

Vickers Indentation Simulation of Buffer Layer Thickness Effect for DLC Coated Materials

Abdul Wasy, Balakrishnan G., Yi Qi Wang, Atta Ur Rehman, and Jung Il Song

Abstract—Vickers indentation is used to measure the hardness of materials. In this study, numerical simulation of Vickers indentation experiment was performed for Diamond like Carbon (DLC) coated materials. DLC coatings were deposited on stainless steel 304 substrates with Chromium buffer layer using RF Magnetron and T-shape Filtered Cathodic Vacuum Arc Dual system. The objective of this research is to understand the elastic plastic properties, stress strain distribution, ring and lateral crack growth and propagation, penetration depth of indenter and delamination of coating from substrate with effect of buffer layer thickness. The effect of Poisson's ratio of DLC coating was also analyzed. Indenter penetration is more in coated materials with thin buffer layer as compared to thicker one, under same conditions. Similarly, the specimens with thinner buffer layer failed quickly due to high residual stress as compared to the coated materials with reasonable thickness of 200nm buffer layer. The simulation results suggested the optimized thickness of 200 nm among the prepared specimens for durable and long service.

Keywords—Thin film, buffer layer. Diamond like Carbon, Vickers indentation, Poisson's ratio, Finite element.

I. INTRODUCTION

DIAMOND-like carbon (DLC) coatings are used as protective coatings on metal, ceramics and polymer substrates. DLC coatings got industrial interest due to their excellent optical, electrical, chemical and physical properties, such as good antireflective, insulator and chemically inert with very high elastic modulus, hardness, wear resistance and thermal stability.

The hardness of coated materials depends on the sp^3/sp^2 content of DLC coating layer. The hardness increases with increase in sp^3 structure. The highest sp^3 content of 88% has reported by Ferrari et al. [1]. The young modulus ranges from 150 GPa to 750 GPa depending on the sp^3 to sp^2 content and deposition method. The young modulus ranges from 330 GPa for 40% sp^3 structure to 630 GPa for 80% sp^3 structure with hardness range of 33 to 63 GPa [2] respectively.

High residual stresses are major resistance in good adhesion strength of DLC coatings. To reduce the residual stress as much as possible, different researchers adopted various techniques as given in literature [3]-[4]. The introduction of soft metal buffer layer or mixing of additives are common techniques. The amount of residual stress also depends on the method of deposition.

Abdul Wasy, Atta Ur Rehman, Balakrishnan G., Yi Qi Wang and Jung Il Song are with the Department of Mechanical Engineering, Changwon National University, Changwon, 641-773, Republic of Korea (e-mail: jisong@changwon.ac.kr).

Physical vapor deposited (PVD) and chemical vapor deposited (CVD) are common deposition categories. Each category further contains different deposition methods and instruments which may yield different hardness and stress ranges. The presence of high stresses, either tensile or compressive greatly affects the mechanical properties of coating materials such as fatigue [5]-[6], adhesion strength and fracture toughness.

Chakravarthy et al [7] calculated energy release rate for the crack perpendicular to the interface between the coating and the substrate with its tip in the homogeneous substrate. Romeo and Ballarini [8] studied the radial crack behavior by considering the material stiffness across interface of coating and substrate. It was concluded that, when local stress intensity decreased and reached to zero then crack will propagate toward stiffer material and when local stress intensity will increased and reached to infinity, the crack approaches through interface from stiffer material to weaker one.

Number of coated material models were reported, having crack phenomenon due to substrate and coating material properties, interface adhesion, residual stresses, type of loading and crack geometry. Smith et al [9] explained the failure of coated material model in two modes; one is the crack initiation and second is the crack propagation and delamination. The cracks are usually initiate in brittle layer due to high residual stresses and then propagate through thickness up to interface as radial cracks and through surface as ring and lateral cracks causing delamination. The crack arrest or propagation depends upon the loading conditions, interface adhesion and material properties of coating [10] and substrate.

