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Research paper

Hyperelastic mechanical behavior of chitosan hydrogels for nucleus pulposus replacement—Experimental testing and constitutive modeling

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ABSTRACT

Chitosan hydrogels (CHs) have been considered as a potential implant material for replacement and repair of the Nucleus Pulposus (NP) within the intervertebral disk. The nonlinear mechanical behavior of a CH material is investigated experimentally and computationally in this study. A series of confined and unconfined compression tests are designed and conducted for this hydrogel. Hyperelastic strain energy density functions (SEDFs) are calibrated using the experimental data. A hyperelastic constitutive model is selected to best fit the multi-axial behavior of the hydrogel. Its general prediction ability is verified using finite element (FE) simulations of hydrogel indentation experiments conducted using a spherical tip indentor. In addition, digital image correlation (DIC) technique is also used in the indentation test in order to process the full-field surface strains where the indentor contacts the hydrogel. The DIC test results in the form of top-surface strains compared well with those predicted by the FE model. Results show repeatability for the examined specimens under the applied tests. Confined and unconfined test results are found to be sufficient to calibrate the SEDFs. The Ogden model was selected to represent the nonlinear behavior of the CH material which can be used in future biomechanical simulations of the spine.

1. Introduction

Hydrogels are cross-linked polymeric networks that are absorbed in solutions with high water content (Anseth et al., 1996). These highly-soft materials exhibit mechanical behavior that is characterized by large strains for a relatively small magnitude of applied mechanical loading. Their mechanical behavior is similar to rubber-like materials, including time-dependent viscoelastic behavior. Therefore, hydrogels can be bio-compatible and suitable for applications in the human body, e.g. contact lenses or as part of

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implants or tissue scaffolds. One such application is the repair or replacement of the Nucleus Pulposus (NP) as part of treatment in cases of intervertebral disk degeneration and related pathology. Several studies have been performed to use different classes of hydrogels in the replacement or repair of the NP tissue (Sebastine and Williams, 2007) in order to treat disk degeneration. This study is aimed at designing and conducting mechanical test setups in order to characterize the nonlinear behavior of chitosan hydrogels for potential use as addition or substitution of the native NP.

The difficulties in measuring the mechanical properties of hydrogels have led to diverse testing in the literature (Joshi et al., 2005, 2006; Borges et al., 2010; Vernengo et al., 2008; Cloyd et al., 2007; Moerman et al., 2009; Wang et al., 2004). Toward this goals, it is important to conduct sufficient tests for the nonlinear calibrations and verification of suitable mathematical constitutive models under general multi-axial states of stresses. The soft and nonlinear behavior of hydrogels makes it difficult to measure its mechanical properties as these change as a function of the stresses and strains in the material under compression state. Low-magnitude tensile experiments cannot be applied on highly-swollen hydrogels (Anseth et al., 1996). In addition, hydrogels can have a wide range of liquid-content levels, from low weight percents (WP) to extremely high WP; their mechanical properties are highly sensitive to this variable. Currently, limited testing material standards and guidelines exist for the mechanical testing of hydrogels (Anseth et al., 1996).

Hydrogels exist in many living organisms and can be manufactured synthetically in the lab. In order to characterize highly swollen hydrogels, it is important to initially determine if the gel is in a solid or a full liquid state. From elasticity point of view, liquids cannot carry shear stress. On the other hand, solids can take both shear and hydrostatic stresses. From a biological point of view, hydrogels are more fluid-like continuum than a solid, in part because their high rate diffusion and absorption of water.

According to Iatridis et al. (1996), the NP is a solid hydrogel that can swallow water up to 200% of its own weight. The mechanical properties of the hydrogel were generated from shear and creep tests, and their complex shear modulus was also calculated. They reached a conclusion that the native NP has solid continuum characteristics more than fluid based on the ratio between complex storage and shear moduli. Cloyd et al. (2007) tested the NP and some synthetic hydrogels in unconfined state that involved a relaxation type test. The hydrogels were deformed up to 25% in axial strain. The stress–strain curves showed a nonlinear response and nonlinear curve fits were made for the stress–strain results.

