Heat conduction in poly(N-isopropylacrylamide) hydrogels

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ABSTRACT

Biological tissues are combination of liquids and fibrous solids, therefore polymer hydrogels can be used as model heat transfer system for soft tissues. The 3D network structure of the polymer prevents the convection and the dominant mechanism becomes heat conduction. The main purpose of the present work is to study the effect of network structure on the thermal conduction behavior of soft gels. Cylindrical poly(N-isopropylacrylamide) gel which shows lower critical solution temperature below which the system is completely miscible, whereas above LCST phase separation accours (LCST, [1]). The LCST temperature of the p(NIPA) gel is denoted by T_{ph} throughout this article and is equal to 35 °C.

One dimensional heat transfer through the length of the gel cylinder has been experimentally and theoretically studied. The temperature at the bottom of the gel cylinder was suddenly raised above the phase transition temperature. That part of gel cylinder becomes opaque where the temperature exceeds T_{ph}. This defines a visible front that moves forward in the opposite direction to temperature gradient. It was found that our novel technique can be successfully applied to determine heat conductivity and heat diffusivity of loosely cross-linked hydrogels. The kinetics of the moving turbid front was determined and analyzed on the basis of standard 1-d transient heat equation and Stefan's law [2]. It was found that the presence of network structure increases thermal diffusivity compared to pure water. It was also observed that latent heat influences thermal diffusivity.

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1. Introduction

Energy transport is an important subject in a number of biomedical and bio-technological process [3—6]. For example heat conduction plays a decisive role is the treatment of cancer by magnetic hyperthermia [7], which includes local heating of the tissue to temperatures up to 40 or 42 °C which damages the cells. The heat surplus must be efficiently removed to prevent overheating of neighboring cells, which could cause protein denaturation. Extreme heating like ablation is used to destroy tumors and cardiatic tissues. Also cooling of organs destined for transplantation reduces cell and tissue damages. Heat conduction studies in biological objects are rare and our knowledge is relatively scarce, especially about the effect of 3D fibrous network on heat conduction. Another effect which should also be taken into account is the latent heat which occurs as a result of protein denaturation which occurs around 42 °C.

The main purpose of the present work is to study the influence of these two foregoing effects by using model hydrogels. A novel method is presented in order to determine the unidirectional heat conductivity and heat diffusivity of soft materials similar in structure to the extracellular matrix of biological tissues. Unidirectional heat conduction in cylindrical poly(N-isopropylacrylamide) (PNIPA) gels was studied. The PNIPA gel undergoes LCST phase transition at 35 °C [6,7]. Due to the coil—globule transition of network chains, this gel may be considered as a rough model of protein denaturation which occurs around 42 °C.

Conductive heat transfer exists in a material due to the presence of temperature difference. If the heat transfer experiment is achieved in a cross-linked polymer gel, then the convective motion of fluid is hindered by the presence of network chains. The only mechanism is heat conduction, which involves direct transfer of energy via molecular contacts. By using PNIPA gels we are able to visualize the phase transition temperature front that moves forward in the opposite direction to temperature gradient. When the temperature exceeds a critical value of T_{cr} = 35 °C, the originally transparent gel becomes opaque. This opacity which is separated from the clear gel phase with a sharp interface makes it possible to

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follow the evolution of 35 °C temperature front during the heat transfer experiment [8,9].

In order to determine the basic parameters that characterize the phenomenon and evaluate thermal material properties, the problem of moving boundary has to be solved for this special case. The experimental data are interpreted on the basis of two different approaches. Standard 1-d transient heat equation describes the relation of heat flux to the temperature gradient as well as the evolution of temperature distribution in space, but the latent heat, due to the phase transition of PNIPA gel as well as the possible difference in the heat capacity of the two gel phases are completely neglected [10—12]. The Stefan's approach takes into consideration all of these effects, since it was originally developed for melting and freezing of water in semi-infinite medium [10—13].

2. Theoretical background

The transport of thermal energy in tissues is a complex process including conduction, convection, radiation, metabolism, evaporation and phase transition [3,4]. A variety of models has been proposed, where the tissue is represented as homogenous continuum vascular network. Pennes proposed a model to incorporate the effect of metabolism and blood perfusion into the standard thermal equation [3].

These terms are considered to be as maintaining the isotherm condition of the model gel.