The finite element analysis is a quick technique to determine the approximate results. Current advancement of contact finite element simulation enabled the numerical analysis of hardness including work of Karl et al. [11], Talijat et al. [12], Wang et al. [13] and Murakami et al. [14]. Finite Element Analysis has the benefit of investigating the influence of an inelastic material constant on the hardness considering the other material constants fixed.

Tsui and Pharr in 1999, Burnett and Ricker by, 1987; Joslin and Oliver, 1990 performed number of experimental and theoretical studies to analyze the problem of extracting the Young's modulus of thin coating films considering substrate properties. The, numerical simulation has been used as an supplementary tool for better understanding of the indentation process by number of researcher as Chen and Vlassak in 2001 and Antunes et al. in 2004.

In this research work, tetrahedral amorphous DLC coating

deposition work was carried out on stainless steel 304 substrate using dual T-Shape Filter Cathodic Vacuum Arc and RF Magnetron Sputtering System. There were four models made with variation of Chromium buffer layer thickness from 50 nm to 200 nm sputtered at 200W and Bias of 100 voltages with $4E-4$ torr chamber pressure during deposition. The DLC coating of 500nm were deposited by FCVA. The characterization including the Vickers indentation test, adhesion strength test, wear test and Raman spectroscopy, Atomic Force Microscopy and Field Emission Scanning Electron Microscopy were performed.

This recent research study deals with three-dimensional numerical simulation of Vickers indentation test, to find the optimized thickness of buffer layer for DLC coatings under the high stress strain distribution and penetration depth under same loading conditions for all four models. The simulation work explains the elastic behavior, stress strain distribution and delamination in DLC coated materials with variations in thickness of buffer layer and Poisson's ratio of DLC coating layers. The stress, strain distributions, penetration depth and delamination in DLC coated structures is discussed in detail under variation of loading rate. In addition, a comparison of stress evolution during indentation was studied, in order to quantify the critical indentation depth under which the substrate is not plastically deformed.

II. 3-DIMENSIONAL MODELING

The four models of DLC coated materials were made in "Computer Aided Three-dimensional Interactive Application (CATIA) [15]. This software is being used to model 3D mechanical systems for designing, sketching, of parts, assemblies, mechanisms and simulations etc. This software is widely being used in research and development sector.

The specimens were comprised of substrate, Cr buffer layer and DLC coating. The modeling was completed at Nano scale accurately. In first step, the replica of actual specimen was model having dimensions of 25mm diameter and the thickness of substrate and DLC coating were 2.5mm and 500 nanometers respectively with variation of Chromium buffer layer thickness of 50, 100, 150 and 200 nm..

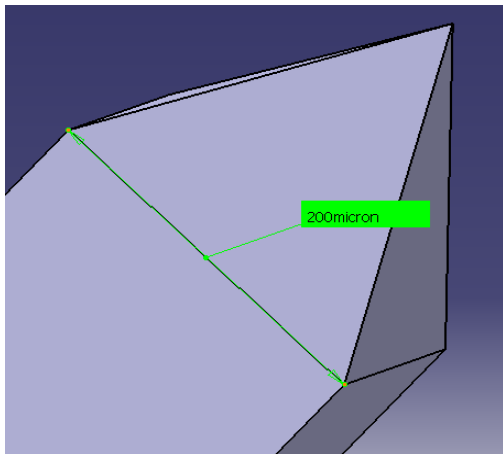


Fig. 1 Vickers indenter

In second step, a small scaled replica was modeled to understand the simulation behavior in batter way. In this model, the specimens have dimensions of 2 mm * 2 mm in length and width respectively, while the thickness of DLC layer have 2mm*2mm, 500 nm with variation Cr buffer layer thickness as 50, 100, 150 and 200 nm of respectively. The small Vickers indenter was modeled having edge length of 200 micron and the external angle of 136° as shown in Fig. 1.

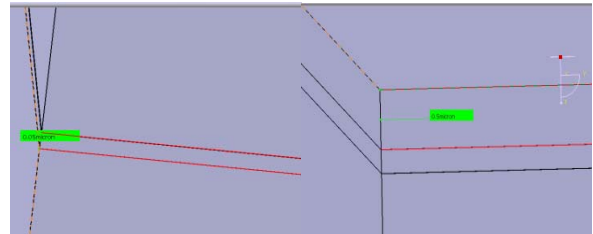


Fig. 2 (a) Cr Buffer layer of 50 nm (b) 500 nm DLC coating

The Fig. 2 represents the nano scale three dimensional modeling. Fig. 2a presents the thickness of buffer layer as 50nm, While Fig. 2b presents the DLC coating layer of 500 nm thickness. The three dimensional modeling assumption includes the uniform and perfect contact between DLC coating to Cr buffer layer and Cr buffer layer to substrate. The substrate, Cr buffer layer and DLC coating materials were homogeneous and pore free.

III. MATERIAL PROPERTIES

The material properties of all components were executed from the literature as given in Table I. Material properties of stainless steel and chromium are commonly available but for Diamond like carbon coating layer, different researchers defined different value depending upon the sp^3/sp^2 structure. The young modulus of DLC coating ranges from 150GPa to 700 GPa [2] depends upon the sp^3/sp^2 structure manufactured by different techniques and methods i.e., PVD, CVD, PECVD etc. The change in sp^3/sp^2 structure also changed the major properties like, young modulus, density etc. For the 88% sp^3 structure, the hardness is 700GPa and density is 3260 Kg/m³ similarly for 70% sp^3 structure the density is 2350Kg/m³ respectively [1].

In this study, the DLC properties are considered with 75% sp^3 structure deposited by FCVA System. Micro-hardness and elastic modulus of ta-C film are 47.74 GPa and 298.46 GPa, respectively [16]. The sp^3 Content is 75% in ta-C films which was find by ratio of intensity peaks I_D/I_G [17].

TABLE I
 MATERIAL PROPERTIES OF DIFFERENT PARTS OF DLC COATED MODEL

Material	Young Modulus (GPa)	Poisson ratio		Density (Kg/m ³)
Diamond	1200	0.17	0.17	3520
DLC	300	0.30	0.20	2360
Chromium	279	0.21	0.21	7190
Stainless Steel	193	0.31	0.31	7750

The material properties of Vickers indenter was taken as 1200 GPa young modulus [18], 0.17 Poisson's ratio and 3520 Kg/m³ density [19] respectively. Hertz Equation (1) [20] can be used to find the combined elastic modulus specimen/indenter (E_r),

$$\frac{1}{E_r} = \frac{(1-\nu^2)}{E} + \frac{(1-\nu_i^2)}{E_i} \quad (1)$$

E_i , E , ν_i and ν are the elastic moduli and Poisson's ratios of the specimen and indenter, respectively.

IV. FINITE ELEMENT SIMULATION

Finite element simulation is a quick and approximate solution of complex problems. Sometime the physical measurement of some parameters at nano and micron level is very difficult. Hence the 3-dimensional Finite Element Analysis is used to predict the stress, strain, indenter penetration and crack propagation behaviors.

The analysis was performed in ANSYS Workbench using structural analysis and Autodyne module. It was observed that, structural analysis module is good for simple materials but the Autodyne give best graphical presentation for stress, strain, indenter penetration and crack propagations behaviors for coated materials.

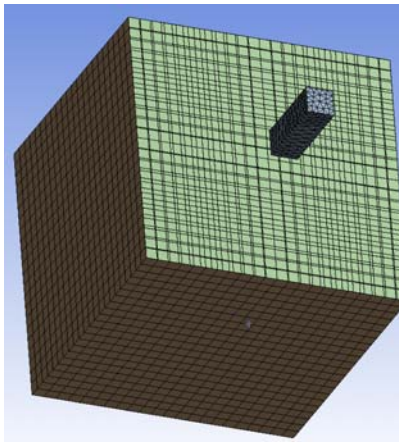


Fig. 3 Uniform Square Meshing

The model was imported in ANSYS Autodyne in .stp format. Material properties were assigned to all materials as given above in Table I. The uniform and perfect bonding was assumed between DLC to Cr buffer layer and Cr to substrate materials. While the virtual body interaction was made between DLC layer and Vickers indenter.

The highly fine and uniform square, explicit meshing was performed with the minimum edge length of 0.01, 0.015 and 0.02nm for all four models having Cr buffer layer thickness of 100, 150 and 200 respectively as meshed model is shown in Fig. 3. The model has 15037 nodes with high smoothing and slow transition. The Meshing statistics are given in Table II.

TABLE II
 MESHING STATISTIC

Contents	Quantity
Number of Nodes	15037
Number of Elements	131036

The boundary conditions were applied. The model was kept fixed from the bottom (x-y plane), the velocity of 1mm/s was assigned to the indenter in downward direction (z axis). The Vickers indentation simulation was performed at different loads ranging from 0.49N to 250N.

The all simulations were run under same boundary conditions for each model of different Cr buffer layer thickness with change in load at indenter. The equivalent stress, total deformation, indenter penetration, eq. elastic strain, eq. plastic strain, were evaluated and discussed below in details.

V. RESULTS AND DISCUSSION

The simulations were performed for two types of material properties of DLC coating layer i.e., the Poisson's ratio of 0.30 and 0.25. The simulation was performed for loading cycle on Autodyne software. The thin film showed the elastic response under Vickers indentation. The plastic behavior appears with increase in thickness of coatings. The results yield the useful conclusions which can be utilized for deposition of buffer layer at optimum thickness with stress, strain, crack growth, penetration and tribological properties.

A. Penetration and Crack Growth

The crack growth and delamination phenomena's are shown in Fig. 4, Fig. 5 and Fig. 6. Fig. 4 represents the SEM spectrograph of ring and lateral cracks produced during Vickers indentation. When the indenter penetrates in the coating layer, the lateral cracks were produced in it. The Fig. 5a shows the lateral cracks and the center point shows the location of indenter penetration. This lateral crack response is similar with experimental results. The Fig. 5b shows the ring crack on the exaggerated scale. These lateral cracks leads toward the delamination of coating from substrate at nearest end as shown in Fig. 5c. If the model is exaggerated at more high scale the material removal from the crack location can be seen as shown in Fig. 5d. This crack growth and delamination phenomena are similar with the experimental behavior of Vickers indentation.

