CORROSION SCIENCE AND TECHNOLOGY, Vol.23, No.5(2024), pp.365~373 pISSN 1598-6462 / eISSN 2288-6524
[Research Paper] DOI: https://doi.org/10.14773/cst.2024.23.5.365

Enhancing the Durability and Surface Properties of Ceramic-Glass via DLC Coating Method

Jae-Un Kim[1,2], Byeong-Seok Lim[1], Byung-Woo Ahn[1], Young-Shin Yun[1], and Han-Cheol Choe[2,†]

[1]*Plasma Coating R & D Center, JNLTECH CO., Ltd., Gwangju, 12345, Korea*
[2]*College of Dentistry, Chosun University, Gwangju, 12345, Korea*
(Received August 25, 2024; Revised September 30, 2024; Accepted October 01, 2024)

The purpose of this study was to investigate how diamond-like carbon (DLC) coatings could improve ceramic-glass durability and surface property. Ceramic-glass is valued for its aesthetic appeal. However, it is brittle and easily damaged. To address this issue, we looked into the use of DLC coatings known for their high hardness and low friction properties. Starting with cleaning the surface with linear ion guns to remove any impurities, a buffer layer was applied to enhance the adhesion of coatings. The DLC layer was deposited using unbalanced magnetron (UBM) sputtering, which maximized the deposition efficiency by controlling magnetic fields. Results demonstrated a significant improvement in mechanical properties of ceramic glass, with DLC-coated surfaces achieved a friction coefficient close to zero, a surface hardness of 22 GPa, and an adhesion strength exceeding 30 N. These findings confirm that DLC coatings can substantially increase the durability and extend the service life of ceramic glass, making them a promising solution for enhancing performances of high-cost brittle materials.

Keywords: *DLC, Wear resistance, Ceramic-glass, Plasma coating, Durability, Surface property*

1. Introduction

Ceramic-glass materials are extensively utilized in high-performance fields such as electronics, optics, and aerospace due to their distinctive combination of mechanical strength, thermal stability, and optical clarity [1,2]. Despite these advantageous properties, their inherent brittleness and susceptibility to surface wear and degradation pose significant challenges, particularly in demanding environments. Addressing these issues is crucial for improving the long-term performance and reliability of ceramic-glass materials [3,4].

A promising method to enhance the surface properties of ceramic-glass is through the application of Diamond-Like Carbon (DLC) coatings. DLC coatings are renowned for their exceptional hardness, low friction coefficient, and excellent chemical inertness. These properties make DLC an ideal candidate for protecting surfaces from wear, corrosion, and other forms of degradation [5-9]. The amorphous structure of DLC, characterized by a combination of sp^2 and sp^3 bonded carbon, provides it with diamond-like qualities, such as high hardness and low friction, while also maintaining the flexibility necessary for coating complex surfaces [10-12].

Although previous studies have shown that DLC coatings significantly improve the durability and performance of various substrates, including metals and polymers [13,14], research on the application of DLC coatings to ceramic-glass materials remains limited. Understanding the interaction between DLC coatings and ceramic-glass substrates is essential, as the unique properties of ceramic-glass may affect the adhesion, uniformity, and overall effectiveness of the DLC coating [15,16].

This study aims to explore the potential of DLC coatings in enhancing the durability and surface properties of ceramic-glass materials, with a particular focus on improving wear resistance, hardness, and corrosion protection. By varying deposition parameters and analyzing the resulting surface characteristics, we seek to develop

†Corresponding author: hcchoe@chosun.ac.kr
Jae-Un Kim: Researcher, Byeong-Seok Lim: Researcher, Byung-Woo Ahn: Researcher, Young-Shin Yun: Researcher, Han-Cheol Choe: Professor

a comprehensive understanding of how DLC coatings can be optimized for ceramic-glass substrates. The outcomes of this research could lead to new applications for ceramic-glass materials in more demanding environments, thereby broadening their use in advanced technological industries. One promising application of this research is in induction cooktop glass panels. By applying DLC coatings to these panels, we anticipate significant improvements in scratch resistance against cookware and enhanced ease of cleaning.

2. Experimental Methods

2.1 Coating Preparation

A commercial ceramic-glass substrate was mounted in a jig, positioned 15 cm from the target. The jig was set to rotate clockwise at 50% of its maximum speed during the process. Before deposition, the vacuum chamber was evacuated to a base pressure below 5×10^{-5} Torr. The substrate temperature was maintained at 150 °C during the deposition process. The preparation of the coating involved three main steps:

1. Surface Cleaning by Ar Ion Bombardment: The substrate surface was cleaned by Ar^+ ion bombardment for 30 minutes with a negative bias of 80 V, effectively removing any thin oxide layer and other adherent impurities.

2. Deposition of Cr/CrN Multi-Buffer Layer: A Cr/CrN multi-buffer layer was deposited onto the substrate for 5 to 15 minutes.

3. Deposition of Carbon-Based Plasma Coatings: Finally, carbon-based plasma coatings were applied using a chromium target (purity > 99.9 wt%) for 60 minutes in a mixed atmosphere of N_2 (99.99%) and C_2H_2 (99.99%).