The relationship between the rate of heat transfer and temperature is known as standard 1-d transient heat equation [3–5,14–17]. The relationship between temperature, T and heat flux, q_x can be written as

$$q_x = -k\frac{\partial T}{\partial x} \tag{1}$$

where k means the thermal conductivity, which is a material dependent quantity. This equation can be applied to both living and nonliving systems. In addition to thermal conductivity, the thermal diffusivity, α is also a widely used characteristic quantity. It is the ratio of thermal conductivity to the heat capacity. If the thermal conductivity can be assumed to be independent from the temperature, the governing equation for unidirectional heat transfer can be written as

$$\frac{\partial T}{\partial t} = \alpha\frac{\partial^2 T}{\partial x^2} \quad \text{where} \quad \alpha = \frac{k}{\rho \cdot c_p} \tag{2}$$

where ρ stands for the density of the material and c_p denotes the heat capacity per unit mass at constant pressure. The dimension of thermal conductivity is W/(mK) and it measures the ability of the material to conduct thermal energy relative to its ability to store it. Materials with large thermal diffusivity respond quickly to temperature difference. Depending on the wide variety of boundary and initial conditions, several solutions of Eq. (1) exist [11,12].

2.1 Heat conduction described by standard 1-d transient heat equation

Eq. (3) has analytic solution if we consider a semi-infinite slab, occupying the space from $x = 0$ to $x = \infty$, with T_0 as the initial temperature distribution. At time $t = 0$, we raise the temperature of the surface at $x = 0$ to T_h, and maintain that temperature. So the initial condition $t = 0$, $T = T_0$, and the boundary conditions for $t > 0$ is $x = 0$, $T = T_h$ and $x = \infty$, $T = T_0$. Analytic solution of Eq. (1) for a single phase takes the form [3,4]

$$T(x,t) - T_0 = (T_h - T_0)\left(\text{erfc}\frac{x}{\sqrt{4\alpha_t}}\right) \tag{3}$$

where erfc is the complementary error function: $\text{erfc}(u) = 1 - \text{erf}(u)$.

Let us fix the temperature T_{ph} at the LCST phase transition temperature and let δ be the penetration length, whose time evolution represents the kinetics of the moving temperature front. Taking into account, that $T(\delta,t) = T_{\text{ph}}$, Eq. (3) can be rewritten as

$$\frac{T_{\text{ph}} - T_0}{T_h - T_0} = \text{erfc}\left(\frac{\delta}{\sqrt{4\alpha_t}}\right) \tag{4}$$

Eq. (3) describes the dependence of the penetration thickness on time, $\delta(t)$ at fixed T_0 and T_h. Since the left hand side of Eq. (3) is determined by the initial conditions, and is constant during the experiments, the time dependence of the $T_{\text{ph}} = 35$ °C front can be described by a simple power law dependence

$$\delta(t) = K_{\text{F}}\sqrt{t} \tag{5}$$

Here subscript F in K_{F} and α indicates that this result has been derived from Fourier's law. This equation says that the penetration length follows square root time dependence, where the slope (K_{F}) is determined by thermal diffusivity, α_{F} as well as the experimental conditions. Combination of Eqs. (4) and (5) results in

$$K_{\text{F}} = \text{erfc}^{-1}\left(\frac{T_{\text{ph}} - T_0}{T_h - T_0}\right)\sqrt{4\alpha_{\text{F}}} \tag{6}$$

By introducing dimensionless temperature

$$T^* = \left(T_{\text{ph}} - T_0\right)/(T_h - T_0) \tag{7}$$

Eq. (6) can be written in a simple form

$$K_{\text{F}} = \text{erfc}^{-1}(T^*)\sqrt{4\alpha_{\text{F}}} \tag{8}$$

2.2. Heat conduction described by Stefan's approach

The general feature of heat conduction in PNIPA gel showing LCST phase transition is that energy is used to induce the phase transition first and then to alter the temperature. The Stefan's description takes into account the latent heat during the phase transition. In case of PNIPA gel, the turbidity front is located at $x = \delta(t)$ position and the temperature of opaque (turbid) and clear gel phases is equal to the phase transition temperature, $T_t(\delta(t),t) = T_c(\delta(t),t) = T_{\text{ph}}$. Here subscript t and c denotes the turbid and the clear phase, respectively. At point $x = \delta(t)$ the net heat current in the gel of cross-sectional area A_0 equals to the sum of heat current. The heat generated during phase change over the time equals to the heat flow in the clear phase. The energy balance can thus be expressed as follows:

$$-k_t\frac{\partial T_t}{\partial x}A_0 + q_{\text{ph}}A_0\frac{d\delta}{dt}\rho_t = -k_c\frac{\partial T_c}{\partial x}A_0 \tag{9}$$

where q_{ph} denotes the latent heat of the phase transition per unit mass and ρ_t represents the density of the opaque gel, while k_c and k_t denote the thermal conductivity of the clear and the turbid phase, respectively. The time dependent temperature profile, $T(x,t)$ that characterizes the thermal state of both gel phases, can also be obtained from Fourier's law: