International Journal of Heat and Mass Transfer 120 (2018) 830–837

Contents lists available at ScienceDirect

International Journal of Heat and Mass Transfer

journal homepage: www.elsevier.com/locate/ijhmt

Mathematical model for mixing in a paper-based channel and applications to the generation of a concentration gradient

Ilhoon Jang [a], Gangjune Kim [a], Simon Song [a,b,*]

[a] Department of Mechanical Engineering, Hanyang University, Seoul 04763, South Korea
[b] Institute of Nano Science and Technology, Hanyang University, Seoul 04763, South Korea

ARTICLE INFO

Article history:
Received 10 October 2017
Received in revised form 5 December 2017
Accepted 15 December 2017

Keywords:
Mathematical model
Paper-based channel
Mixing
Interdiffusion
Concentration field
Convection-diffusion equation

ABSTRACT

A paper-based channel is a useful platform for the facile development of analytical devices implementing various chemical or biological reactions. To improve the analytical performance for various applications, it was necessary to analyze the detailed mixing characteristics within a paper-based channel. In this paper, we proposed a mathematical model to predict a concentration field created as a result of the imbibition of multiple fluids within a porous material. Interestingly, we found that the model exhibited a constant interdiffusion width within a paper-based channel even though the flow front velocity decreased over time. We were able to verify that our model accurately predicted the concentration field by comparing the experimental and numerical results for mixing in a 2 inlet-channel. Finally, we designed and fabricated paper-based channels to generate two (linear and non-linear) concentration gradients based on predictions made by the model. Both the experimental and numerical results were in good agreement, demonstrating that our model was accurate and useful for developing a paper-based analytical device utilizing the mixing characteristics of a sample and reagent flow system.

1. Introduction

Paper-based devices have been developed as diagnostic tools implementing complex chemical and biological reactions through the development of various detection methods [1–3] such as colorimetric, electrochemical, fluorescent, chemiluminescent, electrochemiluminescent, and photoelectrochemical methods since being first introduced as analytical devices in 2007 [4]. Recent studies with regard to paper-based channels have suggested a flow control method [5–8] with elucidated flow characteristics [9–11] to improve detection performance and realize a multi-functional device.

Understanding the mixing characteristics within paper-based channels is essential to improving the accuracy of detection functions for target materials and optimizing channel structures. For example, sample and reagent mixing or the interactions of multiple working fluids should be predicted prior to the design of paper-based channels to reduce trial-and-error. In cell-related research, the effects of paper characteristics such as thin, flexible, strong structures, and large void spaces should be known with regard to mixing. This is because mixing properties are often the key parameters for maintaining the concentration gradients of oxygen, nutrients, metabolites, signaling molecules, etc. that are critical to cell culture studies [12,13]. In addition, the importance of various concentration gradients including linear and non-linear concentration gradients has been reported in the studies of bacterial migrating [14] and cancerous cells [15].

Mixing within conventional microfluidic devices like a polymeric or glass microchannel is generated through molecular diffusion between two or more fluid streams, which is generally dominated by laminar convection. As a result, a concentration gradient is established in such a device within the interdiffusion region of fluids [16]. The interdiffusion width (σ) is known to increase in the streamwise direction, as shown in Fig. 1, and can be analytically estimated under the assumption of constant mixing diffusivity (D_m) and average velocity (U) as follows [17–19]:

$$\sigma \sim \sqrt{\frac{x}{U} D_m}, \tag{1}$$

where x is the distance downstream from the inlet junction. The mixing diffusivity is dominated by molecular diffusion in the conventional microfluidic channel. According to this relationship, a wide interdiffusion width requires a long channel, which is undesirable for microfluidic devices. To make the channel short, several prior studies have used microchannel networks such as Christmas

* Corresponding author at: Dept. of Mechanical Engineering, Hanyang University, 222 Wangsimni-Ro, Seongdong-Gu, Seoul 04763, South Korea.
E-mail address: simonsong@hanyang.ac.kr (S. Song).

https://doi.org/10.1016/j.ijheatmasstransfer.2017.12.078
0017-9310/

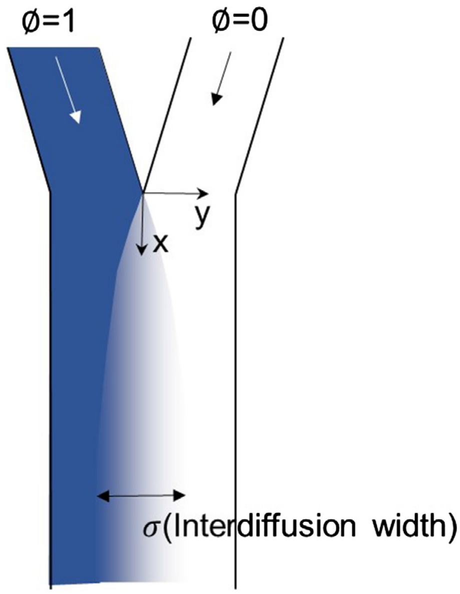

Fig. 1. Schematic for flow mixing in a 2-inlet channel. ∅ indicates the concentration.

trees [20] or flow splitters [21] and have successfully generated concentration gradients within a short distance through a steady pressure-driven flow. However, this approach would be unsatisfactory for utilization in a paper-based channel because the flow through a porous structure is driven by capillary forces and mixing is expected to be characterized via mechanical dispersion in addition to molecular diffusion, unlike within conventional microfluidic devices.

