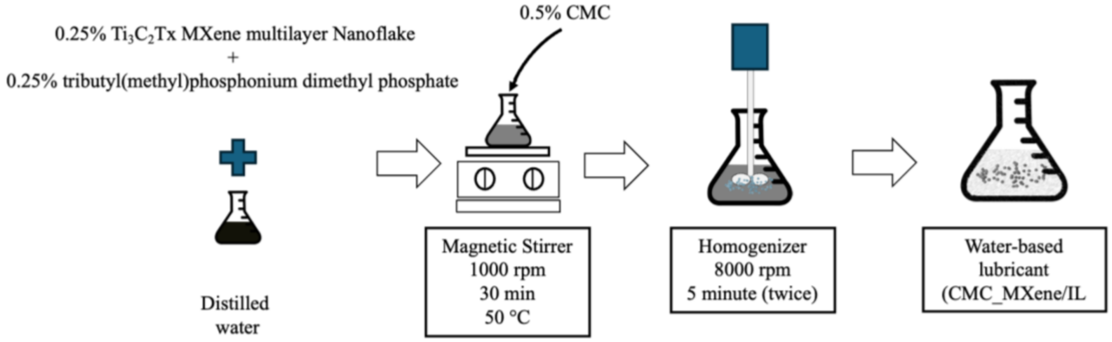


Figure 2: Preparation ow water-based lubricant.

2.2 Testing

Ironing Process:

Ironing has been selected to evaluate the combined effect of coating and WBL. Testing was conducted using an ironing tool coated with a double layer of DLC/TiAlN, which was also applied with WBL CMC_Mxene/IL. The setup of the machine is shown Figure 3. The workpiece used stainless steel SUS304 had a thickness of 1 mm and a diameter of 8 mm. The wear on the tool will be observed using a digital microscope. The ironing process had a 20% reduction of thick workpieces. The ironing tool used has a diameter of 4.4 mm and a nose radius of 1 mm. The die used has a diameter of 6 mm and an upper radius of 1.5 mm. The operation is carried out at a consistent speed, with each stroke executed at a rate of 250 strokes per minute (SPM).

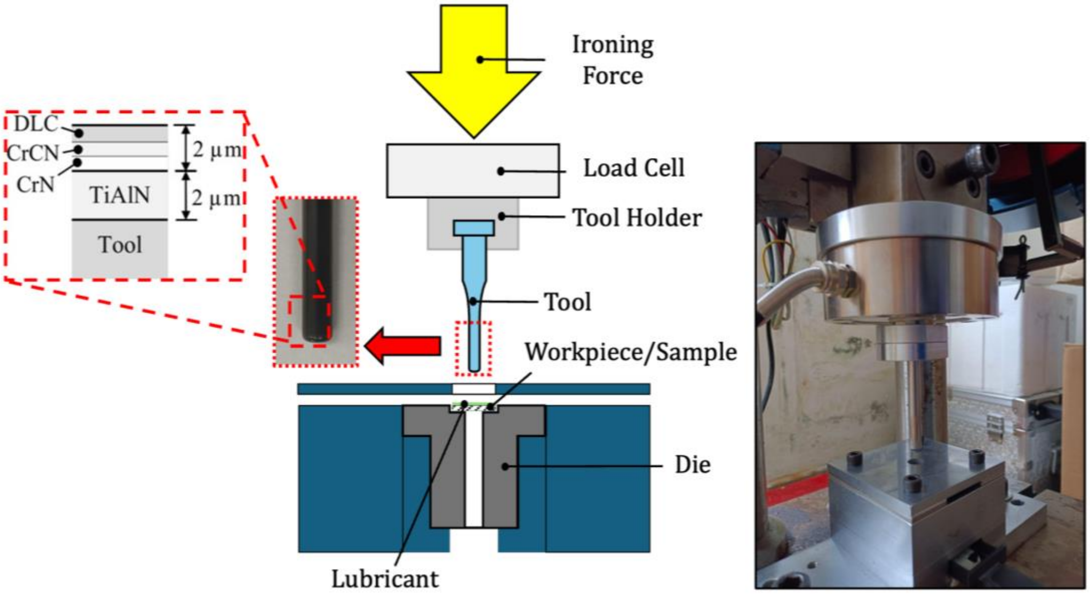


Figure 3: Ironing setup on the press machine.

