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Variation of adhesive force in the interface of Pd and SrTiO₃ as a consequence of residual stresses

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Abstract

Initially, Pd thin films were deposited over a hard substrate using electron beam physical vapor deposition. The growth and the surface roughness of the films were analyzed and their effects upon conventional indentation test were discussed. Afterwards, an experimental method is described which can measure the critical fracture force in thin films using oscillating indentation. Initially, repetitive contacts at a single point with the purpose of identifying the fracture time provide the fracture force versus fracture time plot. Non-linear curve fitting of the data reveals the theoretical fracture force by a single indentation, which is called critical fracture force. Arguments are put forth to show the relation between piling up height and applied force. Discrepancies were observed in the plot of the ratio between total indentation depth and piling up height versus applied force when higher loads than critical fracture force were applied. Discrepancies appear as a result of indenting the substrate. Nanoscratch test facilitated the possibility to measure adhesion strength and adhesion energy of the films considering measured critical fracture force as maximum applied force. The relation between residual compressive stresses, adhesion strength, plastic deformation, and pilling up area was discussed using dislocation theories. Indentation with high applied load leaves behind large plastic deformation and reduces the accuracy and reliability of the test results. Hence, lower loads (in the order of nN) were applied using Atomic Force Microscopy in the friction mode. Pulling off force was mapped in each thickness

of Pd films. The results confirm that the area around a hillock exhibits higher pulling off force due to the local stress relaxation as a consequence of hillock formation. By repeating the mapping process over different area with various applying forces, the plot of the pulling off force versus applied load was drawn representing discrepancies in the results at higher loads. This phenomenon is associated with the plastic deformation in the films.

Keywords: Oscillating Indentation, Nanoscratch Test, Friction Force Microscopy, dislocation theory

1. Introduction

The successful performance and reliability of metallic thin films is often limited by their mechanical integrity and their adhesion to the substrate. Therefore, many applications of thin films and coatings require knowledge of their mechanical properties. Generally, thin metal films on rigid substrates sustain high stresses during growth process. The importance of stress has not decreased in all the years of intense investigation and it is still one of the main reasons for malfunction of thin film applications. Severe damages such as cracking [1], buckling [2], lifting [3], curling [4], and peeling [5] has been reported as a consequence of residual stresses in thin films. In fact, residual stress develops in thin films when the spacing of two neighboring atoms deviates from local equilibrium spacing. Numerous reports could be found in the literature describing the mechanism behind stress generation in thin films [6-10]. It is well established that residual stresses affect upon adhesive properties of the film [11]. Hence these stresses should be considered while measuring thin film mechanical properties. One powerful method for measuring thin film mechanical properties is nanoindentation [12-15]. Marshal and Evans [12] introduced a model in which an indentation creates a plastic impression on the film surface. The residual strain in the plastic impression causes the film to buckle upwards in order to minimize the strain energy, leaving a circular crack at the interface beneath the indentation, which results in a circular blister on the film. In their model, if the accumulated compressive stress in the film plus the stress associated with indentation is greater than the critical stress required for buckling, then the film will buckle from the interface and a ring crack forms in the interface and expands until the driving force for crack extension drops below a critical value [16]. However, this method is highly impractical for very thin films, particularly ones that are soft or have a very strong interface with the substrate. In fact, the strain energy in the Marshal and

Evans [12] model basically is too small to induce delamination. In order to solve this problem, various researchers [17-20] have deposited thick overlayers capable of accumulating large amount of strain energy over the interface. In our previous work [21], a new method was introduced based on the oscillating indentation to accurately measure the critical fracture force in Pd thin films. In this article, the critical fracture force has been described as a critical force that indenter fractures the film and reaches to the interface without indenting the substrate. This method indents generally at low indentation loads to prevent the plastic deformation while repetitive contact of the indenter with the surface using oscillating indentation modifies the final contact area. Here, it is tried to apply this method to measure the adhesive properties of Pd thin film.