Mechanical dispersion and flow unsteadiness are well-known features in the flow of porous materials. Mechanical dispersion refers to the mechanical mixing occurring due to the velocity variation during convective transport, and the mixing in paper-based channels, which are porous materials, depends on mechanical dispersion rather than molecular diffusion [22]. Imbibition caused by capillary forces can create an unsteady flow as the flow front proceeds. According to the Lucas-Washburn relation (Eq. (2)) [23,24], the speed of the flow front within a paper-based channel can be expressed as described in Eq. (3) [9,10,25–27]:

$$l = \sqrt{2D_c t}. \tag{2}$$

$$U = \frac{dl}{dt} = \sqrt{\frac{D_c}{2t}}, \tag{3}$$

where l is the flow distance, D_c is the convective diffusivity that characterizes the paper-based channel flow, t is time, and U is the speed of the flow front within a paper-based channel with constant width that is identical to the average velocity in Eq. (1). Due to velocity unsteadiness, gradient generation within a paper-based device is likely to require sophisticated designs and flow control methods compared to conventional microfluidic devices.

A recent study with regard to gradient generation in a paper-based channel used symmetrical consecutive T-junctions to generate different concentration chambers for cell cultures [28]. This device showed that the concentration gradient was generated by a capillary-driven flow and geometrical structure without any additional instruments and remained steady after being established. Despite the advantage of an easily generated gradient, this study did not demonstrate a method by which to control or vary the concentration profile. If a concentration field could be predicted with a mathematical model, various concentration gradients could be readily generated in a paper-based channel.

We proposed a mathematical model that considered mixing characteristics in a paper-based channel. The model was developed by modifying a well-known convection-diffusion equation and placed emphasis on mechanical dispersion and flow unsteadiness through porous materials. The model was validated by comparing experimental and numerical results for mixing within a 2-inlet paper-based channel. We also introduced a design process for the generation of a concentration gradient in the paper-based channel. Finally, we demonstrated the accuracy and usefulness of the model by generating linear and non-linear concentration gradients in a paper-based channel.

2. Mathematical model for mixing in paper-based channels

A two-dimensional (2-D) concentration field is described with a convection-diffusion (C-D) equation for most laminar microchannel flows within a conventional microfluidic chip. The equation predicts a concentration field (∅) by superposing longitudinal convection and transverse diffusion terms [13] as follows:

$$\frac{\partial \varnothing}{\partial t} = D_m \frac{\partial^2 \varnothing}{\partial y^2} - U \frac{\partial \varnothing}{\partial x}. \tag{4}$$

The mixing diffusivity (D_m) represents both molecular diffusion and mechanical dispersion as follows [29]:

$$D_m = D_e + \alpha U \tag{5}$$

where D_e is molecular diffusivity and α is mechanical dispersivity. For a conventional microfluidic channel, however, the molecular diffusion generally overwhelms the mechanical dispersion whereas the former is negligible as compared to the latter in a paper-based microfluidic channel due to porous structure. In addition, the velocity is no longer constant and should be replaced with Eq. (3) in a paper-based microfluidic channel. Therefore, Eq. (5) can be approximated as:

$$D_m \doteq \alpha U = \alpha \sqrt{\frac{D_c}{2t}}. \tag{6}$$

Then, a C-D equation for mixing within a paper-based channel is obtained by substituting Eq. (3) and Eq. (6) into Eq. (4); the results can be expressed as follows:

$$\frac{\partial \varnothing}{\partial t} = \sqrt{\frac{D_c}{2t}} \left(\alpha \frac{\partial^2 \varnothing}{\partial y^2} - \frac{\partial \varnothing}{\partial x} \right). \tag{7}$$

This mathematical model indicates that diffusion in the transverse direction may have been affected by both the velocity and dispersivity unlike a mixing flow through a conventional microchannel.

3. Experimental and numerical methods

A paper-based channel was fabricated via a wax printing method [30] on chromatography paper (Grade 1, Whatman). The channel wall and other channel components, such as the flow regulator for flow rate control and flow barrier for flow blockage, were drawn with a CAD program (AutoCAD, Autodesk) and printed by a solid wax printer (Phaser 8760 color printer, Xerox) under high-quality grayscale. The paper was placed on a hotplate (MSM-20D, WiseStir) for 2 min at 150 °C to reflow the wax and was cooled to room temperature.

The experimental setup is illustrated in Fig. 2a. A paper-based channel was enclosed within a petri dish containing sufficiently wet tissue paper to minimize evaporation of the working fluids,