2.2. Characterization of Coating

The surface and cross-sectional morphologies of the DLC coating were observed using a field emission scanning electron microscope (Hitachi, S-4700, Japan). The chemical composition of the coating was analyzed using energy-dispersive X-ray spectroscopy (EDX, Hitachi, EX-200, Japan) along with INCA quantitative analysis software. The phase structure was examined using X-ray diffraction (XRD, X' Pert PRD, PANalytical, Netherlands).

The hardness of the coating was measured with a nanoindentation tester (HM2000, Fischer, Germany). The indentation load was set to 10 mN with a dwell time of 5 seconds. The maximum penetration depth was approximately 0.25 micrometers, which is less than one-tenth of the coating thickness, minimizing the substrate effect. At least five separate measurements were performed for each sample to obtain an average value.

The wear characteristics of the coating were evaluated using a pin-on-disk wear tester (CSM, THT) under ambient conditions at room temperature. The test conditions included a normal load of 2 N, a rotational speed of 200 r/min, a test ball made of 100Cr6 steel with a diameter of 4.4 mm, and a wear track radius set to 8 mm. The adhesion strength of the coating was assessed using a scratch tester (JSLT022) by generating scratches with a diamond tip while applying a constant load ranging from 0 to 30 N, allowing for both qualitative and quantitative evaluation of the thin film's failure patterns.

The surface energy was measured using a contact angle goniometer (KSA100, Kruss, Germany).

3. Results

Fig. 1a shows an FE-SEM (Field Emission Scanning Electron Microscope) image of the surface of the DLC coating. The surface appears to have a relatively uniform texture with fine granular features. The uniformity indicates a consistent coating without significant defects such as cracks or voids, which is crucial for the protective performance of the DLC layer [6]. Fig. 1b image provides a cross-sectional view of the coating, showing the different layers of the coating on the ceramic-glass substrate. Below the DLC layer, there are distinct layers that act as buffer and multi-buffer layers. These layers are designed to improve adhesion between the DLC and the ceramic-glass substrate and to manage the stresses that can occur due to differences in thermal expansion between the layers. Schematic diagram (Fig. 1c) illustrates the layered structure of the coating, aligning with what is observed in the cross-sectional view. Each layer has a specific function: the buffer layer serves to manage plasma interactions, the multi-buffer layer is included to enhance the adhesion and mechanical properties, and the DLC top layer provides the primary protective and functional characteristics, such as hardness, low friction, and chemical resistance.

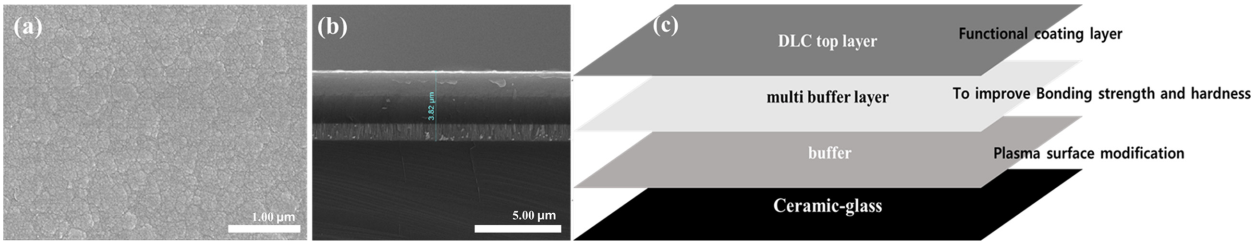


Fig. 1. FE-SEM images of (a) the DLC coating surface, (b) the cross-sectional view, and (c) a schematic diagram of the thin coating film

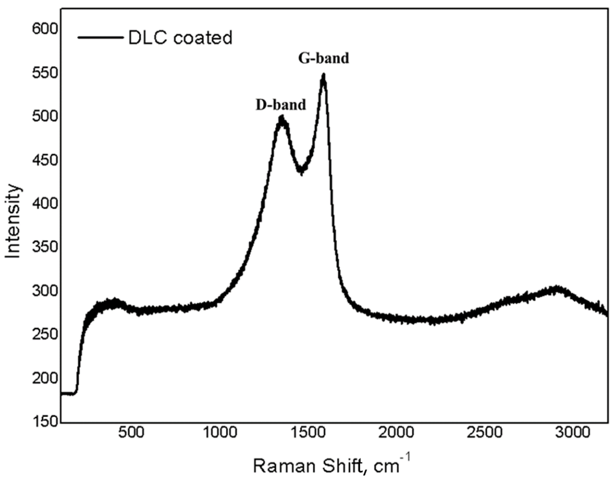


Fig. 2. RAMAN spectrum of the DLC-coated ceramic-glass surface showing the characteristic D and G bands

