MODELING AND SIMULATION OF NANOINDENTATION INTO POLYMER THIN FILMS BY FINITE ELEMENT METHOD

Ch. Srinivasa Rao[1]   C. Eswara Reddy[2]
[1]Department of Mechanical Engineering, PVP Siddhartha Institute of Technology, Kanuru, Vijayawada, 520007, A.P .INDIA.
[2]Department of Mechanical Engineering, College of Engineering, Sri Venkateswara University, Tirupati, 517502 A.P. INDIA.

ABSTRACT

Nanoindentation technique is being widely used to measure the hardness and elastic modulus of thin film coatings. It is being used extensively to measure the nanomechanical properties of hard thin films. However, the determination of the surface mechanical properties of polymeric materials with such techniques is relatively new. In this paper, an attempt has been made to simulate the nanoindentation process into polymer thin films by Finite Element Modeling (FEM) Technique. FEM has been done to extract both elastic and visco elastic properties of polymeric thin films. The experimental values of elastic moduli available from the literature for bulk Polymethyl Methacrylate (PMMA) and ultra-high molecular weight polyethylene (UHMWPE) are used to simulate the nanoindentaion into their thin films. In linear elasticity case, the load-displacement data obtained by simulation is used to calculate the Young's modulus and hardness of the thin films and it is found that the thin films are superior in surface hardness than to bulk materials. The Stress and Strain calculation with plastic deformation of the thin film was simulated by FEM. The model is extended to measure time-dependent behavior (Visco elastic) of polymeric films. In Visco elastic case, the load-displacement data is obtained as a function of time and is compared with that of the analytical results. A good correlation is found between the simulated results and analytical results. The obtained load-displacement data is used to calculate visco elastic properties of polymer thin films i.e., the relaxed shear modulus and Creep compliance. The thin films properties are measured with no substrate influence, and considering the indentation experiments made with in the thumb rule of 10% of the film thickness.

Key words: FEM, Nanoindentation, Loading/unloading characteristics, PMMA, UHMWPE

1. Introduction to Nanoindentaion of Bulk and Thin Polymers.

Thin polymer films are being widely used in electronic industries. Thermo- mechanical properties and residual stresses generated in polymeric films on Silicon substrates are of special interest in microelectronic packaging. Nanoindentation is being widely used to measure the mechanical properties, such as modulus, hardness and adhesion, with the emphasis on Thin Films due to its nanometer displacement resolution. However, the determination of the surface mechanical properties of polymeric materials with such techniques is relatively new. The research in the area has been primarily focused on polymer thin films, as an outgrowth of the microelectronics industry. The most extensively used method to extract Young's modulus and Poisson's Ratio of thin films is by Nanoindentation [1]. The materials response is assumed to be elastoplastic during loading and fully elastic in unloading [1]. One of the challenges in studying mechanical properties of thin films is that the traditional methods used to evaluate mechanical properties of bulk materials are not applicable for thin films. Thus there is a lacuna standard test method in the evaluation of mechanical properties of thin films. New methods, such as depth-sensing nanoindentation, micro bridge test, uniaxial tensile test and ultra sonic method are being developed [2, 3] for the measurement of mechanical properties of thin films. Among those methods, depth-sensing nanoindentation technique provides a continuous record of variation of indentation load with penetration depth into the specimen and this technique has been an area of considerable attention in recent years due to its high resolution at low load scale. Giannakopoulos and Suresh [3] have developed a step by- step method to obtain the mechanical properties of materials from the nanoindentation experimental data. Quasi-creep tests were performed to determine the viscoelastic material constants of Polydimethylsiloxane films and polystyrene (PS) bulk polymer by fitting the experimental data to a three-parameter model [4]. Then the work was extended to a finite difference analysis of

Corresponding author: *chepuri_srao@yahoo.co.in*

the data [5]. The expressions for contact load and pressure distribution during spherical indentation of a linear viscoelastic material have obtained by replacing the elastic modulus in the Hertz solution by hereditary integrals involving the relaxation response function [6]. A five-step indentation scheme is proposed to extract the elastic and visco elastic properties of polymeric materials using a sharp indenter [7]. Nanoindentation was used to study the visco elastic deformation of low modulus, ultra thin polymeric films deposited on a high modulus substrate [8].