Nanoindentation with a scratch test module was applied to measure critical stress and adhesion energy. During the scratch test, the load was ramped from 0N until the critical fracture force in each thickness measured by oscillating nanoindentation. The scratch length was set as 1000 μ m at a moving velocity of 50 μ m/s. Moreover, in order to overcome against plastic deformation such as piling up and sinking in area, the measured critical fracture by oscillating nanoindentation was converted into the critical applied load in Atomic Force Microscopy (AFM) using geometrical ratio between Berkovich indenter and AFM tip.

2. Experimental Method

The materials used in this study were 101B grade (Crystal GmbH) both sides polished (100) oriented SrTiO₃ (STO) substrate and 99.99% pure palladium. Pd and STO have been selected for this study since Pd/STO shows the behavior of a soft film over hard substrate. Moreover, application of Pd as electrode material and STO as insulator is

widespread in electronic devices such as planoparallel capacitors. delamination of electrode metallic layers results in malfunction of these devices which might be attributed to residual stress. This is the reason to study the effect of residual stress upon interface properties of Pd and STO. Electron beam evaporator was used for palladium evaporation. Graphite crucibles were used for resistive evaporation. The evaporation chamber was evacuated with mechanical and turbo molecular vacuum pumps until the base pressure of the chamber was 3.1×10^{-7} torr [22-23]. Palladium was evaporated at 1.2 kV accelerating potential and 140 mA emission intensity. Three thickness of Pd film (50, 100, and 150 nm) were deposited over STO substrate at room temperature. The ex situ film thickness was measured by a Dektak 3030 profilometer (Veeco). The crystalline structure was determined by X-Ray Diffraction (XRD) analyses in θ - 2θ scan by a 4-circle X-ray diffractometer with Cu-K α radiation (MRD PHILIPS). Topographic images were recorded with Digital Instruments Multimode- Veeco AFM in tapping mode ($\nu = 300$ kHz). The nanoindentation tests were performed by a MML NanoTest^{TM} nanoindenter with a Berkovich indenter (face angle 65.27°), whose tip radius is 100 nm. From each indentation cycle, hardness, elastic module, and Poisson's ratio values were determined from the shape of unloading curve by means of Oliver and Pharr method [24]. Average elastic module and hardness were calculated by repeating indenting measurement with various forces at different points of the sample. Scanning electron microscopy (SEM) (ESEM Quanta 200 FEI) has been utilized for measuring the contact area after nanoindentation. Afterwards, repetitive contacts at a single point were obtained by oscillating the specimen against indenter that is mounted on a freely swinging pendulum. The process has been described in [21]. Afterwards, stress was measured in ψ -tilt mode from 0° to 180° ($2\theta = 38^\circ - 42^\circ$) in order to plot the d-spacing versus $\sin^2\psi$. Afterwards, by considering the measured Poisson ratio and elastic

module, residual stress was measured for each Pd film [25, 26]. To reduce the plastic deformation (piling up and sinking in regions), AFM MFP 3D (Asylum) was used to indent Pd films applying very low loads. Measurements are performed in buffer medium PBS using cantilever SNL (Veeco) with nominal spring constant equal to 0.06 nN/nm and a monolithic diamond tip (ND-DYIRS-4) with around 100nm tip radius. Cantilever spring constant was calculated again using the thermal noise method and 102.8pN/nm was obtained. Moreover, AFM force curves are detected by pushing the AFM tip onto a substrate and recording the deflection and the stroke of the cantilever. Pulling off force was mapped in a square with a 10×10 grid in 5×5 μm^2 area for each thickness of Pd film when maximum vertical force did not exceed 15nN. The average pulling off force was studied in each thickness of Pd films.

3. Results and Discussion

3.1 Oscillating nanoindentation

Surface roughness is a very important issue while measuring the adhesive properties of a film using nanoindentation. Since the contact area is measured indirectly from the depth of penetration, the natural roughness of the real surfaces causes errors in determination of the area of contact between the indenter and the substrate [27-28]. Hence, as much film is smoother, the accuracy of the indentation results is higher. Previously it has been shown [22] that thin Pd films transfer the morphology of the underneath layer or substrate toward the final surface. Hence, surface roughness of Pd films depends upon substrate roughness as well as deposition conditions. Therefore, both sides polished (100) oriented STO with the root-mean-square (RMS) roughness less than 3nm has been selected as substrate. Low roughness of STO results in

negligible contribution of the substrate roughness in the surface roughness of Pd films. Furthermore, deposition condition thoroughly modifies the surface quality of the films. It has been found experimentally that at vacuum level less than 5×10^{-5} Torr, and low accelerating potential and emission intensity (low deposition rate) reduces the surface roughness. In fact low vacuum level avoids the collision of the ad-atoms with the atmosphere in the chamber and more homogeneous thin films are expected. Accelerating potential and emission intensity defines the size of the impinging atoms. Basically smaller ad-atoms provide lower surface roughness. After impinging the ad-atoms over the substrate, surface diffusion controls the movement of ad-atoms over surface. At low substrate temperature (room temperature), surface mobility of ad-atoms is limited. Therefore, process such as coalescence which increases the surface roughness does not or not completely occurs. During impinging and growth, residual stresses generate in the films. Dislocations, defects and stacking faults reside in the lattice as a consequence of these stresses. Normally, deposition temperature by enhancing the surface mobility of the ad-atoms avoids of these lattice imperfections. Thus it could be expected that deposited Pd films at room temperature exhibits smooth surface and contains small grain size which are surrounded by grain boundaries with high density of dislocations. Fig 1 represents highly textured Pd film toward (111) crystal direction. Basically, face centered cubic thin film materials tend to growth toward (111) preferred orientation. Fig 2 shows the smooth surface of 50nm Pd thin film. Although by increasing the thickness of the films, surface roughness would be increased; RMS roughness of the films did not exceed 30nm. Hence, possible errors that arise due to surface roughness of the films are negligible during measurement of the contact area using nanoindentation. Contact area is a vital parameter while measuring hardness and elastic module of the films. Normally mechanical properties such as hardness and

elastic module are measured from conventional force-indentation depth curves, as it would appear for a loading and unloading cycle into an elastic-plastic material. In fact, the ways in which the plasticity affects the interpretation of elastic unloading data can be dealt with the shape of the perturbed surface in the elastic analysis. Tabor et al. [29-30] demonstrated that the shape of entire unloading curve and the total amount of recovered displacement can be accurately related to the elastic modulus and the size of impact compression for conical indenter. However, there would be some important parameters which we should be aware of, such as high temperature stability and isolation of vibration which are the essential environment requirements during conducting the low force in order to measure the mechanical properties of thinner films. Fig 3 represents hardness and elastic module of the deposited Pd films calculated using Oliver and Pharr method [24]. This method is a commonly used method for obtaining hardness and elastic module of the films. However there are some shortcomings in this method. First, at very low loads, there is a considerable offset error in the load-indentation depth curve. Therefore, hardness and modulus tend to decrease at very low loads due to effect of the elastic-plastic deformations [31]. However, by applying higher loads, the substrate effects must be considered. STO hardness has been measured equal to 8.9 ± 1.1 GPa. While measuring the hardness, we assumed that STO hardness is 9 GPa. During hardness measurement of soft films over hard substrate, since the film is extremely soft compared to the substrate and it yields at much smaller loads in comparison with the substrate, the film itself accommodates all plastic deformation and only when the indenter is close enough to the interface, substrate starts to yield [32-33]. This reveals the importance of measuring the critical fracture force during indentation [21]. Another potential problem is that this method was deduced from a purely elastic contact model for monolithic material. During indentation, only sinking in is assumed

