



## Contact fatigue behavior of $\alpha$ -Al<sub>2</sub>O<sub>3</sub>-Ti(C,N) CVD coated WC-Co under dry and wet conditions

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### ABSTRACT

The response to cycling contact fatigue load of a WC-6%Co carbide coated with a Ti(C,N)/ $\alpha$ -Al<sub>2</sub>O<sub>3</sub> CVD multi-layer was investigated in dry and wet conditions. Imprints in dry conditions were characterized by small thin cracks forming a circumference at the maximum radii of the imprint. The damaged coating was totally present in the final imprint of the dry test. Wet indentations showcase an area in the imprint where the  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> layer has been removed throughout a ring but was kept at the center of the indentation, suggesting that the coating damage under cycling contact load in wet conditions is dominated by  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> degradation, associated with a fretting effect or tangential loads accelerating the fatigue-corrosion of the alumina layer.

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### 1. Introduction

Coated cemented carbide inserts are used in the machining of metal alloys for the automotive and aerospace industry. WC-Co substrates provide the balance between hardness and toughness, whereas the Ti(C,N)/ $\alpha$ -Al<sub>2</sub>O<sub>3</sub> coatings produced by chemical vapor deposition (CVD) increase the surface resistance to wear and chemical degradation [1,2]. A critical feature of Ti(C,N)/ $\alpha$ -Al<sub>2</sub>O<sub>3</sub> CVD coatings in cemented carbides is the formation of a network of microcracks in the coating during the cooling step of the CVD process (due to the large difference of thermal expansion coefficient between carbide and coating). Additionally, a major problem in interrupted machining is the formation and propagation of microcracks due to the oscillating conditions of force and temperature acting at the cutting edge. These microcracks produce macrocracks and chipping, leading to failure of the tools [3]. In a previous work, contact damage induced by spherical indentation on coated cemented carbides has been studied [4,5]. For single- and multilayered CVD coatings, it was found that film cracking under contact loading is governed by load transfer from the plastically deformed substrate into the coating as well as the effective fracture strength and residual stress condition of the film [5]. Furthermore, the contact damage tests were in complete agreement with previous micromechanical

tests in terms of crack propagation paths in carbonitride systems [6]. Recently, a new experimental set-up designed by the authors was used to investigate the cyclic contact fatigue behavior of cemented carbides under dry and wet conditions [7]. By immersing the cemented carbides in industrial cutting fluid under oscillating loads, the combination of chemical attack and cyclic contact load was used to visualize the advantage of Cr-containing binders in the corrosion-fatigue resistance of commercial cemented carbide grades. In this work the same experimental set-up is used to investigate the crack propagation in Ti(C,N)/ $\alpha$ -Al<sub>2</sub>O<sub>3</sub> CVD coated carbide under dry and wet conditions.

### 2. Experimental

WC-6 wt%Co cemented carbides (grain size: 0.8  $\mu$ m, HV30:1570, K<sub>IC</sub>: 9.9 MPa·m<sup>1/2</sup>) coated with a starting layer of TiN (0.5  $\mu$ m) followed by a Ti(C,N) (3  $\mu$ m), a Ti(C,N,O)<sub>x</sub> bonding layer (1  $\mu$ m) and a top  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> layer (3  $\mu$ m) by CVD in an industrial hot wall reactor were investigated. Cyclic indentation tests at the macroscopic length scale were assessed by spherical indentation (Hertzian tests) using hardmetal indenters of 2.5 mm of curvature radii. Tests were conducted in a servo hydraulic testing machine (Instron 8511) using a sinusoidal wave with 7 Hz frequency to a total number of 1·10<sup>5</sup> cycles. The force range ( $\Delta F$ ) was constant with an indentation force ratio of 0.17 ( $\Delta F = 200$  N/

1200 N), allowing the indenter to be in contact with the material surface during the entire experiment. For wet experiments, an industrial cooling fluid (8 vol% stock solution in tap water) was used, Table 1 [7].

### 3. Results and discussion

The imprints on the insert's surface at the end of the cycling contact load tests are shown in the LOM images in Fig. 1. A remarkable difference is observed between dry (air) and wet (lubricant) conditions. In dry conditions, the indentation only shows circumferential cracks at the maximum radii of the imprint on the  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> coating, whereas in the presence of the cooling cutting fluid, a dark ring with absence of the  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> coating layer is observed (EDS profiles in Fig. 1). Also, the external diameter in wet conditions is larger than in dry ones.