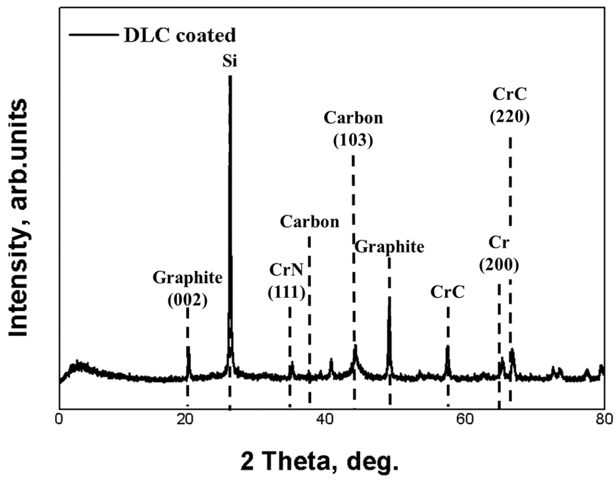


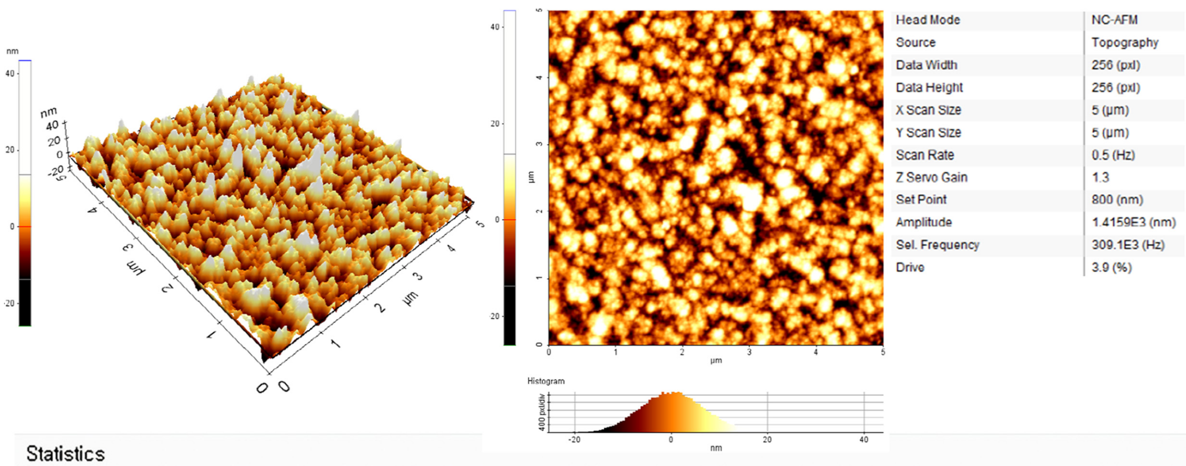
Fig. 3. X-ray diffraction (XRD) pattern of the Diamond-Like Carbon (DLC) coated ceramic glass substrate

The Raman spectrum in Fig. 2 presents the typical features of a Diamond-Like Carbon (DLC) film, characterized by the presence of two prominent peaks: G Peak (around 1580 cm^{-1}) is usually associated with the graphitic (sp^2) carbon structure. It indicates the presence of sp^2 bonded carbon atoms arranged in a graphitic structure. D Peak (around 1350 cm^{-1}) peak is related to the disorder in the carbon structure, often linked to sp^2 hybridized carbon atoms in a disordered or defect-rich state. The relative intensity and shape of these peaks can give insights into the degree of disorder and the sp^2/sp^3 ratio in the DLC coating. A higher G peak relative to the D peak suggests a higher graphitic content. The broadness of these peaks may suggest the presence of a mixture of amorphous carbon phases, where both sp^2 and sp^3 bonded carbon atoms are present [16,17].

Fig. 3 depicits XRD pattern of the Diamond-Like Carbon (DLC) coated sample on ceramic glass substrate. The diffraction peaks observed in the 2θ range of 10º to 80º correspond to various crystalline phases present in the sample. The prominent peak at approximately 13.83º is attributed to the (002) plane of graphite, indicating the presence of a graphitic carbon structure within the DLC coating. The peak at 35.51º is identified as the (111) plane of chromium nitride (CrN), suggesting the formation of CrN phases, likely due to interactions between the DLC layer and the underlying substrate. The peak at 42.71º corresponds to the (103) plane of carbon, while the peak at 52.71º is associated with another graphitic carbon plane, indicating a multi-phase carbon structure. The peaks at 56.07º and 63.46º are indexed to the (200) plane of chromium (Cr) and the (220) plane of chromium carbide (CrC), respectively, which implies the presence of these phases, possibly from the deposition process or substrate material interactions [18,19]. These results collectively indicate a complex microstructure within the DLC coating, comprising both graphitic and carbide phases, alongside residual metallic and nitride components.

Fig. 4 shows AFM topography images reveal the nanoscale surface morphology of the DLC-coated ceramic-glass substrate. The Ra value is 5.486 nm, reflecting the arithmetic average of the absolute values of the surface height deviations from the mean plane. This is a common parameter used to evaluate surface finish. The surface roughness values, especially the low Ra and Rq values, indicate that the DLC coating has a relatively smooth surface, which is beneficial for applications requiring low friction and wear resistance. Overall, the AFM data suggests that the DLC coating was successfully deposited with a nanoscale surface texture that is consistent with the expected properties of such coatings.

Fig. 5 shows scratch test analysis of the DLC-coated ceramic-glass surface. The graph shows the friction force, normal load, and friction coefficient during the scratch test. Despite the applied load increasing up to 30 N, no significant delamination occurred, indicating excellent adhesion of the DLC coating. The friction coefficient remained low, around 0.1, highlighting the coating's superior lubricating properties compared to commercial hard coatings. The inset and lower images illustrate the



Statistics

Region	Min(nm)	Max(nm)	Mid(nm)	Mean(nm)	Rpv(nm)	Rq(nm)	Ra(nm)	Rz(nm)	Rsk	Rku
Whole	-26.003	43.444	8.721	0.000	69.447	6.999	5.486	66.023	-0.392	3.916

Fig. 4. Atomic force microscope (AFM) topography images of DLC-coated ceramic-glass surface

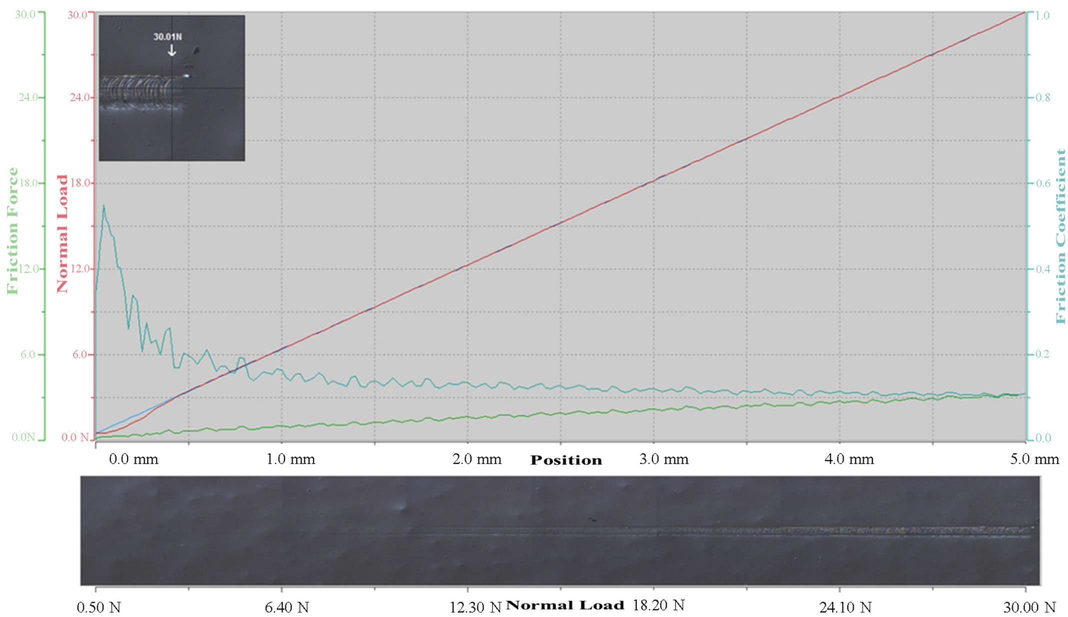


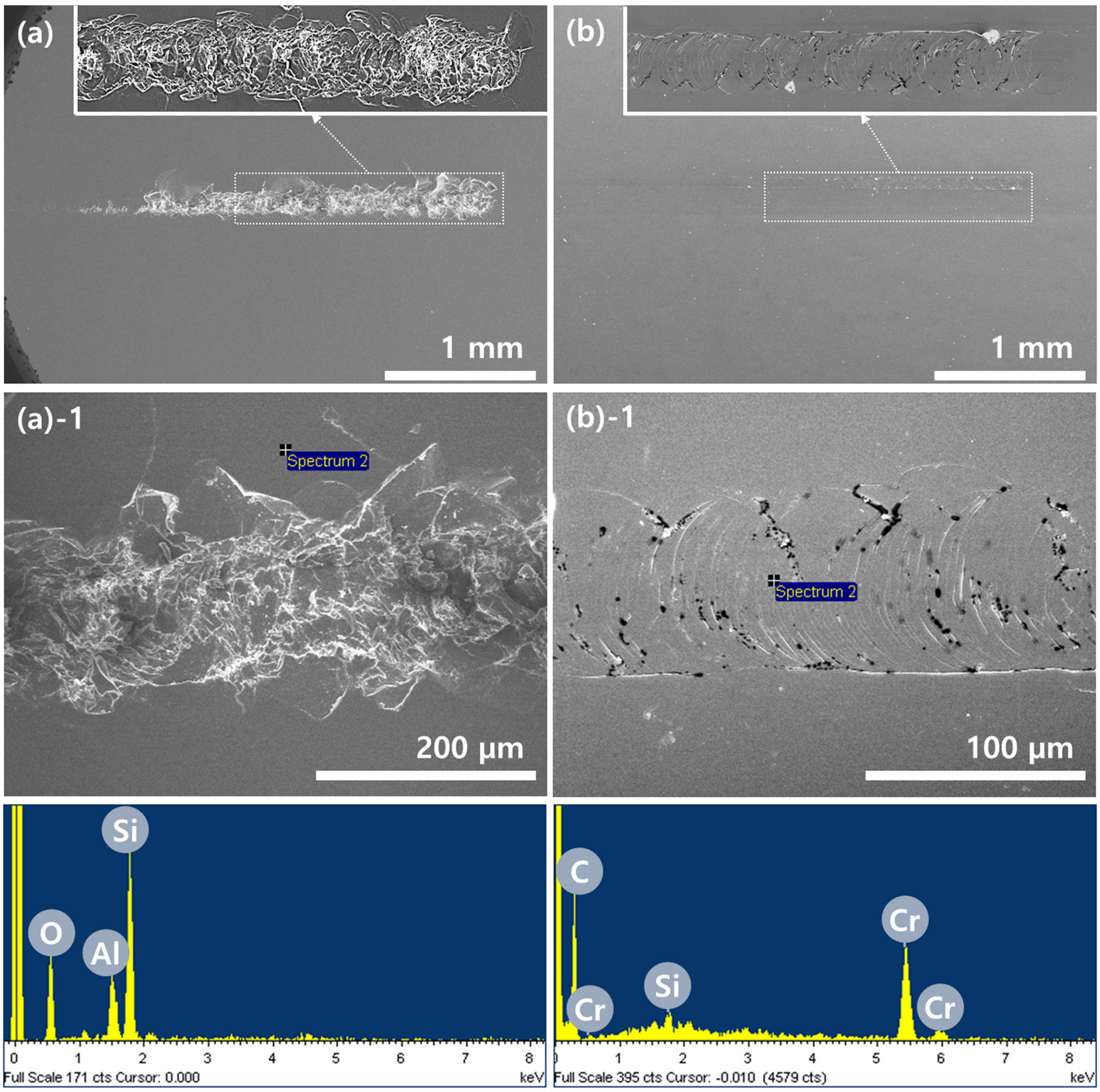
Fig. 5. Coating adhesion and friction coefficient measurement by scratch tester

scratch track, confirming the coating's resilience under high load conditions.

In Fig. 6a, the scratch track of the bare surface is shown, while Fig. 6b displays the scratch track of the DLC-coated surface. Fig. 6a-1 presents the elemental analysis of the bare surface before coating, and Fig. 6b-1 shows the elemental analysis of the DLC-coated scratch track.

The low-magnification SEM images indicate that the bare surface Fig. 6a exhibits severe damage, as evidenced by significant wear and surface deformation. In contrast, the DLC-coated surface Fig. 6b shows only minor microcracks with no evidence of surface delamination.

Elemental analysis further supports this observation. In Fig. 6b-1, a predominant carbon peak is detected, indicating the successful deposition of the DLC coating, which is primarily composed of carbon. The presence of carbon in high concentrations confirms the integrity and adherence of the DLC layer even after the scratch test,



Element	Weight%	Atomic%
O K	49.05	62.63
Al K	10.52	7.97
Si K	40.43	29.41
Totals	100.00	

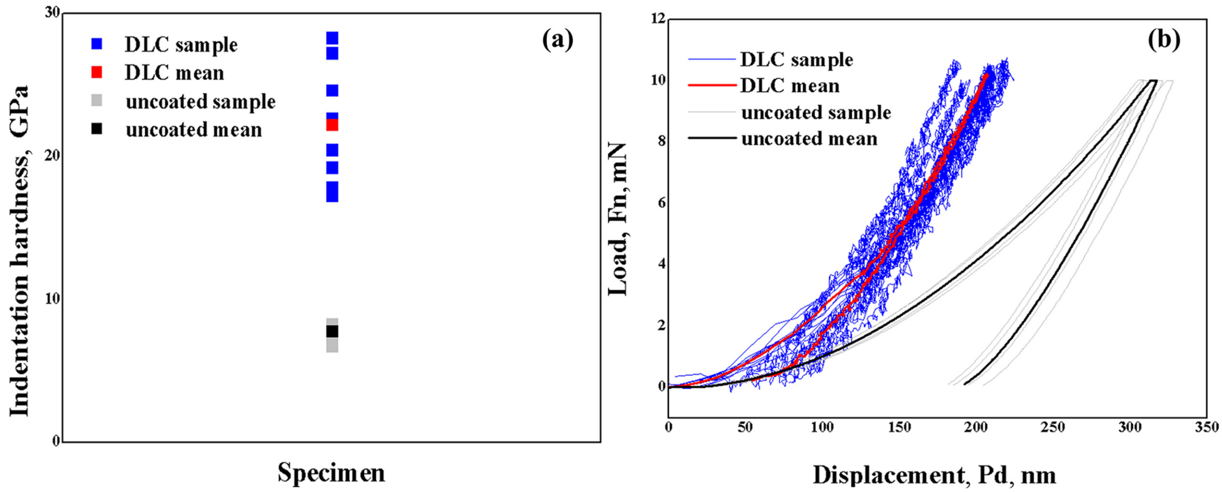
Element	Weight%	Atomic%
C K	49.06	80.08
Si K	2.23	1.55
Cr K	48.71	18.37
Totals	100.00	

Fig. 6. SEM images and EDX analysis of the scratch tracks: (a) Bare surface; (b) DLC-coated scratch track; (a)-1 EDX of the uncoated surface; (b)-1 EDX of the DLC-coated scratch track

whereas the bare surface primarily consists of oxygen, silicon, and aluminum, as seen in Fig. 6a-1.

