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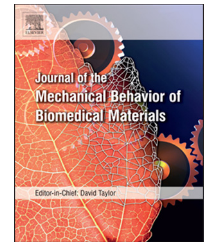

Research Paper

Influence of specimen thickness on the nanoindentation of hydrogels: Measuring the mechanical properties of soft contact lenses

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ABSTRACT

Nanoindentation offers a convenient method for the testing of thin hydrogel specimens, such as contact lenses, to directly assess their mechanical properties. Here we investigate the mechanical properties of poly(hydroxyethyl methacrylate) (pHEMA) specimens of a range of uniform thickness values and demonstrate that, with 50 and 100 μm radius spherical indenters, a significant increase in apparent elastic modulus is seen when the specimen thickness is smaller than 500 μm at indentation depths <1 μm. This is a manifestation of the well known indentation thickness effect but occurring at larger critical thicknesses than seen with other materials. A simple empirical relation is determined for the variation in apparent elastic modulus with normalised thickness. The empirical thickness correction function obtained from pHEMA specimens was subsequently used to correct for the thickness variation within a range of contact lenses supplied by a number of different manufacturers fabricated from both pHEMA and silicone polymers, with a range of optical strengths and hence thickness profiles. The correction function is seen to compensate for the variation in apparent elastic modulus with lens thickness for all four contact lens types, irrespective of lens material. The measured Young's modulus of the contact lens material, corrected for thickness, was compared with that quoted by the manufacturers of the contact lenses, obtained by conventional bulk mechanical testing, to find good agreement.

1. Introduction

Hydrogels are a generic term for highly crosslinked polymers that have a nanoporous internal structure that is filled with aqueous media. Such materials tend to be highly compliant with elastic modulus values many orders of magnitude lower than engineering polymers. Many of the soft tissues within the human body can be classified as hydrogels as also are

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many biomedical materials such as alginates, agarose gels and contact lens materials such as poly(hydroxyethyl methacrylate) (pHEMA). In order to fully understand the compatibility of hydrogel biomedical materials with soft tissue, it is necessary to develop mechanical testing methods to accurately characterise the mechanical behaviour of both materials. In many biomedical applications there is the need to characterise relatively small volumes of materials, in which case nanoindentation has many attractions (Ebenstein and Pruitt, 2006; Akhtar et al., 2011). This is particularly the case for contact lens materials where the thickness of the sample is $<500\,\mu m$ and nanoindentation based techniques have the following advantages:

- It is possible to measure the mechanical properties of a whole manufactured lens.
- It is easier to test the lens in a hydrated environment.
- It is possible to study variation in mechanical properties across a lens.

1.1. *Contact lens materials*

Soft contact lenses are manufactured from hydrogels, which are able to achieve an appropriate balance of materials properties including: optical properties, oxygen permeability, surface wettability, whilst displaying similar mechanical properties to corneal tissue. Most commercial contact lenses are fabricated from two distinct families of hydrogel materials.

- Conventional hydrogels: these are most commonly based on copolymers of poly(hydroxyethyl methacrylate) (pHEMA) and other hydrophilic monomers, such as methacrylic acid (MA) or *N*-vinyl pyrrolidone (NVP); they are usually crosslinked with ethyleneglycol dimethacrylate (EGDMA) (Maldonado-Codina and Efron, 2003).
- Silicone hydrogels: these are based on silicone polymers, including methacryloxypropyl(trimethylsiloxy)silane (TRIS) copolymerised with hydrophilic monomers, such as pHEMA or copolymers of flurocarbons with poly(dimethyl siloxane) (PMS) (Kunzler, 1996; Nicolson and Vogt, 2001).

Silicone hydrogels were brought onto the market in the late 1990s in order provide the anterior eye with more oxygen and to solve many of the hypoxia-related problems observed with conventional hydrogels, which had been on the market since the 1960s. Consideration of appropriate mechanical properties for contact lens materials are a vital part of the lens design because they are directly related to factors such as comfort, optical performance, handling and the conformal fit of the lens to the ocular surface.

Young's modulus and tensile strength are routinely measured by contact lens manufacturers and researchers (Maldonado-Codina and Efron, 2004; Tranoudis and Efron, 2004). Hydrogels are known to display more complex mechanical behaviour than described by simple infinitessimal strain deformation theory because they can withstand large elastic strains and show time-dependent deformation. Various authors have proposed models based on hyperelasticity (Ravi et al., 2006), rubber elasticity (Peppas et al., 2006), viscoelasticity (Ahearne et al., 2005) and poroelasticity (Galli et al., 2009), to more accurately describe the behaviour of hydrogels. However, because of the low loads and consequent deformations they are exposed to during use, manufacturers of contact lenses appear to rely on a nominal Young's modulus as the appropriate metric. Young's modulus is also the default metric used to compare the mechanical properties of biomaterials with the tissue they will be in contact with during service.

1.2. *Indentation and nanoindentation of hydrogels*

Hertz's solution for the indentation of an elastic half-space by a sphere, relates the displacement, h, as a function of indentation load, P, with:

$$h=\left(\frac{9P^2}{16RE^{*2}}\right)^{1/3} \tag{1a}$$

here R is the radius of the indenting sphere and E^* is the contact modulus, defined by

$$\frac{1}{E^*}=\frac{1-\nu_1^2}{E_1}+\frac{1-\nu_2^2}{E_2} \tag{1b}$$

where E and ν are Young's modulus and Poisson's ratio, with the subscript 1 indicating the material of the indenter and subscript 2 that of the half-space being indented respectively (Johnson, 1985). Hydrogels are orders of magnitude more compliant than conventional engineering materials, thus one can approximate the indentation of a hydrogel to that caused by a rigid indenter, in which case Eq. (1b) can be simplified thus

$$\frac{1}{E^*}=\frac{1-\nu_2^2}{E_2} \tag{1c}$$

From this it is clear that indentation and nanoindentation experimemnts measure a contact modulus. Thus in order to obtain a measure of Young's modulus from indentation data, Poisson's ratio must either be measured independently or an assumed value taken.

The use of contact atomic force microscopy (AFM) and nanoindentation to characterise highly compliant materials such as hydrogels and animal tissues must be treated with some caution because of the inherent assumptions carried out during the analysis of data. For most cases of AFM analysis of contact stiffness, it is assumed that the contact can be modelled using Hertz's elastic solution (Eq. (1)); with the substrate taken as an infinite half-space and that the local strains are small enough to allow the infinitesimal strain approximation in linear elasticity theory. This approach was reviewed by Dimitriadis et al. (2002), who showed that it is necessary to use relatively large indenting spheres to ensure that the elastic strain beneath the indenter remains in a linear elastic regime with practical indentation depths.

This contact problem can be partly circumvented using the approach of Oliver and Pharr (1992), who used Seddon's analytical solution for a flat punch indenter. However, in this case the relationship between indentation depth and contact area must be known over the working range of the indenter.