Identification of critical load for scratch adhesion strength of nitride-based thin films using wavelet analysis and a proposed analytical model

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Abstract

The use of Acoustic Emission (AE) signal of scratch test to evaluate the thin films’ adherence led to unavoidable large uncertainties due to the problematic determination of the frequency band and the improper installation of the acoustic sensor, especially for coatings thinner than 100 μm. In this paper, thin film-substrate adherence in high-speed steel is evaluated by Wavelet Analysis (WA) of the force-displacement curve in micro-scratch tests performed on the samples. The measured load displacement signals of deposited thin films were used as inputs for the wavelet module. We propose an analytical model combined with wavelet analysis, which is based on experimental data. Scratch adhesion strength could be identified by applying wavelet technique to the measured force displacement data to detect the location of the critical load during micro-scratch testing for TiSiN coating on high-speed steel. An analytical model is also proposed to predict the adhesion stress and to support the wavelet analysis technique where the stress is difficult to be assessed. Thin films were studied by optical microscopy, micro scratch testing machine and X-ray diffraction. The WA results are compared with model predictions in order to establish indication of scratch adhesion strength aimed at improving the manufacturing process.

1. Introduction

A nano-composite TiSiN coating is a combination of a hard transition metal nitride (TiN) nano-crystal and an amorphous interfacial phase of Si₃N₄ [1,2]. It has attractive mechanical properties such as high hardness, oxidation resistance and adhesion strength [3–7] that is used in cutting tool, among other things [8]. In an application where the process involves sliding contact between at least two materials, scratch test is a practical way to evaluate the material performance and mechanical behavior of the system. The scratch testing can be done by applying certain loads upon an indenter, which is moved horizontally, or vertically onto the coated surface until a coating failure occurs. Failure of thin coating under scratch is usually triggered by initiation and propagation of cracks on the coating substrate interface and consequent flaking of the coating material from the substrate [9,10].

Some computational models have been used including Finite Element Analysis (FEA), Molecular Dynamics Analysis (MDA) and Smoothed Particle Hydrodynamics (SPH) to monitor damage initiation and crack propagation during scratch testing [11–13]. For example, FEA simulation technique was utilized to study displacement and stresses during scratch but has differed 38% with the experimental data [11]. Meanwhile, MDA and SPH are also insufficient to meet requirement for the application in practical engineering problems [12,14].

Acoustic emission wavelet analysis—on the other hand seems to be a good technique for assessing the coating adherence failure at phase interfaces [15]. However, this technique does not use the measured force-displacement of the scratch test. Rather it needs an acceleration sensor to be fixed at proper distance away from the starch indenter to detect the elastic wave signal. Practically, determination of the location and frequency band as well as installation of the acoustic sensor imposes unavoidable large uncertainties of the obtained results especially for coatings lower than 100 μm. According to the current literature, it is difficult to have an analytical model which includes the effects of three types of friction i.e. sliding, adhesion and ploughing friction in one model for scratch adhesion strength.

In another word, there is no comprehensive analytical model for scratch test that interprets the nonlinear relationship between friction components, and interaction between coating thickness and surface roughness with adhesion strengths. That’s why our proposed wavelet analysis uses real experimental data, which includes all effects of the
interacting parameters in order to investigate the combined effects of
the interaction between film thickness, surface roughness, indenter
and friction.

In this manuscript, to avoid large errors from acoustic sensors
especially for those coatings lower than 5 μm, Wavelet Analysis (WA)
was done on the measured force–displacement signals. In other
words, the detection of the critical load location during micro-scratch
testing for TiSiN coating on high speed steel is now depending on
more accurate values.

The measured load–displacement signals of deposited TiSiN coating
are used as inputs for WA instead of the acoustic signal that suffers
from unavoidable large errors. In addition, an analytical model will be
developed to predict coating adhesion strength where it is difficult to
access or get information from WA.

2. Sample preparation and characterization

2.1. Deposition process

The TiSiN thin films were coated on high speed steel (HSS-AISI
(M3:2)) substrates using a multi-target magnetron sputtering
physical vapor deposition (PVD) of high purity Ti and Si targets
which coupled to the DC and RF power source, respectively. Both targets
were fixed above the substrate holder at about 125 mm distance.
The substrates, [2 × 2 × 2] cm cube, were ground and polished to a
mirror finish of surface roughness, Ra < 0.05 μm. The substrates were
ultrasonically cleaned in acetone and subsequently cleaned with
distilled water and dried with nitrogen gas before placing in the PVD
sputtering chamber.

