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Mechanical Characterization of Viscoelastic-Plastic Soft Matter Using Spherical Indentation

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Abstract: In this study, effects of the plastic deformation and the time-dependent deformation behavior on the fundamental relations in the Oliver & Pharr method are studied by using finite element analysis based on a viscoelastic-plastic model developed for polymers. The study eventually yields an experimental protocol and using which, the instantaneous modulus of the viscoelastic-plastic materials may be reliably determined. Experiments have been performed on four polymers to verify the conclusions from the numerical analysis.

Keywords: Nanoindentation; soft matter; viscoelastic properties; adhesion effects

1 Introduction

Nanoindentation tests are very attractive for characterizing the mechanical properties of various materials at the micro or nano-scale [Briscoe et al (1998), Liang et al (2004), Selvadurai and Yu (2005), Chen et al(2002), Qian et al.(2008)]. As a standard method in the commercial software, the procedure proposed by Oliver and Pharr (OP method) [Oliver and Pharr (1992)] by improving the method of Doerner and Nix [Doener and Nix (1986)] is widely adopted. However, recent studies [Hochstellar et al (1999), Ngan and Tang (2002), Ngan et al (2005), Cheng et al (2005a), Cheng et al (2005b), Tranchida et al (2006), Tranchida et al (2007a)] revealed that the mechanical properties of time-dependent materials determined using the OP method might exhibit significant errors. Hochstetter et al. [Hochstellar et al (1999)] showed that the OP method suffers from the effect of the time-dependent deformation behavior of polymers. They recommended using a holding segment and a fast unloading procedure to overcome the effect of creep. At the same time, they presented a new scheme to evaluate the projected contact area. Recently, Ngan

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et al. [Ngan and Tang (2002), Ngan et al (2005)] and Cheng et al. [Cheng et al (2005a)] have proposed novel procedures to determine the initial unloading slope for the indentation of viscoelastic materials. Cheng et al. [Cheng et al (2005b)] showed that for the indentation of viscoelastic materials, the indentation loads at a given indentation depth depend on the loading rates; but the initial unloading slopes corresponding to different loading rates are the same. This indicates that the contact depth determined using the procedure suggested by Oliver and Pharr depends on the loading rates. Thus, the Young's modulus given by the OP method in this case may be not reliable. Recently, et al. [Tranchida et al (2006), Tranchida et al (2007a)] have performed comprehensive analysis on the applicability of the OP method. They concluded that it was incorrect to use the OP method to the characterization of the polymers exhibiting viscoelastic-plastic deformation since the method is based on elastic analysis. Furthermore, they suggested that viscoelastic models should be applied to analyze the nanoindentation data [Tranchida et al (2006)]. The objective of the present study is to address the following important issue.

Ngan et al. [Ngan and Tang (2002), Ngan et al (2005)] and Cheng et al. [Cheng et al (2005a)] have proposed novel procedures to determine the initial unloading slope which can be reliably applied to both linearly viscoelastic materials and nonlinear viscoelastic materials. Taking into consideration that in the indentation of some polymeric materials and biological materials, the indented solids may undergo both *viscoelastic* and *plastic* deformation, it is interesting and useful to study the applicability of the procedures proposed by Ngan et al. [Ngan and Tang (2002), Ngan et al (2005)] and Cheng et al. [Cheng et al (2005a)] to these circumstances. In addition, in the presence of viscoelastic deformation, Cheng et al. [Cheng et al (2005a)] show that the OP method with or without the correction of Ngan et al. cannot be used to correctly evaluate the contact area. Thus, they propose to use the correlation between the contact depth and indentation depth to determine the contact area, but their method [Cheng et al (2005a)] is only for viscoelastic materials and ideal indenter geometries. In the case that the plastic deformation occurs or the indenter has tip defects, the method [Cheng et al (2005a)] may be invalid. Bearing this point in mind, it is still necessary and useful to investigate to what extent the contact area can be well determined by using the OP method, when the indented material simultaneously undergoes the viscoelastic and plastic deformation. Based on the objective above, research as outlined below has been performed.

First, computational modeling of the spherical indentation is carried out by using a viscoelastic-plastic model developed for polymers. The intention is to examine the applicability of the two fundamental relations in the OP method when the indented materials undergo viscoelastic and plastic deformation. Experiments are performed on four kinds of polymers, to validate the conclusions drawn from the numerical and theoretical analysis.