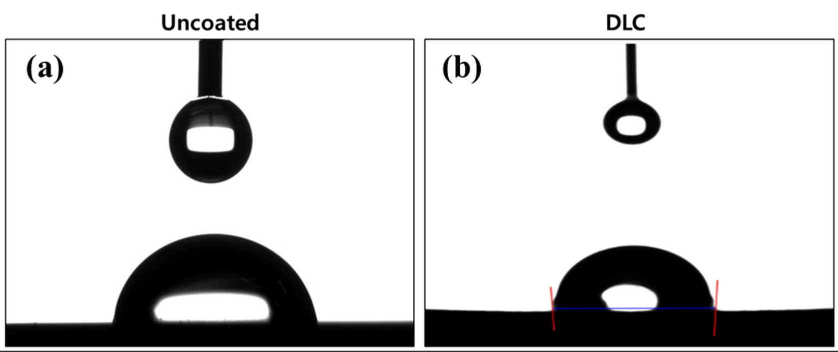
Fig. 7a the blue squares represent the indentation hardness values for the DLC-coated sample. The mean hardness (indicated by the red square) is around 22 GPa. This high hardness is typical of DLC coatings, which are known for their excellent hardness properties due to their high carbon content and dense structure.

The data clearly show that the DLC coating significantly improves the hardness and mechanical properties of the ceramic-glass substrate. The DLC-coated sample exhibits higher indentation hardness and greater resistance to



Specimen	Uncoated	DLC
Indentation Hardness (GPa)	7.79 ± 0.66	22.20 ± 3.92
Elastic Modulus (MPa)	79.34 ± 2.49	146.47 ± 17.5

Fig. 7. (a) Indentation hardness of DLC-coated and uncoated ceramic-glass (b) Load-displacement curves from nanoindentation tests indicating the mechanical response of both DLC-coated and uncoated ceramic-glass samples under applied load



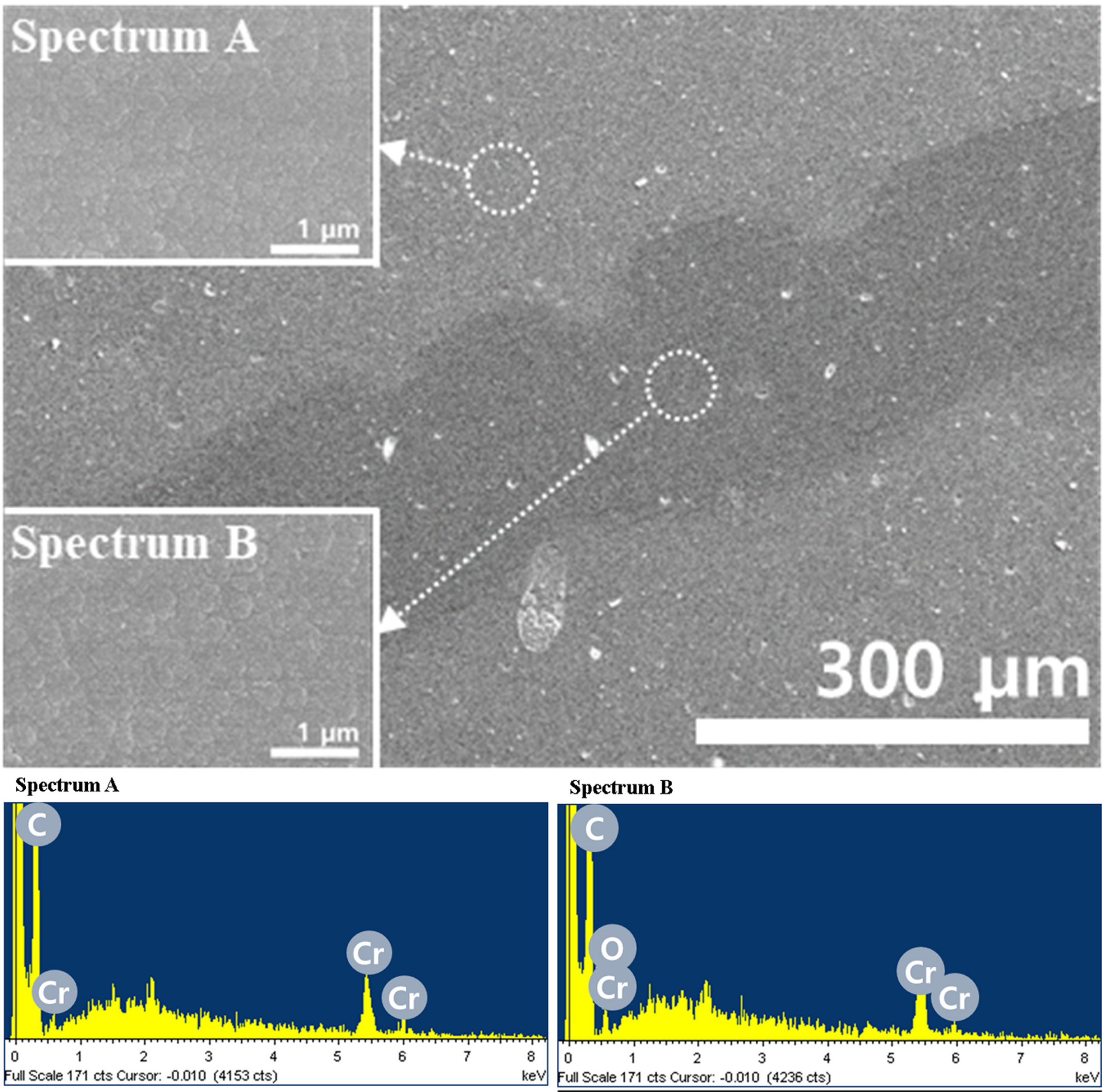
Properties \ Contact angle (°)	1	2	3	4	5	Average
Bulk	73.28	74.08	77.14	78.08	78.77	76.27 ± 2.07
DLC	97.42	92.08	92.74	94.92	92.49	93.93 ± 1.79

Fig. 8. Contact angle measurements of water on (a) uncoated ceramic-glass and (b) DLC-coated ceramic-glass

deformation under load, making it more suitable for applications requiring high wear resistance and mechanical durability.