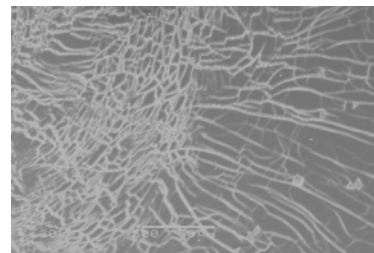


Fig. 4 Radial and lateral crack growth in DLC coated materials

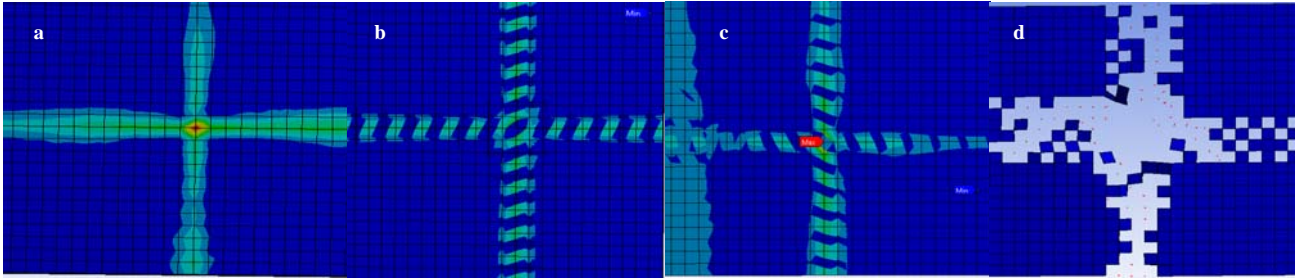


Fig. 5 Crack formation, growth, and delamination in DLC and Chromium layer

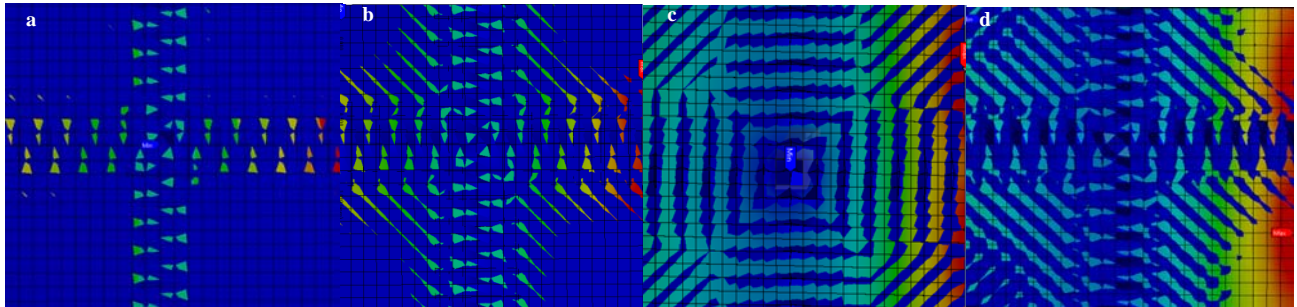


Fig. 6 Stress Distribution in DLC coated specimens

Fig. 6 presents the Scanning Electron Microscopy captured by JP/JSM 5200, JEOL, Japan at 500X resolution. This spectroscopy presents the radial racks in DLC coated material with 200nm Cr buffer layer during Vickers indentation.

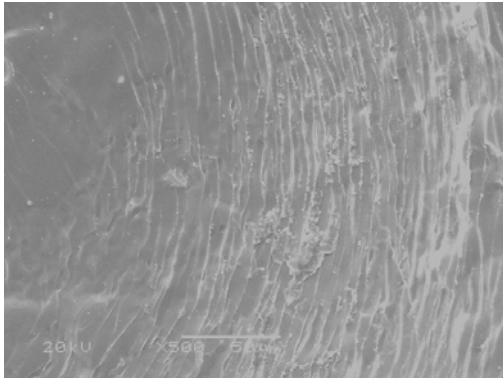


Fig. 7 Radial cracks in DLC coating with 200nm Cr layer

B. Stress Distribution

Delamination of DLC coating is still a big research issue. The stress distribution with increase of load on DLC coated specimens is shown in Fig. 7. From Fig. 7 (a to d), it can be seen that, as the load was increasing the amount of stresses were increasing concentrically around the indentation point. This stress distribution increases outward with increase in force and causes the ring cracks. The lateral cracks propagates from indentation point to ward specimen edges and the Fig. 7 (a, b) shows the high stress concentration on the end of lateral cracks. This high stress concentration causes crack propagation at high loads and leads toward delamination at nearest end as shown on Fig. 7d.

The Fig. 8a shows the stress distribution only on chromium buffer layer. It can be seen that, the stress increases from indentation point to towards end Fig. 8b presents the lateral crack propagation with stress distribution on Cr buffer layer supported by substrate materials. While Fig. 8 (c-d) graphically presents the effect of increase in load which is increasing the stresses on the edges of Cr layer with DLC coating. When the stress increased beyond yield strength it causes delamination.

C. Strain Distribution

The strain distribution under loading effect is shown in Fig 9(a-c). The Fig. 9a showed the concentric propagation of the equivalent strain during Vickers simulation. Fig. 9b showed the total deformation predicted across the indentation location. This fig presented the maximum deformation occurred during Vickers indentation experiment. The Fig. 9c showed the directional strain distribution. This strain distribution is propagating outward representing the formation of ring cracks.

D. Delamination

The delamination initiation phenomenon is discussed above. The following Fig. 9d represents the initiation of delamination stresses under the effect of Vickers indentation on buffer layer. When the indenter penetrates in the thin film, the cracks produced on the indenter tip and the near around locations. As the load increases, cracks grow and the stress intensity increase at nearest ends which cause the film to peel off.

In literature, the value of first principal stress of 20 GPa [16] at 49N is given. From the FEM numerical simulation results, it can be evaluated that, the maximum value of the

first principal stress appeared as 21.2 GPa, 6.01GPa, 4.2 GPa and 2.74 GPa for buffer layer of 50nm, 100nm, 150nm and 200nm thickness respectively under the load of 50 N. These results also suggest the failure of thin film with 50 nm buffer layer at high stresses. The stress strain curve for each model is shown in Fig. 10. The stress strain curve of 200 nm showed more ductile response as compared to others. The specimens having 100 and 150nm bear stress up to certain limit then suddenly failed. 50nm buffer layer delaminated due to high residual stresses at small loads.

The effect of applied load and indenter penetration and the stress is shown in Fig. 11. The specimen having 200nm buffer layer have minimum penetration as compared to other models. The model with 50nm buffer layer has maximum penetration depth under same loading conditions which predict its poor wear resistance.

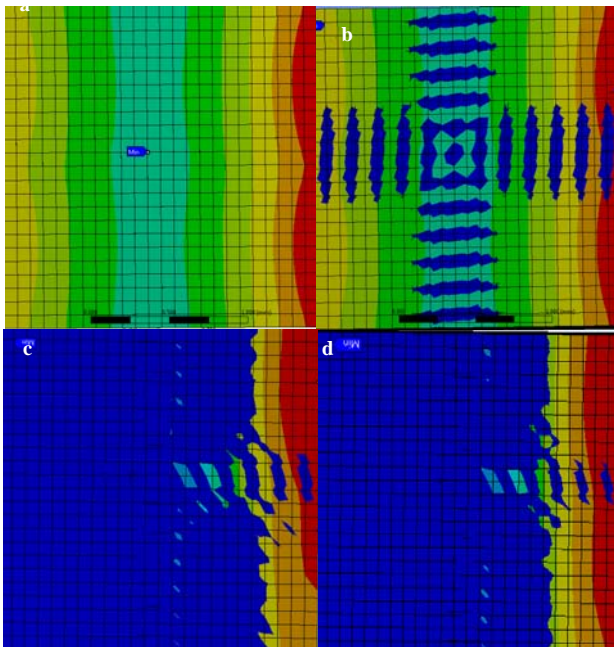


Fig. 8 Stress distributions in (a) Chromium buffer layer (b) Cr with substrate (c-d) Cr with DLC coating

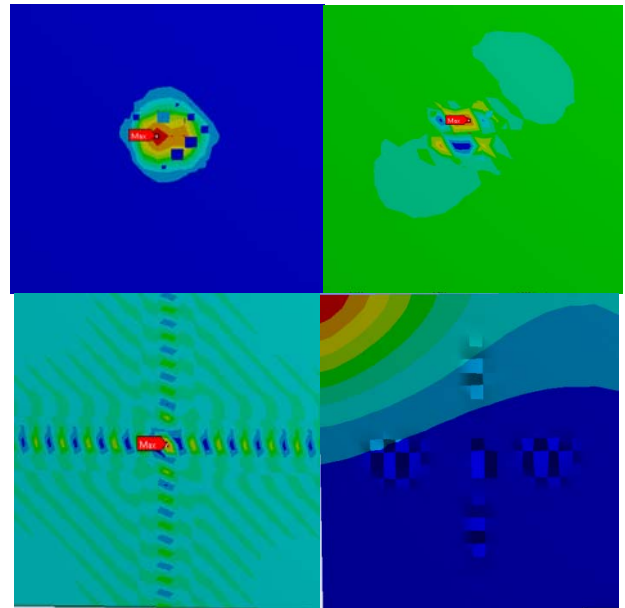


Fig. 9 (a) Equivalent Strain (b) Total Deformation and (c) Directional Strain Distributions (d) Crack and delamination initiation on Buffer layer

