Elsevier Editorial System(tm) for 43rd International Conference on Metallurgical Coatings and Thin Films
Manuscript Draft

Manuscript Number:

Title: Small Scale Fracture Behaviour of Multilayer TiN/CrN Systems: Assessment of Bilayer Thickness Effects by Means of Ex-situ Tests on FIB-milled Micro-cantilevers

Article Type: Full Length Article

Section/Category: Symposia E, H, TS1,TS3 and TS5

Keywords: Multilayered TiN/CrN coatings, small scale fracture behavior, layer architecture, bilayer period, micro-cantilevers

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Abstract: TiN/CrN multilayered PVD coatings are known to exhibit outstanding micromechanical properties and wear resistance. On the other hand, information on their small scale fracture behaviour is rather scarce. The present work aims address it by testing to failure FIB-milled microbeams of multilayer TiN/CrN systems with different bilayer periods (8, 19 and 25 nm). In doing so, these micrometric specimens are first FIB notched, and thus deflected by means of a nanoindentation system. It is found that multilayer architecture translates into a beneficial synergic effect regarding critical load for reaching unstable failure; and thus, on energy absorption at fracture. Such behaviour is associated with small scale crack deflection as main toughening mechanism.

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*Cover Letter

Surface Coatings and Technology
Editor

Barcelona, May 14th 2016

Dear editor

Enclosed please find the manuscript entitled "**Small Scale Fracture Behaviour of Multilayer TiN/CrN Systems: Assessment of Bilayer Thickness Effects by Means of Ex-situ Tests on FIB-milled Micro-cantilevers**" to be considered for its publication in Surface Coatings and Technology. I hereby declare that this work is original and not intended for submission elsewhere.

I believe that this paper will be of interest for the community of researchers working on hard coatings deposited on soft and ductile substrates in order to estimate the proper way to observe the fracture behavior of multilayer TiN/CrN systems by ex-situ tests on FiB-milled micro-cantilevers.

Thank you in advance for taking into consideration.

Looking forward hearing from you,

Yours sincerely

Dr. J. J. Roa

*Authors Certification Form

AUTHOR'S CERTIFICATION

This is to certify that I have obtained the necessary authorization for publication of the enclosed paper # H2-1-3 in the Proceedings of the ICMCTF 2016 Conference and in Surface and Coatings Technology/Thin Solid Films and that the paper is original and unpublished and is not being considered for publication elsewhere.

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Research Highlights

- Fracture behavior of multilayer TiN/CrN systems by means of micro-cantilever deflection.
- Multilayer architecture translates into a beneficial synergic effect.
- Crack defection is the main toughening mechanism.

*Manuscript

Small Scale Fracture Behaviour of Multilayer TiN/CrN Systems: Assessment of Bilayer Thickness Effects by Means of Ex-situ Tests on FIB-milled Micro-cantilevers

Small Scale Fracture Behaviour of Multilayer TiN/CrN Systems: Assessment of Bilayer Thickness Effects by Means of Ex-situ Tests on FIB-milled Micro-cantilevers

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Abstract

TiN/CrN multilayered PVD coatings are known to exhibit outstanding micromechanical properties and wear resistance. On the other hand, information on their small scale fracture behaviour is rather scarce. The present work aims address it by testing to failure FIB-milled microbeams of multilayer TiN/CrN systems with different bilayer periods (8, 19 and 25 nm). In doing so, these micrometric specimens are first FIB notched, and thus deflected by means of a nanoindentation system. It is found that multilayer architecture translates into a beneficial synergic effect regarding critical load for reaching unstable failure; and thus, on energy absorption at fracture. Such behaviour is associated with small scale crack deflection as main toughening mechanism.

Keywords: Multilayered TiN/CrN coatings, small scale fracture behavior, layer architecture, bilayer period, micro-cantilevers

1. Introduction

Rapidly growing trends in developing and deploying advanced processing technologies, manufactured components and products are expected to demonstrate superior quality and enhanced functional performance. Within this context, material removal processes continue to dominate among all manufacturing process. In this regard, the functional performance of components from material removal processes is heavily influenced by the quality and reliability of the produced surfaces, in terms of both topography as well as metallurgical and mechanical state of the subsurface layers [1,2,3,4,5,6].

Ceramic coatings are commonly applied to various kinds of cutting and forming tools, providing surfaces with enhanced tribological properties in terms of low friction, high hardness and improved wear resistance [7,8,9,10,11].

From this viewpoint, extensive research conducted in the last two decades has validated the approach of multilayered architectures for optimizing the performance of coatings based on transition metal nitrides. Among these, TiN/CrN multilayer films are the most successful example. Work done on TiN/CrN coatings includes microstructural characterization, hardness and stiffness assessment, chemical stability, residual stress evaluation, and friction/wear behaviour, among others [12,13,14,15]. However, information regarding small scale mechanical response of these multi-layered coatings is quite limited [16]. This is particularly true concerning fracture behaviour of notched FIB-milled microbeams, the main subject of this study.

Over the last decade, great advances have been made in understanding the small scale mechanical properties on coating systems at micron and submicron length scales by means of nanoindentation technique to bend micro-cantilevers, confined in the coating as well as in the interface, produced by focused ion beam (FIB) machining [17,18]. This technique is a miniaturized version of the conventional macroscopic methods to evaluate fracture behaviour, yielding responses close to those found for macroscopic samples [19,20,21,22,23]. Nevertheless, special care should be taken when implementing FIB technique to mill micro-cantilevers, as it can cause Ga^+ implantation as well as radiation damage. Furthermore, material removed may be redeposited along the periphery of the beam trajectory, causing a modification of the stress state induce during the test.

In this study, a systematic micromechanical study to correlate the micromechanical properties (hardness and elastic modulus previously investigated [15]) with the coating fracture behaviour has been conducted in three multilayer TiN/CrN systems with different bilayer period (9, 19 and 25 nm). Additionally, experimental work has been performed on corresponding single layers, for comparison purposes. Finally, fracture mechanisms, in terms of crack-layer architecture interactions, observed after bending of micro-cantilevers are compared with those discerned when subjecting the coated systems to macroscopically imposed loads.

2.- Experimental procedure

Five different coatings (TiN/CrN with different bilayer periods: 8, 19 and 25 nm; as well as TiN and CrN monolayer) were deposited in an industrial scale METAPLAS MZR323 system. More information related to the deposition process can be found in Ref. [15]. Substrate material is a commercial high speed steel (HSS, M2/EN-1.3343), thermally hardened prior to deposition and polished to a 0.5 μm of an arithmetic mean surface roughness (R_a).

Three micro-cantilevers were manufactured per each specimen by using the FIB equipment. Shaping of triangular cantilevers was done with a 30 kV Ga^+ source in a dual beam Zeiss Neon 40 focused ion beam – field emission scanning electron microscope (FIB/FESEM) with currents of around 500 pA in final machining stages, following the method described by Armstrong et al. [24]. The most important milling step is the final stage, in which the notch is produced. To make the notch as thin as possible, milling was done using a low ion current, 20 pA.

Figure 1a exhibits a scheme of the cantilevers milled by using FIB. The final cantilevers had a length (L) of ~ 10 μm, a width (W) of ~ 3 μm, a triangular cross section with a maximum depth or also known as height (H) of ~ 1.5 μm, and a notch thickness of around 150 nm (**Figure 1b** and **1c**).

The dimensions of each micro-cantilever tested in this work were measured individually using FESEM images collected both before and after testing (the latter are needed to capture the point of load application). The data for all the fifteen micro-cantilevers tested are summarized and reported in **Table 1**.

Each cantilever was imaged using the FESEM column of the Zeiss Neon 40 FIB/FESEM before testing for accurate measurement of the test geometry. Similar inspection procedure was conducted after testing, to study the deformation and/or fracture that had occurred. The different micro-cantilvers were tested by using the nanoindenter system XP (Agilent Technologies). A Berkovich diamond indenter was employed to bend the different micro-cantilevers at a constant rate of 10 nm/s until rupture. Finally, load-displacement responses of those micro-cantilevers were continuously recorded at the same time.

3. Results and discussion

Representative load-displacement or load-defection curves for each coating type studied is presented in **Figure 2**. All curves exhibit an initially linear trend. In general, stiffer systems (TiN and multilayer with lower bilayer periods) showed higher slopes [15]. Very interesting, the load-deflection response of multilayer films tend to exhibit subsequent discontinuities (pop-ins) related to nonlinear deflection. This is not the case for both single-layer coatings. In all the cases, after peak load is reached (known as critical load or P_c), unstable crack propagation until rupture is discerned. Wider or narrower separation of individual deflection points close to critical load level is associated with effective separation of the cantilever from the remainder of the tested specimen.

Regarding bilayer thickness effects, not only stiffness but also critical load to fracture rises, as bilayer period decreases. On the other hand, the different pop-ins observed in the load-deflection curves may be related to continuous small length-scale deflections of cracks nucleated at the notch tip. This is completely supported by the fracture scenario observed by FESEM inspection after rupture (e.g. **Figure 3**). Furthermore, it is in complete agreement with the crack-layer architecture interaction previously discerned for these coatings at the subsurface level, when subjected to monotonic spherical indentation. The referred crack deflection is associated with length scales of microstructural column width and thickness ratio of the individual TiN and CrN layers for a particular bilayer period. Accordingly, smaller bilayer periods are expected to translate into higher capability of energy absorption before unstable crack extension takes place.

Crack deflection phenomena may be related to two factors: i) mismatch in terms of thermal expansion coefficient between each layer (TiN and CrN), which causes residual stresses, and ii) interface adhesion strength. Both factors may induce debonding and sliding effects in the multilayer interface, as commented by Evans and co-workers [25]. Then, the fracture along the multilayer interface is governed by the combination of elasticity and plasticity mismatch across the interface. It can be modelled by two

Dundurs parameters which takes into account the elastic mismatch. These mistmach parameters can be estimated by assuming two completely isotropic layers, as follows [26]:

$$D_{\alpha} = \frac{\overline{E_1} - \overline{E_2}}{\overline{E_1} + \overline{E_2}} \tag{1}$$

and

$$D_{\beta} = \frac{1}{2}\frac{\mu_1(1-2\upsilon_2) - \mu_2(1-2\upsilon_1)}{\mu_1(1-\upsilon_2) + \mu_2(1-\upsilon_1)} \tag{2}$$

where $\overline{E}$ is determined as $\overline{E} = E/(1-\upsilon^2)$, μ is $\mu = E/2(1+\upsilon)$, the subindex 1 and 2 are related to TiN and CrN layers respectively, E is the Young's modulus for the material of each layer (E_{TiN} = 500 ± 15 GPa and E_{CrN} = 320 ± 8 GPa, as determined by means of nanoindentation in Ref. [15]) and ν is the Poisson's ratio - taken as 0.25 for both materials. Then, by using equations (1) and (2), the D_a and D_β parameters for the systems of interest are 0.15 and 0.07, respectively. As it is found that $D_\alpha \neq D_\beta$, a nucleated crack will be expected to grow until reaching the TiN/CrN interface, but once there it will move along the interface. This will be the crack path until the system reaches a critical energy value which induces the crack to propagate readily into the adjacent layer, until reaching the next interface. Such fracture mechanisms may be understood as an autocatalytic process due to the fact that indenter is continuously bending the micro-cantilever. Hence, critical values are subsequently reached, until rupture occurs. These observations have been previously observed by Folsom and co-workers for multilayer beams comprised of indented glass plates systems [27,28]. Furthermore, Suresh [29] attributed the tortuous crack path through the multilayers to the non-linearity stress distribution along the coating, being this the main responsible for explaining the different pop-ins observed in the loading-deflection curves shown in **Figure 2**.