The mechanical behavior of the NP is composed of viscoelastic and non-linear hyperelastic components, has been shown by Iatridis et al. (1996) and Cloyd et al. (2007). Some viscoelastic models have been suggested (Iatridis et al., 1996, 1997). More tests, such as confined (Johannessen and Elliott, 2005a) and unconfined (Cloyd et al., 2007), take out to estimate the compressive modulus of the native NP. Umehara et al. (1996) execute local indentation test to evaluate the local compressive modulus of the NP and its distribution.

Other unconfined experiments have been conducted (Joshi et al., 2006; Borges et al., 2010; Vernengo et al., 2008; Stammen et al., 2001; Bertagnoli et al., 2005) on the NP material and its synthetic replacement candidates with an applied maximum strain ranging from 25% to 83%. Some of the candidates hydrogels are poly(vinyl-alcohol) (Joshi et al., 2006; Stammen et al., 2001), poly(vinyl-pyrrolidone) (Joshi et al., 2006), poly(acrylonitrile) (Bertagnoli et al., 2005), hyaluronan elastin-like polypeptide (Leckie, 2010), poly (N-isopropylacrylamide) (Vernengo et al., 2008) poly(ethylene-glycol) (Vernengo et al., 2008), alginate (Cloyd et al., 2007) and agarose (Cloyd et al., 2007). The nonlinear response was generated and often represented using piecewise-linear functions (Joshi et al., 2006; Vernengo et al., 2008; Stammen et al., 2001).

Mechanical tests of gels under fully or partially confined conditions were usually performed to generate the compression behavior under hydrostatic stress close to native loading environment. Another objective was to examine the hydrogel compressibility behavior or lack thereof. Confined experiments, used to test gel-type materials as NP candidates for repair and replacement, were also conducted in in-vitro conditions. These gels were placed in HDPE (High Density Polyethylene) and rubber containers (Joshi et al., 2006; Borges et al., 2010; Bertagnoli et al., 2005; Johannessen and Elliott, 2005b), and in porcine (Leckie, 2010), bovine (Hegewald et al., 2009) lumbar spine units (LSUs), and LSUs from human cadavers (Joshi et al., 2005, 2006; Arthur et al., 2010; Bertagnoli et al., 2005).

Most of the above tests were carried out with the aim to compare the gel mechanical performance to that of the native NP. However, little effort has been directed toward generating 3D hyperelastic constitutive models calibrated from these tests (Mathews et al., 2008). Direct mechanical tests on the hydrogel to calibrate nonlinear isotropic hyperelastic material models can make use of the data from confined and unconfined experiments. These tests can be viewed as uniaxial stress–strain responses. Moreover, in-direct tests, such as indentation where a spatial rich environment of multi-axial stress states are present, can be used to calibrate constitutive functions in an implicit manner “inverse problem”. Toward that goal, digital image correlation (DIC) is an optical technique that can be employed with indentation to measure surface strains on a deformed body. Little work has been carried out on using DIC to characterize hydrogels. Moerman et al. (2009) employed DIC to measure deformations in silicone gel under indentation in order to study the feasibility of using DIC to calibrate constitutive equations for tissue materials. A finite elements (FE) analysis of that experiment was made. The silicone gel was modeled as neo-Hookean hyperelastic minimizing the error between the FE results and the experimental data allowed to find the material’s parameters. They showed that a 3D DIC together with a FE model can be used to determine the bulk mechanical properties of materials that behave by neo-Hookean hyperelastic constitutive law.

Several previous studies have examined the suitability of chitosan-type gels as a bio-compatible candidate for replacement and repair of NP. Majeti and Kumar (2000) reviewed many applications for chitosan polymers and pointed to their ability to transform into hydrogels. Lee and Mooney (2001) described the properties of chitosan that