Surface & Coatings Technology xxx (2016) xxx–xxx

Contents lists available at ScienceDirect

Surface & Coatings Technology

journal homepage: www.elsevier.com/locate/surfcoat

Scratch resistance of superhard carbon coatings – A new approach to failure and adhesion evaluation

M. Zawischa [a,b,*], S. Makowski [a,1], N. Schwarzer [c], V. Weihnacht [a]

[a] Fraunhofer Institute for Material and Beam Technology IWS, Dresden, Germany
[b] Technische Universität Dresden, Institute of Manufacturing Technology, Dresden, Germany
[c] Saxonian Institute of Surface Mechanics, Ummanz, Germany

ARTICLE INFO

Article history:
Received 7 June 2016
Revised 20 July 2016
Accepted in revised form 24 July 2016
Available online xxxx

Keywords:
Scratch test
Ta-C
DLC
Delamination
Adhesion
Failure mechanisms

ABSTRACT

Scratch testing was methodically studied by examining the adhesion of tetrahedral amorphous carbon coatings (ta-C). Usually, critical loads L_C are used as a parameter to characterize the adhesion strength of coatings in scratch testing. However, the comparability of the critical loads for different materials and coating thicknesses is limited. The critical loads result from different coating failure mechanisms that require a careful investigation with respect to plastic substrate deformation and coating thickness.

In this work, the different failure modes of ta-C coatings occurred in strong dependence on the geometrical proportions (indenter radius and coating thickness) for a wide range of hardness and Young's modulus: Adhesive failure in the center of the groove was located at cohesive bending cracks for coating thickness of 1 to 3 μm. For increasing coating thickness failure by spallations was found instead of bending cracks. At the edge of the groove outside the scratch-induced substrate plasticity, wing-shaped delaminations were observed for all test and coating configurations.

A new method was introduced that quantifies the adhesive failure by means of segmentation of an optical image. The relative area of delamination was found to correlate with calculated shear stresses at the interface plane outside the heavily deformed scratch track. Furthermore, it was possible to distinguish the effect of two different plasma pretreatments on adhesion regardless of the coating thickness. Thus, this new method expanded the information gained from the standard scratch test and allows a better interpretation of coating adhesion than the L_C-evaluation.

1. Introduction

A sufficient adhesion to the substrate material is necessary to benefit from the good mechanical and tribological properties of hard PVD coatings such as hydrogen-free tetrahedral amorphous carbon coatings (ta-C). Those coatings show a very high indentation hardness from 40 GPa to 80 GPa and excellent low friction properties in lubricated and unlubricated conditions. Adhesion of such coatings can be critical due to the high mismatch of elasticity between coating and substrate as well as high compressive internal stress which is typical for PVD coatings. Therefore, the evaluation of the adhesion is an important part of the coating development. This study provides a methodical investigation of the scratch test based on the empirical analysis of scratch experiments on more than 100 ta-C-coated specimen regarding failure mechanisms and adhesion evaluation.

The scratch test is a comparative technique in adhesion testing where a diamond indenter with spherical tip is drawn over the surface of the specimen under progressive load. The first appearance of failures such as cracks and delaminations is identified by critical loads that are used for the adhesion assessment. The scratch test is defined in the standards EN 1071-3 and ASTM C1624 and is widely used for its low requirements concerning time, equipment and test specimens [1,2]. The critical loads depend on numerous test and specimen parameters like coating thickness, substrate material or indenter size [3–10], and are characteristic for a specific combination of coating and load. Only if all influences coincide, the critical loads of different specimens are comparable, which is an extensive impairment of the scratch test. The true stresses leading to failure, however, cannot be assessed. Furthermore, a careful investigation of the failure mechanisms, that are already basically schematized [7,11,12], is necessary for the correct interpretation of the results. When scratching a hard coating deposited on a ductile substrate material coating failure is mainly caused by plastic deformation of

* Corresponding author at: Winterbergstraße 28, 01277 Dresden, Germany.
E-mail addresses: martin.zawischa@iws.fraunhofer.de (M. Zawischa), stefan.makowski@iws.fraunhofer.de (S. Makowski).
[1] Winterbergstraße 28, 01277 Dresden, Germany.

http://dx.doi.org/10.1016/j.surfcoat.2016.07.109
0257-8972/

the substrate material, which is in contrast to common applications of coated surfaces.

The dimension of the cohesive and especially the adhesive damage of different coatings can vary even in case of similar critical load, coating thickness and failure mechanisms. Large-area coating removal provides the impression of an insufficient adherent coating to the observer even for high critical loads. Adhesion evaluation by means of critical loads does not consider the different adhesive failure size so far. Therefore, it is advantageous to assess objective information on the dimension of adhesive coating failure.

2. Experimental

2.1. Material

Adhesion evaluation by scratch testing was studied for ta-C coatings on steel substrates. Density, hardness, and Young's modulus of the coating material are directly related to the amount of sp^3-hybridized carbon [13]. Thus, a varying fraction of sp^3 results in different values of these properties. Adhesion was investigated for different ta-C coatings that were deposited by an industry-scale laser-induced pulsed vacuum arc technique (LaserArc™) [14].

The coating thickness was measured for each specimen with the calotte grinding method [15] and differed from 0.4 μm to 9.7 μm, with the majority in the range from 1 μm to 3 μm. Young's modulus of the coatings was determined using laser-induced surface acoustic wave measurement [16] and ranged from 200 GPa to 900 GPa, with the majority in the range from 500 GPa to 700 GPa. The hardness of the investigated coating material can be estimated as the tenth of the Young's modulus [17]. Coating thickness and hardness/Young's Modulus are not directly related to each other.

