EFFECT OF CONTACT GEOMETRY ON THE FAILURE MODES OF THIN COATINGS IN THE SCRATCH ADHESION TEST

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SUMMARY
To use scratching for assessing adhesion of a hard, thin coating to its substrate, adhesive failure must be confirmed since many coatings fail by other modes in this test. This study shows, theoretically and experimentally, that indenter-induced bending stress is very likely to cause cohesive failures. To induce adhesive failure but suppress cohesive failure in the scratch adhesion test, the compressive coating stress should be high and the bending induced stress low. This can be achieved by using an indenter with large radius and a high normal load. To assess coatings with good adhesion to the substrate, the radius of the most commonly used Rockwell C indenter may not be large enough.

Keywords: Scratch adhesion test; Thin coatings; Adhesive failure; Cohesive failure; Coating stresses

1 INTRODUCTION
The scratch test is the most practical method of assessing the adhesion of a hard, thin coating to a substrate [1-3] as it is reliable, simple to use and no special specimen shape or preparation is required. Adhesion is measured by drawing a spherically tipped diamond indenter over the coated surface under increasing normal load until a critical value is reached at which coating failure occurs. Provided the failure is adhesive, this critical normal load is taken as a measure of the coating-substrate adhesion, or, the work of adhesion is derived from the critical normal load [4-7].

During scratch adhesion testing, a compressive stress field is induced ahead of the indenter. When the mean compressive stress over an area in the coating exceeds a critical value, the coating detaches from the substrate to lower the elastic energy stored in the coating [8]. The work of adhesion at the coating-substrate interface is equal to the energy release rate from coating detachment and this rate is a function of the mean compressive coating stress over the detaching area at the instant of detachment. Thus, the critical mean compressive coating stress responsible for the detachment is a measure of coating-substrate adhesion.

Thin, hard coatings that are assessed by the scratch adhesion test are usually brittle. While a coating can withstand the compressive stress induced by the indenter to a certain extent, it may fracture if a high tensile or shear stress field is induced simultaneously. Therefore, a range of non-adhesive failure modes may occur before, or along with, adhesive failure during testing [9, 10]. If the scratch test is to be used for the assessment of coating-substrate adhesion then it is an adhesive failure that must be induced with no interference from any non-adhesive failure. The failure modes in the scratch adhesion test depend on many factors. Of these, the geometry of the scratch indenter is critical. Despite the widespread use of the scratch adhesion test, the correlation between failure modes and the indenter geometry is still poorly understood.

This study aims to better understand the effects of indenter geometry on the failure modes, so that proper scratch parameters can be chosen to ensure an adhesive failure is induced in the scratch adhesion test.

2 STRESSES IN A COATING INDUCED BY A SCRATCH INDENTER

2.1 Compressive stress in coating
When a hard indenter scratches a good commercial coating, it detaches if the compressive stress ahead of the indenter reaches a critical value [9]. Our earlier analysis showed that the mean compressive stress over a semi-annulus area ahead of the scratch indenter (with width of L as shown in Fig. 1), mc, can be expressed as [11]

\[
\sigma_{mc} = K_1 H_f^{0.5} E_f^{0.3} E^{0.2} \frac{a}{R} \]

(1a)

where K1 is a constant, Hf is the hardness of the coating (film), Ef is the Young’s modulus of the coating, E is the Young’s modulus of the substrate, a is the contact radius and R is the indenter radius. The value of K1 depends on the ratio of R to L and L is the width of the detachment, which is usually at least 20 times the coating thickness [8]. Our recent calculation found that K1 is approximately proportional to (R/L)0.7. Therefore,

\[
\sigma_{mc} = K_2 H_f^{0.5} E_f^{0.3} E^{0.2} \left( \frac{a}{L} \right)^{0.7} \frac{a}{R} \]

(1b)

where K2 is a constant independent of deformation geometry.

2.2 Bending-induced stresses in coating
As well as the compressive stress field, several others are also induced in the coating by the scratch indenter and may cause coating failure. These include the tensile stress field below the contact, the tensile stress field at the contact trailing edge, the residual tensile stress field below the surface as the contact recovers its elastic component, and the stress fields at the scratch ridges due to bending of the coating. Many researchers [e.g. 9, 12] have studied the first three stress fields and the failure modes they cause can be distinguished from the adhesive failure induced by the compressive stress field. However, the bending-induced stress fields during scratching have not been studied before.
As shown in Fig. 2, because of the scratch indenter induced displacement of material, the coating is subjected to a bending deformation. Bending-induced stress-strain fields in the coating are very complicated, however, the strains can be estimated from the local surface curvature using beam theory. The bending-induced normal strains in the coating are related to the radius of local surface curvature, $r$, by [13]

$$\varepsilon = \varepsilon_{\text{max}} \frac{z}{t}$$  \hspace{1cm} (2a)

with

$$\varepsilon_{\text{max}} = \frac{t}{r}$$  \hspace{1cm} (2b)

being the normal strain at the surface ($z = t$, tensile) or at the interface ($z = -t$, compressive) and $2t$ is the coating thickness. In turn, $r$ is related to the surface profile of the coating, $z(x)$, by [14]

$$r = \left(1 + \frac{dz}{dx} \right)^{3/2} \frac{d^2z}{dx^2}$$  \hspace{1cm} (3)