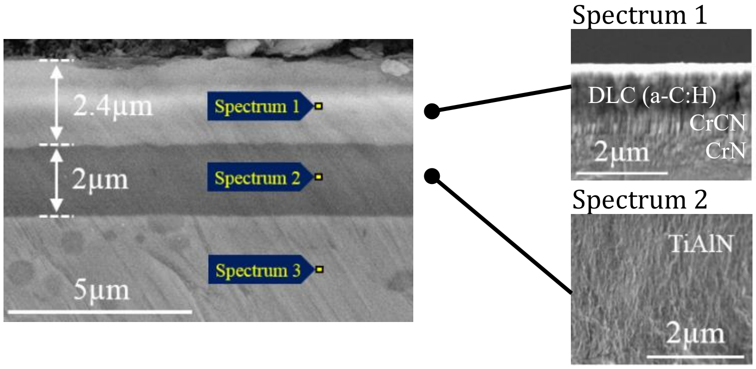


Figure 4: A cross-sectional view of the tool surface with deposited DLC/TiAlN coating. (Note: Spectrum 1 – DLC, Spectrum 2 – TiAlN, and Spectrum 3 – Cutting tool surface).

Friction Analysis:

Tribological tests were performed to evaluate the wear characteristics and friction coefficients of tribological pairs when lubricated. In this project, reference has been made to the ASTM G77-98 standard, which is a recognized method for evaluating the resistance of materials to sliding wear against metals, using a block-on-ring apparatus as in Figure 5.

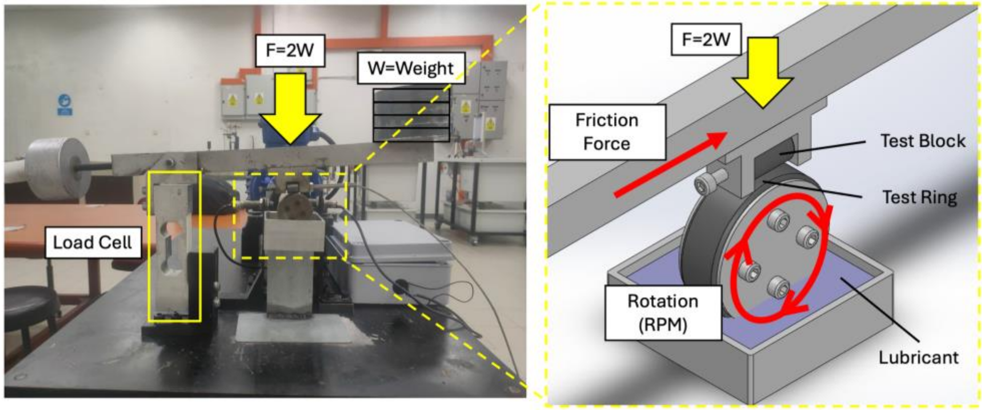


Figure 5: Tribological test setup following ASTM G77 test standard.

3.0 RESULTS AND DISCUSSION

Ironing Force:

Figure 6 shows the forces required for the ironing process under three different conditions: one involving dry friction, second involving DLC/TiAlN coating and three involving DLC/TiAlN coating together with a WBL that includes CMC as a surfactant, MXene, and an ionic liquid. (IL) as an additive. It shows the required force for every 5 strokes and provides a calculated average force for each condition.

Under dry friction conditions, the required force remains consistently high throughout all strokes, typically exceeding 400 N. Especially around 15th and 25th stroke, the force often approaches or even exceeds 500 N. This pattern indicates that there is significant friction between the tool and the workpiece in the absence of lubrication or protective coating. The ironing process using DLC/TiAlN coating shows the little lower than dry friction condition. Using WBLs and the DLC/TiAlN coating significantly reduces the force required for each stroke during the blanking process. This combination ensures that the applied force remains consistently low, mainly oscillating between 350 N and 450 N, except at 40th stroke 450 N. Such stability shows the effectiveness of the coating and lubricant in reducing friction and thus increasing the efficiency of the emptying operation. Compared to scenarios involving dry friction, this lubricating condition consistently requires less force, thereby increasing the interaction between the tool and the workpiece. This shows that the use of both lubricants and coatings can provide advantages in terms of extending tool life and improving the quality of the workpiece. This result underlines the advantages of applying advanced coatings like DLC/TiAlN, combined with an effective lubricant system, in reducing the operational forces and possibly leading to better energy efficiency and less wear in the blanking process.

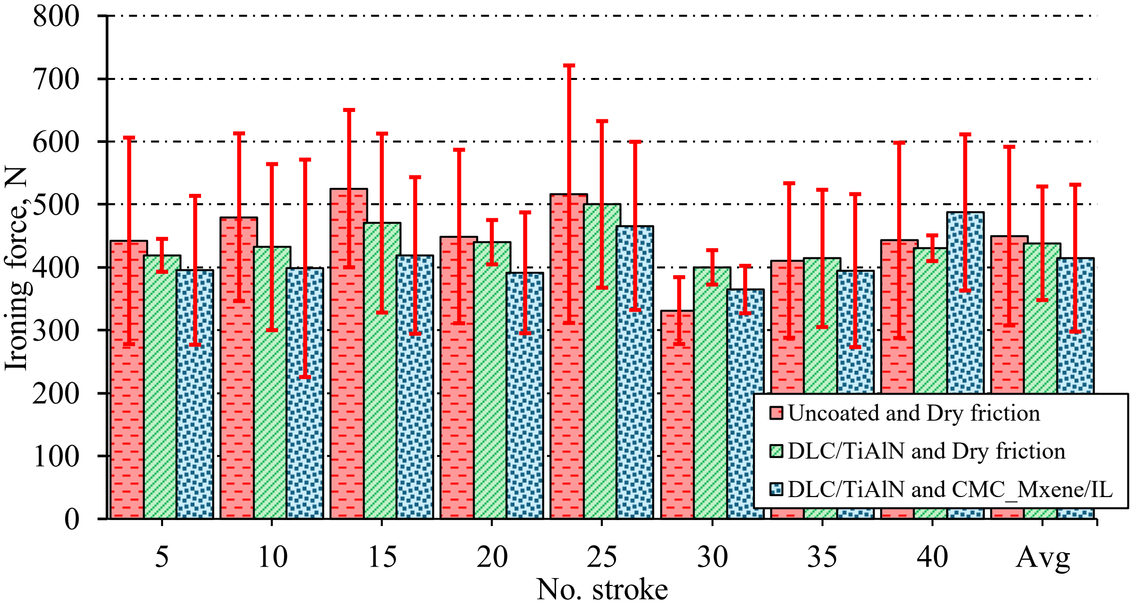


Figure 6: Force during blanking process every 5 strokes for dry frication and using DLC/TiAlN coating with lubricant CMC_Mxene/IL.

Wear on The Tool Surface:

The wear patterns observed on the punches during the ironing process show significant differences when comparing different lubrication and coating conditions. There is a significant difference among the dry friction condition (Figure 7) , tools coated with DLC/TiAlN (Figure 8) and tools coated with DLC/TiAlN with WBL that contain MXene/IL as an additive (Figure 9). In dry friction conditions, the surface of the tool shows significant rough wear. The severe scratching and plowing at most area indicate high material transfer between the tool and the workpiece, leading to extensive damage. This phenomenon is often caused by insufficient lubrication, which leads to direct metal-to-metal contact, which in turn causes adhesive wear and material removal from the surface of the punch. The grooves formed are deeper and exhibit a consistent pattern, indicating continuous wear during operation. In contrast, tools coated with DLC/TiAlN and lubricated with WBL infused with MXene/IL exhibited different wear mechanisms. This coating provides a protective barrier that reduces the intensity of wear. Although localized delamination and damage can be observed in the Figure 8 the overall level of wear is much lower than that caused by dry friction. The lubricant forms a barrier that prevents direct metal-to-metal contact between the tool and the workpiece, while MXene/IL additives may increase the effectiveness of the lubricant, thus reducing friction and wear. The presence of irregular wear patches indicates localized deterioration, which may result from isolated failures in the coating. However, the lack of extensive rough wear suggests that the combined effect of the coating and lubrication system is successful in reducing wear.

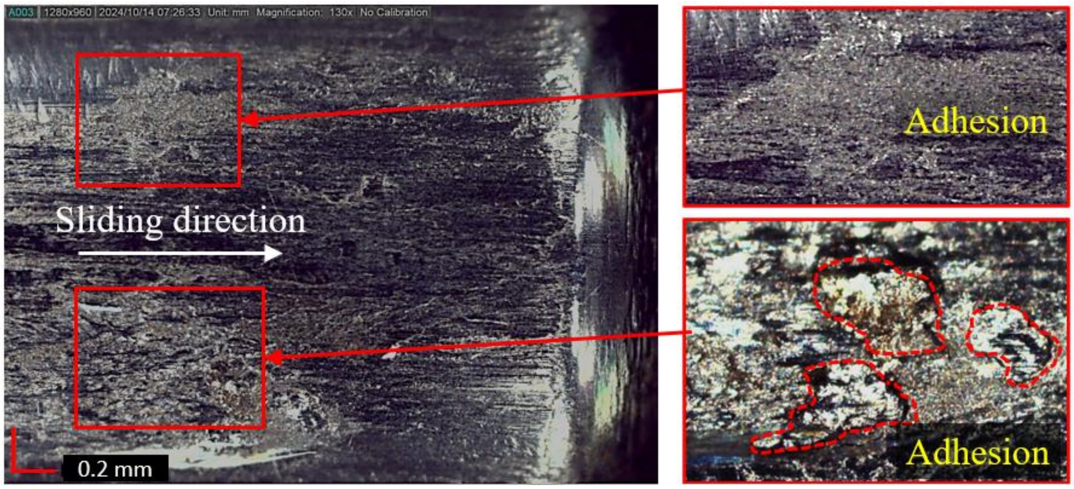


Figure 7: Wear visible on punch nose of uncoated tool performed in dry friction condition.

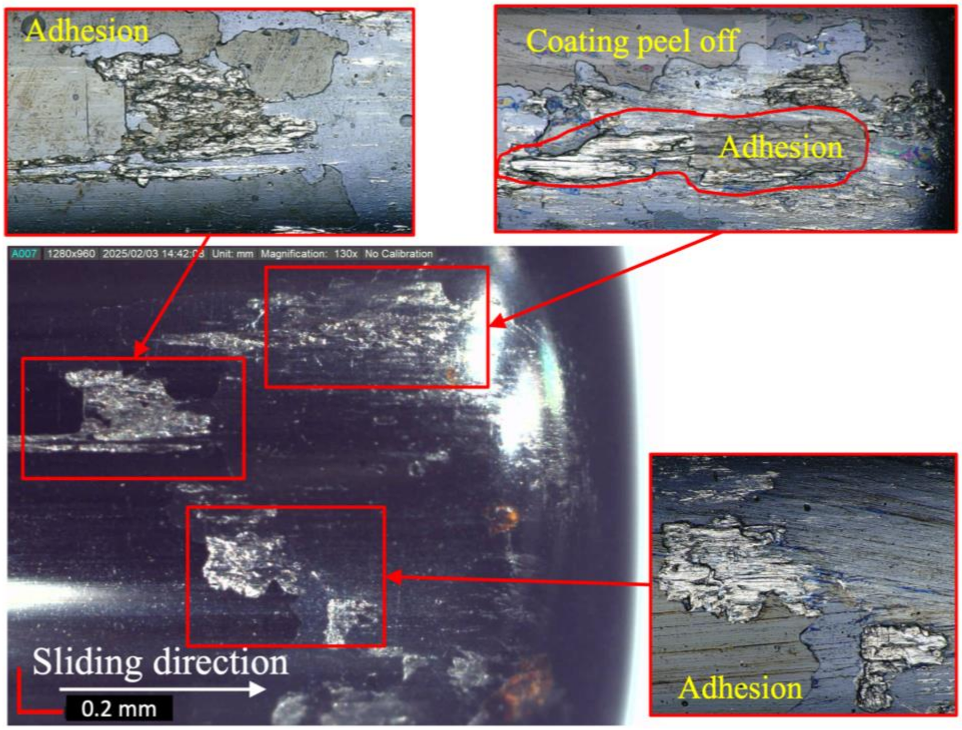


Figure 8: Wear occurred to punch nose for DLC/TiAlN coated punch with dry condition.

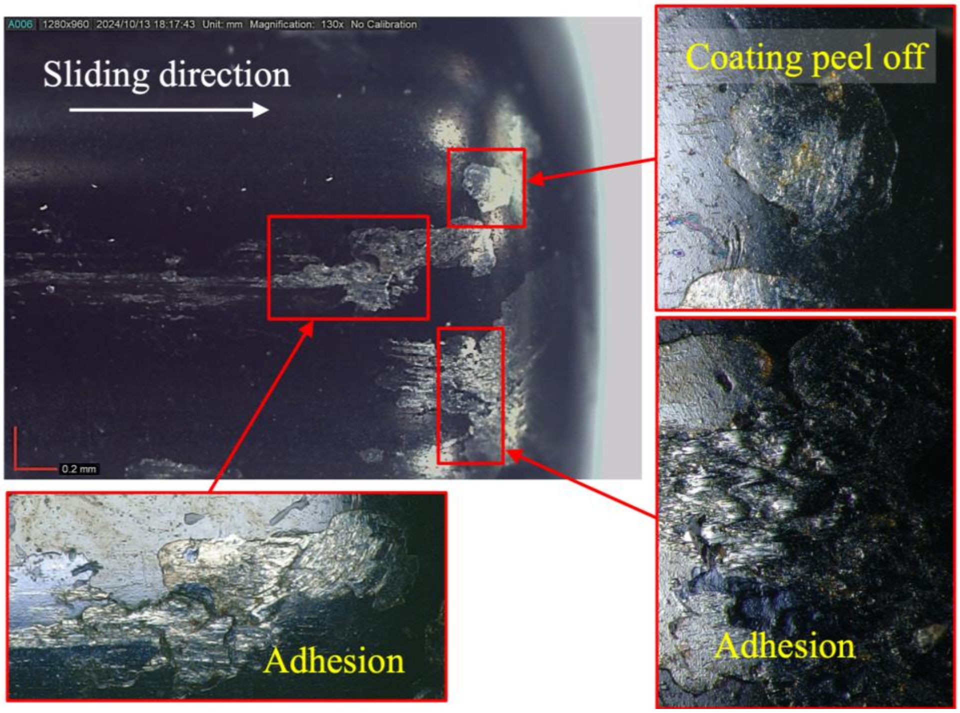


Figure 9: Wear occurred to punch nose for DLC/TiAlN coated punch lubricated with CMC_Mxene/IL WBL.

DLC/TiAlN and CMC_Mxene/IL Testing:

The combinations of CMC_Mxene/IL and DLC/TiAlN were subjected to a comprehensive friction analysis. This evaluation aimed to understand the tribological behavior of these advanced material pairings, which are pivotal in applications demanding high wear resistance and low friction coefficients. The setup was the same as the previous test but using a DLC/TiAlN coating on the block. The COF over cycles is presented in Figure 10, and the average COF is summarized in Figure 11, which includes measurements from the last 5000 cycles assumed to represent a steady state. The results show that using a DLC/TiAlN coating reduces COF from 0.110 (uncoated) to 0.081 (coated), amounting to a decrease of about 26%. This could be of wear-resistant qualities of the DLC coating could be the reason for this improvement. These qualities are enhanced by the superior adhesion strength provided by the TiAlN interlayer coating (Sulaiman et al., 2019).

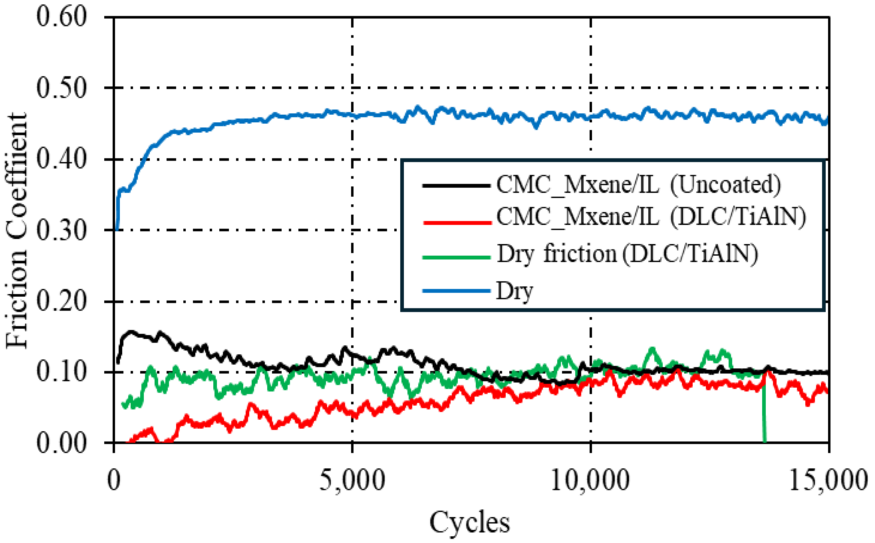


Figure 10: Measurement of friction for CMC_Mxene/IL with DLC/TiAlN coated and uncoated.