2 Computational study on the applicability of the fundamental relations in the OP method to the viscoelastic-plastic materials

Finite element analysis was performed by using a viscoelastic-plastic model [Kichenin et al (1996), ABAQUS (2006)]. For one-dimensional problem, the model is schematically shown in Fig. 1. E_v and E_p in the figure are the elastic moduli of the elastic-viscous network and the elastoplastic network, respectively. η is the viscosity coefficient. σ_y is the initial yield stress, g represents the work-hardening of the material, which is a function of the plastic strain. When σ_y is taken as infinity, the model degenerates to the well-known three-element linearly viscoelastic model. For the sake of simplicity, only perfectly plastic cases (i.e., g =0) are investigated in the present study. Piling-up of the indented material is more significant in plastic cases than that of the materials with the same ratio of the Young's modulus to the yield strength but exhibits plastic hardening. Detailed information of the model is reported elsewhere[15–16]. It is noted that Huber et al. [Tyulyukovskiy and Huber (2006),Huber et al (2008)] have proposed systematic inverse approaches to determine the material properties using indentation tests based on an elastic-viscoplastic model suited for metallic materials. The material model used herein developed for polymers, which can represent the viscoelastic-plastic deformation behavior of some typical polymers to the first order [Kichenin et al (1996)]. The relaxation time, τ for the model given by Fig. 1 is

$$\tau = \eta / E_v \tag{1}$$

The following property range is investigated in the present study: σ_y/E_p varies from 0.1 to 0.01 and E_v/E_p varies from 0.1 to 10. This property range should include many common polymers. The OP method based on the Sneddon's solution [Sneddon (1965)] deals with the contact of an axisymmetric indenter with an elastic half-space. We focus on spherical indentation in the present simulations using an axisymmetric model. Because the common diamond indenter is much harder than the typical polymers, the indenter is assumed rigid in the simulations. The indenter tip radius is taken as $R = 2\mu$m. The radius and the height of the substrate are taken as $50\mu\text{m}\times 50\mu\text{m}$. The boundary conditions on the indented solid are specified such that the outer surface nodes are traction-free with fixed lower surface nodes. A total of 10,000 four-node bilinear axisymmetric reduced-integration elements with hourglass control are used to model the semi-infinite substrate of the indented solid. The convergence of the simulations has been examined by comparing the results

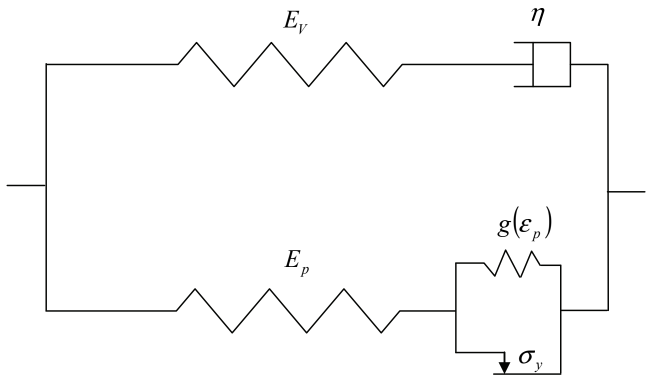


Figure 1: The viscoelastic plastic model used in the present analysis

with a refined mesh. The OP method is based on the following two fundamental relations[Oliver and Pharr (1992)]

$$E_r = \frac{\sqrt{\pi}S}{2\gamma\sqrt{A}} \tag{2}$$

$$A = f(h_c), \quad h_c = h_m - \kappa\frac{F_m}{S} \tag{3}$$

where E_r, S, A are the reduced modulus, the initial unloading slope, and the projected contact area, respectively. The contact area A is related to the contact depth h_c by the area function f. The correction factor γ in equation (2) used in most cases is due to King [King (1987)] in order to correct the effects of the indenter geometry. In recent years, there are some novel interpretations on this factor[Hay et al (1999), Cao et al (2007)] h_m and F_m are the maximum indentation depth and the load, respectively. κ is a constant, depending on the indenter shape; for a blunt tip, for instance the spherical indenter, κ is 0.75.

