Nanoscratch evaluation of adhesive strength of Cu/PI films

K. Tanaka, K. Gunji & T. Katayama
Department of Mechanical Engineering, Doshisha University, Japan

Abstract

A scratch test has provided a simple, rapid means of assessing the adhesion strengths of thin films on substrates. However, it is not standardized how to detect the interfacial failure. Copper thin films deposited on Polyimide (PI) substrates are used for Flexible Print Circuit (FPC). It is needed to improve and measure the adhesive strength of Cu films on PI substrates to avoid interfacial fractures caused by cyclic bending. In this study, in order to improve the adhesive strength, the surfaces of PI substrates were modified by ion beam using argon or oxygen with a linear ion source and the adhesion strengths of Cu films on PI substrates were measured using nanoscratch tests. Cu thin films were deposed by Physical Vapor Deposition (PVD) using an electron beam. The friction coefficient rapidly decreased and normal displacement rapidly increased at the delaminated point at the same time. The slope of the friction coefficient increased at the delaminated point, whose normal load is called the critical load. The scratched surfaces were observed from films (Cu) side and substrate (PI) side. A transparency of PI substrates allowed us to observe the interfaces between Cu film and PI substrate. The scratched mark observed from substrate side started at just the point where the slope of the friction coefficient changed. As the roughness of the surface of polyimide substrates increased, critical load increased.
Keywords: nanoscratch test, adhesive strength, copper films, polyimide substrates.

1 Introduction

There are a lot of methods to measure the adhesive strength of interfaces between coatings and underlying substrates, which include pull-off test, peel test,

WIT Transactions on Engineering Sciences, Vol 55, © 2007 WIT Press
www.witpress.com, ISSN 1743-3533 (on-line)
doi:10.2495/SECM070291

four point bending test, scratch test and indentation test. The scratch adhesion test has provided a simple, rapid means of assessing the adherence of coatings. The mechanism or stress field of scratch test have been studied by Bull et al [1] and many researchers [2-4]. Scratch test is the test by drawing a diamond indenter along the coated surface. The applied normal load is increased continuously until delamination is occurs. This critical normal load is considered as a semi-quantitative measure of the coating-substrate adhesion. There are several ways of detecting the occurrence of delamination. Mutoh et al [5] used Acoustic Emission, Shibutani et al [6] used microscopes, and Li and Bhushan [7] used the coefficient of friction to detect the delamination. Observation of scratched surface can detect the delamination with fractures of films. In order to detect the initial fracture in interfaces between films and substrates, it is very important to observe the behavior of friction coefficient. Therefore, it is necessary to clarify the relationship between fracture mechanism and the changes of friction coefficient.

Copper (Cu) films prepared on Polyimide (PI) substrates are used in electronics field, such as, for the Flexible Print Circuits. The interfacial fractures caused by cyclic bending have been a serious problem. To obtain the reliable devices, it is needed to improve and measure the adhesion strength of films on substrates.

In this paper, nanoscratch test was used to measure the adhesive strength of Cu thin films prepared on PI substrates by Physical Vapor Deposition (PVD) and the influence of modification on PI surface by ion beams using argon or oxygen with linear ion source was discussed.

2 Experimental details

2.1 Specimens

The substrates used for this study are Kapton® EN films (thickness: 50μm) manufactured by Du Pont. This film is the PMDA-ODA type polyimide, which is optimized for direct coating without adhesive. Prior to the deposition process, surfaces of the substrates were modified by ion beam modification system developed by Ektessabi et al [8]. The system using a linear ion source, which has a couple of electrodes, enables us to modify large surface area. Cu thin films were prepared on as-received and ion-irradiated surfaces of PI substrates by PVD, whose thicknesses were set for 170μm and 350μm. Oxygen and Argon ions were irradiated to the surfaces of PI substrates. The accelerating voltage was between 750V to 1500V. The roughness of as-received and ion-irradiated surfaces was measured by Atomic Force Microscope (AFM, Veeco Instruments, NanoScope3). The sample names, ion accelerating voltage, names of irradiated ions and the RMS roughness of the surfaces of PI substrates are shown in table 1.