In addition, there are bending-induced shear strains which are related to $r$ and its variation with spatial location, $x$, along the coating, given by [13]

$$\gamma = \gamma_{\text{max}} \left(1 - \frac{z}{t} \right)^2$$  \hspace{1cm} (4a)

with

$$\gamma_{\text{max}} = \frac{t}{r} \left(1 - \nu \right) \frac{dr}{dx}$$  \hspace{1cm} (4b)

being the strain at the neutral plane ($z = 0$), where $\nu$ is the Poisson’s ratio of the coating.

The bending-induced normal stresses, $\sigma$, are thus

$$\sigma = E_f \varepsilon = E_f \frac{z}{r}$$  \hspace{1cm} (5)

when the stresses do not exceed the elastic limit of the coating, where $E_f$ is the Young’s modulus of the coating.

The bending-induced shear stresses, $\tau$, are

$$\tau = G_f \gamma = \frac{E_f}{2(1+\nu_f)} \gamma = \frac{E_f}{2(1+\nu_f)} \left(\frac{1}{r^2} - \frac{z^3}{r^2} \right) \frac{dr}{dx}$$  \hspace{1cm} (6)

when the deformation is elastic, where $G_f$ is the shear modulus of the coating.

Consequently, by measuring the shape of the deformed surface $z(x)$, the bending-induced normal and shear stresses can be estimated.

According to Eqs. 1, 5 and 6, the stresses in a coating resulting from scratching are determined by the mechanical properties of the coated system ($H_f$, $E_f$ and $E$), the contact geometry ($a/R$) and the deformed surface geometry ($r$ and $dr/dx$). The stress fields in the coating will then determine its failure mode. Adhesive failure results when a critical compressive stress is reached in front of the indenter. Cohesive failure occurs when the stress intensity within the coating exceeds a critical value. Although adhesive and cohesive failures may appear to occur simultaneously, it is the mode that reaches the critical stress first that initiates the failure. Therefore, the contact geometry and the deformed surface geometry determine the failure mode in the scratch adhesion test. In the present study, we tried to find by experiment the relationships between the failure modes, the contact geometry and the deformed surface geometry.

## 3 EXPERIMENTAL

### 3.1 Specimens

Experiments were carried out on two coated systems: (1) a 2.3 $\mu$m thick, multilayered (10 CrN layers and 10 Cr interlayers) CrN/Cr coating, deposited by a commercial PVD process, on 304 stainless steel; (2) a 2.8 $\mu$m thick TiN coating deposited by unbalanced magnetron sputtering onto a specialty alloy steel (Uddelholm "Chipper Steel") used for wood chipper blades, that was hard and tough after quenching and tempering. The hardness and elastic modulus of the coatings and the elastic modulus of the substrates were measured by depth sensing indentation using a Berkovich diamond indenter at a peak load of 10 mN. The hardness of the substrates was determined by Vickers microhardness testing at 2.94 N. The measured values are listed in Table 1.

<table>
<thead>
<tr>
<th>Material</th>
<th>$H$, GPa</th>
<th>$E/(1-\nu^2)$, GPa</th>
</tr>
</thead>
<tbody>
<tr>
<td>CrN/Cr</td>
<td>23.8</td>
<td>274</td>
</tr>
<tr>
<td>TiN</td>
<td>21.4</td>
<td>320</td>
</tr>
<tr>
<td>Stainless steel</td>
<td>3.2</td>
<td>209</td>
</tr>
<tr>
<td>Blade steel</td>
<td>7.2</td>
<td>246</td>
</tr>
</tbody>
</table>

*Table 1: Measured hardness and plain strain modulus*

### 3.2 Scratch testing

Scratch testing was carried out using a commercial scratch tester (CSEM Revetest). Spherical diamond indenters with cone angle of 120° and tip radii $R$ of 50, 200 and 500 $\mu$m were used. During each test, the normal load was increased from zero until severe coating failure occurred. Scratching was carried out at 3.2 mm/min. The loading rate was 32 N/min. when using 50 and 200 $\mu$m indenters, and 200 N/min. when using the 500 $\mu$m indenter. Both specimen surfaces and indenter tip were cleaned with ethanol before each scratch test.

