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Length scale dependence in elastomers – comparison of indentation experiments with numerical simulations

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ABSTRACT

Probing depth dependent deformation at nano- and micrometer length scales has been observed in indentation experiments of polymers. Unlike in metals, where size effects are observed in plastic deformation and are attributed to geometrically necessary dislocations, the origin of size dependence in polymers is not well understood. As classical continuum theories are unable to describe such phenomena, higher order gradient theories have been developed to capture such size dependent deformation behavior. The present study adopts the penalty finite element approach for a couple stress elasticity theory under axisymmetric conditions to numerically simulate and analyze the probing depth dependent deformation. Polydimethylsiloxane (PDMS) and natural rubber have been used as model materials to analyze the depth dependent deformation at different probing depths. Simulations were performed on PDMS using spherical indenter tips of different radii to show the influence of strain/rotation gradients on elastic modulus. To capture the experimentally observed increase in hardness with decreasing probing depth, simulations applying a conical indenter tip were performed and compared with experimental data.

1. Introduction

Depth sensing indentation testing has been widely applied to determine hardness, elastic modulus, as well as to study deformation mechanisms at micro- and nanometer length scales. Numerous indentation experiments conducted on metals [1–3] and polymers [4–16] demonstrated that the hardness is significantly higher at small probing depths. In Refs. [11,13,17] the length scale dependence in polymers has been experimentally observed in elastic deformation, which is in contrast to metals, where length scale effects were observed in plastic deformation and usually attributed to geometrically necessary dislocations [2]. Although there is mounting experimental evidence for size effects in polymers [4–20], compared to metals, length scale dependent deformation in polymers is arguably not well understood and there are only few length scale dependent theories suggested in the literature [5,6,21,22] for polymers.

Length scale dependent phenomena cannot be explained nor predicted by classical, local continuum theories. This has led to the development of phenomenological continuum theories with higher order gradients in the displacements where the material length scales are introduced into the deformation energy [23–26]. Despite considerable theoretical and computational contributions on the analysis of length scale dependent phenomena with higher-order gradient theories, there are only very limited numerical studies [27,28] in the literature that examine the probing depth dependent deformation in polymers.

It has also been observed that the determined elastic moduli of some polymers like epoxy [12], polydimethylsiloxane (PDMS) [13] and natural rubber [14] increases with decreasing depth when Sneddon's theory together with a pyramidal tip is applied, whereas no change in the determined elastic moduli of these polymers have been observed with depth when the Hertz theory along with a spherical tip was applied. It was hypothesized that the differences between the determined elastic moduli using spherical and pyramidal tips are attributed to the higher order gradients of displacements, as it is argued that these gradients remain essentially constant with a spherical tip, while applying a conical/pyramidal tip these gradients increase with decreasing probing depth.

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The goal of this paper is to compare the experimental results with numerical simulations based on the axisymmetric couple stress approach developed in Ref. [29] and to examine whether the change in hardness with respect to the probing depth can be predicted by the couple stress elasticity theory. Simulations were carried out with spherical and conical indenter (equivalent to pyramidal tip) tips on PDMS as a model material and compared with the experiments to interpret the probing depth dependent deformation behavior by considering the rotation gradients. Also, the simulations with a conical tip were performed on natural rubber and compared with the experimental data. In the next section, results obtained from the indentation type experiments are briefly discussed. In the section thereafter, the influence of the rotation gradients with respect to different tip geometries is suggested and a rotation gradient model to predict such a size dependent behavior (when conical tips are applied) is discussed. The penalty approach of the applied couple stress axisymmetric elasticity theory of [29] is then applied to perform numerical simulations on PDMS and natural rubber at different probing depths using different tip geometries.

2. Experiments

The universal hardness H_U, also known as Martens hardness, is obtained from load-displacement data and can be calculated in accordance with ISO 14577-1 [30] as follows

$$H_U = \frac{F_{max}}{A_s}, \tag{1}$$

where A_s is the nominal surface area of the indenter tip penetrating beyond initial contact with the material sample under the maximum applied force F_{max}. For the Berkovich indenter tip, the nominal surface area is given as

$$A_s = C\,h^2, \tag{2}$$

where h corresponds to the probing depth at F_{max} and C is a constant equal to 26.43. It should be noted that the expression “indentation” is usually applied to tests inducing permanent residual impressions on the sample. The experiments on elastomers in Refs. [11,13,14], however, showed almost pure reversible, elastic deformation after fully removing the load. Therefore, we adopt “probing depth” in this article to describe the experimental results and their corresponding numerical simulations. It can be seen in Fig. 1 that the ratio of the inelastic deformation work (area enclosed by the curve) to the total deformation work is small indicating the