The compressive intrinsic stresses of ta-C coatings deposited by LaserArc™ were not measured on the studied steel specimens but investigated in previous studies using the Stoney method on silicon wafers coated with comparable deposition parameters. The intrinsic stresses were found to range from 3 GPa to 8 GPa correlating with the fraction of sp^3.

The substrate material was hardened chromium steel (EN 1.3505, Young's Modulus 205 GPa, hardness approx. 60 HRC) with a polished surface. Two different pretreatment plasma processes for cleaning and heating of the specimens labelled “pretreatment A” and “pretreatment B“ were used before the actual coating deposition. A chromium interlayer with a thickness of 100 nm to 150 nm was deposited as bonding layer between ta-C and steel. In total specimen from 108 different coating processes were investigated. Some specimens were lapped before the scratch test to remove growth defects which typically resulted from the deposition process. Lapping is not expected to alter the stress state of the coating due to low process temperatures. The surface topography has a detectable influence on the critical loads but less on the failure mechanisms [4]. To study their influence in detail, several data plots in this work contain both surface conditions which are indicated by diamond shaped (non-lapped) and circle shaped (lapped) labels.

The described specimens were used to study the scratch failure mechanisms in detail to achieve conclusions about scratch resistance and coating adhesion.

2.2. Scratch test

A macro scratch tester with a maximum normal load of 100 N and a tangential force sensor was used. The scratch tests in this work were carried out with a load rate of 50 N/min and a sliding speed of 5 mm/min using an orientated Rockwell-C indenter with a tip radius of 200 μm. Scratch tests using indenter with 50 μm, 100 μm and 500 μm radii were performed additionally. The indenter was subjected to high wear due to the very hard ta-C coating. Therefore, the indenter wear was checked every two scratches by observation of an imprint of the tip in a soft material. That way, cracking and spalling of the tip could be detected but not the gradual flattening of the tip. The latter has an influence on contact pressure and therefore the dimension of critical load, but only a negligible influence on the appearance of the failure mechanisms. The critical loads were determined by optical microscopy. The critical load L_{C2} that identifies the first adhesive failure was used for adhesion evaluation in this study according to EN 1071-3. From a large number of tests it was found that coating failure and critical load were satisfyingly reproducible in case of an intact indenter (L_{C2} standard deviation below 1.5 N). But even minor spalls on the indenter influenced the failure appearance considerably.

2.3. Calculation of the stresses

The evaluation of scratch tests was complemented by examination of the stress situation leading to coating failure. Scratch-induced stresses were calculated by the commercial software FilmDoctor by SIOMEC [18]. This software applies analytical calculation of the ideal elastic state of stress at the contact region of indenter and specimen that is based on the extended Hertzian approach for coatings [19]. The calculation requires properties of coating and substrate material (coating thickness, elastic constants and yield strength) and the loading conditions as input values. These parameters are shown in Table 1. Two scenarios were used for calculation: Scenario 1 describes a typical contact situation for the scratch tests in this study. Scenario 2 considers actual specimen properties and results from scratch tests. The load situation in the moment of adhesive failure was described by the experimentally obtained critical load L_{C2} and the tangential force at L_{C2}. By this manner, the shear stresses at the interface coating/substrate which are decisive for the adhesive failure according to crack opening mode II and III (fracture mechanics) were determined. Within the evaluations performed here however, only elastic properties have been taken into account. It could be shown that this simplification did not matter with respect to the reproducibility of the results, which probably was, because of the insignificance of plasticity within our tests (in relation to the purely elastic stress and strain contributions). Therefore, the calculated stresses in the contact region are probably not consistent with the real stresses.

Preliminary calculations with the FilmDoctor iStress module [18], a system to design instrinsic stresses against external or internal loading situations, showed that intrinsic coating stresses cause very small shear stresses at an ideal interface. In that case, their influence on the interfacial shear stress situation in a scratch test is negligible. However, at discontinuities in the interface (i.e. detachment, crack) intrinsic stresses appreciably influence the shear stress situation. As it is not practicable to determine the real interfacial structure of a specimen that is possibly responsible for scratch failure, the intrinsic stresses were not taken into account for further calculations.

The Cartesian coordinate system for the calculation and all illustrations in this article is defined as follows: The abscissa (x) indicates the scratch direction; the ordinate (y) is perpendicular to it in the surface plane. The applicate (z) is perpendicular to the surface plane and points

Table 1
Input parameters for the calculation of the stress situation.

		Scenario 1	Scenario 2
ta-C coating	Young's modulus E	650 GPa	E (coating)
	Poisson coefficient ν	0.20	0.20
	Thickness t	2.0 μm	t (coating)
Cr-interlayer	Young's modulus E	248 GPa	248 GPa
	Poisson coefficient ν	0.21	0.21
	Thickness t	0.1 μm	0.1 μm
Steel substrate	Young's modulus E	205 GPa	205 GPa
	Poisson coefficient ν	0.29	0.29
Load	Normal load F_N	20 N	L_{C2}
	Tangential load F_T	0.15*FN	F_T at L_{C2}
	Indenter radius R	200 μm	200 μm

Please cite this article as: M. Zawischa, et al., Surf. Coat. Technol. (2016), http://dx.doi.org/10.1016/j.surfcoat.2016.07.109