In most tests, the critical normal load, $P_c$, at which coating failure initiated was detected by a sudden increase in acoustic emission and confirmed by microscopic examination. When the acoustic emission increase was not significant enough, the value of $P_c$ was determined by microscopic examination alone. The critical normal load values, along with the measured values of the corresponding critical contact radii, are given in Table 2.

Figs. 3 and 4 show the failure modes of the two coatings produced by different radius indenters. With the 50 $\mu$m indenter, the failure mode was mainly cohesive spallation along the scratch ridges. The 200 $\mu$m indenter caused failure by ridge spallation, a mixture of cohesive failure and adhesive failure. With the 500 $\mu$m radius indenter, the failure occurred by large scale buckling ahead of the scratch indenter, a typical adhesive failure.
profile range from a few microns from the ridge top. Therefore, we used the side ridge and the maximum variation of the radius occurs when the value of 1/r_{amin} and (1/r^2 × dr/dx)_{max} versus a/R for the coated specimens. Therefore, from these few results at small a/R values and using the same equation forms obtained from uncoated specimens, similar relationships between 1/r_{amin} and a/R, and between (1/r^2 × dr/dx)_{max} and (a/R)^{3} for the coated specimens, are also shown in Figs. 6 and 7.

Table 2: Critical normal load and contact radius in scratch test

<table>
<thead>
<tr>
<th>Coating</th>
<th>R_{a} μm</th>
<th>P_{c} N</th>
<th>a_{c} μm</th>
<th>a_{c}/R</th>
</tr>
</thead>
<tbody>
<tr>
<td>CrN/Cr on stainless steel</td>
<td>50</td>
<td>~ 1</td>
<td>14</td>
<td>0.28</td>
</tr>
<tr>
<td></td>
<td>200</td>
<td>20</td>
<td>58</td>
<td>0.29</td>
</tr>
<tr>
<td></td>
<td>500</td>
<td>59</td>
<td>115</td>
<td>0.23</td>
</tr>
<tr>
<td>TiN on blade steel</td>
<td>50</td>
<td>~ 2</td>
<td>15</td>
<td>0.30</td>
</tr>
<tr>
<td></td>
<td>200</td>
<td>43</td>
<td>62</td>
<td>0.31</td>
</tr>
<tr>
<td></td>
<td>500</td>
<td>153</td>
<td>111</td>
<td>0.22</td>
</tr>
</tbody>
</table>

For comparison of the deformed geometries scratch testing was also carried out on the substrate materials using the same indents and conditions as for the coated surfaces. No fracture was observed on the scratched solid materials surfaces. The deformed surface geometry of all scratched specimens was measured by a WYKO NT 2000 optical surface imaging system. The contact width 2a, scratch depth below original surface h and the total scratch depth h_{t} at some normal loads were measured. The deformation geometry of coated surfaces were also measured after scratching at loads below the critical load for coating failure since a failed coating might alter the surface geometry. The values of the scratch strain, a/R, and the extent of “piling-up” caused by scratching, h_{c}/h, were then calculated.

5 DISCUSSION

Using theoretical equations 5 and 6, the empirical equations in Figs. 6 and 7, and the measured mechanical elastic properties of the coatings (Table 1), we obtain the relationship between the bending-induced normal stress and shear stress and the contact geometry a/R.

For the CrN/Cr coating on stainless steel, before reaching the coating elastic limit, the bending-induced mean normal and shear stresses are (assuming Poisson’s ratio ~ 0.3):

\[
\sigma_{mb} = \frac{1}{2} \int_0^l \sigma \, dz = 17.2 \frac{a}{R} \text{ (GPa)} \tag{7a}
\]

\[
\tau_{mb} = \frac{1}{2} \int_0^l \tau \, dz = 1.18 \left( \frac{a}{R} \right)^3 \text{ (GPa)} \tag{7b}
\]

For the TiN coating on blade steel, the mean stresses are

\[
\sigma_{mb} = \frac{1}{2} \int_0^l \sigma \, dz = 29.3 \frac{a}{R} \text{ (GPa)} \tag{8a}
\]

\[
\tau_{mb} = \frac{1}{2} \int_0^l \tau \, dz = 2.46 \left( \frac{a}{R} \right)^3 \text{ (GPa)} \tag{8b}
\]

The bending-induced shear stress in both coatings is very small, thus, it is unlikely to cause coating failure. However, when the value of a/R is not so small, the normal stress can be sufficiently high to cause cracking. Hard coatings usually have fracture toughness, K_{f}, values less than 10 MPa m^{1/2} [15, 16]. If we express fracture toughness in the form of stress intensity factor [17]

\[
K_{f} = \sigma \sqrt{ab} \tag{9}
\]

and assume that the critical size for cracking, b, is equal to the coating thickness, the critical a/R value to cause coating fracture < 0.22 for the CrN/Cr coating on the stainless steel and < 0.12 for the TiN coating on the blade steel. These values are very likely to be exceeded in a scratch adhesion test. For example, when scratching a thin coating deposited on a steel with hardness of 7 GPa and using the geometry of a Rockwell C indenter (120° cone angle and 200 μm tip radius), the a/R value will exceed 0.22 when the normal load is more than about 25 N.

