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### **Abstract**

Sliding tests both in micro and nano level were done on hard coatings developed on silicon substrates and 304SS substrates using magnetron sputtering. Different types of failure morphologies manifested themselves and were found to be a function of substrate, indenter, loading nature apart from the coating. Linear profiles of the failed segments along with analysis of the scratch track were done which provided a qualitative depiction of adhesive, wear and abrasive properties of the coatings useful for their heavy duty and microelectronic applications

# Sliding indents of hard coatings: analysis of failure morphologies and intermediate fracture energies

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## ABSTRACT

Sliding tests both in micro and nano level were done on hard coatings developed on silicon substrates and 304SS substrates using magnetron sputtering. Different types of failure morphologies manifested themselves and were found to be a function of substrate, indenter, loading nature apart from the coating. Linear profiles of the failed segments along with analysis of the scratch track were done which provided a qualitative depiction of adhesive, wear and abrasive properties of the coatings useful for their heavy duty and microelectronic applications

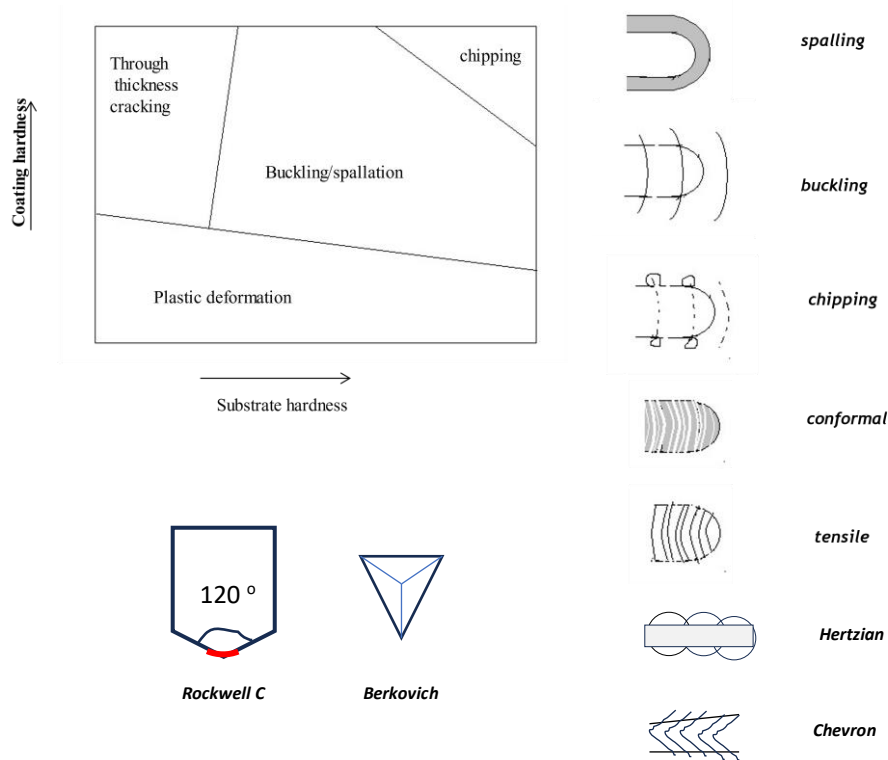
Keywords: Scratch, hard coatings, nano-scratch, adhesion, critical load

## INTRODUCTION

The determination of coating strength based on indentation techniques is one of the most basic practices exercised by material scientists. Apart from static indentations performed by indenters of different size and shapes, sliding indentation or popularly known as scratch tests has proved its own significance as it gives a two-dimensional representation of the chronological coating response subject to an increasing load. The load at which the coating fails (critical load, **L<sub>c</sub>**) is taken as the measure of the adhesive property. However, the failure in most cases is not specific to a particular load or scratch length. There happens to be different stages of coating failure which again vary based upon the coating properties, the substrate as well as the scratch conditions like scratch velocity, loading rate and shape of the indenter [1-6]. The nature of substrate and thickness have influenced wear and abrasive properties of the coating as well [7-9]. The sliding indentation method has been used on hard coatings to assess their adhesive strength and wear properties. Nanocomposite hard coatings like Si-C-N, Ti-B-Si-C-N have multiple phases which emerge based on the deposition conditions and effect the coating properties [10-21].