occurring on material surface and therefore, depth of contact is always less than real depth. However, in the case of soft thin films, when piling up occurs, depth of contact is higher than real depth. Hence this method is not accurate for plastic material where material plastically piles up around the indenter. These problems are the origin of the error bars in fig 3. Furthermore, metal films are tolerant of defects in their grain boundaries, whose influence can be blunted by plastic deformation effects. Hence, variation of hardness versus thickness of metal films is small. In fact, by increasing the thickness, ad-atom surface mobility enhances and hence, the density of dislocation in grain boundaries decreases. This is the reason that at lower thickness, higher density of GB dislocations results in higher hardness. Normally, hardness versus thickness of metallic films decreases until reaching to the bulk hardness.

Going beyond the determination of elastic modulus and hardness, a new method is introduced to measure fracture force at the interface of the soft thin films and substrate using repetitive contacts at a single point by oscillating the specimen against indenter [21]. Oscillating indentation with different applied loads was performed on the deposited Pd films and indentation depth versus time was drawn (not shown). The results reveal the fatigue crack growth and an abrupt film delamination. The fatigue crack growth was detected since the diamond indenter tip begins to move away from the surface caused by expanding the crack volume as evidence by a decrease in the indentation depth. Fig. 4(a) represents the fracture force versus fracture time. It could be seen that at lower loads, fracture time increases. Theoretically, at higher indentation time, hardening occurs in the film which results in the asymptotic shape of the fracture force versus fracture time plot [21]. The crossover of the plots in fig 4(a) and Y-axis identifies critical fracture force. Theoretically, critical fracture force is a theoretical fracture force that reaches the indenter to the interface between film and substrate in the

first stage of indentation. Fig. 4(b) represents the critical fracture force versus thickness of Pd thin film. As shown in fig. 4(b), critical fracture force increases at higher thickness of Pd thin film. Fig 5(a) shows SEM image of 150nm Pd film which is indented with measured critical fracture force (1.3mN load for 150nm Pd film (fig 4(b))). Pilling up area could be detected around the indentation area. Fig 5(b) reveals the height profile along the line which is drawn in fig 5(a). h_t is the total depth of indentation and h_i is piling up height. It has been shown previously [21] that the ratio between h_t and h_i is constant by increasing the applied load until a critical value. Indeed, this critical value is critical fracture force. Inconsistency could be detected at loads higher than critical one which is due to the substrate deformation irrespective of the tip rounding. Fig 5(c) points out this variation as well. Simply, this could be a way to test the accuracy of the measured critical fracture force using piling up heights.

3.2 Effect of residual stress on the interface adhesion strength

Basically, large percentage of the plastic deformation in soft thin films is associated with piling up region and this area expands at higher loads. Randal [34] showed that at low loads, little pile up is present, although at higher loads, pile up area increases and a substrate relaxation occurs when indentation depth goes beyond the film thickness. In fact, applied load generates stresses around indentation area which will be released by piling up phenomenon. Lee et al. [35] showed a dependency between pile up height and applied stress into the specimen. Since thin films normally undergo residual stresses, pile up height might be dependent to the residual stresses in the films. Fig 6 represents the residual stresses in the deposited thin films measured using X-ray diffractions. In fact, stress was measured from the slope of the d-spacing versus $\sin^2\psi$ plot. Afterwards,

by considering the measured Poisson's ratio and elastic module using nanoindentation, residual stress was measured for each Pd film [36]. Underwood [37] showed the effect of residual stresses upon the shift in the shape of the pattern around indentation. The pattern around indentation consists of piling up and sinking in area. Therefore, any mechanical properties measured using nanoindentation, are dependent to the residual stresses in the film. Marshal and Evans [12] showed that residual stresses in the films modify the crack extension force depending on the buckling of the films. Therefore, the effect of the residual stress upon the critical stress for the delamination (σ_c) and the fracture energy release rate (G_c) for interface delamination (adhesion energy) is studied. Nanoindentation with a scratch test module was applied to measure critical stress and adhesion energy. The applied force for interface delamination, defined as the critical loads (P_c) and the critical scratch track width (d_c) were measured from the scratch morphology as shown in fig 7. Critical stress for the delamination (σ_c) (adhesion strength) was measured using equation (1) as follow [38-40]:

$$\sigma_c = \left(\frac{2P_c}{\pi d_c^2} \right) \left[\frac{(4+\nu_f)3\pi\mu - 8(1-2\nu_f)}{8} \right] \quad \text{Eq. (1)}$$

where μ is the measured friction coefficient of indenter sliding given by nanoscratch tester about 0.025 and ν_f is the Poisson's ratio measured in each thickness using nanoindentation. Thereafter, adhesion energy was measured using equation (2).

$$G_c = \frac{\sigma_c^2 t}{2E_f} \quad \text{Eq. (2)}$$

where t is the thickness of the film and E_f is the elastic module measured by nanoindentation. Fig 8 represents the variation of the adhesion strength and adhesion energy versus residual stress in the films. It can be seen that at higher compressive

residual stress and higher thickness of Pd film, adhesion strength and adhesion energy increased. However, since the contribution of the residual stress mixed with the contribution of the thickness, separation of the two parameters is extremely difficult. Huang et al. [41] showed that residual compressive stress enhanced the adhesive properties of SiN/glass. Hence, an increase in residual compressive stress and thickness enhances the adhesive properties. Basically, compressive residual stress generates during thin film deposition. During indentation, these stresses blunt the cracks' tip and suppress their propagation. This is why typical force-displacement curves of the films which undergo compressive stress shift toward lower displacement. In fact, compressive residual stress can aid the nucleation of the dislocations during deposition. Elastic mismatch stress in epitaxial films provides a driving force for threading dislocations that extend from free surface of the film to the interface between film and substrate [42-44]. The propagation of threading dislocations in the film layer results in misfit dislocation lines along the interface. This process relaxes the elastic mismatch strain and increases the total length of the dislocation [45] when the thickness of the film is higher than Matthews' critical thickness [42]. During indentation with the forces lower than critical fracture force, the density of the misfit dislocation increases which leads to an increase in the adhesion strength of the films. Indeed, indentation may lead to appearance of perfect Lomer-Cottrell dislocation which results in interlocking between misfit dislocations in the interface between film and substrate. Hence, adhesion strength increases due to the interface hardening. There would be two possible solutions avoiding these dislocations. First solution is already discussed by applying critical fracture force onto the films with the fast ramping load. This can avoid the interface dislocation due to the fact that fast indentation avoids the relaxation in the interface, particularly when thin film ruptures under the applying force. Second solution would be

applying very low loads using small tips. This is nowadays feasible using Atomic Force Microscopy in friction mode.

3.3 Pulling off mapping by Atomic Force Microscopy

As mentioned before, using AFM provides the possibility to conduct force measurement with low applying forces. Before conducting force measurement, it is necessary to accurately determine cantilever spring constant. Various methods are described in the literature for calibration of AFM cantilevers [46-50]. In our experiments the so-called thermal noise method was used, as it was found to deliver most reliable results. Spring constant has been measured equal to 102.8 pN/nm. Fig 9 shows the indentation area using AFM diamond tip over 50nm Pd films. Fig 9 (a), (b), and (c) correspond to the indentation with 250nN, 150nN, and 50nN, respectively. Fig 9 (d), (e), and (f) are the counter plots of the fig 9 (a), (b), and (c), respectively. It could be seen that pile up area around the indentation area became smaller at lower applying loads. The height and area of the piling up region corresponds to plastic deformation around the indentation area. It could be seen that maximum height of pile up area decreased from 70 nm until 60 nm by reducing the applied force from 250nN to 150nN. However, further decreasing the applied force from 150 nN to 50nN almost suppressed the appearance of the pile up area. It coincides by the fact that still clear indentation area could be seen which reveals that the film underwent plastic deformation. The surface after indentation with 50nN (f) is smoother in comparison with the indentation at higher forces (d and e). Hence, in agreement with the variation of the pile up height induced by indentation, it could be seen that in low indentation depth and low applying loads, pile up height is negligible. It