The experiments were conducted by varying four parameters
i.e. the RF power, DC power, N2/Ar gas flow rate ratio and deposition
time. The deposition process was initiated by evacuating the cham-
ber to 2.67 mPa. Then argon gas was purged into the chamber to fa-
cilitate plasma formation for etching possible oxides/contaminations
from the targets. The top surface of the substrates was also etched
by applying negative substrate bias of 100–150 V. A Ti interlayer
deposition was deposited which then followed by TiSiN thin
film coating. The deposition of TiSiN was initiated by purging in N2
gas into the chamber. The working pressure was maintained
in the range of [0.93: 1.19] Pa during the coating process. The
substrates were taken out after coating when the temperature in
the chamber was at room temperature and then kept in a dry box
for further investigation.

2.2. Material characterization

The microstructure analysis of the deposited thin film was carried
out using a field emission scanning electron microscopy (FESEM)
model (Zeiss Auriga). Typical microstructure and morphology images
are illustrated in Fig. 1.

Micro-scratch testing of the TiSiN coating was performed using a
Micro Test system (Micro Materials Ltd., Wrexham, UK). A diamond
probe with a nominal radius of 25 μm was drawn across the surface of
the coating at 1.20 μm/s. Pre-processing is necessary to remove the
effect of roughness, topography, slope, and instrument bending on the
data according to [16]. A typical scratch test and failure point LC2 is
indicated in Fig. 2. Three stages in failure mode of typical PVD coating
were identified in the scratch test. LC2 is marked as ‘4’ whereby the
delamination is obviously seen in the micro image.

The scratch procedure involved three sequential scans over 1000 μm
scan distance using a multi-pass wear test mode in the instrument’s
software. The three scans were (1) an initial topography scan of
2.00 mN constant load, (2) a scratch scan where the applied load was
ramped after 50 μm at 3.00 mN/s to the maximum load of 1000 mN
and (3) a final topography scan over the scratched track at 2.00 mN
load. At least three tests were carried out on each sample.

The coating thickness was measured using the Micro Test System
as a surface profile meter scanning with a minimum load of 2.00 mN
(sufficiently low that no wear occurs at this load) across boundaries of
the coated and uncoated regions. The uncoated region was exposed
when the tape-covering portion of top surface of substrate was
removed. The difference in the step height between these regions
provides the coating thickness. An average value from at least three
measurements was taken on each sample.

The crystal structure of the deposited TiSiN coating was character-
ized by an X-ray diffraction pattern (XRD) model (Siemen D500) with
Cu Kα (1.54 A) radiation in a [θ:2θ] scan mode. The XRD with initial
angle of 2θ = 10–100°, a step size of Δθ = 0.10°, and step time of Δt =
3 s. The XRD pattern is plotted in Fig. 3. The macrostrain was determined
using XRD line broadening analysis by employing an approximation
method [17] followed by Rietveld analysis [18,19] using HighScore
Plus developed by PanAnalytical company.

The strain and size analysis was performed using line profile analy-
sis. Williamson–Hall plots were prepared to quantify the broadening.
Then B*cosθ was plotted against sinθ for (111), and (022) correspond-
ing to 2θ = 36.8, and 61.96°, respectively. The information on how to
calculate the macrostrain are extensively discussed and explained in

Fig. 1. FESEM micrographs of a typical sample.
open literature [20–22]. The average microstrains and size were determined too. The microstrain analysis based on selected XRD peaks is 0.4%.

Nanoindentation test [19,23] was performed using a Micro Materials Nano test. Indentation to 1 m-Newton (mN) was carried out at room temperature with a Berkovich indenter at 0.05 mN/s, and 5 s hold period at peak load. Ten indentations were investigated on each thin film. It is reported in literature that thin films grown by physical vapor deposition coating methods are rather stressed. The residual stress in the films can be either tensile or compressive this is initiated by the non-equilibrium microstructure formations. These behaviors are mainly associated with the high-energy plasma particle bombardment. It is stated that elastic energy is stored in the films as a result of the stress, and once the stored energy is larger than the adhesive energy, the condensate films would peel off. The peeling of the deposited film from its substrate is an indicator of the critical film thickness [17].

The adhesion data was obtained in terms of critical load, \( L_{c2} \), in the scratch test. The critical load was recorded based on the scratch profile of the load–displacement graph. It was verified with an optical image of the scratch track since it was discovered that some failure modes may occur without a noticeable change in the scratch profile. Fig. 2 shows an example of a scratch profile of TiSiN coatings along with its optical image of the scratch track. The onset of failure as denoted by \( L_{c2} \) is indicated by the first spike on the scratch graph. \( L_{c2} \) indicates the point of adhesion failure and signifies the interfacial adhesion strength in this coating/substrate system and denoted as the scratch adhesion strength [16].