2.0 BACKGROUND AND OBJECTIVE:

Computational simulation such as Finite Element Analysis (FEA) can be used to analyze the mechanics of deformation for thin films confined to substrates during nanoindentation. Bressan, Tramontin and Rosa [9] have modeled nanoindentation into bulk materials and thin films using finite element approach with ABAQUS. Procedures have been developed based on finite-element modeling of nanoindentation data to obtain the mechanical properties of thin films that deduce the yield strength, young's modulus and layer hardness from indentations as deep as 50% of the layer thickness or more[10]. Load-displacement characteristics of titanium thin films deposited on Silicon substrate were obtained by finite element modeling of nanoindentation with elastic-plastic approach [11]. FEM has been used to simulate the mechanical behaviour of bulk materials and thin films as a function of stress and strain calculations [12]. Effect of material thickness and nonlinearity on the mechanical properties of Polyelectrolyte thin films are studied by comparing the numerical simulation results with analytical results [13]. The Elastic modulus and hardness of bulk polymers like polymethylmethacrylate (PMMA) and, ultra-high molecular weight polyethylene (UHMWPE) are determined from the corresponding load-displacement material response [14].

In this paper, PMMA and UHMWPE thin polymers are indented with in the thumb rule of 10% thickness of the thin films to avoid substrate influence. The objective of the present paper is to simulate the nanoindentation process into PMMA and UHMWPE polymeric thin films in order to extract the elastic and visco elastic properties. Simulation has been carried out using FEM for the spherical nanoindentation of polymeric thin films to analyze the deformation field behavior as a function of stress – strain and time variation.

3.0. INDENTATION THEORY FOR LINEAR ELASTIC AND VISCOELASTIC MATERIALS

The Hertzian elastic contact model is the most widely used closed-form solution to calculate Young's Modulus of any material under generalized contact conditions. For small indentations (h_{max}<<R), the paraboloidal indenter is well approximated as a sphere for which the Hertzian elastic solution is available. The relation between the depth of a spherical indenter and the corresponding applied load was formalized by Sneddon [15] based on the Hertzian formulation:

$$P_{sphere} = \frac{4}{3}\frac{E}{\left(1-\nu^2\right)}\sqrt{Rh^3} \text{ ----------- (1)}$$

Where, ν is the Poisson's Ratio of the sample. Thus, E can be calculated directly from the Equation (1).It is important to note that Equation (1) tacitly assumes linear elastic behavior of a semi-infinite, indented material. The Hertz contact pressure distribution when a frictionless, rigid, spherical indenter of radius R is pressed onto a linear elastic solid is given by:

$$P_l = \frac{2E^*}{\pi R}\left(a^2 - r^2\right)^{1/2} \text{ -------------- (2)}$$

Here, the subscript 'l' refers to the linear elastic case. Also, r is the radial coordinate measured from the indenter axis, and $E^* = E/(1-\nu^2)$, where E^* is the reduced modulus. Further, the indentation depth $h_l(t)$ is related to the contact radius 'a' and load $P(t)$ by:

$$h_l = \frac{a^2}{R} = \left[\frac{3P(t)}{4E^*\sqrt{R}}\right]^{2/3} \text{ ---------------- (3)}$$

The hardness of the thin films is found by

$$H = \frac{P_{Max}}{A} = C_y \sigma_y \text{ ---------------- (4)}$$

Where P_{Max} = Load at maximum indentation depth, A= projected contact area,

C_y=constant=3, and σ_y is the yield stress.