The different failure modes based on coating and substrate hardness is given **Fig 1a** and schematically represented in **Fig 1b** [6]. Total delamination is represented by spallation, while buckling corresponds to partial delamination. Chipping on the other hand is found to appear along the edges of the scratch track. A tensile frictional stress behind the stylus causes tensile cracking, while tensile bending moment within the coating leads to conformal cracks. Circular Hertzian cracks are also seen in some instances which arise due to union of two symmetrical cracks at the edge of the scratch line are found in the case of a

spherical or Rockwell C indenters. Symmetrical radial cracks from the center called Chevron cracks is another form of coating failure observed during scratching. The geometry of Rockwell C and Berkovich indenter is given in Fig 1c.



**Fig 1** (a, b) The different scratch failure modes based on coating and substrate hardness. and (c) geometry of Rockwell C and Berkovich indenter.[6]

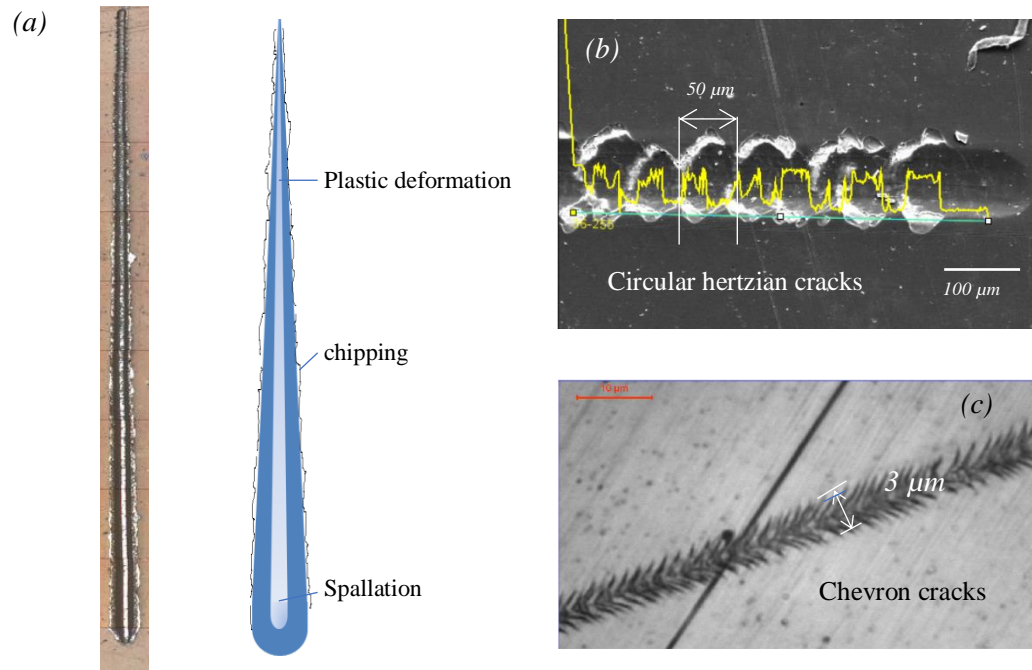
Studies involving cohesive zone model (CZM) dealing with contact friction and interfacial plasticity and damage have been performed. Energy-based model of adhesive contact for initiation and growth of interface cracks in the shearing mode has also been proposed [2].

#### MATERIALS & METHODS

Si-C-N and Ti-B-N coatings were deposited on Si(100) and 304SS substrates by magnetron sputtering (HHV, Bangalore, India) in Ar/N<sub>2</sub> atmosphere from SiC (sintering a Silicon and Carbon powder compact taken in 1:1 ratio) and TiB<sub>2</sub> (Ti and B in 1:2 ratio) targets. The sliding indents were carried out by Scratch Test Tr-101 (Ducom, Bangalore, India) that used a Rockwell C indenter which is a 120° diamond cone with 0.2mm diameter spherical tip. The scratch tests were done at rate of 10 N/mm at a speed of 0.2 mm/s. The critical load (L<sub>c</sub>) was determined from the point of change of slope in the coefficient of friction vs. normal load, tractional force, and scratch length curves. ImageJ software was used to get profiles of the scratch tracks. Nanoscratch tests were performed by Berkovich indenter (MTS, USA).

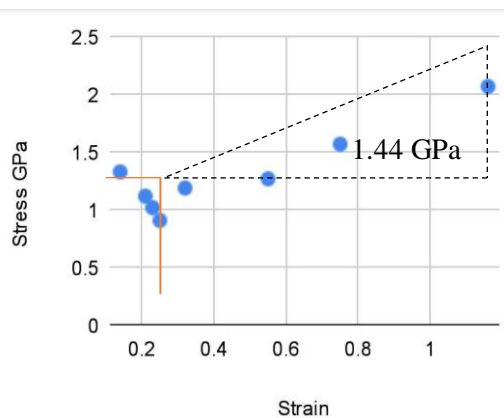
## RESULTS & DISCUSSIONS

The different types of failure occurring in case of the Ti-B-N and Si-C-N coatings is given in **Fig 2**.



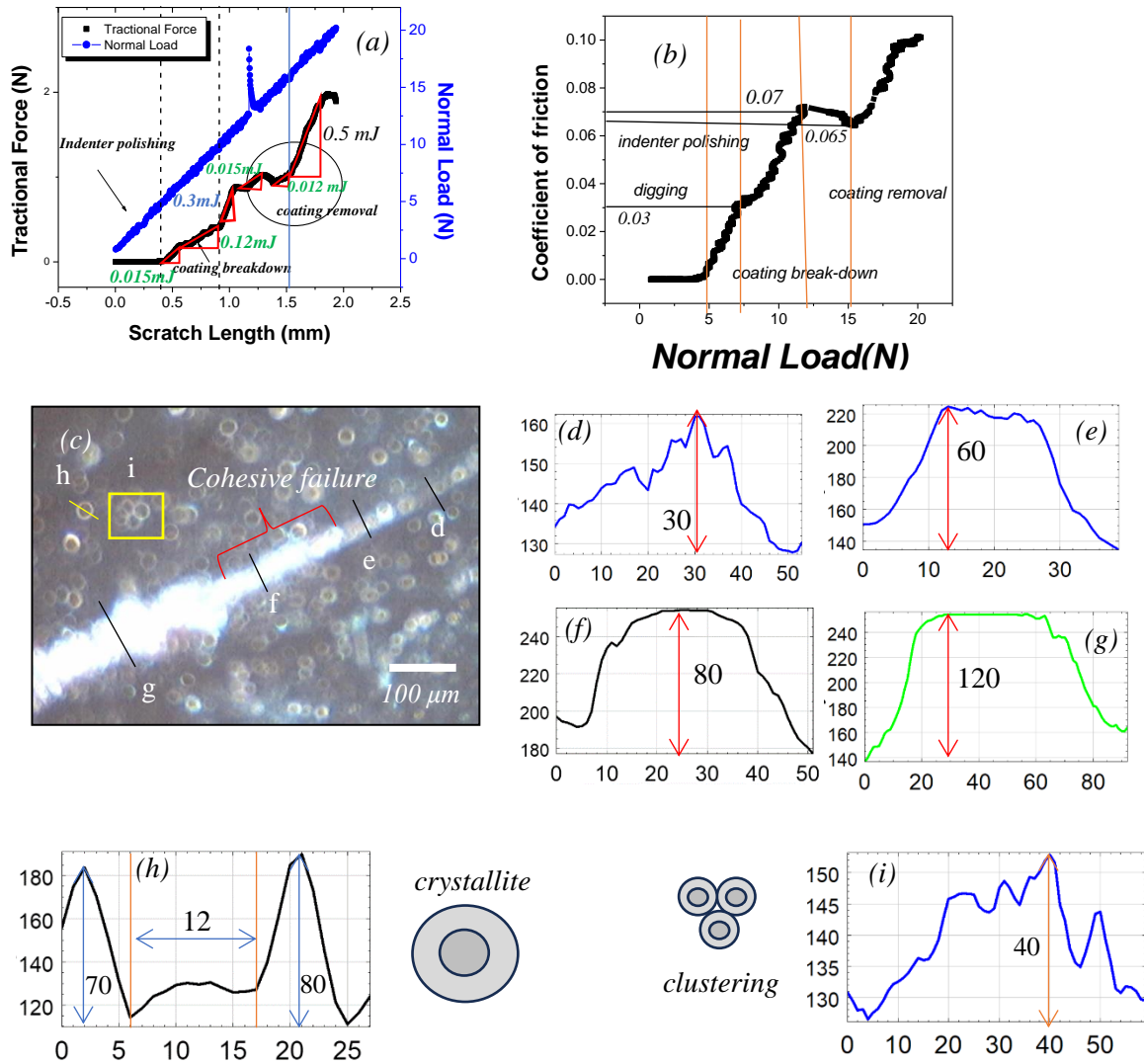
**Fig 2.** Scratch tests on Ti-B-N [22] on (a) 304 SS with (b) Si (100) and (c) Si-C-N coatings on 304 SS showing different failure modes

The process of scratching involves large elastoplasticity, cracking, fracture, wear) and the nonlinearity in geometry of a contact surface due to severe plastic deformations at the surface. The adhesive failure starts at **0.8GPa** corresponding to a strain of **0.25**. The area under the stress-strain curve post adhesive failure is the interfacial toughness which is calculated to be **1.44 GPa**.



**Fig 3.** Stress-Strain plot from scratch test

Ti-B-N coatings deposited on 304 SS showed plastic deformation followed by chipping from the sides and ultimately causing spallation (Fig 2a). For deposition on Si(100) substrates, circular Hertzian cracks were seen (Fig 2b). Si-C-N coatings on 304SS showed chevron cracks. The difference being nano scratch was used for Si-C-N/304 SS. This shows the brittle failure observed for nano scratch has negligible influence from the substrate.



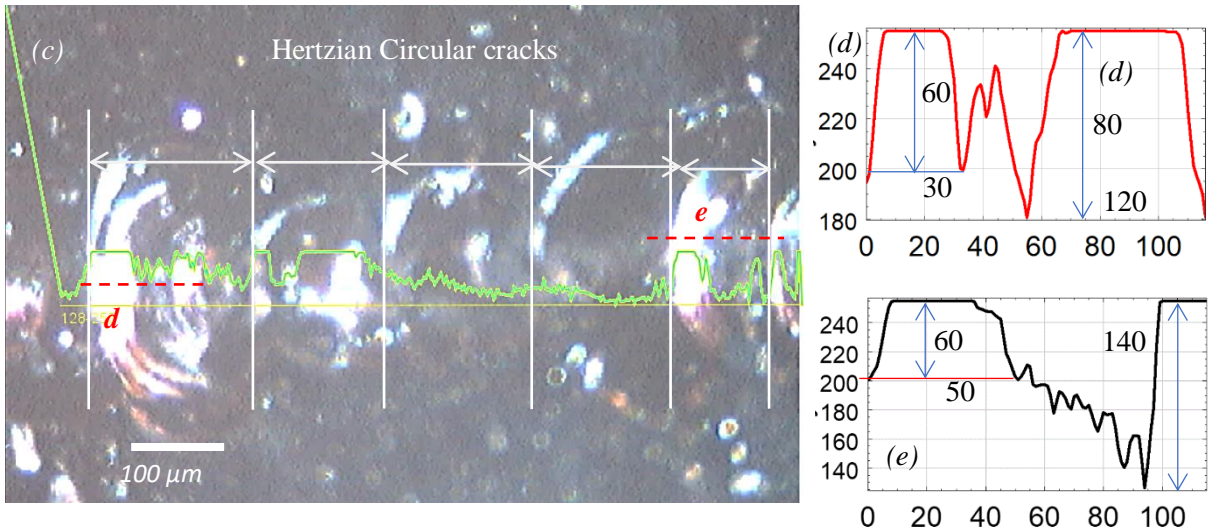
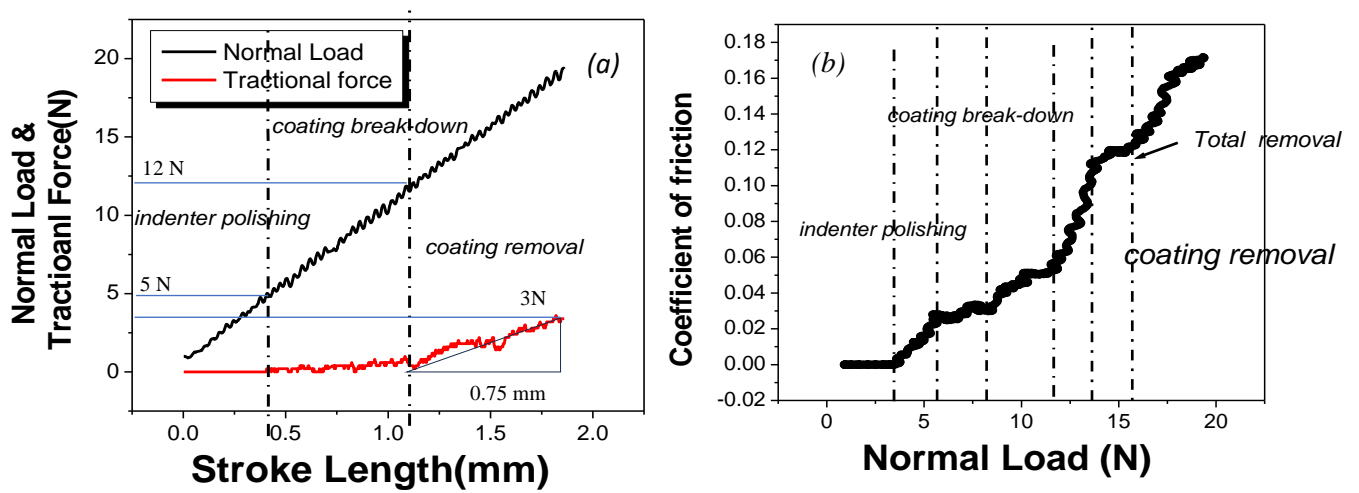
**Fig 4.** Scratch tests with 10N/mm loading rate for silicon substrates showing plots of (a) Normal load and Tractional force vs. stroke length, (b) C.O.F vs. normal load and (c) the optical micrograph of the scratch track showing sudden and brittle failure (d -g) cross-sectional profiles of the scratch track at different stages (h) line profile of a crystallite and (i) clustered crystallites

