Responses to reviewers' questions:
Reviewer: Dr. István Biró, PhD

Dear Reviewer,

Thank you for review my dissertation. I have carefully addressed and answered all the questions raised. For clarity, the questions presented in **bold**, and my responses in regular font, as shown below:

1. Statement 1 says that zirconium is preferred as a counter material for TiN and TiC coatings. What applications requiring CVD hard coatings could benefit from this observation, and how?

Thank you for your question. The preference for zirconia as a counter material in tribological testing of CVD coatings is particularly relevant in applications requiring high-performance wear-resistant coatings. This observation is beneficial for evaluating the tribological behaviour of coated components, such as CVD-coated cutting tools, because zirconia exhibits a non-adhesive nature (between the coating surface and the counter ball surface), resistance to surface heating, and comparable hardness to CVD TiN-based coatings. These properties make zirconia an ideal material for evaluating coating performance without external interference.

Based on these findings, the following industrial applications could benefit:

1. Cutting Tools and machining industry:
    - CVD coatings enhance the wear resistance and mechanical properties of cutting tools such as drills, mills, and inserts.
2. Mold and die industry:
    - Increased resistance to wear and deformation in injection molding and stamping dies.

By use zirconia as a counter material in tribological testing, these industries can achieve a more accurate assessment of coating durability, enabling the selection of optimal coatings for harsh working conditions.

2. How can the coating's thickness affect a cutting tool's cutting ability?

Based on our investigation of the performance of bilayer TiN/TiC CVD coatings and TiN-based CVD coatings in both single and multilayer configurations, as well as their effects on adhesion performance, tribological behavior, wear resistance, and hardness, we can summarize the impact of coating thickness as follows:

- Too thin (<0.5 μm): Provides insufficient wear protection, leading to rapid coating depletion under scratch tests.
- Optimal thickness (2–10 μm): Achieves a balance between wear resistance, hardness, and adhesion performance.
- Excessively thick (>10 μm): Increases brittleness, reduces heat dissipation efficiency, and leads to edge rounding, which negatively affects cutting precision and tool

sharpness. We did not investigate CVD coatings thicker than 10 μm, but this observation is discussed in the literature review.

These observations are also supported by literature findings. Yang et al. (2023) reported that a well-crystallized Ti(C,N) layer demonstrated superior adhesion, but thicker coatings (16.62 μm) exhibited increased porosity and delamination risks. Additionally, the hardness decreased by approximately 30% when the coating thickness increased by 17%. Similarly, Ben Hassine et al. (2025) indicated that excessive thickness can lead to the formation of residual stresses, crack formation, and reduced adhesion performance. Coating thickness plays a crucial role in friction and wear resistance. Sirtuli et al. (2025) compared different CVD coatings and found that a κ-Al_2O_3-TiN/Ti(C,N) multilayer with a total thickness of 6.2 μm provided better initial notch wear resistance than a slightly thicker (7.2 μm) α-Al_2O_3/Ti(C,N) coating. This suggests that an optimal thickness is necessary to balance friction reduction and mechanical stability. Vallejo et al. (2025) found that TiN coatings exhibited a decrease in wear resistance for thin films with thicknesses below 0.1 μm, emphasizing the importance of sufficient material volume for wear durability. However, Sirtuli et al. (2025) noted that while thicker coatings can resist wear, excessive thickness does not necessarily improve performance significantly and may even accelerate wear progression due to stress concentration at the coating-substrate interface. The hardness is directly linked to coating thickness, but an excessive increase in thickness can lead to a decline in hardness. Aliofkhazraei et al. (2021) observed that hard multilayer coatings followed a Hall-Petch relationship, where hardness increased with decreasing thickness up to a critical point, beyond which it declined due to dislocation activity.

Refs:

Aliofkhazraei, M., Walsh, F. C., Zangari, G., Köçkar, H., Alper, M., Rizal, C., & Assareh, S. (2021). Development of electrodeposited multilayer coatings: A review of fabrication, microstructure, properties, and applications. Applied Surface Science Advances, 6, 100141.

Ben Hassine, M., Andrén, H.-O., Iyer, A. H. S., Bäcke, O., Stiens, D., Janssen, W., Kümmel, J., & Halvarsson, M. (2025). Growth of a hard, novel CVD multilayer coating: Ti(C,N) on (Ti,Al)N on TiN. International Journal of Refractory Metals and Hard Materials, 127, 106966.

Sirtuli, L. J., Boing, D., Bushlya, V., & Norgren, S. (2025). Study of initial notch wear during turning of stainless steel with CVD Al2O3/Ti(C,N) coated cemented carbide tools. International Journal of Refractory Metals and Hard Materials, 129, 107116.

Vallejo-Otero, V., Crespo-Monteiro, N., Gamet, E., Ollier, N., Donnet, C., Valour, A., & Jourlin, Y. (2025). Advancements in nitridation of TiO2 layers: Mechanisms, techniques, and applications for TiN thin films. Journal of the European Ceramic Society, 45(10), 117330.

Yang, L., Xiong, J., Chen, X., Li, X., Deng, C., Zhang, D., & Yi, L. (2023). Study on the growth and wear characteristics of CVD coating deposited on Ti(C,N)-based cermets with different C/N ratios of Ti(C,N) powders. Ceramics International, 49(11), 18023-18034

3. Regarding the diagrams: What is the reason (i.e. scientific meaning) of data points being directly connected by higher-order functions (see Fig. 41, 42 and 46) or straight lines (see Fig. 54 and 55)?

Thank you for this question. The method of connected data points in the figures was selected based on the nature of the experimental results and the clarity in present these figures:

Figures 41, 42, and 46 show time-dependent results where higher-order curves used to reflect the non-linear behavior of processes like diffusion-controlled oxidation, elemental depletion, and hardness degradation vs oxidation duration. Typically, these processes develop progressively and non-linearly due to compositional changes accumulate over time. Using smooth curves in these diagrams allows for clearer the visualization, including acceleration or stabilization directions.

Figures 54 and 55 present relationships between time-independent physical parameters (such: Ra and HV vs. volume loss). Straight lines were used to maintain clarity, avoid overfitting, and make it easier to follow and identify overall trends and intersection points between the data sets. Therefore, the straight lines were more suitable.

Veszprém, 17 June, 2025

PhD student

Osamah Ihsan Ali