should be noted that by further decreasing the indentation load ($<1\text{nN}$) indentation area disappears due to the elastic behaviour of the film. Hence, measurement of the exact indentation area would not be possible. Moreover, applying indentation with low forces avoids tip rounding. Thereafter, to investigate the adhesive properties of the film, pull off test was done over a 10×10 grid ($5\text{ }\mu\text{m}\times 5\text{ }\mu\text{m}$ area) of each Pd film. Each grid has been selected equal to $0.5\text{ }\mu\text{m}\times 0.5\text{ }\mu\text{m}$. It should be noted that it might be possible to decrease the grid size, but the attention should be made about plastic deformation around each indentation. Fig 9 (f) shows that plastic deformation after indentation with 50nN is a sphere with around 100nm radius. This area on the surface is even smaller at lower applying loads ($<5\text{nN}$), although in the area far from the surface (deeper area) it is not possible to calculate it. It should be considered that the distance between indentation points should not be lower than the plastically deformed area. Therefore, in order to enhance the accuracy, the distance between indentation points was selected about $0.5\text{ }\mu\text{m}$.

Indeed, when the AFM is retraced, the adhesion force causes a kink in the force curve. Fig 10(a) shows the grids in an area in the 50nm Pd film. Crosses in fig 10(a) represent the points that the force curve was drawn. The squares in fig 10(a) correspond to an area in which hillocks are presented. Fig 10(b) reveals the pulling off map of the fig 10(a). Pulling off force varied from $4.23\times 10^{-10}\text{ N}$ until $6.96\times 10^{-10}\text{ N}$. It should be noted that the indentation on the area around hillocks entirely represents high pulling off force. This might be in connection with the presence of hillocks. It has been previously showed that hillocks appear on the surface of the metallic film as a result of stress relaxation [51]. It could be concluded that the area around a hillock undergoes lower residual stress. Therefore, the density of dislocation is lower than not-relaxed area. This in turn results in an area with lower hardness. When an area exhibits softer properties, while retracing

the AFM tip, film sticks to the tip which results in a larger pile up area around indentation imprint. Therefore, pulling off force increases in this area. Fig 11 represents the average pulling off forces of each thickness of Pd films versus indentation load. Apparently, after a critical applied load, discrepancy appears in plot of the pulling off force versus load. This irregular variation of the points (showed by the squares in fig 11) might be related to the plastic deformation in the films. Basically, as much indentation load increases, sticking increases during retracing. Consequently, pile up area generates around the indentation and its height becomes larger as much the indentation load increases. In principal, other parameters such as tip rounding, porosity, and generation of the stacking faults during indentation might impact on the accuracy of the results. Ability in applying loads even in the range of pN is a significant advantage of using Atomic Force Microscopy. Further study is essential to clarify the main reason for observed discrepancies in fig 11.

4. Conclusion

In this study, Pd thin films were grown using electron beam physical vapor deposition. Surface roughness and crystal structure of the deposited film were studied and their effects upon accuracy of the force-displacement curves were discussed. Moreover, the mechanical properties and interface adhesion energy of 50nm, 100nm, and 150nm Pd thin film were measured using oscillating nanoindentation. Critical fracture force was introduced as a critical force to reach into the interface without impacting the substrate. It was measured in each thickness of Pd films and their connection with the pile up area was drawn. It was found that at higher applied forces than critical fracture force, the plot of h_f/h_i versus applied force represents irregular variations as a result of indenting the