3. Wavelet analysis and analytical model

3.1. Proposed model for adhesion stress

Previous models presented in [24–29] do not take into consideration the effects of substrate elastic modulus and the difference between the thermal expansion coefficients of thin films and substrates which play significant roles on the residual and interfacial adhesion stresses [9,10]. These models are considered and the critical load \((L_{c2})\) can be simply written as in Eq. (1),

\[
L_{c2} = \frac{t_c \alpha_d}{\mu_c} \quad \text{(1)}
\]

where \( \sigma \) represents the interfacial adhesion stress; \( t_c \) is the coating thickness, \( \Delta \) is the track width, \( \nu \) is the Poisson’s ratio and \( \mu_c \) is the coefficient of friction between indenter and the coating.

From the theory of thermal stress [9] the interfacial forces due thermal bending of bimetallic plate at free loading conditions are given by Eq. (2) as;

\[
P = \frac{2(E_l I_s - E_s I_c) (\alpha_c - \alpha_s) \Delta T}{(t_s + t_c) \left( \frac{t_c + t_s}{2} + \frac{2(E_l I_s + E_s I_c)}{t_s + t_c} \left( \frac{1}{E_l I_s L_s} + \frac{1}{E_s I_c L_c} \right) \right)} \quad \text{(2)}
\]

where \( \alpha_l \), \( \alpha_s \) and \( E_l \), \( E_s \) are the thermal expansion coefficients and Young’s moduli of the film and substrate, respectively. While \( t_s \), \( t_c \) is and \( L_s \) and \( L_c \) are the thickness and moment of inertia of the thin and substrate respectively. \( \Delta T \) is the difference between room and deposition temperatures whereas \( L \) is the sample length. Assuming that the thermal coefficients \( \alpha_l \), \( \alpha_s \) are constant, the maximum interfacial stress acting on thin film, after the deposition process, will be generated on the contact surface of thin film and substrate. Equating Eqs. (1) and (2) the critical adhesion stress can be predicted as;

\[
\sigma = \frac{2(E_l I_s - E_s I_c) (\alpha_c - \alpha_s) \Delta T}{(t_s + t_c) \left( \frac{t_c + t_s}{2} + \frac{2(E_l I_s + E_s I_c)}{t_s + t_c} \left( \frac{1}{E_l I_s L_s} + \frac{1}{E_s I_c L_c} \right) \right)} \quad \text{(3)}
\]

This equation will be used to predict the film adhesion stress and compare the results with the XRD measurements.

3.2. Wavelet analysis for critical load

Wavelet analysis is a new signal processing method, although its mathematical underpinning is backdated to the work of Joseph Fourier in the nineteenth century [30]. The attention of researchers gradually turned from frequency-based analysis such as fast Fourier transformation (FFT) to scale–based analysis (i.e. wavelet analysis) when it started to become clear that an approach for measuring average fluctuations at different scales might prove less sensitive to noise. The purpose of this
section is to show how wavelet analysis can clearly detect a discontinuity in the load–displacement curve of the microscratch test at the positions of initial and critical loads. Although the single apparently follows a smooth curve, it actually contains a discontinuity due to the sudden change of the tangent modulus from elastic to plastic state. The discontinuity might appear at the first or the second derivative of the load displacement curve. The un-noisy component of the signal may undergo abrupt changes shown as a sharp change in the first or second derivative. The main characteristic of a critical load is that the change in load is localized in time or in its corresponding scratch distance. Therefore, the purpose of using the wavelet analysis is to determine the site of the change (e.g. position), the abrupt change in wavelet’s first or second derivative, and the amplitude of the change of the load signal. Some researchers claim that short wavelets are often more effective than long ones in detecting a signal abrupt change. In the initial analysis scales, the support is small enough to allow fine analysis [31]. The hopes of discontinuities that can be identified by the smallest wavelets are simpler than those that can be identified by the longest wavelets.

In this work, we followed an empirical approach to determine the most suitable type of wavelet used to analyze the load–displacement signal of scratch test. Accordingly, to identify a signal discontinuity (which means that the initiation of the crack at critical load (Lc2) Fig. 2), different types of wavelet have been tested in order to choose the best one. Namely; Biorthogonal wavelet filter (Bior), Reverse Biorthogonal spline wavelet filter (Rbio), Symlet wavelet filter (Sym), Daubechies wavelet filter computation (Db), Coiflet wavelet filter (Coif), DMeyer wavelet filter (Dmey) and the Harr wavelet filter (haar) were implemented. The presence of noise, which is after all a
where, the coefficients $C_{jk}$ summarize with no redundancy the entire signal content and $\psi_k(x)$ is the used wavelet filter.