The projected contact area is found by

$$A = C_0 h_c^{\,2} \text{ ----------------------- (5)}$$

Where C_0=24.5 for a Spherical indenter and h_c is the actual depth of contact and is given by

$$h_c = h_{max} - \varepsilon\frac{\left(P_{Max}\right)}{S} \text{ --------------- (6)}$$

Where 'ε' is a geometrical constant; ε=0.75 for a Spherical indenter and 'S' is the slope of the unloading curve

According to Lee and Radok [16], when Poisson ratio ν is time-independent, equations (2) and (3) can be utilized to obtain the following relations.

$$p(r,t) = \frac{2}{\pi R\left(1-\nu^2\right)}\int_0^t G(t-\xi)\frac{d}{d\xi}\left[\sqrt{a^2(\xi)-r^2}\right]d\xi \text{ --(7)}$$

$$h^{3/2}(t) = \left[\frac{a}{\sqrt{R}}\right]^3 = C_1\int_0^t J(t-\xi)\frac{dP(\xi)}{d\xi}d\xi \text{ ------(8)}$$

Where C_1= constant $= \frac{3\left(1-\nu^2\right)}{4\sqrt{R}}$, G(t) and J(t) are the relaxation and creep compliance functions in uniaxial compression. ξ is the integration variable for time The integrals on the right hand side of equations (7) and (8) can be evaluated by assuming a Prony Series representation. When load is applied gradually on the indenter and indentation depth is monitored as a function of time, the measured load-displacement data can be used to calculate the creep compliance (for ν =const) by

$$J(t) = \frac{\frac{8}{3}\sqrt{R}h_0^{3/2}(t)}{2P_0(1-\nu^2)} \text{ ----------------(9)}$$

Where $h_0(t)$ is the monitored displacement, R is the tip radius, P_0 is the maximum applied load.
If a stepwise indentation depth is applied and the load is monitored as function of time, then from the measured load-displacement data, one can calculate the relaxation modulus (for ν =const) by using

$$J(t) = \frac{2P(t)(1-\nu^2)}{\frac{8}{3}\sqrt{R}h_0^{3/2}} \text{ -------------- (10)}$$

4.0 FINITE ELEMENT MODELING

The modeling consists of three stages.

(i)Geometrical construction and Inputs.

The geometric construction consists of the selection of quadrant as diamond tip and the specimen as a cylinder and the contact is frictionless. Size of the PMMA thin films is 1 μm and it is 2 μm for UHMWPE. Radius (R) of the indenter is 100 nm.

(ii) Discretisation and Meshing

The indenter and Specimen are meshed by eight noded 2D solid structural element PLANE 82 with a total of approximately 2800 elements and 8000 nodes. Interaction of indenter and Specimen is considered as a contact pair, for which element Target 169 is applied to the specimen and Contact 172 to the indenter Tip. The mesh of the 2D model is shown in figure-1.

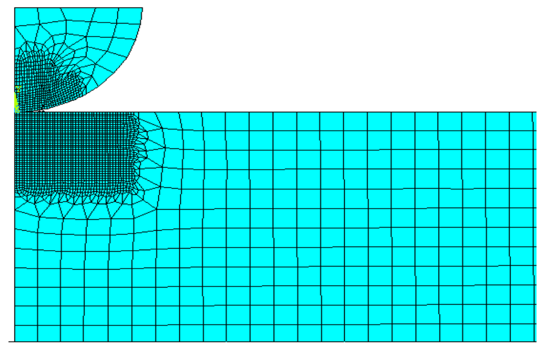


Fig-1.Mesh of the FE model

(iii) Loading and boundary conditions:

The boundary nodes in the axial direction are restrained to move in X-axis. The nodes at the bottom line of the half space are restrained to move in both X and Y directions.

Elements are finest in the central contact area and became coarser outwards. A displacement control technique is used to apply the load to the indenter. A Vertical displacement is applied to the chord portion of the indenter.