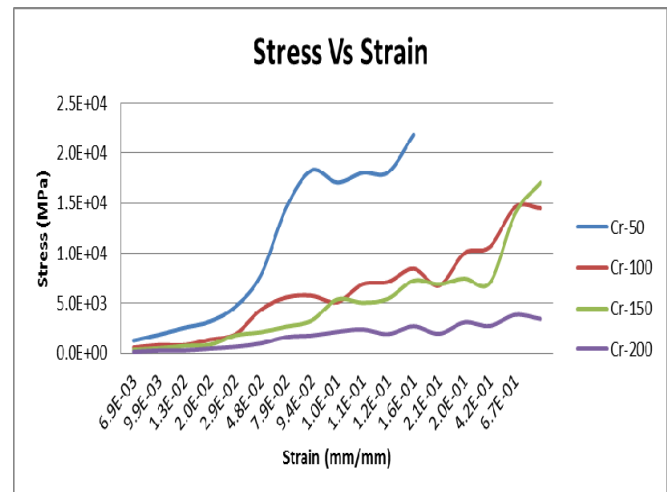


Fig. 10 Stress-Strain for DLC coated materials with buffer variation

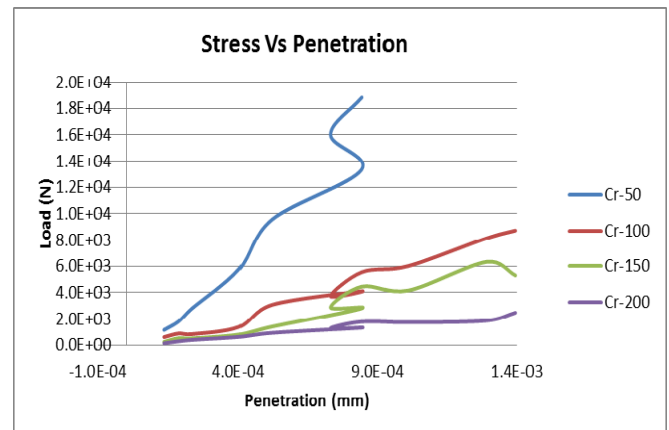


Fig. 11 Stress and Penetration for DLC coated materials with buffer variation

VI. CONCLUSION

The von misses and maximum principal stress criterion was used to predict the crack initiation and propagation during the Vickers indentation simulations. Indenter penetration is more for 50nm buffer layer as compared to 200nm under same loading conditions and the penetration increase with increase in Poisson's ratio under all other fixed conditions. Similarly, DLC coated materials having less Poisson's ratio bears more stress as compared to materials with high Poisson's ratio of DLC coating. Thin films were peeled off due to high residual stress. The amount of stress was very high in 50nm buffer layer model as compared to 200 nm which cause the failure of material at small loads.

Ring cracks produced under the indenter tip and propagate outward in concentric manner. These ring cracks combined with lateral cracks, which cause delamination of thin film from nearest edge, notch or discontinuity. Thin films showed the elastic behavior because plastic behavior increases with increase in thickness. Substrate has to bear stresses but strain did not propagate beyond the buffer layer and substrate interface.

ACKNOWLEDGMENT

This research work was funded by a National Research Foundation, Korea under grant No: FGM-2011-0002804 and ERC-2012-0009455 and supported by National Brain Korea 21 Project.