To observe the crack propagation through the material, cross sections were inspected both in the LOM and in the SEM (Fig. 2).

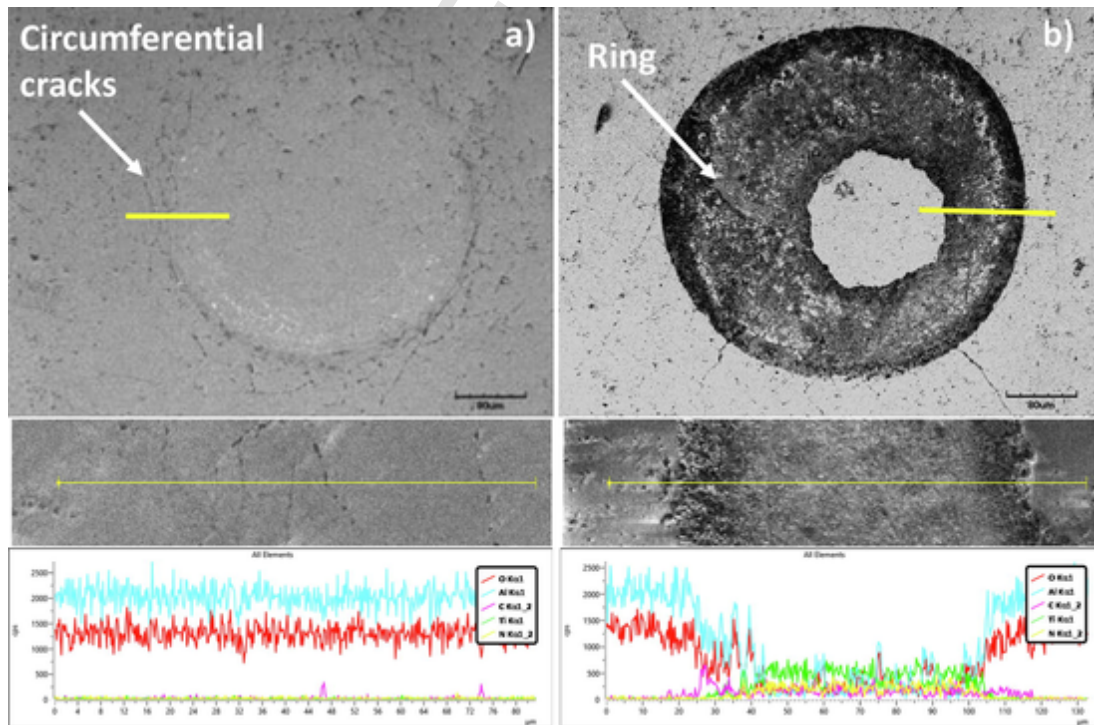
In dry conditions cracks initiating from the  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> layer through the rest of the coating were observed, sometimes propagating into the substrate. In wet conditions the absence of the  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> layer on the ring previously shown on the top view LOM images (Fig. 1) is clearly seen in the SEM image of Fig. 2b. The  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> layer has either been corroded or detached from the rest of the coating in that area, as veri-

fied by EDS (Fig. 1). The ring area is heavily damaged, having a high density of cracks in the coating which is left. Some damage in the substrate beneath the coating can be observed for both conditions, particularly in the center of the imprints, which is the area that has been always in contact (under the maximum stress condition) with the WC sphere during the contact-fatigue tests (Fig. 2). Cracks are observed in the  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> layer for both conditions, which propagate through the coating and often through the substrate on the laterals of the indentation (the circumference). In Fig. 2 it can be observed that these cracks tend to follow the WC grain boundaries in the near-surface (max. 5 $\mu$ m). SEM-FIB cuts along the indentation are shown in Fig. 3. Cracks propagating from the  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> layer, through the Ti(C,N) layer and also into the substrate are observed in both conditions. In dry conditions (Fig. 3a) a network of microcracks is observed in the  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> layer, which seems to be connected to the grain size of the polycrystalline layer. However, in wet conditions (Fig. 3b) this microcrack network is present, but part of the layer has been removed during the test. The degradation of such layer appears to be gradual, resulting from the interaction between cyclic loads and the liquid media. The susceptibility of environmental assisted mechanical degradation of alumina is a well-established phenomenon taking place under sustained or variable stresses, i.e. under static or cyclic fatigue [8,9]. Furthermore, besides the Hertzian contact stresses related to the macroscopic contact geometry, two experimental facts should be underlined: a) the existence of repetitive low amplitude relative displacements between the contact surfaces of coating and indenter within the residual imprint (it indeed would resemble a fretting-fatigue scenario yielding local shearing stresses at the surface and subsurface levels) and b) the evidence of a pre-existing cracking-network within the  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> layer [3]. Both facts indicate that the relative differences observed may be rationalized on the basis of environmentally-assisted cracking under cyclic load of ceramics.