4.1 Simulation Procedure:

General purpose commercial software ANSYS 8.0 Multiphysics is used to simulate the nanoindentation into polymer thin films. Both the material films and the spherical indenter exhibit axial symmetry, and are thus considered in a two-dimensional simulation. Simulation is carried out for linear elastic and, separately, visco elastic cases for both the materials with the inputs as shown in Table-1. A displacement controlled analysis is implemented to simulate the nanoindentation in order to extract the reaction force 'P' corresponding to the indentation depth 'h' at each time point. A Vertical displacement is applied to the chord portion of the indenter and the reaction force at the bottom line of the half space is measured.

Assumptions

The simulation starts with the following assumptions as inputs and is shown in Table-1

Table-1. Inputs to the model

Material	Young's Modulus (E) GPa	Poisson's ratio ν	Material behavior
Diamond (Indenter)	1140	0.04	Elastic
PMMA (Thin Film)	6.75 [14]	0.38	(i)Elastic, (ii)Visco elastic
UHMWPE	0.61 [14]	0.36	(i) Elastic, (ii)Visco elastic

Three load cycles are applied to both PMMA and UHMWPE. Each cycle consists of two load steps. In first step, the load is ramped to peak value and in the later case; it is unloaded to zero value. Each load steps consists of 30 substeps. Minimum and maximum no of sub steps is 10 and 50.

For the visco elastic case, the materials analyzed are assumed to have the standard solid behavior and obey linear, isotropic visco elasticity. The maximum time period for which visco elastic response simulated is 100 seconds. Since for this duration the change in ν is negligible. The analysis is carried for ramp and hold load history in which the indenter is displaced to 100 nm by ramped loading and is held constant for 100 seconds. Each load step consists of 20 sub steps and maximum and minimum number of sub steps is 50 and 10 respectively. Maximum number of equilibrium iterations is 15.

After simulation, the out put obtained in linear elastic case is the Reaction force (F_Y) as a function of indentation depth and for visco elastic case, the same is as a function of time along with the stress distribution and strain variation in thin films. More appropriate post processing has made to plot the load-displacement, load-time and the displacement-time curves.

5. RESULTS AND DISCUSSION

Case-1: Linear elasticity

The load-displacement curves for PMMA and UHMWPE with maximum indentation depth of 60 nm and 200 nm respectively are shown figures-2 and 3.

By measuring the slope of the load-displacement curves, the average values elastic modulus and hardness of the thin films are calculated for the three indentation cycles. It is found that the hardness of the thin films is more than to bulk materials and it is due to reduction in scale size. Table-2 gives the elastic moduli and hardness value for thin films and bulk materials.

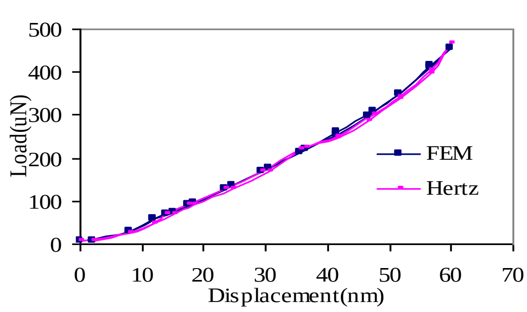


Fig-2. Load-displacement curve (PMMA)

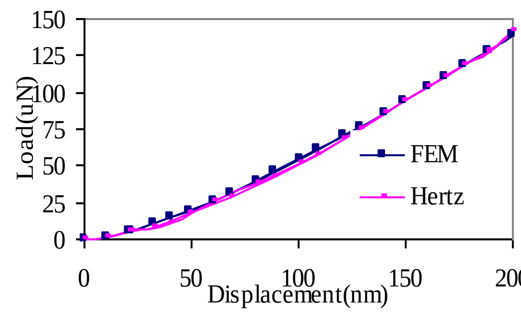


Fig-3. Load-displacement curve (UHMWPE)

The von mises stress distributions and strain variations for PMMA are shown in figures-4, 5 and figure-6 for the deformed, combined deformed and undeformed cases.