2.2 Nanoscratch test

The adhesive strengths of Cu films on PI substrates were measured using nanoscratch tests. The nanoscratch tests were performed by a Triboindenter®

(Hysitron Inc.). A triangular pyramid diamond tip of 90° angles was used for scratch tests in the face forward direction and edge forward direction as shown in Figure 1.

Table 1: Sample name, ion accelerating voltage, ion, RMS roughness.

Sample Name	Ion Accelerating Voltage (V)	Ion	RMS Roughness (nm)
As received	-	-	1.6
Ar1500	1500	Argon	1.3
Ar750	750	Argon	2.6
O1500	1500	Oxygen	0.7
O1250	1250	Oxygen	0.9
O1000	1000	Oxygen	1.2
O750	750	Oxygen	2.8

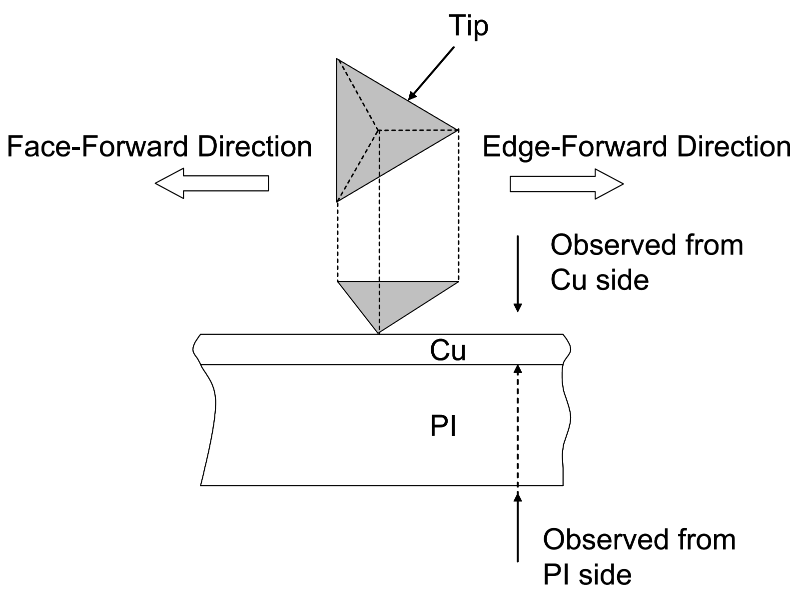

Figure 1: Nanoscratch direction and observe side.

The film surfaces of 170μm in thickness were scratched in the face forward direction (which will be called ‘thin-face test’) and edge forward direction (thin-edge test) at a scratch speed of 0.1μm/s. The film surfaces of 350μm in thickness were scratched in the edge forward direction at the same speed (thick-edge test). The applied normal load was increased continuously from 0μN to 1000μN for thin-face test and thin-edge test, and 0μN to 700μN for thick-edge test. The

 © 2007 WIT Press
www.witpress.com, ISSN 1743-3533 (on-line)

scratched surfaces were observed with Scanning Electron Microscope (SEM) and optical microscope from both film and substrate side to observe the interface between Cu films and PI substrates as shown in Figure 1.

3 Results

Figures 2 and 3 show typical scratched surfaces observed by SEM and the relationship between the friction coefficient and normal load for thin-face test and thin-edge test. In the initial stage, the friction coefficient remained constant with some amplitude of vibration (stage 1). After that, the slope of friction coefficient changed and the friction coefficient increased linearly (stage 2). Then the slope of friction coefficient changed to smaller value than that of stage 2 (stage 3). Finally, the friction coefficient rapidly increased (stage 4) in the thin-face test. From the SEM observation, neither cracks nor fracture occurred during stage 1 and stage 2. Then the crack initiated during stage 3. At the beginning of stage 4, the indentation tip reached to the PI substrate and the Cu film peered off the substrate in the thin-face test.

