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Research Paper

Evaluation of friction properties of hydrogels based on a biphasic cartilage model

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ABSTRACT

Characterizing hydrogels using a biphasic cartilage model, which can predict their behavior based on structural properties, such as permeability and aggregate modulus, may be useful for comparing active lubrication modes of cartilage and hydrogels for the design of articular cartilage implants. The effects of interstitial fluid pressurization, inherent matrix viscoelasticity and tension–compression nonlinearity on mechanical properties of the biphasic material were evaluated by linear biphasic (KLM), biphasic poroviscoelastic (BPVE) and linear biphasic with anisotropy cartilage models, respectively. The BPVE model yielded the lowest root mean square error and highest coefficient of determination when predicting confined and unconfined compression stress–relaxation response of hydrogels ($n=15$): 0.220 ± 0.316 MPa and 0.93 ± 0.08; and 0.017 ± 0.008 MPa and 0.98 ± 0.01 respectively. Since the differences in error between models were not statistically significant, the simplest model we considered, KLM model, was sufficient to predict the mechanical response of this family of hydrogels. The coefficient of friction (COF) of a hydrogel–ceramic articulation was measured at varying loads and pressures to explore the full range of lubrication behavior of hydrogel. Material parameters obtained by biphasic models correlated with COF. Based on the linear biphasic model, COF correlated positively with aggregate modulus (spearman's rho$=0.5$; $p<0.001$) and velocity (rho$=0.3$; $p<0.001$), and negatively with permeability (rho$=-0.3$; $p<0.001$) and load (rho$=-0.6$; $p<0.001$). Negative correlation of COF with load and positive correlation with velocity indicated that hydrogel–ceramic articulation was separated by a fluid film. These results together suggested that interstitial fluid pressurization was dominant in the viscoelasticity and lubrication properties of this biphasic material.

1. Introduction

Researchers have investigated the structure–function relationship of articular cartilage in order to understand causes and effects of pathologies such as osteoarthritis, which is thought to be mechanically induced (Accardi et al., 2011; Caligaris et al., 2009). The interstitial fluid, which constitutes 65–75% of articular cartilage (Mak, 1986b; Mow et al., 1980), is

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now known to contribute significantly to its viscoelastic mechanical response and lubrication mechanisms (Caligaris and Ateshian, 2008; Mak, 1986a, 1986b; Mow et al., 1980; Mow et al., 1993). By taking into account an incompressible fluid phase and a solid matrix phase, a biphasic cartilage model has been shown to successfully explain how interstitial fluid pressurization supports the collagen–proteoglycan network in withstanding high contact loads (Mak, 1986a, 1986b; Mow et al., 1980; Mow et al., 1993). Mow et al. idealized the collagen–proteoglycan network as a linear elastic matrix in the biphasic model (KLM) in which the time-dependent response of cartilage was only due to interstitial fluid flow (Armstrong et al., 1984; Mow et al., 1980). Because collagen fibrils and proteoglycan gel are known to be viscoelastic (Suh and Bai, 1998), Mak expanded on the linear biphasic model by introducing relaxation of the solid matrix as a second source of time-dependent response, which resulted in the biphasic poroviscoelastic (BPVE) cartilage model (Mak, 1986a, 1986b). Although these models were successful in predicting cartilage response in confined compression configuration, peak to equilibrium load intensity ratio observed in unconfined compression experiments was not possible to account for using isotropic matrix assumptions (Cohen et al., 1998; Soltz and Ateshian, 2000). More recently, anisotropy was introduced to the solid matrix phase of the linear biphasic model in which differences in stiffness in compression and tension enabled better prediction of mechanical response of articular cartilage (Cohen et al., 1998; Soltz and Ateshian, 2000).