Micro-Scratch tests performed on Si-C-N coatings deposited on Si(100) substrates at a loading rate of 10 N/mm as shown in **Fig 4**. The energy dissipated during the scratch process at different stages can be evaluated by the area under the tractional force and scratch length plots. A higher resistance offered gets reflected in a higher slope of the plot. Slopes of different magnitudes can be seen along the total scratch length. Energy from each zone were calculated which varied from **0.015 mJ** during the initial contact between the indenter and sample to **0.5 mJ** during adhesive failure. An overall increase in the COF value with abrupt changes whenever the configuration of the indenter-sample contact changed can be noticed.

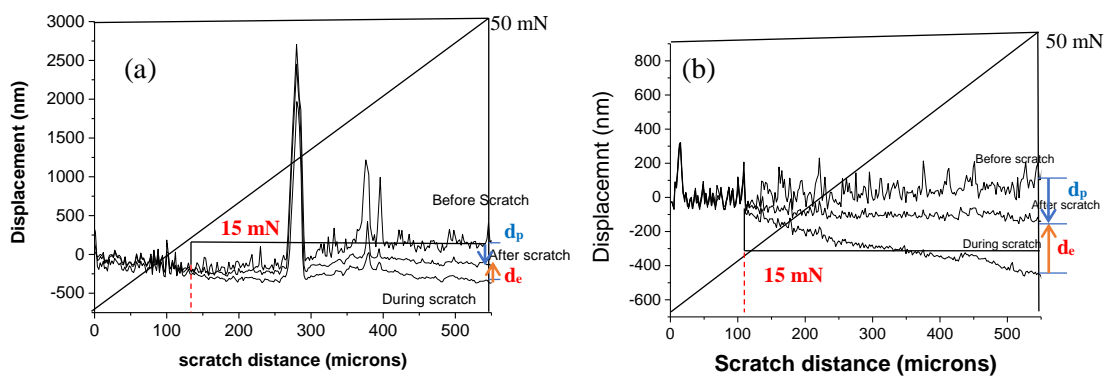
A clear distinction can be between the different failures occurring during the sliding event from the optical image of the scratch track (**Fig 4c**). The abrasion started with material removal confined within the scratch track to a maximum as cohesive failure, which was followed by removed material going out of the scratch track indicating the initiation of adhesive failure. The line profiles of the different regions as marked in the figure are shown in **Fig 4(d-g)**. The formation of crystallites and their clustering can also be seen alongside the scratch track (**Fig 4 h, i**).