Similarly, the von mises stress distributions and strain variations for UHMWPE are shown in figures-7, 8 and figure-9 for the deformed, combined deformed and undeformed cases.

Table-2. Properties of thin films and bulk materials.

Material	E (GPa)	H (MPa)
PMMA thin film (FEM)	3.70	53.8
PMMA Bulk[14]	3.75	51.6
UHMWPE thin film (FEM)	0.58	13.2
UHMWPE bulk [14]	0.61	12.1

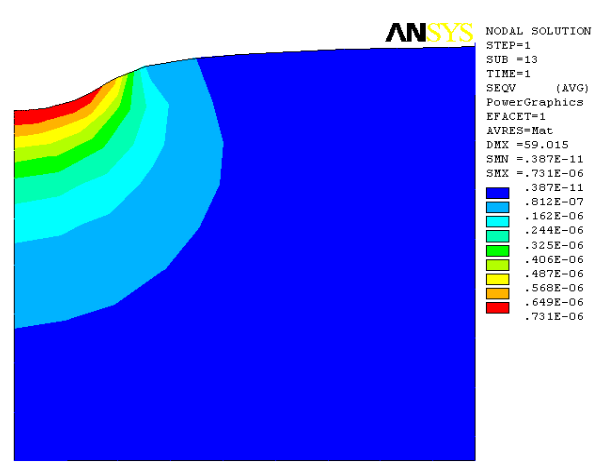


Fig-4. Von mises stresses (PMMA)

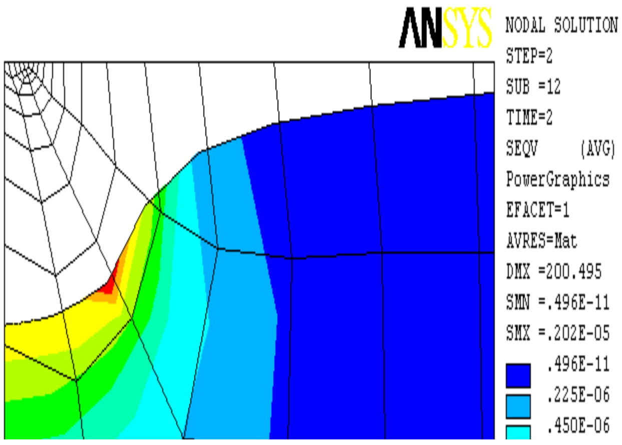


Fig-5. Von mises stresses for combined deformed and undeformed model (PMMA)

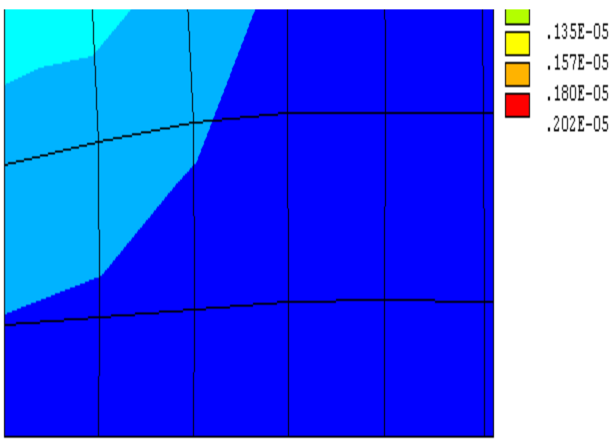


Fig-6. Strain contours (PMMA)