2.1 Examining the applicability of equation (2)

The fundamental relation given by equation (2) can be derived from the elastic contact theories. Based on Sneddon's solution [Sneddon (1965)], Pharr et al. [Pharr et al (1992)] have shown that equation (2) is valid for the contacts of an axisymmetric rigid indenter with arbitrary profile with an elastic half-space. Recently, Cheng and Cheng [Cheng et al (2005a), Cheng et al (2005c)] further showed that this fundamental relation is also true for the indentation of linearly viscoelastic solids provided the unloading rate be sufficiently fast. However, in the case that the indented materials exhibit both viscoelastic and plastic deformation, the applicability of equation (2) needs further investigation. In the present study, this fundamental relation is examined by using finite element simulations based on the material model given by Fig. 1. A trapezoidal displacement-controlled loading-holding-unloading protocol is used. In the sequel, the word of 'holding' always represents 'holding-at-the- maximum-depth' for the displacement-controlled indentation. t_h, t_l and t_u represent the holding, loading and unloading time, respectively. t_l/τ, t_h/τ and t_u/τ vary in a wide range. In the simulations, a wide property range as indicated above has been studied. The three sets of material properties (table 1) are used as examples to show the findings from the simulations. The results from simulations are reported in fig.2, the ratio of the maximum indentation depth to the indenter radius is taken as $h_m/R = 0.1$. Fig. 2 gives the initial unloading slopes corresponding to different unloading rates and the holding time. The results in the figure are normalized by the convergent solutions, which correspond to a very fast unloading rate ($t_u = 0.01\tau$). The results in Fig. 2 confirm the finding of [Hochstetter et al. (1999)] i.e., the holding time has a significant influence on the determination of the initial unloading slope. The results show that when the holding time is much longer than relaxation time (Fig. 2(a)) even for cases where the unloading time is comparable with the relaxation time, the errors in the initial unloading slope are still small. When the holding time is shorter (Fig. 2(b)), a faster unloading rate (e.g.$t_u < 0.1\tau$) is necessary to obtain a converging S.

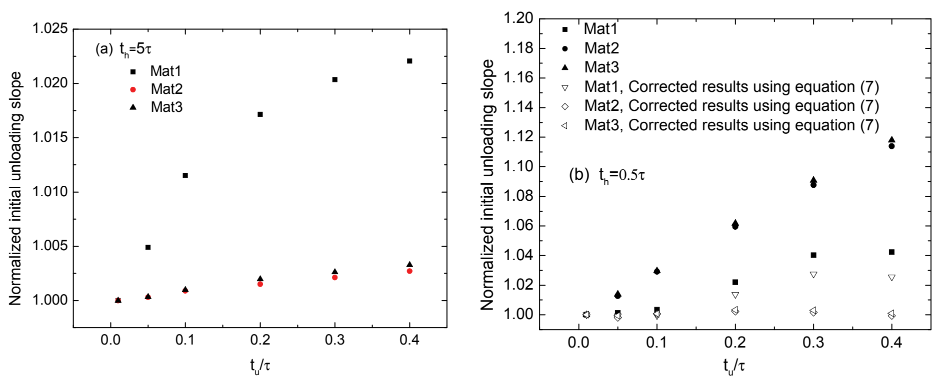


Figure 2: Effects of the holding-at-the maximum-displacement time and the unloading rate on the initial unloading slope. (a). t_h=0.5s; (b). t_h=0.5s

Using the converged initial unloading slope and the projected contact area obtained directly from the deformed finite element mesh at maximum indentation depth, the instantaneous modulus was determined via equation (2). The results are 2100MPa (Mat1), 2080MPa (Mat2) and 2001MPa (Mat3), respectively matching the exact solution (2000MPa) remarkably well. The computational results [Cheng et al (2005a)] in this study show that equation (2) is even valid for the indentation of the viscoelastic-plastic materials, if the suggested protocol could be followed. Besides the fast unloading protocol, Ngan et al.[Ngan and Tang (2002), Ngan et al (2005)]and Cheng et al. [Cheng et al (2005a)] have proposed novel procedures to determine the initial unloading slope for the indentation of viscoelastic materials. For a displacement-controlled indentation, Cheng et al. [Cheng et al (2005a)] showed that the following relation holds true

$$E_r = \frac{\sqrt{\pi} S_e}{2\sqrt{A}} \tag{4}$$

where S_e is given by

$$S_e = \frac{dF}{dh}\big|_{h=h_m} + \frac{dF/dt\left.\right|_{t_m^-}}{v_h} \tag{5}$$

where $\frac{dF}{dh}\big|_{h=h_m}$, $dF/dt\left.\right|_{t_m^-}$, v_h are the initial unloading slope, the relaxation rate of the load immediately before the unloading and the unloading rate, respectively. Equation (5) is also examined by using finite element analysis and the three sets of material properties as given in table 1. It is interesting to find from the computational results that although equation (5) derived from the indentation of linearly viscoelastic materials, it works very well also for the indentation of viscoelastic-plastic materials as represented by Fig. 1 (see Fig. 2(b) for detail). Equation (5) is used in explaining the importance of holding time. The term $dF/dt\left.\right|_{t_m^-}$ in equation (5) depends on the holding time; and for a long holding time, $dF/dt\left.\right|_{t_m^-}$ can be very small. In this case, a fast unloading rate may be not necessary. The numerical analysis above shows that equations (2) and (5) may be applicable to the spherical indentation of the viscoelastic-plastic materials. Now the key issue is accurately determining the projected contact area A appeared in equation (2). This point will be addressed in detail below.

2.2 Examining the applicability of the fundamental relation given by equation (3)

In the OP method, the contact depth is determined from equation (3). Cheng et al. [Cheng et al (2005a)] point out that equation (3) is not responsible for the inapplicability of equation (3) Ngan et al. [Ngan and Tang (2002)]. The reason responsible for the inapplicability of equation (3) according to Cheng et al. [Cheng et al

(2005a)] may be given as follows. In the indentation of time-independent materials, one-to-one relationship exists between the maximum indentation load and the depth in a single loading-unloading loop. However, in the indentation of time-dependent materials (e.g. polymers) the maximum indentation depth may correspond to different indentation loads depending on the loading rate and the holding segment for a displacement-controlled indentation (see Fig. 7 in ref. [Cheng et al (2005a)]). Thus, equation (3) does not give the unique contact area. In order to solve this problem, we may consider two extreme cases for the indentation of visoelastic plastic materials as given by Fig. 1. First, if the loading-unloading time is much shorter than the relaxation time, the material may behave like a time-independent elastoplastic material. The Young's modulus of the material in this case represents the instantaneous modulus. Second, if both the loading and unloading rates are very slow, such that the loading and unloading segments are much longer than the relaxation time, the material may also behave like a rate-independent elastoplastic material. In this case, the Young's modulus of the material can be regarded as the long-term modulus. In the present study, the instantaneous modulus is the main concern; thus, we will investigate the possibility to achieve the fast loading and fast unloading protocol corresponding to the first case. In equation (3), the initial unloading slope may be determined either from the fast unloading procedure or from equation (5) as proposed by Cheng et al. [Cheng et al (2005a)]. In theory, well determination of the contact area by using equation (3) requires the use of the maximum indentation load corresponding to a sufficiently fast loading rate and the beginning point of the holding segment (if the holding segment is used). In the OP method, the maximum indentation load at the end of the holding segment is applied; this is incorrect in theory and may lead to significant errors. In practice, we face the question of how fast the loading rate should be. We attempt to answer this question by using the finite element analysis. Based on the computational results, it is found that when the loading time is much shorter than the relaxation time, e.g. $t_l < 0.1\tau$, the maximum indentation load at the beginning of the holding segment converges to a constant value (see Fig. 3). Summarizing the analysis above, we suggest a protocol as illustrated by Fig. 4. The key issue in the protocol is that the maximum indentation load (F_m in the figure) corresponding to the fast loading procedure, which appears at the beginning point of the holding-segment, is used in the determination of the contact area.

The initial unloading slope is determined from the unloading curve resulted by a fast unloading rate or equation (5). Using the protocol, the OP method may provide a good estimation on the instantaneous modulus of polymers. We verified this specific protocol by using numerical experiments. The instantaneous modulus determined by using the protocol, equations (2) and (3) for the three sets of material

Table 1: Materials used in the simulations to examine the two fundamental relations in the OP method (for all the cases, Poisson's ratio is taken as 0.4)

Materials	E_v (MPa)	E_p(MPa)	σ_y(MPa)
Mat 1	200	1800	18
Mat 2	1800	200	2
Mat 3	1800	200	20