Critical load ($L_{c2}$), and scratch adhesion strength of nitride-based thin films during scratch test was determined by wavelet signal analysis and compared with optical microscopy results. The performance of TiSiN coatings was observed and showed different locations of critical loads corresponding to the different coating thickness. The displacements at which the critical load occurred have been identified by finding of the largest wavelet coefficients ($C_{jk}$) of the analyzed signal. By performing optical microscopy observations, the critical load was assigned to optical micro-image surface quality. Thus, by using the traditional wavelet-based, on rupture and test parameters, information about thin films critical load ($L_{c2}$) could be obtained during signal transformation. WA permits a clear detection comparing with visual observation because the transition from elastic to plastic stress is a transformation. WA was applied to force–displacement signal of the microscratch test results shown in Fig. 4. WA with different bases was employed in calculations in order to find out the best wavelet type. Dmey, Symmlet and Rbio WA coefficients $cd1$ have given very clear detection of critical load position when compared with other wavelets such as Harr, Coif1.3, rbio1.3 and Daubechies (not shown). Among the best wavelet types, Rbio was selected and the data were transformed into a graph shown in Fig. 4. The figure shows higher coefficient values that correspond to higher energy release form elastic to plastic state. These interface separations appear to be what observed in the micrographs (Fig. 2) which starts at the same location as predicted by WA. Fig. 6 shows the predicted critical load, $L_{c2}$, as calculated from Eq. (4) developed by the authors. The results reveal that the critical load increases concurrently with the thickness of thin film. This can be attributed to the proportional increase in the interfacial force as the coating thickness increases as it is depicted from the equation. The result also reveals that the critical load increases concurrently with the thickness of thin film. This can be attributed to the proportional increase in the interfacial force as the coating thickness increases as it is depicted from the equation.

WA results of with rbio1.3 wavelet as shown in Fig. 4 are clear and more accurate at the second level of transformation than those obtained by sys2 and dmey or using naked eyes. The highest values in amplitude coefficients $cd2$ of rbio1.3 WT corresponded quite well with the separation positions and critical load determined from experiment as shown in Fig. 2.

### 4.2. Adhesion stress

Fig. 5 shows the predicted adhesion stress as calculated from Eq. (3). The adhesion stress slightly decreases from approximately [from 4 to 3] GPa as the thin film thickness increases from approximately [0.7: 1.7] μm. Subsequently, it abruptly increases to 5 GPa with increase in the thickness. It is also observed that the approximated value of compressive residual stress obtained from XRD data increases from ~3.5 to ~18 GPa. Then reduced after reaching a thickness of 1.7 μm. Increasing the thickness of thin film might induce residual stress and it is opposite in trend to the adhesion stress. The adhesion stress is induced by the difference in coefficient of thermal expansion (CTE) between the TiSiN coating (i.e. CTE ranges [8.4:9.3] parts per million per kelvin) and the high speed steel substrate (i.e. CTE is 13 parts per million per kelvin). This relationship between the compressive residual stress and adhesion stress are in good agreement with Zhang et al. [32].

The previous (Fig. 6) shows the trend of critical load in relation to the thickness of thin film. The trend is simply improving the integrity of the thin film up to 1.7 μm thickness. It is significantly observed after reaching this thickness; the critical load is rapidly improved. The rapid increment can be explained by the need for the increased force of the indenter to break into the thicker thin film. It is also likely due to relaxation of residual stress [33] in the thin film structure as indicated by the downward trend in Fig. 7, after reaching 1.7 μm thickness. In view of CTE at this thickness, it has sufficiently provided better resistance towards thermal cycling, and consequently reduces the mismatch due to thermal expansion between TiSiN thin film and the substrate [34,35].

### 5. Conclusions

Previous published works on FEA, MDA, SPH and Acoustic Emission (AE) wavelet analysis do not deal directly with the measured
force–displacement of the scratch test to evaluate the thin films’ adherence. Therefore, large uncertainties due to the problematic determination of the frequency band and the improper installation of the acoustic sensor especially for thin films lower than 100 μm cannot be avoided. To overcome such problem, the measured load displacement signals of deposited thin films were used as inputs for the wavelet module. Wavelet analysis proved to be a good technique for assessing thin film critical load of nitride-based thin films at which the coating starts to deform from elastic to plastic deformation. The critical load, Lc2 and scratch adhesion strength were also predicted by the proposed analytical model and the results agreed well with those obtained by WA identification. The coating starts to deform and breakdown when it reaches the critical load Lc2 in the range of forces applied in the present work [200:580] mN. Adhesion stress, compressive residual stress and critical load improved with thickness in the range [1.7:2] μm. To conclude Wavelet Analysis is an effective approach for signal analysis since measuring average fluctuations at different scales might prove less sensitive to noise.

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