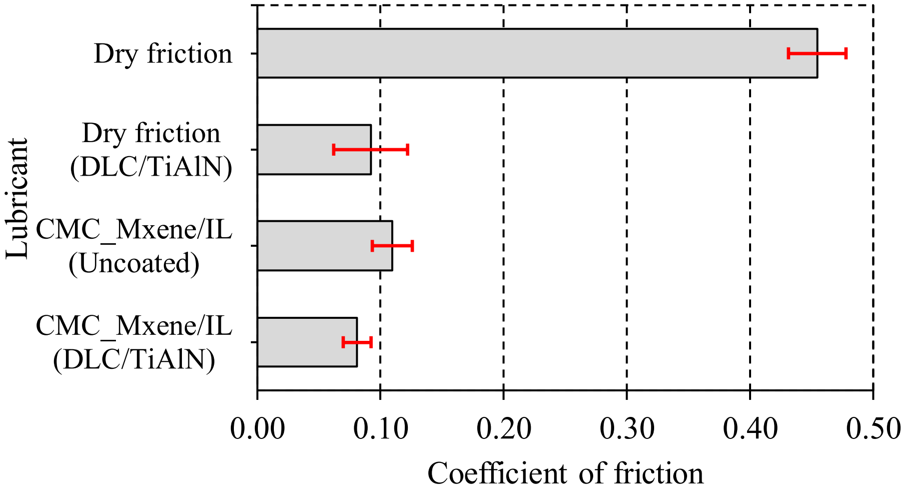


Figure 11: Average COF for CMC_Mxene/IL with DLC/TiAlN coated and uncoated at 500 rpm.

The average temperature and maximum temperature for WBL CMC_Mxene/IL with coating DLC/TiAlN and without coating is plotted in Figure 12. For temperature measurements, where WBL CMC_Mxene/IL with DLC/TiAlN coated sample experiences a bit higher maximum temperature than the uncoated with lubricant sample. This increase in temperature can be attributed to coating durability and wear resistance, which limits frictional heat dissipation by preventing significant lubricant damage or heat dissipation through wear. As a result, more heat accumulates at the interface, leading to a higher temperature but given the friction coefficient is reduced with the CMC_MXene/IL lubricant and the DLC/TiAlN coating.

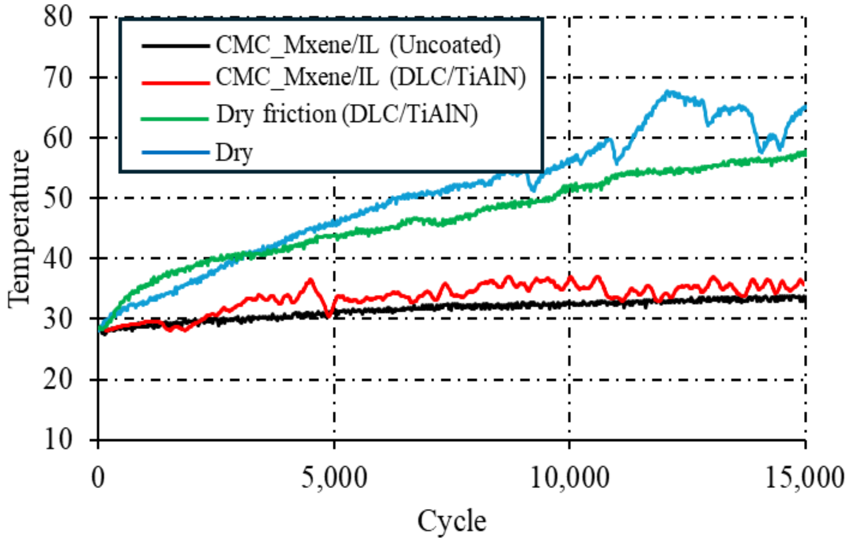
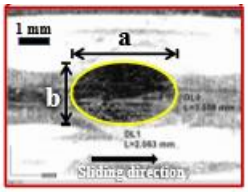