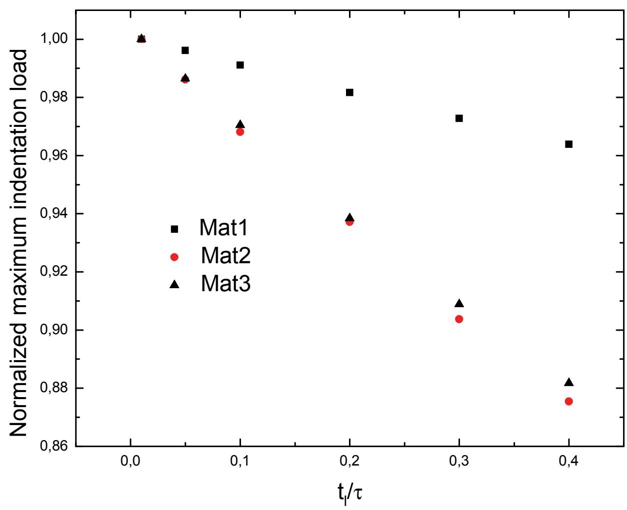


Figure 3: Effect of the loading rate on the maximum indentation load (at a given indentation depth)

parameters as given above are 2062MPa (Mat1), 2045MPa (Mat2) and 2041MPa (Mat3), respectively. Comparing the exact solution (2000MPa) to results obtained using the OP method based on the protocol as identified above, gives a quite reasonable evaluation of the instantaneous modulus. It is noted that many polymers have the relaxation time of tens or hundreds seconds, thus in practice it is possible to achieve the required fast loading condition by applying various commercialized indenters in the market.

2.3 *Examining the effects of piling-up on equation (3)*

Cheng et al. [Cheng et al (2005a)] suggest to use the simple correlation between the indentation depth, h, and the contact depth, h_c, (i.e., h (t) $=2h_c$ (t) for spherical indentation) to determine the contact area. This relation is only valid for the

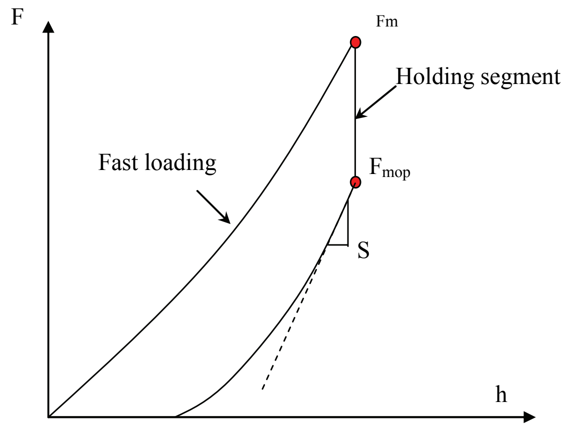


Figure 4: The experimental protocol suggested in the present study (displacement-controlled indentation)

indentation of linearly viscoelastic materials and ideal indenter geometries. When plastic deformation occurs or the indenter deviates from the ideal shape, the correlation used by Cheng et al. [Cheng et al (2005a)] may be invalid. In this case, the fast loading protocol suggested above may be useful. In theory, the fast loading procedure is not always necessary due to the plastic deformation. When the plastic deformation occurs, the extent of sinking-in of the indented material may be largely reduced. In this case, the ratio of κF_m/ S to h_m (see equation (3)) can be very small; and the effects of the uncertainties in the maximum indentation load caused by the time-dependent deformation behavior of the materials may be negligible. It is also noted that the indented material may exhibit piling-up when it undergoes plastic deformation. In this case, equation (3) shows that the OP method will underestimate the projected contact area and thus overestimate the reduced modulus. Hence, evaluating the applicability of equation (3) to viscoelastic-plastic materials requires a careful examination of the development of piling-up under the spherical indentation. For the spherical indentation of time-independent elastoplastic materials: the extent of piling-up depends on the materials properties, and the ratio of the indentation depth to the indenter radius [Oliver and Pharr (2004)]. According to the computational results, the following information has been obtained. First, the larger the ratios of the indentation depth to the tip radius and E_p to σ_y are, the larger the ratios of the contact depth to the indentation depth will be. This phenomenon is similar to that observed in the indentation of time-independent elastoplastic materials. Second, for the same ratios of the indentation depth to the tip radius and E_p

to σ_y, the larger the ratio of E_p to E_v is, the greater the ratio of the contact depth to the indentation depth will be. We have examined a critical case for which $E_p/\ \sigma_y$, =100, E_p / E_v =0.9 and h/R=0.2, the ratio of the contact depth to the indentation depth is smaller than 1.03; thus piling-up is indeed small.