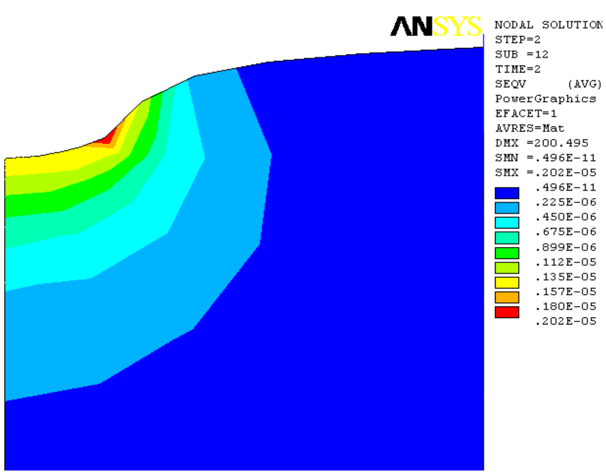


Fig-7. Von mises stresses (UHMWPE)

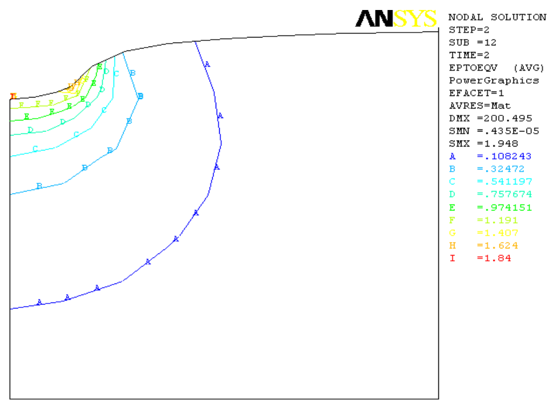


Fig-8. Von mises stresses for combined deformed and undeformed model (UHMWPE)

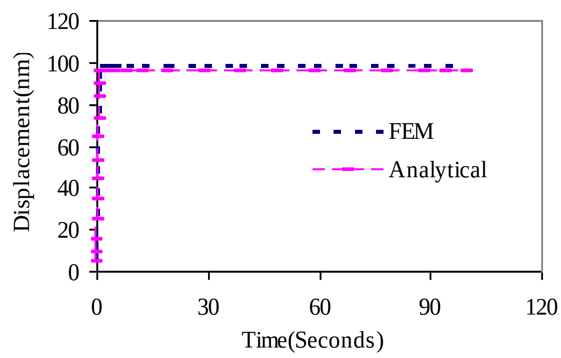
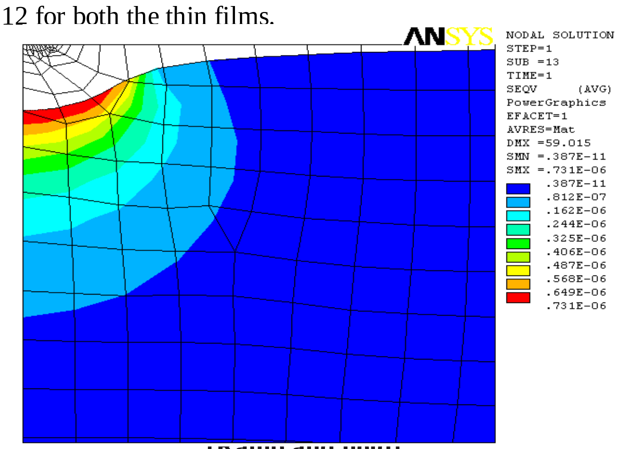


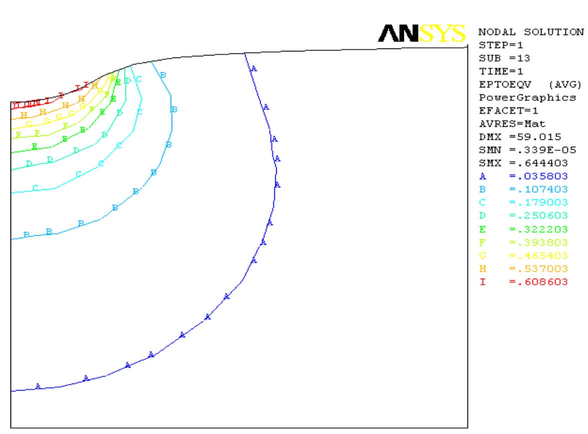
Fig-9 Strain contours (UHMWPE)

Case-2: Linear visco elasticity

The FE results for ramp and hold loading case is compared for PMMA with the analytical solution of Lee and Radok (equation 8), where the displacement is ramped to 100 nm and is held constant for 100 seconds (figure-10). With the obtained load-displacement data the stress relaxation modulus and creep compliance values are calculated and are shown in figures-11 and 12 for both the thin films.