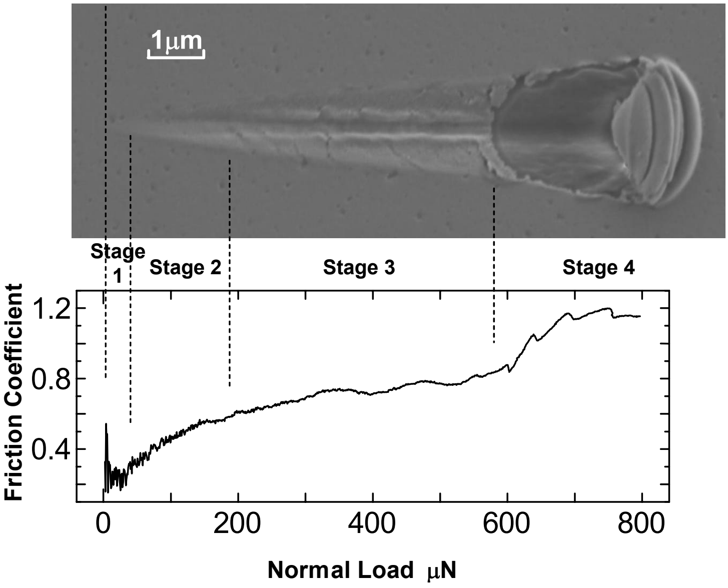

Figure 2: Typical SEM image and relationship between normal load and friction coefficient of thin-face test (as-received).

Figure 4 shows a typical result for thick edge test. The observed photos by optical microscope from Cu and PI side are also shown in Figure 4. In the initial stage, the friction coefficient remained constant with some amplitude of vibration until the normal load reached about 250μN (stage 1) and dropped suddenly. After that, the slope of friction coefficient changed and the friction coefficient increased linearly (stage 2). From the SEM observation, neither cracks nor fracture occurred during stage 1 and stage 2. The scratch damage observed from PI side occurred at the beginning of stage 2. Figure 5 shows another result for thick edge test. For this specimen, sudden drop of the friction coefficient between stage 1 and 2 was not clear, but after stage 1, the slope of friction coefficient changed and the friction coefficient increased linearly (stage 2), showing the same behaviour described for Figure 4.

4 Discussions

Schematic drawings of four stages described above are shown in Figure 6. For all the tests during both stage 1 and stage 2, no cracks or no fractures of films were observed. The scratch damage observed from PI side with optical microscope started at the beginning of stage 2. The observation from PI side allowed us to observe the interface between films and substrates because of a transparency of PI substrates. This indicates that the change of friction coefficient between stage 1 and stage 2 shows the occurrence of interfacial fractures. For thin-face test and thin-edge test, cracks of films were observed during stage 3. It is indicated that this cracks released energy and had the slope of friction coefficient decreased. For thin-face test, fractures of films and interfacial fractures between Cu films and PI substrates were observed in stage 4. For thin-edge test, there was no boundary between stage 3 and stage 4 in the relationship between normal load and friction coefficient. This is because the edge of the tip cleaved the PI substrates for thin edge test. On the contrary, for thin edge test, the film fractured in buckled in stage 4.

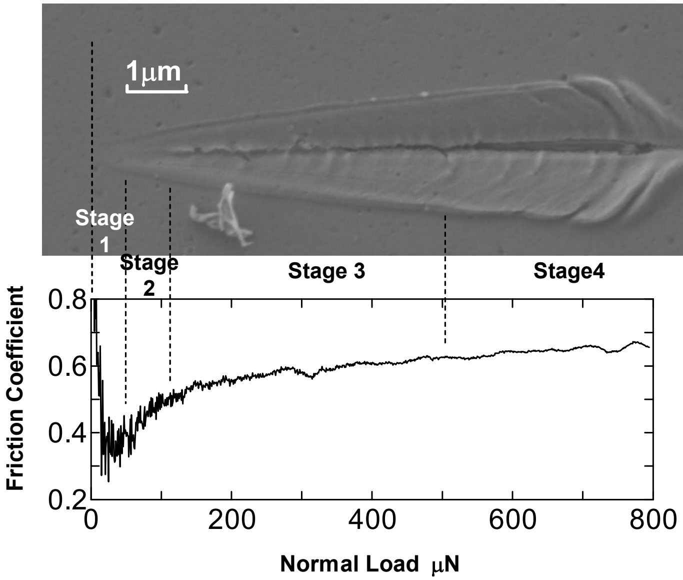

Figure 3: Typical SEM image and relationship between normal load and friction coefficient of thin-edge test (as-received).

 © 2007 WIT Press
www.witpress.com, ISSN 1743-3533 (on-line)

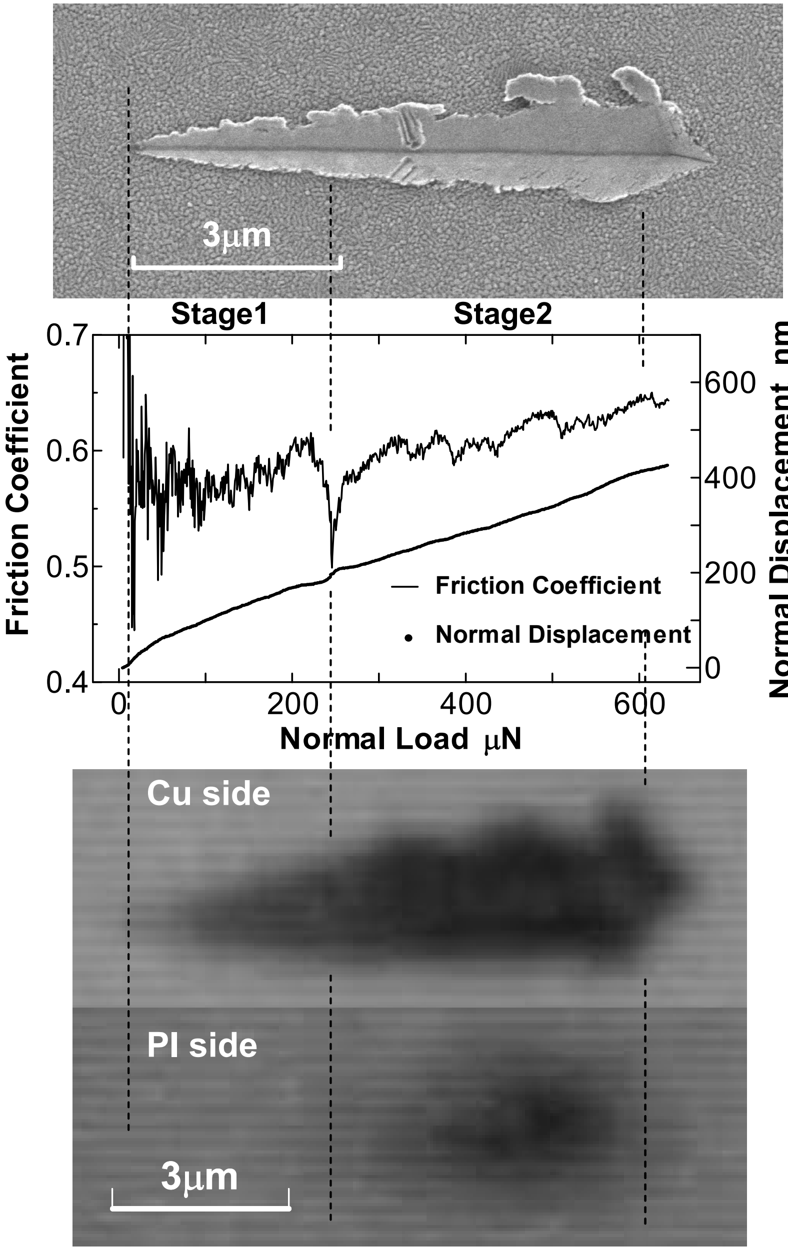

Figure 4: Typical SEM image, relationship between normal load, normal displacement and friction coefficient, and optical micrographs from Cu and PI side of thick-edge test (as-received).