DLC-coated ceramic-glass (Fig. 8b) contact angle is significantly higher compared to the uncoated surface. This suggests that the DLC coating increases the hydrophobicity of the ceramic-glass, preventing the water droplet from spreading out as much. The DLC coating effectively increases the hydrophobicity of the ceramic-glass surface, as evidenced by the increase in contact angle. This is consistent with the known properties of DLC coatings, which often impart hydrophobic characteristics to surfaces.

Fig. 9 shows the SEM analysis of a wear track on the DLC-coated surface after friction and wear testing. The high-magnification insets (Spectrum A and Spectrum B) reveal that the surface morphology remains consistent with the typical DLC structure, with no noticeable changes



Element	Weight%	Atomic%
C K	80.50	94.70
Cr K	19.50	5.30
Totals	100.00	

Element	Weight%	Atomic%
C K	70.64	86.75
O K	7.70	7.10
Cr K	21.66	6.14
Totals	100.00	

Fig. 9. FE-SEM micrographs of the wear track on the DLC-coated surface after tribological testing, with high-magnification insets (Spectrum A and Spectrum B) showing the selected areas analyzed by EDX

before and after testing. This indicates that the DLC coating successfully retains its characteristic features even under tribological stress. Spectrum A confirms that the region is predominantly carbon-rich, as expected for a DLC coating, while Spectrum B reveals an increase in chromium and oxygen, likely resulting from material transfer or minor oxidation during testing.

These findings, along with the minimal observed morphological changes, demonstrate the excellent wear resistance of the DLC coating, confirming that the wear rate is extremely low. The retention of the typical DLC surface structure despite wear testing highlights the coating's effectiveness in maintaining its protective properties under demanding conditions.

4. Conclusions

In this study, a DLC coating was successfully deposited onto a ceramic-glass substrate, leading to significant enhancements in surface properties, including hardness, wear resistance, and hydrophobicity. The SEM analysis of the scratch tracks revealed that the bare substrate surface exhibited severe damage under stress, whereas the DLC-coated surface showed only minor microcracks without delamination, demonstrating the coating's superior protective performance. Elemental analysis further confirmed the successful deposition of the DLC layer, with a high carbon content observed in the coated samples.

Raman spectroscopy displayed typical features of DLC coatings, with the G and D peaks indicating the presence of both sp^2 and sp^3 hybridized carbon structures. XRD analysis identified multiple phases, including graphitic carbon, chromium nitride, and chromium carbide, highlighting the complex microstructure within the DLC layer. The AFM results showed a smooth nanoscale surface with low roughness values, supporting the coating's potential for low-friction applications.

The scratch test confirmed excellent adhesion and low friction coefficients, reinforcing the DLC coating's suitability for applications requiring robust mechanical durability. The indentation hardness measurements revealed that the DLC coating increased the hardness by approximately three times compared to the uncoated substrate, reaching around 22 GPa. This substantial improvement underscores the enhanced mechanical strength provided by the DLC layer. Additionally, contact angle measurements indicated a significant increase in hydrophobicity, making the surface less prone to contamination and easier to clean, as dirt and other substances are less likely to adhere and can be easily removed.

Further SEM analysis of the wear track after tribological testing revealed that the DLC-coated surface maintained its structural integrity, with no significant morphological changes observed. This indicates the coating's exceptional wear resistance, ensuring its effectiveness even under harsh conditions.

Overall, the study demonstrates that the DLC coating offers considerable functional advantages, including improved mechanical strength, wear resistance, surface hydrophobicity, and exceptional durability under stress, making it a highly effective protective coating for ceramic-glass substrates in demanding applications.

Acknowledgments

This work was supported by the Technology Development Program (00223520) funded by the Ministry of SMEs and Startups (MSS, Korea).

