AE AND ELECTROCHEMICAL NOISE ANALYSIS FOR FRACTURE STUDY OF HARD SURFACE FILM

AKIO YONEZU, HIDEO CHO, TAKESHI OGAWA and MIKIO TAKEMOTO
Department of Mechanical Engineering, College of Science and Engineering,
Aoyama Gakuin University, 5-10-1, Fuchinobe, Sagamihara, Kanagawa, 229-8558, Japan.

Abstract

We propose a new hybrid technique of AE, corrosion potential and FEM for studying the fracture mechanism of hard surface films during micro-Vickers indentation. This method makes it possible to determine the fracture strength of the film when hard film is deposited on soft substrate. Both the AE and corrosion potential fluctuation of PVD-TiN film deposited on AISI-304 steel in hydrochloric acid (1 mol/l HCl + 0.01 mol KSCN) were simultaneously monitored during Vickers indentation test. We observed characteristic potential change in mV scale due to non-faradic reaction during loading to 20 N. Timing of the potential fluctuations coincided well with strong AE events. Both the AE and potential fluctuation were found to be produced by the Mode-I fracture of the film bending during loading. The number of equidistance step-wise terrace or rippled cracks observed in pyramidal indentation agreed well with the number of potential fluctuation. It was found that potential fluctuation analysis, jointly utilized with AE analysis, can reveal the fracture process of the film.

Keywords: Hard surface film, Indentation test, Electrochemical noise analysis, Finite element method, Fracture strength

1. Introduction

Hard surface coatings are widely used to improve the functionality of the metals. Hard films such as ceramic are brittle and often suffer damages as cracking and exfoliation. Evaluation of fracture strength of the hard thin films is important for a practical application. Though various test methods such as scratch and external loading have been utilized so far, property of the film tends to change depending on the test method and mechanical characteristics of the substrate. It is generally difficult to determine the intrinsic fracture strength of the film.

One of the authors once attempted tensile and compressive loading of the specimens with CVD-TiN film deposited on steel substrate [1, 2]. They studied the crack dynamics of the film using quantitative source inversion technique. They also used finite-element method (FEM) to determine the fracture strength of the film and residual stress [1], and revealed extremely large compressive residual stresses. External tensile loading hardly generates crack in the film due to large compressive residual stresses. Ikeda et al. [3-5] reported that external loading of WC-Co plate with CVD-diamond resulted in the unstable fracture of the substrate. This implies that the external loading method cannot be used for evaluating the intrinsic fracture strength of films.

We previously performed micro-indentation tests to TiN film deposited onto soft and ductile stainless steel substrate AISI-304 [6, 7], and monitored AE to study the fracture mechanism of the films. Fracture mode analysis [8, 9] revealed that a number of AEs by Mode-I fracture were
produced during loading. Indentation test of the hard film on the soft substrate makes the evalua-
tion of the fracture strength of the film possible, since the film tends to suffer surface Mode-I
cracks due to tensile stresses induced by film bending. It was made unclear whether the AEs
were produced by Mode-I tensile crack or friction or cracking by compressive loading. In order
to determine the fracture strength of the film, we must clarify the film fracture process and
mechanism during indentation testing.

Monitoring of corrosion potential fluctuation or electrochemical noise (EN) analysis has been
successfully utilized to study the initiation of localized corrosion and stress corrosion cracking
(SCC) [10-12]. The potential fluctuation is caused by non-faradic anodic or cathodic local cur-
cent [12]. If we use EN analysis, we can monitor the process of film cracking. Thus, we
simultaneously monitored EN and AE during indentation testing and analyzed the details of the

In this study, we first monitored AEs during indentation testing in air of TiN film deposited
onto WC-Co cermet and AISI-304 steel. Effect of hardness of the substrate on the damage char-
acteristics of the film was studied. Next we utilized AE and EN analysis during indentation test
in specially designed solution. We investigate the fracture behavior of the film. Finally, we esti-
mated the fracture strength of TiN film using FEM [6, 7].

2. Materials and Experimental Procedures

The hard films tested are TiN films of a few µm thick. The TiN films were deposited by PVD
method onto metastable austenitic stainless steel (AISI 304) and WC-Co cermet. Here, sample
with AISI-304 substrate has a middle layer of 0.5-µm thick titanium deposited with ion bomb-
bardment in order to improve the interfacial strength. These samples are designated as
TiN/Ti/AISI304 and TiN/WC-Co, and coated by hollow cathode discharge (HCD) and arc ion
discharge (AID), respectively. Film thickness and specimen size are shown in Table 1.

<table>
<thead>
<tr>
<th>Film/Substrate</th>
<th>TiN/Ti/AISI304</th>
<th>TiN/WC-Co</th>
</tr>
</thead>
<tbody>
<tr>
<td>Film thickness µm</td>
<td>4</td>
<td>2</td>
</tr>
<tr>
<td>Material shape mm</td>
<td>20 x 20 x 2</td>
<td>20 x 20 x 5</td>
</tr>
</tbody>
</table>

We performed micro-indentation test by an electromagnetic type servo-testing machine
equipped with a Vickers indenter and two eddy current sensors. This machine can measure the
indentation force (F) and indentation depth (h) or F-h curve with high resolution. Maximum in-
dentation force (F_{max}), were optionally changed depending on the purpose. All tests in air were
performed with loading rate (dF/ds) of 5 mN/s and holding time (t_v) of 10 s at room temperature.

We monitored AEs using four resonant-type small sensors (PICO: PAC). They are mounted
on the film surface to monitor the Lamb wave AEs. We also used a wideband frequency sensor
(WD: PAC), which was mounted on the bottom surface of the specimen, as shown in Fig. 1. The
WD sensor signal was set on the trigger channel. Sensor outputs were amplified to 60 dB by pre-
amplifiers (PAC), and digitized by an A/D converter (Gage Applied Inc.) and fed to a personal
computer. AEs are digitized at an interval of 40 ns with 2048 points. The threshold value for
channel 1 (trigger channel) was set at 25 µV.
We also monitored the corrosion potential fluctuation induced by film fracture in a solution. A 20-mm diameter plastic cell was glued on the specimen surface. This cell is connected to another cell with Ag/AgCl reference electrode (GS-5015C: TOA-DKK). The solution was hydrochloric acid (1 mol/l HCl + 0.01 mol KSCN) [14], which can monitor the damage occurrence sensitively. Output of the electrode was digitized by a digital voltmeter (RE6871E: ADVANTEST) at resolution of 1 µV and time interval of 76 ms. This indentation test in solution was performed with $F_{\text{max}} = 20$ N, $dF/ds = 1$ mN/s and $t_v = 60$ s at room temperature.
3. Results and Discussion

3.1 Analysis of micro-crack behavior during Vickers indentation test

Figure 2 shows the $F$-$h$ curves and cumulative AE counts. The specimen TiN/Ti/AISI304 produced a number of AEs from small force, while TiN/WC-Co produced no AEs. Figure 3 shows the typical AE waveforms detected during loading of TiN/Ti/AISI304 specimen. It is noted that polarities of first $S_0$ mode Lamb waves are all positive as indicated by arrows, indicating Mode-I opening fracture type. Open triangles in Fig. 2 indicate the AEs from Mode-I opening fracture types estimated by the fracture-type classification method [8, 9]. Most AEs detected are produced by Mode-I crack with opening vector in the direction parallel to the film surface. We detected only one AE from Mode-II crack at around 2.5 N.

Figure 4 shows the micrographs of indenter contact region. There observed a number of steps in the pyramidal indentation of the specimen TiN/Ti/AISI304. These steps are film cracks, known as the ripple cracks. This type of crack appeared to produce AEs during the loading process. In contrast, two small median cracks from the corners of indentation were observed for TiN/WC-Co.
3.2 Stress analysis of thin films subjected to Vickers indentation test

In order to study the influence of the substrate on the film damage, we analyzed stress distribution by FEM as shown in Fig. 5. Here the film thickness was set as 4 µm. The yield strength ($\sigma_{ys}$) of the film is assumed to be 7 GPa. This value was estimated by three times the Vickers hardness value [13]. Figure 6 shows stress $\sigma_x$ distribution in the direction parallel to the specimen surface for indentation depth $h$ from 2 to 10 µm. Horizontal axis ($r$) designates the distance from the center of indentation. Open triangles on the curves indicate the contact boundary. Figure 6(a) indicates that the $\sigma_x$ is compressive in the contact region, while it changes to tensile stress outside the contact region. This stress distribution, with large tensile stress higher than the tensile strength of the film, strongly indicates that crack is produced by the tensile stress induced by bending of the film. A number of Mode-I cracks, appeared as ripple crack, are produced outside the contact point with progression of indentation. In contrast to this, only the compressive $\sigma_x$ is induced for TiN/WC-Co, as shown in Fig. 6(b). The cermet substrate does not suffer plastic de
formation. Thus, the crack morphology for TiN/WC-Co specimen is similar to that observed for monolithic ceramics.