According to the biphasic cartilage model, drag forces produced by interstitial fluid flow through pores in the extracellular matrix separate articulating surfaces by hydrostatic load support and facilitate fluid film lubrication. The fluid film lubrication, which is dictated by the bulk properties of the lubricant, produces a low coefficient of friction (Accardi et al., 2011; Caligaris and Ateshian, 2008; Caligaris et al., 2009; Katta et al., 2008). Once the interstitial fluid is exuded out as a result of extended static loading, the articulating cartilage surfaces come into contact and the coefficient of friction in boundary lubrication is then determined by substances adsorbed on the surfaces (Caligaris et al., 2009; Gleghorn et al., 2010; Katta et al., 2008). Components of synovial fluid, such as hyaluronic acid, lubricin and glycosaminoglycans, aid in lubricating the articulating surfaces (Accardi et al., 2011; Caligaris et al., 2009). Because multiple lubrication modes may occur within the joint, researchers employed methods to assess the tribological response of articular cartilage under varying velocity and load conditions. For instance, Stribeck analysis, which was originally developed to explain lubrication mode transitions of hard bearings by displaying coefficient of friction on a “Stribeck curve”, was used in studying lubrication of articular cartilage (Gleghorn and Bonassar, 2008; Gleghorn et al., 2010).

Hydrogels are complex hydrophilic polymers that are swollen with water (Gong, 2006; Peppas and Merrill, 1977). Due to their structural similarity to articular cartilage, hydrogels have been considered for replacing damaged articular cartilage in the joints (Baykal et al., 2012; Bodugoz-Senturk et al., 2009; Covert et al., 2003; Katta et al., 2007). Hydrogels have been characterized by various testing configurations, such as confined and unconfined compression creep tests and indentation tests (Bavaresco et al., 2008; Bodugoz-Senturk et al., 2009; Spiller et al., 2008). However, correlating the mechanical behavior of hydrogels to their structural properties, such as water content and stiffness, will be useful during the material design phase. Furthermore, employing the articular cartilage-modeling framework for this purpose will also enable direct comparisons to articular cartilage (Spiller et al., 2008). Based on this approach, Spiller et al. utilized a linear biphasic cartilage model to compare the mechanical properties of their hydrogel with articular cartilage in terms of aggregate modulus and permeability (Spiller et al., 2008).

Understanding the relationship between active lubrication modes of articular cartilage and its mechanical response is useful for the design of articular cartilage implants (Accardi et al., 2011). Therefore, identifying material properties of hydrogels using biphasic cartilage model is a necessary step in evaluating the cause of similarities and differences in active lubrication modes of cartilage and hydrogels. Design guidelines produced by this approach to mimic the behavior of articular cartilage may improve tribological properties of hydrogels. The objectives of this study were to obtain hydrogel material properties using a biphasic cartilage model, and to investigate the relationship between the frictional properties and the material properties of hydrogels. Because hydrogels display cartilage-like viscoelastic behavior (Bodugoz-Senturk et al., 2009; Gong, 2006; Spiller et al., 2008), we hypothesized that the solid phase of hydrogels would be intrinsically viscoelastic to complement the viscoelasticity caused by interstitial fluid pressurization. In addition, we assumed isotropic matrix properties for simplicity in this study. Our hypotheses were: (1) the BPVE model would yield a lower error than KLM or a linear biphasic with anisotropy (ANISOTROPIC) cartilage model when predicting mechanical response of hydrogels in confined and unconfined stress–relaxation tests; and (2) coefficient of friction of hydrogel articulation at various speeds and loads that covered the clinically relevant range would correlate more strongly with material parameters obtained by BPVE model compared to parameters obtained by KLM and ANISOTROPIC models.

2. Theory

2.1. *Biphasic modeling framework*

2.1.1. *Confined compression stress relaxation*

The biphasic model equations for confined compression configuration were derived by complementing the momentum and continuity equations of continuum mechanics with Darcy's law, which couples interstitial fluid flow with the pressure gradient, to arrive at the general governing equation (Eq. (1)) (Mak, 1986a; Mow et al., 1980; Setton et al., 1993; Soltz and Ateshian, 2000).

$$\frac{\partial u}{\partial t} - k\nabla \cdot \sigma^e = 0 \tag{1}$$

The governing equation for the linear elastic matrix assumption depends on permeability (k) and aggregate modulus (H_A) (Eq. (2)) (Mow et al., 1980; Soltz and Ateshian, 2000). In the biphasic poroviscoelastic model (BPVE), inherent