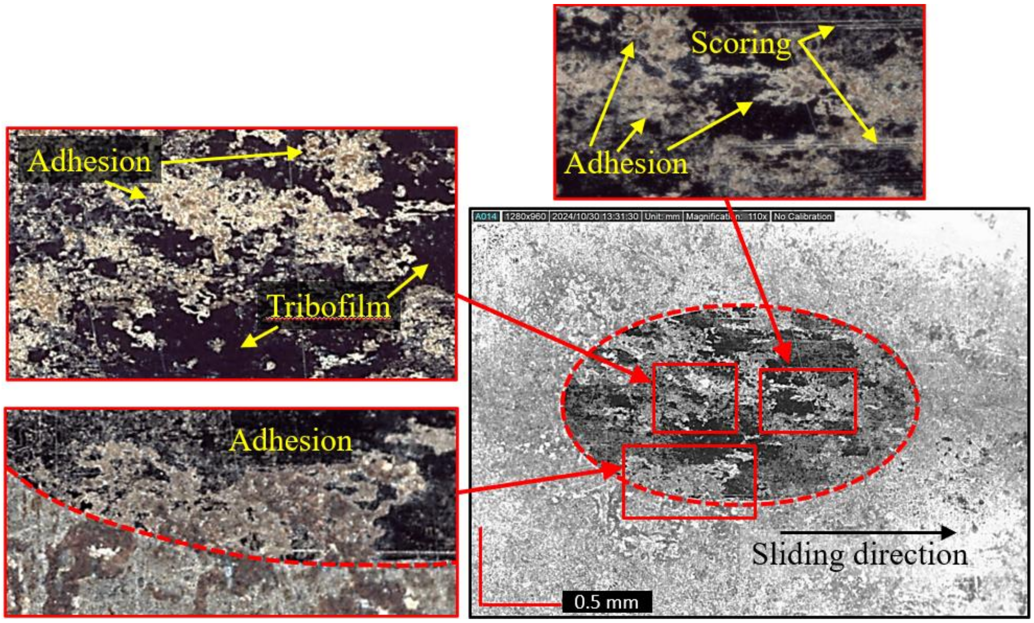
Figure 12: Average and maximum temperature near to sliding surfaces for WBL CMC_Mxene/IL with coating DLC/TiAlN and without coating.

The wear area is determined by referring to Figure 13 and is summarized in Table 1 which includes measurements from Figure 14. The results indicate that using WBL CMC_Mxene/IL with a DLC/TiAlN coating shows a marginally lower wear area compared to surfaces without the coating. Specifically, there is a 7.3% difference, with the DLC/TiAlN coated surface measuring 2.036 mm^2 and the uncoated surface measuring 2.196 mm^2. This reduction can be attributed to low friction coefficient and superior anti-adhesive properties of DLC coatings, which are advantageous for the sliding interaction between two contact surfaces (L. Wang et al., 2022) Additionally, the application of TiAlN over DLC films, along with intermediate layers, has been shown to significantly enhance the substrate's resistance to wear (Sousa et al., 2021).



$$A = \frac{\pi}{4} ab$$

Figure 13: Wear area measurement.