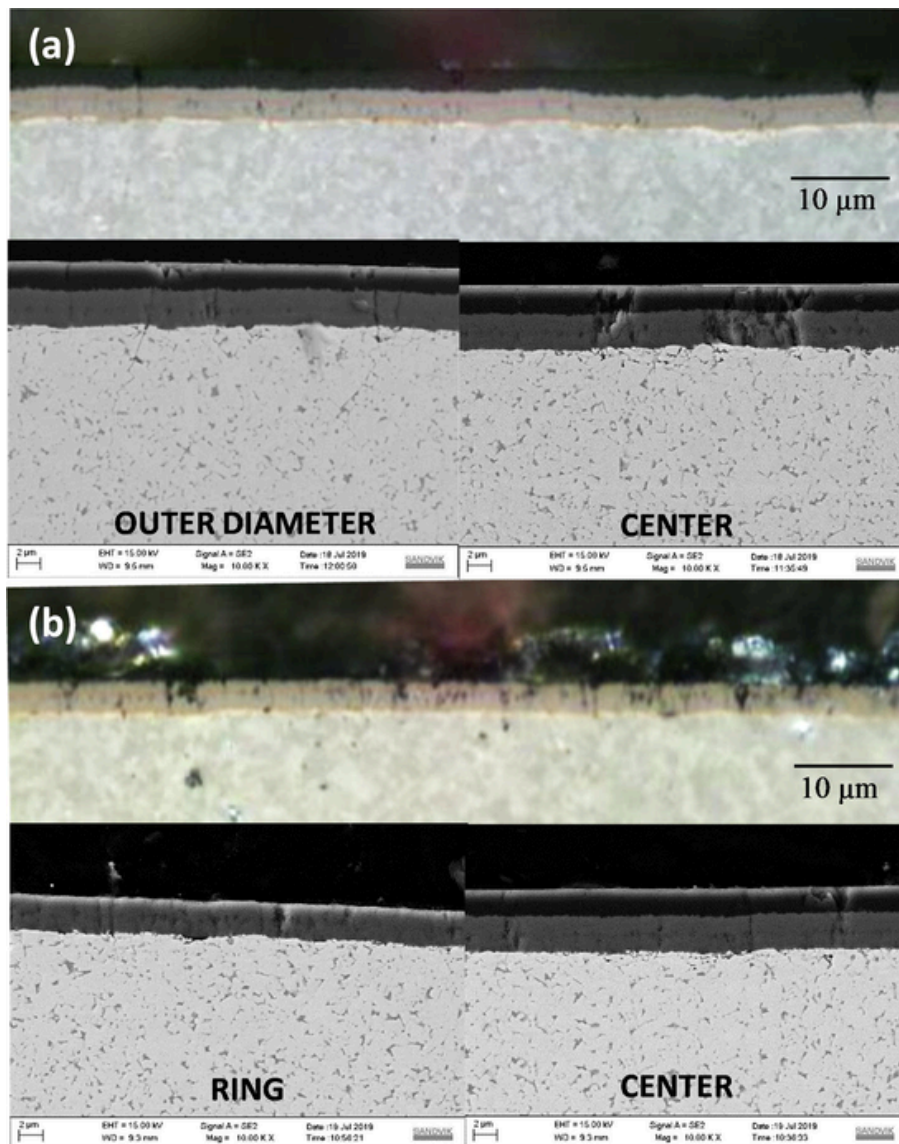
Therefore, exposure of the stressed  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> layer to a liquid media results in applied mode II stress intensity factors higher than the corresponding threshold for driving subcritical propagation of the existing

**Table 1**  
Chemical composition of stock solution for the cyclic indentation tests.

CAS-nr.	Name	Percentage (%)
68608-26-4	Sulfonic acids, petroleum, sodium salts	< 6
122-99-6	Phenoxyethanol	< 6
68132-46-7	Fatty acids, tall-oil, compounds with triethanolamine	< 2
8012-95-1	Highly refined mineral oil	Rest