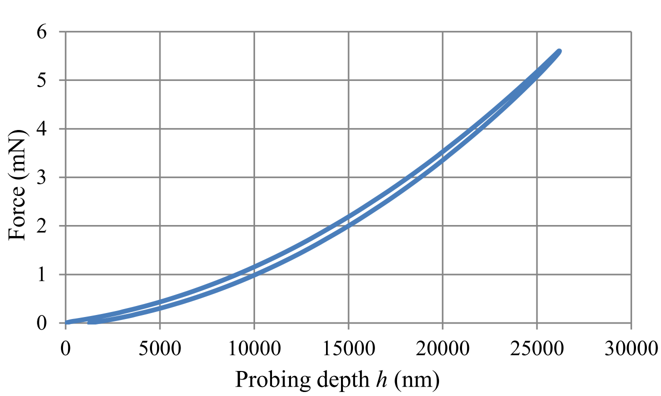

Fig. 1. Force versus probing depth h with the Berkovich tip (unpublished data of experiments in Ref. [11]).

elastic nature of the deformation mechanism during nanoindentation of PDMS.

Unlike the indentation hardness [31], which is evaluated by a projection technique to approximate the contact area of the tip with the sample at the maximum depth, the universal hardness is defined as the force divided by the nominal surface area (the area that is nominally below the initial surface). In this respect, it should be noted that the indentation hardness has been originally developed for ceramics and metallic materials subjected to plastic/permanent deformation and may not be applicable for the highly elastic materials considered here [8,30]. As the universal hardness H_U incorporates both plastic and elastic deformations, it is actually applicable to all materials.

The indentation type experiments conducted on PDMS in Ref. [11] resulted in an increase in H_U (from 0.28 to 0.44 MPa) with decreasing h (from 31.5 to 6.0 μm). Recent experiments [12–14] exploring size dependence associated with probing of polymers with a diamond spherical tip have revealed that for a fixed radius of a spherical indenter tip, the determined elastic moduli showed negligible changes with the changing probing depth, i.e. elastic modulus is independent of h. The elastic modulus E_P of PDMS in Ref. [11] was determined according to the Hertzian theory and Sneddon's theory by applying spherical and pyramidal Berkovich tips, respectively. It was assumed that both spherical (curvature radius of 250 μm) and Berkovich tips are rigid as the elastic modulus of tip material (diamond) is many orders of magnitude higher than that of PDMS.

Hardly any changes in determined elastic moduli of PDMS were observed experimentally when spherical indenter tips were applied at different probing depths. According to the Hertzian theory [32], the elastic modulus of the polymer E_p^{Hertz} can be obtained as

$$E_p^{Hertz} = \left(1 - \nu_p^2\right)E_r, \tag{3}$$

where ν_p is the Poisson's ratio of the polymer and E_r is the reduced elastic modulus obtained from the load-displacement curve as

$$E_r = \frac{3}{4}\frac{F}{R^2}\left(\frac{R}{h}\right)^{\frac{3}{2}}, \tag{4}$$

where F is the applied force, R the curvature radius of the spherical tip, and assuming linear elasticity and a small ratio of contact area to R. In contrast to spherical tips, the elastic modulus determined with a conical tip applying Sneddon's theory [33] $E_p^{Sneddon}$ was found to increase (from 2.64 to 4.68 MPa) with decreasing h (from 31.5 to 6.0 μm). According to Sneddon's theory for frictionless indentation of elastic materials with conical tip, the elastic modulus can be determined from load-displacement relation as

$$E_p^{Sneddon} = \frac{F\pi\left(1 - \nu_p^2\right)}{2\tan\alpha\, h^2}, \tag{5}$$

in which $\alpha = 70.3°$ is a half angle defining a cone equivalent to a Berkovich tip. For reference, the results obtained from these experiments are illustrated later.

3. Non-local, rotation gradient theory

To predict the length scale effects mentioned above, atomistic simulations may be tempting, as length scale effects have been found to be dependent on polymer molecular structure [7,15]. While molecular simulations may provide some basis for general trends [34,35], they have their limitations as (i) deformation in