References

1. M. Gonon and F. Dupla, Glass-Ceramics and Their Applications, *Encyclopedia of Materials: Technical Ceramics and Glasses*, **2**, 709 (2021). Doi: https://doi.org/10.1016/B978-0-12-818542-1.00008-4
2. S. Nikkisha, S. Rohan, P. Pattanaik, A. K. Mishra and D. Darshini, Review Study on Mechanical and Thermal Properties of Ceramic Materials for Future Aerospace Applications, *Materials and its Characterization*, **2**, 107 (2022). Doi: https://doi.org/10.46632/mc/1/2/7
3. A. R. Boccaccini, Assessment of brittleness in glass-ceramics and particulate glass matrix composites by indentation data, *Journal of Materials Science Letters*, **15**, 1119 (1996). Doi: https://doi.org/10.1007/BF00539954
4. G. A. Khater and E. M. Safwat, Some Types of Glass-Ceramic Materials and their Applications, *International Journal of Research Studies in Science, Engineering and Technology*, **7**, 1 (2020). https://ijrsset.org/pdfs/v7-i3/1.pdf
5. M. S. Kabir, Z. Zhou, Z. Xie, and P. Munroe, Designing

multilayer diamond-like carbon coatings for improved mechanical properties, *Journal of Materials Science and Technology*, **65**, 108 (2021). Doi: https://doi.org/10.1016/j.jmst.2020.04.077

6. D. K. Rajak, A. Kumar, A. Behera, and P. L. Menezes, Diamond-Like Carbon (DLC) Coatings: Classification, Properties, and Applications, *Applied Sciences*, **11**, 4445 (2021). Doi: https://doi.org/10.3390/app11104445
7. H. Ito and K. Yamamoto, Mechanical and Tribological Properties of DLC Films for Sliding Parts, *KOBELCO Technology Review*, vol. 35 (2017). https://www.kobelco.co.jp/english/ktr/pdf/ktr_35/055-060.pdf
8. B. Dong, X. Guo, K. Zhang, Y. Zhang, Z. Li, W. Wang, and C. Cai, Combined effect of laser texturing and carburizing on the bonding strength of DLC coatings deposited on medical titanium alloy, *Surface and Coatings Technology*, **429**, 127951 (2022). Doi: https://doi.org/10.1016/j.surfcoat.2021.127951
9. D. Guo, S. Zhang, T. Huang, S. Wu, X. Ma, and F. Guo, Corrosion Properties of DLC Film in Weak Acid and Alkali Solutions, *Coatings*, **12**, 1776 (2022). Doi: https://doi.org/10.3390/coatings12111776
10. R. Zhang and L. Wang, Synergistic improving of tribological properties of amorphous carbon film enhanced by F–Si-doped multilayer structure under corrosive environment, *Surface and Coatings Technology*, **276**, 626 (2015). Doi: https://doi.org/10.1016/j.surfcoat.2015.06.006
11. H. Kovacı, A. F. Yetim, Z. Baran, and A. Çelik, Tribological behavior of DLC films and duplex ceramic coatings under different sliding conditions, *Ceramics International*, **44**, 7151 (2018). Doi: https://doi.org/10.1016/j.ceramint.2018.01.158
12. S. Grigoriev, M. Volosova, S. Fyodorov, M. Lyakhovetskiy, and A. Seleznev, DLC-coating Application to Improve the Durability of Ceramic Tools, *Journal of Materials Engineering and Performance*, **28**, 4415 (2019). Doi: https://doi.org/10.1007/s11665-019-04149-1
13. D. Toboła, T. Liskiewicz, L. Yang, T. Khan, and U. Boroń, Effect of mechanical and thermochemical tool steel substrate pre-treatment on diamond-like carbon (DLC) coating durability, *Surface and Coatings Technology*, **422**, 127483 (2021). Doi: https://doi.org/10.1016/j.surfcoat.2021.127483
14. K. Enke, Dry machining and increase of endurance of machine parts with improved doped DLC coatings on steel, ceramics and aluminium, *Surface and Coatings Technology*, **116-119**, 488 (1999). Doi: https://doi.org/10.1016/S0257-8972(99)00096-1
15. J. I. Kim, W. Y. Lee, K. Tanaka, T. Tokoroyama, M. Murashima, and N. Umehara, Nitrogen-containing diamond-like carbon deposited on glass substrate via filtered cathodic vacuum arc with high-frequency pulsed DC substrate bias, *Surfaces and Interfaces*, **44**, 103695 (2024). Doi: https://doi.org/10.1016/j.surfin.2023.103695
16. A. Poukhovoi, S. Schiporeit, H. W. Becker, L. Calliari, V. Buck, Assessment of spectroscopic methods for the characterisation of DLC films deposited by PECVD, *Journal of Optoelectronics and Advanced Materials*, **14**, 383 (2012). https://joam.inoe.ro/articles/assessment-of-spectroscopic-methods-for-the-characterisation-of-dlc-films-deposited-by-pecvd/
17. A. C. Ferrari and J. Robertson, Raman spectroscopy of amorphous, nanostructured, diamond-like carbon, and nanodiamond, *Philosophical Transactions of the Royal Society A, Mathematical, Physical and Engineerings Sciences*, **362**, 2477 (2004). Doi: https://doi.org/10.1098/rsta.2004.1452
18. H. Pang, X. Wang, G. Zhang, H. Chen, G. Lv, and S. Yang, Characterization of diamond-like carbon films by SEM, XRD and Raman spectroscopy, *Applied Surface Science*, **256**, 6403 (2010). Doi: https://doi.org/10.1016/j.apsusc.2010.04.025
19. G. Wu, L. Sun, W. Dai, L. Song, and A. Wang, Influence of interlayers on corrosion resistance of diamond-like carbon coating on magnesium alloy, *Surface and Coatings Technology*, **204**, 2193 (2010). Doi: https://doi.org/10.1016/j.surfcoat.2009.12.009