Above findings are very interesting because, different from the layer architecture influence assessed on hardness and stiffness, multilayer systems do exhibit an enhanced small scale fracture behaviour as compared to the single layer ones. On the other hand, and as expected, as bilayer period decreases, measured micromechanical properties and energy absorption capability (related to density of length scale deflection events) rise proportionally.

4. Summary

The influence of layer architecture and bilayer period on the small scale fracture behaviour of TiN and CrN base coatings has been assessed. It was evaluated at by testing FIB-milled micro-cantilevers in the films. Load-deflection curves are directly proportional to stiffness of the studied coatings. However, multilayer systems exhibit qualitative differences, in terms of discontinuities (pop-ins) related to nonlinear deflection, with respect to the single layer ones. Such length scale deflections translate into effective toughening, and can be rationalized on the basis of elastic mistmatch between layers and stress gradient distribution present along the one layer system. Very interesting, experimental findings point out a beneficial synergic effect associated with multilayer configuration, which was not previously discerned for other micromechanical properties measured, i.e. hardness and stiffness.

Acknowledgements

The authors greatly acknowledge “Direcció General de Recerca del Comissionat per a Universitats i Recerca de la Generalitat de Catalunya” for recognizing CIEFMA as consolidate Research Group (2014-SGR-130). This work was financially supported by the Spanish Ministerio de Economía y Competitividad (Grant MAT2015-70780). Dr. J.J.Roa would like to thank the Juan de la Cierva Programme for its financial support (Grant no. JCI-2012-14454).

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Table captions

Table 1. Sample description (bilayer period, λ for each system) and sample dimensions (*W*, *L* and *H*, as defined in **Figure 1a**).

Table 1

Sample	λ (nm)	Dimensions (μm)		
		W	**L**	**H**
TiN	-	2.40	10.7	2.01
		2.08	9.74	1.63
		2.67	10.1	1.83
CrN	-	1.88	9.75	1.15
		2.33	10.1	1.37
		2.25	10.7	1.31
TiN/CrN	8	1.64	10.2	1.41
		2.36	9.58	1.72
		1.93	9.33	1.52
	19	2.27	9.24	1.45
		2.67	9.37	1.42
		2.53	9.14	1.52
	25	2.72	9.40	1.79
		2.25	9.24	1.44
		2.67	9.37	1.54

Figure captions

Figure 1. (a) Cantilever scheme, where L, *H* and *W* are the length, height and width of the cantilever, while t is the notch thickness. FESEM single cantilever images for the TiN/CrN with a bilayer period of 25 nm tilt, (b) top-view observation and (c) end geometry (tilt corrected).

Figure 2. Load-displacement response for each specimen investigated here.

Figure 3. Fracture surface observed by FESEM of TiN/CrN specimen with a bilayer period of 25 nm. Zig-zag crack paths, corresponding to small scale deflections are clearly discerned.