**Fig. 1.** LOM top-view image of imprints for dry a) and wet b) tests, together with SEM images and EDS images of the yellow marked lines for both conditions. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)



**Fig. 2.** LOM cross section images of the outer region of the imprints and high magnification SEM images of the center and outer diameter of the imprints for the dry conditions (a), and center and ring for the wet conditions (b).

fissures following interlayer paths, i.e. lateral-like ones parallel to the surface (see Fig. 3). Thus, detachment of the  $\alpha$ - $\text{Al}_2\text{O}_3$  layer finally yields the effective exposure of underneath layers in the presence of the coolant fluid. These then get corroded (dark ring in Fig. 1) under the synergic effect of both liquid media and the fretting-like slip region [10]. The fact that such scenario is not observed in the residual imprints in dry conditions, where a higher threshold for crack extension in alumina should be expected together with the intrinsic chemical stability of the alumina in air, support the above ideas.

#### 4. Conclusions

A testing method to access information about the early stages and mechanisms that govern degradation of coated cemented carbides under cycling load is presented. In both conditions, we observe widened micro-cracks and damage in the carbide substrate in the center of the indentation, where compressive stresses were higher during the contact-fatigue test. In dry conditions, the coating is kept throughout the indentation despite microcracks are opened and the multilayer shows pronounced deformation. Wet indentations showcase an area in the shape of a ring, where the  $\alpha$ - $\text{Al}_2\text{O}_3$  layer has been removed throughout

the ring but is kept at the center of the indentation. It shows that the faster degradation of the  $\alpha$ - $\text{Al}_2\text{O}_3$  layer under wet cycling contact loads contributes to the faster widening of pre-existing CVD cooling cracks, which then evolve rapidly into observed large comb-cracks in wet milling operations [3,5].

#### CRedit authorship contribution statement

**D.A. Sosa:** Methodology, Formal analysis, Investigation, Writing - review & editing. **V. Collado Ciprés:** Formal analysis, Investigation, Writing - review & editing. **J. García:** Conceptualization, Methodology, Writing - original draft, Writing - review & editing, Visualization, Supervision, Funding acquisition. **E.L. Dalibón:** Formal analysis, Investigation, Writing - review & editing. **L. Escalada:** Formal analysis, Investigation, Writing - review & editing. **J.J. Roa:** Methodology, Formal analysis, Investigation, Writing - review & editing. **F. Soldera:** Formal analysis, Investigation, Funding acquisition. **S.P. Brühl:** Formal analysis, Investigation, Writing - review & editing, Supervision, Funding acquisition. **L. Llanes:** Formal analysis, Investigation, Writing - review & editing, Supervision, Funding acquisition. **S.**