3.3 Monitoring of micro-cracks using electrochemical noise analysis

Figure 7 shows the $F-h$ curves and cumulative AE counts. 254 events were detected during the loading and 17 events during the unloading. We analyzed the fracture mode of 60 events with large amplitude of So-Lamb wave. Timing of Mode-I type AEs, with all positive polarities, were shown by open triangles near the $F-h$ curve. The Mode-I AEs were detected from small loads and produced at an almost constant rate with increasing load. This behavior is quite similar to that found in Fig. 2 for TiN/Ti/AISI304.

![Fig. 7 $F-h$ curve with cumulative AE count for TiN/Ti/AISI304 in HCl solution.](image)

![Fig. 8 Variation of potential noise with cumulative AE count during indentation test.](image)
Figure 8 shows overall changes of corrosion potential or electrochemical noise (EN) and cumulative AE count as a function of loading time. Loading history is also shown by the dashed line. The corrosion potential shifts from 300 mV vs. Ag/AgCl of TiN to active direction, with a number of RD-type potential fluctuations (rapid drop to active potential and gradual recovery). First drop was observed at a low load of $F = 2.5$ N, corresponding to the loading time of 380 s.

Shown in Fig. 9 is the potential fluctuation in the range from 303 to 296 mV. Note the position in the loading history in Fig. 8. RD type fluctuations with large and small amplitudes were frequent. Figure 10(a) shows typical RD type fluctuation, which is caused by local anode current flows due to the film fracture [12]. Figure 10(b) shows changes of cumulative counts of RD-type fluctuations with the drop velocity of above 2 mV/s, AE event counts and indentation force with time. Most ENs and AEs were detected during loading. We observed a good coincidence between the two, AE and EN. This strongly suggests that the ENs were produced by film cracks occurring outside the contact region. FEM results (Fig. 5) suggested that ripple cracks were generated by the film bending, since large bending stress occurred outside the contact region.

![Fig. 9 Potential fluctuation during the indentation testing.](image)

![Fig. 10 Typical RD-type EN detected (a) and cumulative RD-type EN and AE counts (b) during the indentation testing.](image)
Figure 11 shows an example of RD-type ENs and the timing of AEs. It is noted that an RD-type EN was detected just or slightly after an AE timing. Similar behavior was observed over the entire potential range.

Figure 12 compares SEM photographs of the indentation induced in air (b) and in solution (a). A partial film delamination was observed near the edge of pyramidal indentation in solution, suggesting the film damage by solution diffused into the film cracks. We counted the number of cracks on each pyramidal plane in Fig. 12(a). Figure 13 shows the number of cracks found in the contact region as a function of distance from the center of indentation, \( r \). Total number of cracks counted was as 28 to 35 on one pyramidal plane. The total number of cracks reaches 128 and coincides well with the cumulative RD-type ENs number of 112 shown in Fig. 10. Thus, we conclude that both AEs and RD-type ENs were produced by the film fracture of TiN.

4. Conclusion

In order to study the fracture mechanisms and strengths of PVD-TiN film, both acoustic emission (AE) and electrochemical noise (EN) were monitored simultaneously during indenta-
Fig. 13 Number of cracks as a function of the distance from the center of contact region.

Effect tests. Effects of substrate hardness on the film fracture and the fracture process of TiN film deposited on AISI-304 steel in 1 mol/l hydrochloric acid solution were analyzed in detail. Results are summarized below.

1. TiN film deposited on WC-Co cermet developed small median cracks at the corners of pyramidal indentation. TiN film on AISI-304 steel, however, sustained a number of film cracks (ripple cracks) but no median cracks. In the latter material, AEs from Mode-I film cracks were detected from low indentation loading and increased with the load. FEM analysis predicted that Mode-I film cracks was produced by large tensile stresses due to the film bending.

2. Simultaneous monitoring of AEs and ENs for the TiN film on AISI-304 steel in the solution of hydrochloric acid revealed that the number of EN coincides with that of AEs. Rapid shift of the potential to active direction (RD-type EN) is induced by the Mode-I film fracture near the indentation. EN analysis, jointly utilized with AE analysis, can reveal the fracture process of the film.

Acknowledgement

This work was performed as a part of the Center of Excellence (COE) Program, funded by the Ministry of Education, Culture, Sports, Science and Technology.

References


