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DEPARTMENT OF MECHANICAL ENGINEERING

HEAT AND MASS TRANSFER IN GELS

Progress Report

Submitted by

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Introduction

Stimuli-responsive or smart gel has the ability to undergo large volume change up to thousands-fold or even more when it is subjected to change of the environments: alterations in pH, ionic strength, temperature, electric field and irradiation of light. Extensive progress has been made in the technological application of gel (Shibayama and Tanaka, 1983). In order to analyze and predict the swelling and shrinking behaviour of stimuli responsive gels numerous experimental and mathematical studies have been conducted. In experimental work, a vast number of tailor-made hydrogels have been synthesized and several stimuli have been identified, a short summary of which provided by Roy and Gupta (2003). In tandem, various mathematical models have been developed, ranging from thermodynamics model to biphasic and triphasic model.

Objectives

The research mainly concerns two aspects related to the gels (especially hydrogels), namely fundamental research and applications.

Fundamental research, including:

- Identify the essential physics/chemistry of gels especially during the swelling and shrinking.
- Develop mathematical models of the swelling and shrinking behaviour of hydrogels.
- Conduct scaling analysis to provide an insight into the swelling and shrinking behaviour of hydrogels prior to numerical computation.
- Simulation and Validation of the developed model

Applied research, addressing:

- Application of hydrogels in microfluidic flow as a microvalve.
- Drying of gels and jelly-like materials such as vegetables and fruits.

Literature Review

Various mathematical models have been developed in an effort to understand predict the deformation kinetics and swelling equilibrium of stimuli responsive hydrogel subject to change of the environmental condition such as: pH (Kang et al., 2008; De and Aluru, 2004; De et al., 2002), temperature (Huang et al., 2008; Birgersson et al., 2007; Lele et al., 1997), glucose (Parker and Schwartz, 1987; Albin et al., 1987; Gough et al., 1988), electric field (Li et al., 2007; Zhou et al., 2002), and pressure (Kato, 2005; Kato, 1997). They can generally be divided into two main parts, namely: equilibrium swelling and deformation kinetic models.

The equilibrium swelling models usually follow Flory's mean field theory (Flory, 1953) or alterations thereof. Dissimilar to the equilibrium swelling, the model derived for describing the deformation kinetics of gels have no unified approach. The reason is the fact that numerous physiochemical phenomena such as polymer science, mass and heat transfer and continuum mechanics have to be taken into account when attempting to predict the swelling kinetics. Thus, models vary in their consideration of the different physical phenomena, as well as the types of gel the model are developed for. Most of the kinetic models, however, are for one-dimensional uniform swelling only. Thus, one disadvantage of these models is that they cannot be applied to situations where non-uniform swelling occurs. Recently, Birgersson et al (2008) developed a model that was able to capture the kinetics deformation of a hydrogel slab subjected to the temperature stimuli where the deformation is non-uniform.

Research Performed to Date

There are three studies that are carried out in tandem: modelling of pH-sensitive hydrogels, modelling temperature and alcohol sensitive hydrogel and modelling of hydrogel for microfluidic flow controller.

1. Modelling of pH sensitive hydrogel

   The continued study on modelling pH sensitive hydrogel has been carried out. A mathematical model based on the continuity conservation, momentum conservation, Poisson equation and Nernst-Planck equation that has previously been developed, analyzed and reduced was used to predict the equilibrium swelling and deformation kinetics of the pH-sensitive hydrogel. The simulation has been carried out by using Finite element solver, COMSOL Multiphysics and programming language, Matlab. The model predictions for equilibrium swelling as well as for deformation kinetics were then compared to the experiments. Overall, good agreement is achieved between the model predictions and the experiments. Parametric studies on the fixed charge density and ionic strength have also been carried out. It is found that the swelling ratio, ratio between final volume and initial volume of gels, of this pH-sensitive hydrogel increases as the fixed charge density increase and it decrease as the ionic strength increase. Hence to obtain higher swelling ratio, one need to either increase the fixed charge density of the hydrogels during polymerization or decrease the ionic strength of the bath solution.

2. Modelling of temperature and alcohol sensitive hydrogel

   Previously, the mathematical model has been derived and implemented to predict the equilibrium swelling of hydrogel immersed into water/methanol solution subject to change in temperature and methanol volume fraction. Good agreement was achieved between the model prediction and experiments by Hirotsu (1987). The continued study is conducted by employing to predict the swelling equilibrium of hydrogel immersed in water/ethanol solution and to predict the deformation kinetics of hydrogel subject to change in temperature and alcohol concentration.

   In predicting the equilibrium swelling of hydrogel immersed in water/ethanol solution, the mathematical model results show good agreement with the experiments conducted by Acharya et al (2002). The mathematical model has also been able to capture “re-entrant phenomenon” reasonably well.

   Next, the mathematical model was used to predict the deformation kinetics. Due to unavailability of experiments work in deformation kinetics of gels subject to alcohol concentration, the model was validated with the experiment for temperature sensitive hydrogel. Here, the model was set to predict the deformation kinetics of hydrogel immersed at pure water when temperature is changed. The model predictions were then validated with experiments by Andersson (1998). Overall, good agreement is achieved.

3. Modelling of hydrogel for microfluidic flow controller.

   This is preliminary study where we apply mathematical model to predict the flow behaviour in a micro channel when the hydrogel is implemented as a valve. The pH-sensitive hydrogel that has previously been studied is chosen for this application. Simulation was carried out for two-dimensional geometry. Some typical configurations were studied such as T-channel and Y-channel with single hydrogel at one side, T-channel with two hydrogels in series and T-channel with two hydrogel in parallel. The study will be extended for transient case in three-dimensional geometry.

Future Work

The study will be continued by developing mathematical model for non-uniform deformation. We will then implement the mathematical formulation into two and three-dimensional model to capture non-uniform deformation during swelling and shrinking of hydrogels for various stimuli: pH, temperature and alcohol concentration. We will also extend the model for microfluidic flow controller to 3D transient model to capture the kinetics of gel deformation and fluid flow inside the channel.

Publication

1. J.C. Kurnia, E. Birgersson, A.S. Mujumdar, Y.K. Yew, H. Li, *Reverse-Engineering of Material Properties for Coupled Alcohol and Temperature-Sensitive Hydrogels*.(Manuscript in preparation).
2. J.C. Kurnia, E. Birgersson, A.S.Mujumdar, *Computational Study of Three-Dimensional Finite Deformation of Macro-Porous Hydrogels*. (Manuscript in preparation).
3. J.C. Kurnia, E. Birgersson, A.S. Mujumdar, *Heat and mass transfer in gels*, Workshop on Mathematical Modeling in Minerals, Metals and Materials Processing, 14 March 2009, Singapore.
4. J.C. Kurnia, E. Birgersson, A.S. Mujumdar, L.C. Quah, *Mathematical Modeling of Hydrogels for Microfluidic Flow Control*, International Conference on Materials for Advanced Technologies (ICMAT), 2009 Singapore. (*Accepted for oral presentation; will be published in a special issue of Advanced Material Research*)
5. J.C. Kurnia, E. Birgersson, A.S. Mujumdar, *A Reduced Model for pH-Sensitive Hydrogels*, International Conference on Materials for Advanced Technologies (ICMAT), 2009 Singapore. (*Accepted for oral presentation*).
6. J.C. Kurnia, E. Birgersson, A.S. Mujumdar, *Study of Thermo-sensitive Hydrogels for Microfluidic Flow Control*, ECS meeting, 2009, Vienna, Austria. (*Abstract submitted*).