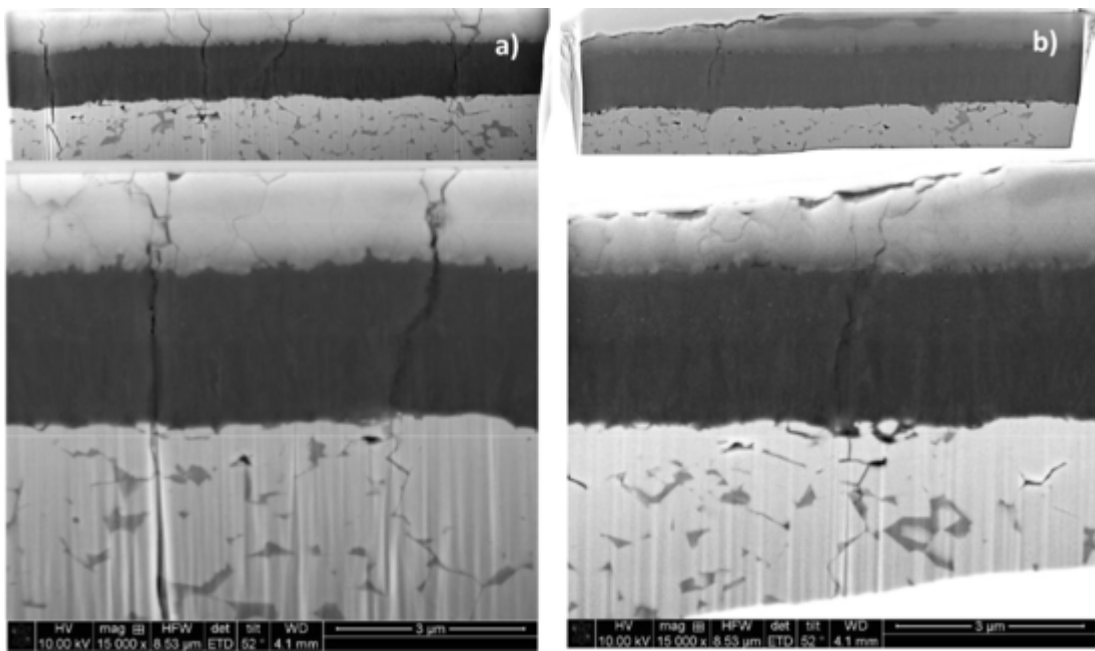


Fig. 3. SEM-FIB images on the outer diameter of the imprint in air (a) and in wet (b) conditions.

**Simison:** Formal analysis, Investigation, Writing - review & editing, Supervision, Funding acquisition.

#### 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.

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.matlet.2020.129012>.

#### References

- [1] J. García, V. Collado Ciprés, A. Blomqvist, B. Kaplan, Cemented Carbide Microstructures: a Review, *Int. J. Refract. Metals Hard Mater.* 80 (2019) 40–68, doi:10.1016/j.ijrmhm.2018.12.004.
- [2] J. García, R. Pitonak, R. Weißenbacher, A. Köpf, F. Soldera, S. Suarez, F. Miguel, H. Pinto, A. Kostka, F. Mücklich, Design and Characterization of Novel Wear Resistant Multilayer CVD Coatings with Improved Adhesion Between  $\text{Al}_2\text{O}_3$  and Ti(C, N), *Adv. Eng. Mat.* 12 (2010) 929–934, doi:10.1002/adem.201000130.
- [3] J. García, M. Moreno, J. Östby, J. Persson, H. Pinto, Design of coated cemented carbide with improved comb crack resistance. In: *Proceedings of the 19th Plansee Seminar, Reutte, Tirol, 2017*.
- [4] E. Tarrés, G. Ramírez, Y. Gaillard, E. Jiménez-Piqué, L. Llanes, Contact Fatigue behavior of PVD-coated hardmetals, *Int. J. Refract. Metals Hard Mater.* 27 (2009) 323–331, doi:10.1016/j.ijrmhm.2008.05.003.
- [5] I. El Azhari, J. García, F. Soldera, S. Suárez, E. Jiménez-Piqué, F. Mücklich, L. Llanes, Contact damage investigation of CVD carbonitride hard coatings deposited on cemented carbides, *Int. J. Refract. Metals Hard Mater* 86 (2020) 105050, doi:10.1016/j.ijrmhm.2019.105050.
- [6] I. El Azhari, J. García, M. Zamanzade, F. Soldera, C. Pauly, L. Llanes, F. Mücklich, Investigations on micro-mechanical properties of polycrystalline Ti (C, N) and Zr (C, N) coatings, *Acta Mat.* 149 (2018) 364–376, doi:10.1016/j.actamat.2018.02.053.
- [7] J. J. Roa, S. Simison, J. Grasso, M. Arcidiacono, L. Escalada, F. Soldera, J. García, A. D. Sosa, Cyclic contact fatigue of cemented carbides under dry and wet conditions: Correlation between microstructure, damage and electrochemical

behavior, *Int. J. Refract. Metals Hard Mater.* 92 (2020) 105279/1-105279/10. Doi: 10.1016/j.ijrmhm.2020.105279.

- [8] S.M. Barinov, N.V. Ivanov, A.V. Kurepin, V.Y. Shevchenko, Effect of Environment on Delayed Failure in a Glass-Bonded Alumina, in: R.C. Bradt, D. Munz, M. Sakai, V.Y. Shevchenko, K. White (Eds.), *Fracture Mechanics of Ceramics*, Springer, Boston, MA, 2002, pp. 235–242.
- [9] K.S. Lee, Y.-G. Jung, I.M. Peterson, B.R. Lawn, D.K. Kim, S.K. Lee, Model for Cyclic Fatigue of Quasi-Plastic Ceramics in Contact with Spheres, *J. Am. Ceram. Soc.* 83 (2000) 2255–2262, doi:10.1111/j.1151-2916.2000.tb01544.x.
- [10] Y.-G. Jung, I.M. Peterson, D.K. Kim, B.R. Lawn, Lifetime-limiting Strength Degradation from Contact Fatigue in Dental Ceramics, *J. Dent. Res.* 79 (2000) 722–731, doi:10.1177/00220345000790020501.