3 Experimental

In order to validate the conclusions above from the numerical analysis, experiments are performed. Polymers ranging from amorphous to semi-crystalline structure and from glassy to rubbery mechanical behavior are selected, namely, low-density polyethylene (LDPE), polystyrene (PS), polyethylene terephthalate (PET), and polypropylene (PP) (GoodFellow). Local surface roughness for all samples was around 2nm as measured by atomic force microscopy (AFM) (Veeco Metrology Group, Dimension^{TM} 3100). Nanomechanical characterization was performed for all polymers with a Hysitron Triboscope (Hysitron Inc.). The Triboscope operated in a closed loop feedback system, which controls the displacement applied to the indenter by means of an electrostatic transducer [Tribscope, (2003)]. A spherical indenter with a 5μm tip radius was applied. Field Emission Scanning Electron Microscope (FE-SEM, LEO 1550VP, GEMINI) images were made to map the tip radius. The area function requires a precise calibration when using the OP method. We had the following difficulties in using quartz as a calibration material. First, it had a relatively high surface roughness (12nm) in comparison to the indentation depth. Second, the maximum indentation depth with the Triboscope was only 100nm for quartz. For this reasons, we used PMMA (polymethyl methacrylate) as a calibration material. Moreover, PMMA is the case where the influence of surface forces is weak [Owens (1969)]. The area-function was determined for a contact depth ranging from 5nm to 300nm. The calibration was repeated several times and no significant change in the area function was observed. We performed indents on polymers using the displacement control mode from 5nm to 200nm.

The zero point load or initial contact force is the force used by the indenter to identify the sample surface. For indentation loads above 25μN, the initial contact load was 2μN and for indentation loads between 5μN and 15μN, a set point load of 0.5μN was used [Tribscope, (2003)]. Each indent was made using a trapezoidal loading function in which the indentation depth was first increased to the maximum set value and then held at the peak value before unloading. In each case, an array of ten indents at five different places was made for all chosen polymers. The maximum thermal drift was 0.027nm/s before starting the indentation. Drift correction was used to minimize thermal effects. All the experiments were performed at room temperature.

The numerical analysis above shows that besides the unloading rate, the loading rate is also very important for the determination of the contact area. If sufficiently fast loading rates are used the loading curve may converge [Tranchida et al (2007b)]; otherwise, the maximum force corresponding to maximum depth is different for different loading rates. To obtain a sufficiently fast loading rate, loading segment times of 0.5s, 5s and 50s were tested. Thus, for the same indentation depth the lower the segment time is, the faster the loading rate will be. A loading segment time of 50s was not sufficient for both rubbery and glassy polymers. For PET and PS a sufficient loading condition was obtained with loading segment time of 5s, whereas for LDPE, and PP it was 0.5s. The fastest possible unloading segment time of 0.5s is applied.

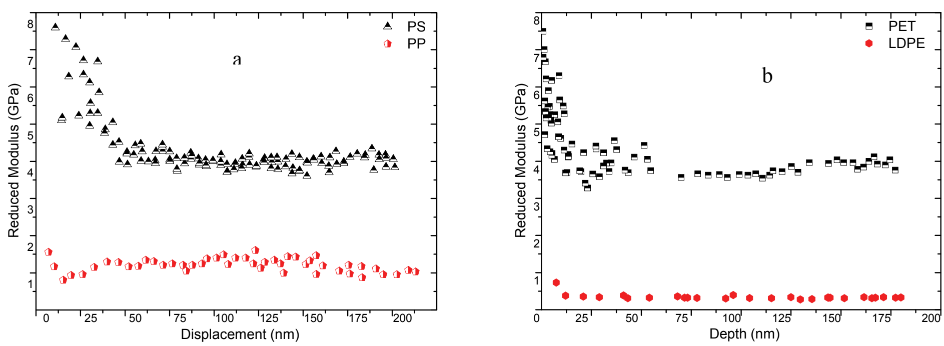


Figure 5: Depth dependent reduced modulus for all polymers, also showing the scatter in data.