REFERENCES

- [1] A. C. Ferrari et al., "Elastic constants of tetrahedral amorphous carbon films by surface Brillouin scattering", *Appl. Phys. Lett.* vol. 75, pp. 1893-1895, 1999.
- [2] M. Bonelli et al., "Structure and mechanical properties of low stress tetrahedral amorphous carbon films prepared by pulsed laser deposition" *Eur. Phys. J. B* 25, pp. 269-280, 2002.
- [3] Clapa M, Batory D., "Improving adhesion and wear resistance of carbon coatings using Ti:C gradient layers", *Journal of Achievements in Materials and Manufacturing Engineering*, 20(1/2), pp. 415-418, 2007.
- [4] Mikula J, Dobrzanski L A., "PVD and CVD coating systems on oxide tool ceramics", *Journal of Achievements in Materials and Manufacturing Engineering*, 24(2), pp. 75-78, 2007.
- [5] Hetrmanczyk M, Swadzba L. and Mendala B., "Advanced materials and protective coatings in aero-engines application" *Journal of Achievements in Materials and Manufacturing Engineering*, 24(1): pp. 372-381, 2007.
- [6] Zalounina A, Andreasen J H., "Theoretical analysis of fatigue crack growth in a coated substrate", *International Journal of Fracture*, 2005, 133(1), pp. 3-10.
- [7] Chakravarthy S S, Jordan E H. and Chiu W K S., "Thin film and substrate cracking under the influence of externally applied loads" *Engineering Fracture Mechanics*, 2005, 72(8) pp. 1286-1298.
- [8] Romeo A, Ballarini R., "A crack very close to a biomaterial interface", *Journal of Applied Mechanics*, 62(3), pp. 614-619, 1995.
- [9] Smith G A, Jennett N. and Housden J., "Adhesion of thin coatings — The VAMAS (TWA 22-2) inter laboratory exercise", *Surface and Coatings Technology*, 197(2/3), pp. 336-344, 2005.
- [10] Xie Chang-Jin, Tong Wei., "Cracking and decohesion of a thin Al₂O₃ film on a ductile Al-5%Mg substrate", *Acta Materialia*, 53(2) pp. 477-485, 2005.
- [11] Karl, E. R., Komvopoulos, K. and Bogy D.B., "Elastic-Plastic Finite Element Analysis of Repeated Indentation of Half-Space by Rigid Sphere", *Trans. ASME, Journal of Applied Mechanics*, vol.60, No.4, pp. 829-841, 1993.

- [12] Talijat, B., Zacharia, T., and Kosel, F., "New Analytical Procedure to Determine Stress-Strain Curve From Spherical Indentation Data", *Int. J. Solids Structures*, Vol.35, No.33, pp. 4411-4426, 1998.
- [13] H.F.Wang and H.Bangert. "Three dimensional finite element simulation of Vickers indentation on coated systems", *Mater. Sic. and Engng*, A163, pp.43-50, 1993.
- [14] Murakami, K. and Matsuda, K., "Analysis of Vickers hardness by the finite element method", *Trans. Japan Society of Mechanical Eng.*, vol.A57, No.539, pp.1665-1673, 1991.
- [15] Munawar Iqbal et al, "Design modification in rotor blade of turbo molecular pump", *Nuclear Instruments and Methods in Physics Research Section A*, vol. 678, pp. 88-90, 2012.
- [16] Xiang Yu et al., "Structural, mechanical and frictional properties of tetrahedral amorphous carbon film by filtered cathodic vacuum arc system" *Surface & Coatings Technology* 201, pp. 4995-4998, 2007.
- [17] H. Mori, H. Tachikawa, "Increased adhesion of diamond-like carbon-Si coatings and its tribological" *Surface and Coating Technology*, 149: pp.225-230, 2002.
- [18] Spear and Dismukes, "Synthetic Diamond – Emerging CVD Science and Technology" Wiley, 1994 NY.ISBN 978-0-471-53589-8.
- [19] "Diamond", *Mindat*. Retrieved, 2009.
- [20] Adonias Ribeiro Franco Jr et al., "The use of a Vickers indenter in depth sensing indentation for measuring elastic modulus and Vickers hardness" *Mat. Res.* vol.7 no.3 São Carlos July/Sept. 2004.