substrate. Nanoscratch test assists with the measurement of the adhesion strength and adhesion energy of Pd films. It has been found that compressive residual stress enhances the adhesive properties of the films. The compressive residual stress can aid the nucleation of the dislocations during deposition. The propagation of threading dislocations in the film layer results in misfit dislocation lines along the interface. This process relaxes the elastic mismatch strain and increases the total length of the dislocation in the interface. During indentation with the forces lower than critical fracture force, the density of the misfit dislocation increases which leads to an increase in the adhesion strength of the films. However, applying forces equal to critical fracture force lead to large plastic deformation and pile up area around the indentation area. This plastic deformation may reduce the accuracy of the indentation process. Therefore, Atomic Force Microscopy was used to apply low loads leave behind a small plastically deformed area. Pulling off force was mapped for each Pd film using applied forces not more than 15nN. It is found that the area around hillocks represents higher pulling off force. This is due to the fact that presence of hillocks releases the residual stresses of the area around it. Hence, AFM tip indents a softer area which results in higher pulling off during retracing. It could be concluded that during nanoindentation, applying forces equal to the critical fracture force can enhance the accuracy of the results. Furthermore, applying low loads using AFM perfectly reduces the plastic deformation around the indentation area.

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Figure Caption:

Figure 1. X-ray diffractions of 50nm, 100nm, and 150nm deposited Pd thin films over STO substrate at room temperature. Single (111) diffraction of Pd film shows that deposited films are highly textured toward (111) preferential orientation.

Figure 2. AFM topographic 2D (a) and 3D (b) image of the 50 nm Pd film represents a smooth surface with RMS roughness less than 10nm.

Figure 3. Measured hardness and elastic module using nanoindentation versus thickness of Pd film

Figure 4. Fracture force versus fracture time for different thickness of Pd thin film (a) and critical fracture force versus thickness of Pd thin film (b) measured using oscillating nanoindentation [21].

Figure 5. SEM image of 150nm Pd film indented with 1.3mN (a) and height profile (b) along the line which is drawn over (a).

Figure 6. Residual stress measurement using variation of d-spacing of (111) crystal plane. Large error bars are due to the 2θ position of (111) crystal diffraction (around 40°). Basically, accuracy of the measurement enhances when the planes at higher 2θ positions (≤ 180) were studied.

Figure 7. SEM surface morphology along the scratch track on 150 nm Pd film facilitating the measurement of the critical scratch track width.

Figure 8. Variation of the adhesion strength and adhesion energy versus residual stress in the films. It can be seen that at higher compressive residual stress and higher thickness of Pd film, adhesion strength and adhesion energy increases.

Figure 9. AFM topographic image of the indentation areas using AFM diamond tip over 50nm Pd films. (a), (b) and (c) correspond to the indentation with 250nN, 150nN, and 50nN, and. (d), (e), and (f) are the counter plots of (a), (b), and (c), respectively.

Figure 10. AFM topographic image of $5\mu\text{m} \times 5\mu\text{m}$ area of 50nm Pd film (a). This area is divided to 10×10 grids when the crosses in each grid represent the indentation area. (b)

exhibits the pulling off map of (a). It can be seen that pulling off force varied from 4.23×10^{-10} N until 6.96×10^{-10} N.

Figure 11. The average pulling off forces of each thickness of Pd films versus indentation load.

Figure 01 (Fig 1.tif)

Figure 02a (Fig 2a.TIF)

Figure 02b (Fig 2b.TIF)

Figure 03 (Fig 3.tif)

Figure 04a (Fig 4a.tif)

Figure 04b (Fig 4b.tif)

a)

Figure 05a (Fig 5a.tif)

Figure 05b (Fig 5b.TIF)

Figure 05c (Fig 5c.tif)

Figure 06 (Fig 6.tif)

Figure 07 (Fig 7.tif)

Figure 08 (Fig 8.tif)

Figure 09 (Fig 9.TIF)

Figure 10a (Fig10a.TIF)

Figure 10b (Fig10b.TIF)

Figure 11 (Fig11.tif)