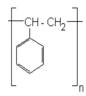
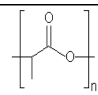
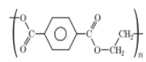
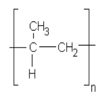
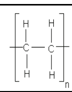
In summary, the protocol used for attaining the instantaneous modulus were a loading segment time of 0.5s or 5s, holding-at-the-maximum-displacement with segment time 10s, and unloading segment time of 0.5s, respectively. Using the protocol suggested in section 2 and the OP method, the instantaneous modulus for PP, PS, PET, and LDPE are determined, fig. 5. The results in fig. 5 show that the reduced moduli obtained herein are consistent with the reference values from the supplier of the materials and the values reported in the literature for indentation depths larger than certain critical values. When the indentation depths are smaller than the critical values, the instantaneous moduli are overestimated; this phenomenon may be due to the effects of adhesion [Cao et al (preparation), Falsafi et al(1997), Wahl et al(2006), Cao et al (2005)].

The key issue in the protocol suggested based on the numerical analysis is to use the fast loading procedure and the maximum load (F_m) appearing at the beginning of the holding segment (fig. 4). The reduced modulus from this non-standard analysis was compared to the OP method, to give an estimate of the error that may occur when using a standard OP analysis (maximum load (F_{mop}) appearing at the end of the holding segment). To make a valid comparison for E_r we used data beyond the critical depth (30nm) to avoid the influence of adhesion. An error of 10% was noticed in the reduced modulus, when using the standard OP method for depths around 40nm. This error decays exponentially at depths of about 100nm for both glassy and rubber polymers. These results further confirm that the protocol is more useful when the viscoelastic deformation is dominant. At comparatively greater depths, polymers undergo plastic deformation so that the effect of the ratio F_m/Son the contact depth is negligible. Thus, we observed that plastic deformation might be an advantage when estimating the reduced modulus of viscoelastic-plastic solids.

4 Discussion

The OP method based on the Sneddon's solution [Sneddon (1965)] deals with the contact of an axisymmetric indenter with an elastic half-space. However, in practice the OP method is always applied to analyze the nanoindentation data from a Berkovich indenter; and in the analysis, the Berkovich indenter is assumed equivalent to a cone. Lim and Chaudhri [Lim and Chaudhri (2006)] argued that such equivalence might be invalid for the indentation of polymers. The analysis in section 2 is for a spherical tip; the conclusions may be true also for other axisymmetric blunt tips. However, caution is shown that the model applied in the present study can represent the deformation behavior of some typical polymers at least to the first order [Kichenin et al (1996)]. Nevertheless, in the case that the deformation of the polymers is very complicated, the model may give a poor approximation and the conclusions drawn herein may be invalid. For instance, in the indentation of polymers using a very sharp Berkovich indenter, the indented materials may exhibit very complex deformation and relaxation mechanism [Tranchida et al (2007), Fujisawa and Swain (2007), Fujisawa and Swain(2008)]; and the model given by Fig. 1 may give poor approximation. In addition, the deformation of the polymers strongly depends on the indenter geometry. It has been reported that for the Berkovich indentation, the area functions from the same indenter are different for different calibration polymers [Tranchida et al (2007a)]. In our study, we also find this phenomenon. However, it interesting to find that for spherical indentation the area functions are basically consistent for different polymers [Balasundaram et al (2009)]. This finding deserves further investigation.

Table 2: Investigated polymers with chemical structure.

Polymers	Chemical structure	Reduced modulus GPa
PS		4.35
PMMA		3.21
PET		3.75
PP		1.42
LDPE		0.27

5 Conclusions

Using finite element analysis based on a viscoelastic-plastic model, the two fundamental relations in the OP method are examined. The analysis shows that the correlation between the initial unloading slope, reduced modulus and the projected contact area (equation (2)) may hold true even the indented material undergoes viscoelastic and plastic deformation simultaneously. The condition is that the contact stiffness should be determined by using the fast unloading protocol or corrected by following the procedures reported by [Ngan and Tang (2002), Ngan et al (2005)] or Cheng et al. [Cheng et al (2005a)]. Recently, Cheng et al. [Cheng et al (2005a)] showed that equation (3) with or without the correction of Ngan et al. [Ngan et al. Ngan and Tang (2002), Ngan et al (2005)] is inapplicable to the indentation of viscoelastic materials. The present study shows that such a failure may be overcome by using the fast loading procedure and appropriate maximum indentation load as shown in fig. 4. Using such a protocol, the OP method may still apply to the indentation of viscoelastic or viscoelastic-plastic materials. Experiments performed on four kinds of polymers i.e., PET, LDPE, PP, and PS validate the protocol identified from numerical analysis.

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