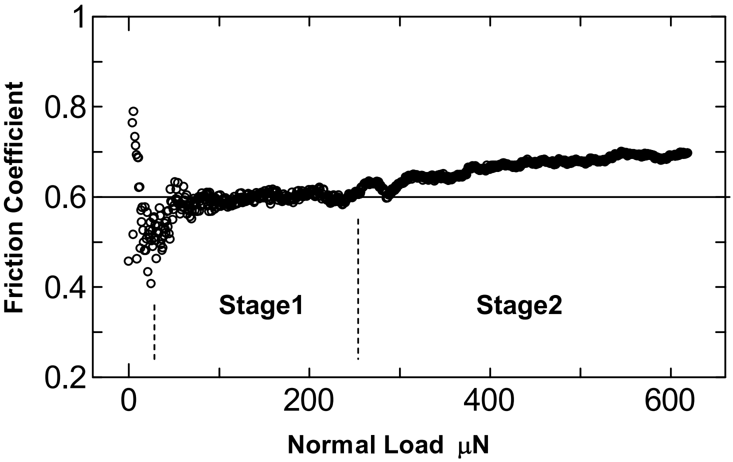

Figure 5: Typical relationship between normal load and friction coefficient.

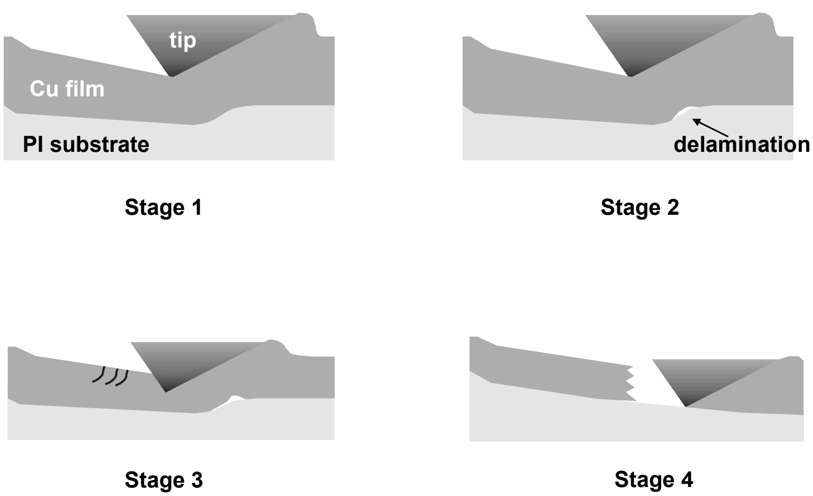

Figure 6: Schematic drawings of four stages.

Considering the discussion above, the normal load at the end of stage 1 can be considered as a critical normal load, a semi-quantitative measure of the film-substrate adhesion. Figure 7 shows the relationship between the critical normal loads and the RMS roughness of un-modified and modified PI substrates measured by thick edge test. The critical normal loads correlate with the RMS roughness of PI substrates. This indicates that the adhesive strength between films and substrates correlate with the roughness of the substrates due to anchor effects. The correlation between the roughness of the surface of the substrates and the adhesive strength was also indicated by Ge et al [9].