Scoring
Adhesion
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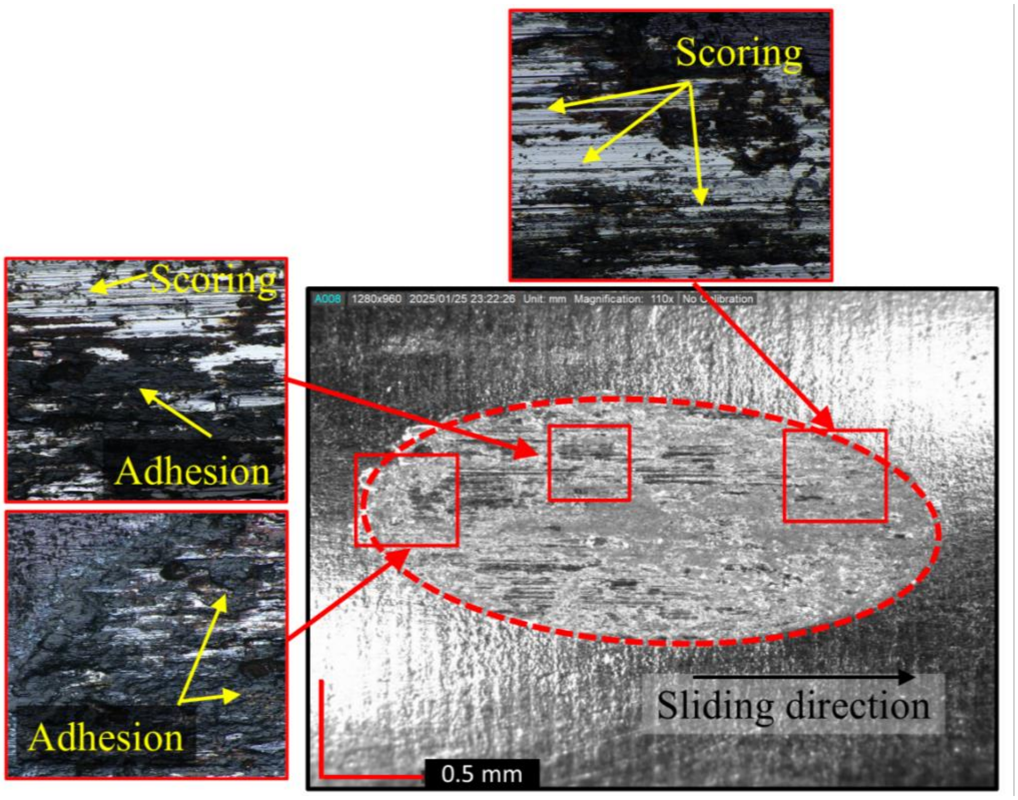
(a)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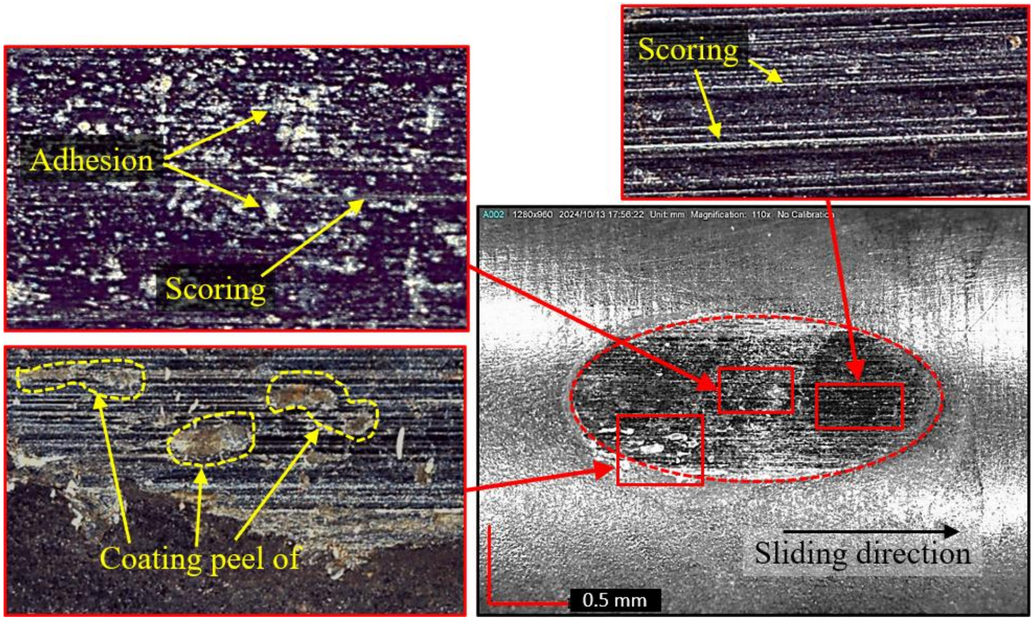

(c)

Figure 14: Wear area on the block using (a) DLC/TiAlN coating with CMC_Mxene/IL, (b) DLC/TiAlN coating with dry condition and (c) Uncoated steel surface.

Table 1: Wear area for CMC_Mxene/IL with coating DLC/TiAlN and without coating.

Conditions	**Wear area (mm^2)**
DLC/TiAlN and CMC_Mxene/IL	2.036
DLC/TiAlN with Dry condition	2.879
Uncoated and CMC_Mxene/IL	2.196
Dry uncoated	14.248

* This wear area was calculated from the measured wear area, indicated by the red-coloured hidden line.

CONCLUSIONS

The tribosystem combining the effects of WBLs formulated with CMC_Mxene/IL and DLC/TiAlN coating has produced positive results, showing a significant reduction in the average forming force compared to the uncoated tool, which lead to less wear appear on tool coating compared to uncoated tool. Further examination through the tribological experiment using block-on-ring test setup on the use of this tribosytem with a combination of DLC/TiAlN coating and CMC_Mxene/IL WBL formulation, has found to reduce friction coefficient by approximately 26%, which is a significant improvement over the uncoated condition. This reduction in friction has a direct effect on wear formation, leading to a decrease in wear area. There is a 7.3% reduction in wear area when using CMC_Mxene/IL with a DLC/TiAlN coating, with measurements showing 2.036 mm^2 compared to 2.196 mm^2 without the coating and 29% % reduction in wear area when compared to coating at dry condition (2.879 mm^2) This data underlines the effectiveness of the coating in improving the durability and performance of the tribosystem.

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