(Ramp and hold)



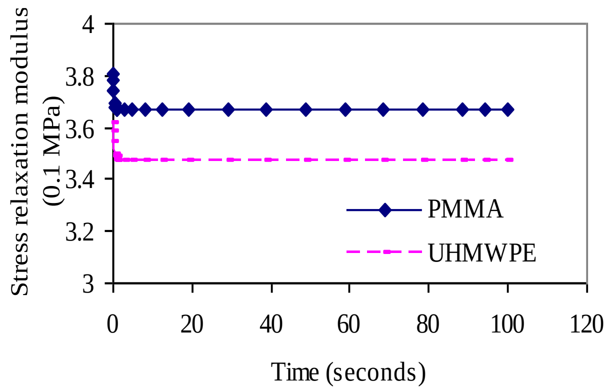


Fig-11. Relaxation response function

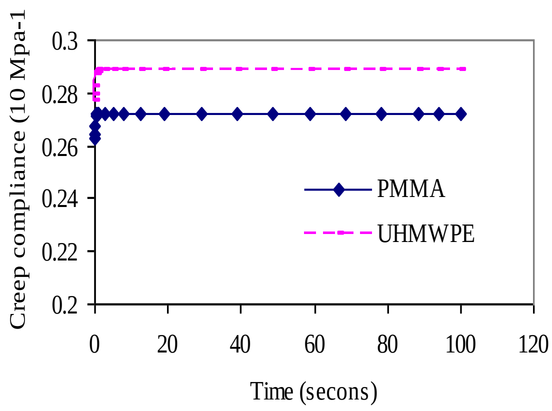


Fig 12. Creep response function

Validation:

The load-displacement data obtained by FEM is compared with the analytical data (Hertz) and a good correlation is found between the two. The hardness of the thin films is more than the bulk materials and is due to reduction in scale and fine grain size. From the measured stress-strain values, the yield strength of PMMA and UHMWPE thin films is found and are approximately 18 and 4.5 MPa respectively and follows the analytical equation-4. Thus the results for linear elasticity case are validated.

The Fe simulated results for visco elasticity case is compared with the analytical results of Lee and Radok and a close agreement is found between the two. Thus the results are validated for linear visco elasticity case also. The obtained load-displacement data is used to calculate the stress relaxation and creep compliance functions and are useful for testing the accuracy and validity of an experimental model.

6.0 Conclusions

The nanoindentation process is successfully simulated into polymer thin films by Finite Element Modeling. An attempt is made to extract elastic and visco elastic properties of polymer thin films PMMA and UHMWPE. Hardness of the thin films are calculated and are superior than to the bulk materials due to fine grain size at the nano scale. Ramp and hold loading is followed for the visco elasticity and the obtained load-displacement data by FEM is compared with analytical results. A good correlation found between the FEM and analytical results. The visco elasticity model used to calculate the stress relaxation and creep compliance functions may be used to test the accuracy and validity of an experimental model.

7.0 Nomenclature

Symbol	Meaning	Unit
P	Load	N
a	Contact radius	nm
h	Displacement	nm
h_{max}	Maximum indentation depth	nm
R	Radius of indenter	nm
A	Contact area	nm^2
ν	Poisson's ratio	
E	Elastic Modulus	GPa
E^*	Reduced Modulus	GPa
H	Hardness	MPa
σ_y	Yield Stress	MPa
G(t)	Stress relaxation modulus	MPa
J(t)	Creep Compliance	MPa^{-1}
ξ	Integration variable for time	

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