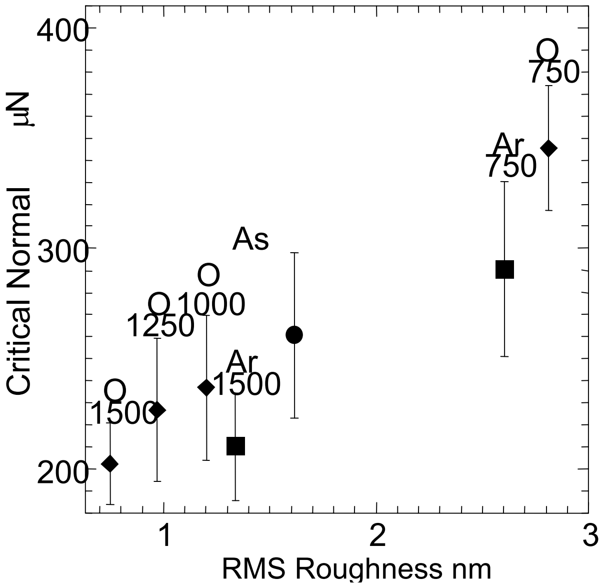

Figure 7: Relationship between roughness of the substrates and critical normal loads.

5 Conclusions

By analyzing the scratch processes of Cu films on PI substrates, it was found that four stages occur as the normal load increases during the scratch test; plastic deformation of Cu films (stage 1), interfacial fractures without fractures of films (stage 2), cracks in films (stage 3), fractures of films and delamination caused by the tip reaching to the PI substrates (stage 4). In the first stage, the friction coefficient remained constant. In the second stage, the slope of friction coefficient increased and the friction coefficient increased linearly. In the third stage, the slope of friction coefficient changed to smaller value than that of the second stage. In the last stage, the friction coefficient rapidly increased because of the scratching in face-forward direction. For the scratch tests in edge forward direction, there was no boundary between the third and the last stage in the relationship between the normal load and the friction coefficient. In our study, the adhesive strength between films and substrates correlated with the roughness of the substrates.

References

[1] Bull, S.J., Rickerby, D.S., Matthews, A., Leyland, A., Pace, A. R. and Valli, J., 'The Use of Scratch Adhesion Testing for the Determination of

Interfacial Adhesion: The Importance of Frictional Drag‘, Surface and Coatings Technology, 36, pp503-517, 1988.

[2] Holmberg, K., Laukkanen, A., Ronkainen, H. and Wallin, K., ‘Tribological analysis of fracture conditions in thin surface coatings by 3D FEM modeling and stress simulations’, Tribology International, 38, pp.1035-1049, 2005.

[3] Damayanti, M., Widodo, J., Sritharan, T., Mhaisalkar, S.G., Lu, W., Gan, Z.H., Zeng, K.Y. and Hsia, L.C., ‘Adhesion study of low-k/Si system using 4-point bending and nanoscratch test’, Materials Science and Engineering, B121, pp.193-198, 2005.

[4] Ye, J., Kojima, N., Ueoka, K., Shimanuki, J., Nasuno, T. and Ogawa, S., ‘Nanoscratch evaluation of adhesion and cohesion in SiC/low-k/Si stacked layers’, Journal of Applied Physics, vol.95, no.7, pp.3704-3710, 2004.

[5] Mutoh, Y., Xu, J., Miyashita, Y., Kuroishi, T. and Sasaki, Y., ‘On Evaluation of Adhesive Strength in Scratch Test of Coating Materials.’ Transactions of the Japan Society of Mechanical Engineers A68-670, pp. 909-915, 2002.

[6] Shibutani, T., Yu, Qu., Shiratori, M. and Akai, T., ‘Mechanism of Damage Process on Interface between Films in Nanoscratch Test’, M&M Shinshu Spring Symposium, No.05-03, pp.43-46 2005.

[7] Li, X. and Bhushan, B., ‘Micro/nanomechanical and Tribological Characterization of Ultrathin Amorphous Carbon Coatings’, Journal of Materials Research, Vol.14, No.6, pp.2328-2337 1999.

[8] Ektessabi, A. I., Yasui, N. and Okuyama, D., ‘Characteristics of an Ion Beam Modification System with a Linear Ion Source’, Review of Scientific Instruments, Vol.73, No.2, pp873-876 2002.

[9] Ge, J., Turunen, M.P.K. and Kivilahti, J.K., ‘Surface modification and characterization of photodefinable epoxy/copper systems’, Thin Solid Films, 440, pp198-207, 2003.