Circular Hertzian cracks were visible for coatings of lower hardness deposited at a substrate temperature of 100 °C. The effect of substrate temperature on the adhesive properties of Si-C-N has been reported earlier [23]. The coating showed a cohesive failure at 5N and adhesive failure at 12N (**Fig 5a**), although it involved failure in multiple stages as clearly depicted in COF vs normal load plot (**Fig 5b**). The energy dissipated due to adhesive failure was **2.25 mJ** as obtained from the area under the tractional force ( $F_T$ ) and scratch distance curve. The linear profile along the scratch track showed coating removed in a periodic manner as per the geometry of the spherical Rockwell C indenter tip. Each of the failed regions as shown distinctively had an advancing zone and a trailing zone, the difference between the two got reduced (in terms of separation and height to width ratio) from lower to higher load applications (**Fig 5 d, e**).

The nano-scratch studies of the coatings were done with a Berkovich indenter, scratching with a speed of **50  $\mu\text{m/s}$**  to a maximum load of **50 mN** on a **600  $\mu\text{m}$**  scratch track for coatings on 304SS and Silicon (**Fig 6**). The three curves indicate before, during and after scratch displacements. The indenter depth increased linearly with the ramping load without abrupt change on the curve due to progressive peeling of the coating indicating good adhesion. The curves after scratching and before scratching are separately observable indicating permanent plastic residual deformation in the coating. Effect of substrate can be seen in this case as with 304SS substrate, the permanent deformation was higher in ratio ( $d_p/d_e$ ) with respect to the elastic recovery due to the ductility associated with 304SS. The critical load which marked the deviation of the three profiles was around 15 mN at a scratch distance of 130  $\mu\text{m}$  for 304SS and 110  $\mu\text{m}$  for Silicon.



**Fig 5.** Scratch test with **10N/mm** loading rate for Si Substrates showing plots of (a) Normal load and Tractional force vs. stroke length, (b) C.O.F vs. normal load and (c) the optical micrograph of the scratch track showing failure by circular cracking and surface intensity profile. The films were deposited at **100 °C in RF mode**. Linear profiles of the regions marked (d) for higher loads (e) lower loads [21]



**Fig 6.** Nanoscratch tests performed on Si-C-N coatings deposited on a) 304 SS and b) Si(100) substrates showing variation in permanent deformation ( $d_p$ ) and elastic recovery ( $d_e$ )

## CONCLUSIONS

The analysis of scratch tests performed on hard coatings for studying properties related to adhesion, wear and abrasion requires a multi-facial approach involving scratch track analysis and load-depth plots going in tandem. The linear profiles showing the material removal and the dimensions of the different film failure features will help in proper investigation of a hard coating's capabilities for usage. Si-C-N hard coatings showed different failures feature in response to sliding indents based upon its cohesive and adhesive. Intermediate energy dissipations were determined leading to full adhesive failure. Coatings on Si failed faster while effect of ductility was found for coatings on 304SS even at lower loads. The intermediate fracture energies were determined related to localized failures during the scratching event.

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## DECLARATIONS

### Compliance with Ethical Standards

The manuscript has not been submitted in parallel either in full or partially to any other journal.

### Conflict of interest

There is no conflict of interest among the authors

### Research Data Policy and Data Availability Statements

Data shall be provided on request

### Credit author statement

A. S. Bhattacharyya being the sole author, was involved in performing the experiments, analysing, and framing the manuscript

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