Figure 1

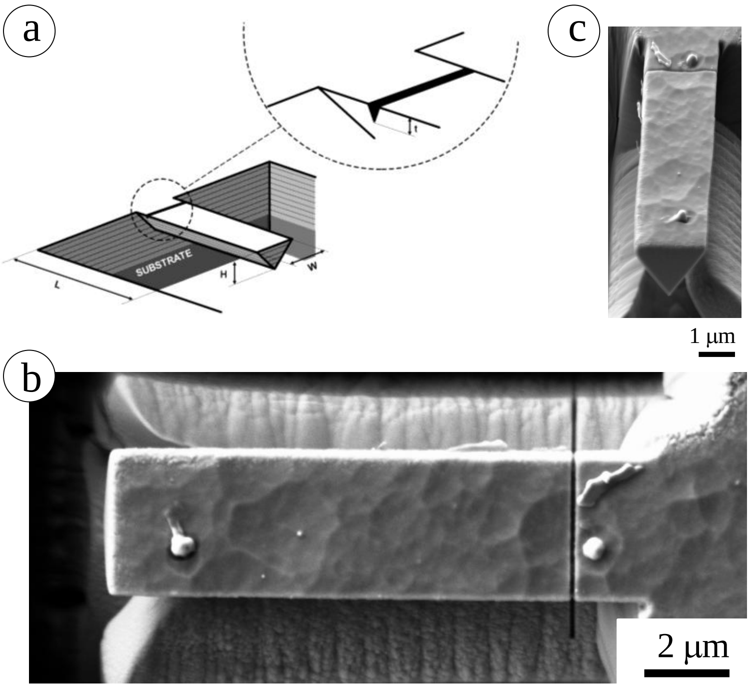

Figure 1

Figure 2

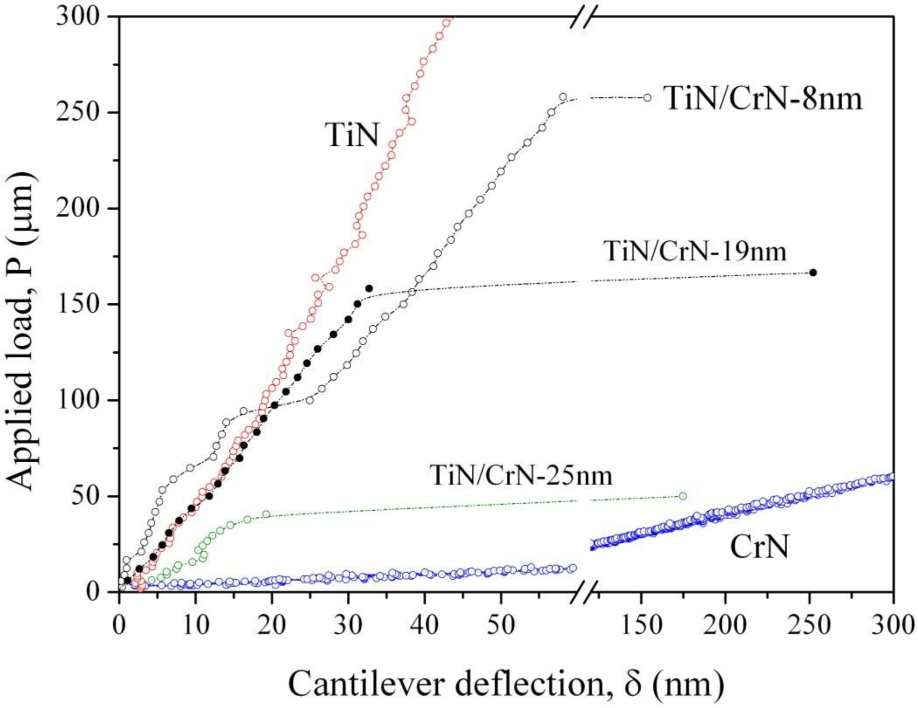

Figure 2

Figure 3

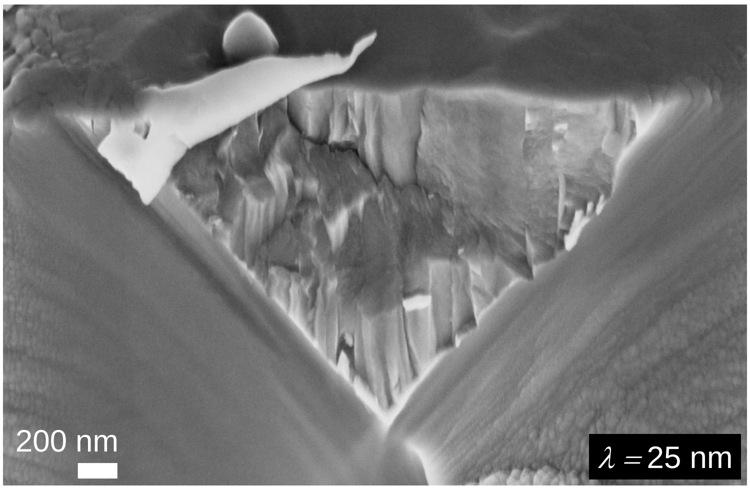

Figure 3