Coatings today usually have good adhesion to their substrates. To cause failure in the scratch adhesion testing of such coatings, it is a common to use an increasing normal load at a certain indenter geometry, or, to use a smaller radius indenter. According to the above analysis, this approach is very likely to cause coating cracking before adhesive failure takes place. This may be the reason why pure adhesive failure is seldom observed nowadays when scratching with the Rockwell C indenter. Instead, failure tends to take place cohesively, as observed by Hedenqvist and Hogmark [10].
To induce adhesive failure but suppress cohesive failure in a coated system, the compressive stress should be high and the bending-induced stress low. This can be achieved by using an indenter with large radius, and a high normal load. Since contact radius $a$ is approximately proportional to the square root of normal load, according to Eq. 1, a large indenter radius $R$ and high normal load will lead to large contact radius $a$ but small $a/R$ value; thus, large compressive stress but small bending induced stresses. Fig. 8 gives an example of the variation of the mean compressive stress $\sigma_{cm}$ with $a/R$ when using indenters with different radii, calculated by our model leading to Eq. 1 [11]. This figure shows that, when inducing a mean compressive stress of 4 GPa (over 50 $\mu$m wide area) in a coated system with the mechanical properties noted, 500 $\mu$m and 200 $\mu$m radius indenters simultaneously cause scratch strains of about 0.09 and about 0.18, respectively.

The compressive stress required to cause adhesive failure in today's coated systems can be very high. For example, the measured compressive stress for a 1 $\mu$m thick diamond coating to detach from its titanium alloy substrate was 7 GPa [18]. However, most hard coatings are quite brittle. To assess the adhesion of these coatings, the radius of the most commonly used Rockwell C indenter might be too small and, thus, an indenter with a larger tip radius should be used.

6 CONCLUSIONS

Theoretical analysis and experimental investigation have discovered that the failure mode of a thin, hard coating on a ductile substrate in the widely used scratch adhesion test is controlled by the geometry of the scratch indenter. While inducing a compressive stress field in the coating necessary for adhesive failure, the scratch indenter simultaneously induces a bending stress field in the coating, which may be high enough to cause the coating to fail cohesively before the desired adhesive failure occurs. The sharper the indenter, the more severe the bending stress field. To assess coatings with good adhesion to the substrate, the radius of the most commonly used Rockwell C indenter may not be large enough. In this case, use of an indenter with a larger tip radius is recommended so that a higher compressive stress field but a lower bending stress field can be induced.

7 ACKNOWLEDGEMENTS

Thanks are expressed to Chessen Group Inc. for providing the CrN/Cr coating, and to NRC-IAR for the TiN coating.

8 REFERENCES:


Fig. 1: Schematic illustration of the adhesive failure on a hard coating-soft substrate system caused by the compressive stress induced by a hard scratch indenter.

Fig. 2: Schematic illustration of the bending induced stresses.

Fig. 3: Failure modes when scratching the multilayered CrN/Cr coating on 304 stainless steel using spherical indenters with radius of: a) 50 µm at about 1 N load; b) 200 µm at about 20 N load; and c) 500 µm at about 200 N load. Scratch direction from left to right.

Fig. 4: Failure modes when scratching the TiN coating on the blade steel using spherical indenters with radius of: a) 50 µm at about 2 N load; b) 200 µm at about 45 N load; and c) 500 µm at about 200 N load. Scratch direction from left to right.
Fig. 5: Comparison of (a) contact width and (b) piling-up between coated and uncoated specimens.

Fig. 6: Maximum local surface curvature as function of scratch strain.

Fig. 7: \((1/r^2 \times dr/dx)_{\text{max}}\) as function of scratch strain.

Fig. 8: Calculated mean compressive coating stress over a semi-annulus area ahead of scratch indenter with width of 50 µm as function of scratch strain when using indenters with three different radii. The material properties used in the calculation were: \(E_c = 300\) GPa, \(E = 200\) GPa, \(H_c = 22.5\) GPa and \(H = 7.5\) GPa.