CHEMICAL ENGINEERING RESEARCH AND DESIGN 90 (2012) 1719–1726

Contents lists available at SciVerse ScienceDirect

Chemical Engineering Research and Design

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

Numerical study on microstructured reactor with chaotic heat and mass transfer and its potential application for exothermic process

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ABSTRACT

Design of microstructured reactors with thermal control function is investigated through numerical simulation. It consists of one middle channel for handling chemicals and two other channels attached to its top and bottom for cooling purpose. Three designs are examined. Reactor *A* uses simple straight channels. In reactor *B*, chaotic flow is applied to the middle channel, and in reactor *C* chaotic flow is applied to all the three channels. Results show that in comparison with the straight channel, the Nusselt number in current design is greatly improved through chaotic flow. Rapid mixing is also achieved. Potential application of the design for continuous exothermic process is analyzed. For reactor *A*, it is not workable as the temperature of the chemical solution continuously increases over the channel. In comparison, for both reactors *B* and *C* the temperature can be well controlled within the required range. As the coolant flow in reactor *C* is also chaotic, it provides a higher heat removal capacity.

Keywords: Microstructured reactor; Chaotic flow; Heat transfer; Fluid mixing; Thermal control

1. Introduction

The microreaction technology has been developing very fast in the past decade. Compared with traditional batch reactors, microstructured reactors provide many significant advantages, such as higher conversion and selectivity, less energy consumption, improved safety and production flexibility (Ehrfeld et al., 2000; Gavriilidis et al., 2002; Pennemann et al., 2004). Many conventional batch mode processes may potentially be replaced with the microreactor-based continuous production, especially in the fine chemical and pharmaceutical industries (Roberge et al., 2005; Lomel et al., 2006; Buchholz, 2010).

At micro scales, the surface-to-volume ratio is notably increased leading to intensified heat and mass transfer. But on another aspect, the fluid viscous effects become more significant and relevant Reynolds number ($Re = UD_h/\upsilon$) is small. Consequently, the flow typically falls in the laminar regime. For simple straight channels, the flow is stable and fluid mixing solely relies on diffusion. Similarly, in the direction perpendicular to the flow, the heat transfer can only be achieved through heat conduction. With absence of turbulence, even in micro-geometries, pure diffusive mixing and heat conduction may still fail to meet relevant requirements in application. Thus, specially designed micro-mixer/heat exchanger is usually required in the microreactor system. Extensive reviews on micromixers can be found in Hardt et al. (2005) and Hessel et al. (2005). Relevant mixing principles include hydrodynamic focusing, multi-lamination, splitting-and-recombination, etc. These mixers have been used for Phenyl Boronic Acid Process (Hessel et al., 2004), selective Friedel–Crafts reactions (Nagaki et al., 2005), pigment fabrication (Pennemann et al., 2005), etc. About application of heat exchanger/reactors for continuous chemical process, more discussions can be found in Anxionnaz et al. (2008) and Tochon et al. (2010).

One important strategy to enhance the mass and heat transfer in laminar flow is chaotic advection (Aref, 1984). Passively, specially designed configurations are used to produce chaotic flow. It can be patterned microstructures fabricated

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Received 15 September 2011; Received in revised form 17 February 2012; Accepted 29 March 2012

http://dx.doi.org/10.1016/j.cherd.2012.03.019

Nomenclature

D_h	hydraulic diameter (mm)
L_N	dimensionless channel length
Nu	Nusselt number
Q	constant wall heat flux (W/m^2)
Re	Reynolds number
T_c	temperature of the chemical solution (°C)
T_w	temperature of water (°C)
T_{wall}	channel wall temperature (°C)
T_m	bulk mean temperature of water (°C)
U	fluid velocity (mm/s)
ν	kinematic viscosity (m^2/s)
ρ	density (kg/m^3)
λ	thermal conductivity (W/(m·K))
φ	concentration
σ_T	standard deviation in temperature (°C)
σ	standard deviation in concentration

on the channel wall (Stroock et al., 2002), serpentine channels with Dean flow effect (Liu et al., 2000; Jiang et al., 2004), or split-and-recombine (SAR) structures (Schönfeld et al., 2004; Xia et al., 2005; Ohkawa et al., 2008). Chaotic flow produces substantial transverse momentum to improve cross-sectional mixing. It also provides transverse convective heat transfer to increase the heat flux (Peerhossaini et al., 1993; Kumar and Nigam, 2007). Relevant applications include food production (Metcalfe and Lester, 2009), fuel cells (Lasbet et al., 2006), etc.

Chaotic flow may also be used in microstructured chemical systems. In many exothermic or endothermic chemical reactions, both mixing and thermal control are important issues. This can be achieved using a “multifunctional heat exchanger” (MHE) which combines mixing, reaction and heat transfer (Ferrouillat et al., 2006). Relevant designs can be examined through computational fluid dynamics (CFD) technique, which has proven to be an important tool to analyze flow behaviors and chemical reactions (Phillips et al., 1997; Marchisio and Barresi, 2003; Aubin et al., 2005; Gentric et al., 2005; Habchi et al., 2010; Mousavi et al., 2010). In this study, microstructured reactors with chaotic heat and mass transfer will be investigated. Their mixing and thermal performance as well as potential application for exothermic process will be analyzed through CFD simulation.

2. Methodology

2.1. Design of the microstructured reactor

Chaotic flow is introduced into the design to intensify both the mixing and heat transfer processes. The channel configurations are modified based on our previous design of chaotic micromixer (Xia et al., 2005). Two-layer interconnected channels are used to facilitate fluid manipulations including stretching-and-folding, splitting-and-recombination to produce chaotic flow. Two configurations are analyzed, as shown in Fig. 1. In both the designs, the top layer and base layer have same structures. One is just reversed and then combined with the other to form the channel. The dashed lines in Fig. 1(a) indicate the same longitudinal locations for alignment. For channel 1, its structure is regular that provides a consistent thermal performance over its length (detailed results will be given in Section 3.1). However, regular periodic structure may result in small isolated zones that hinder complete mixing. In channel 2, section *a* as indicated in Fig. 1(b) is turned in the opposite direction, and the small branch channel *b* is also re-orientated. This irregularity will destroy isolated zones to achieve global homogeneous mixing.

Three microstructured reactors, namely reactor *A*, *B* and *C*, are studied. Each design contains three layers of channels. In reactor *A*, straight channels are used with the dimensions of 33 mm (w) × 12 mm (h) × 317 mm (l). In reactor *B*, the middle channel is replaced with abovementioned channel 2. In reactor *C*, the top and bottom channels are further replaced with channel 1 (see Fig. 2). For both channels 1 and 2, w_c (as indicated in Fig. 1(a)) is 10 mm, d_c is 6 mm. The size of the inlet and outlet is 14.14 mm × 12 mm. The multi-layer structure of the device can be fabricated layer by layer. They are then assembled together using common clamping method with gaskets. Permanent diffusion bonding (Martin et al., 1999) may also be used for metallic materials.

2.2. Numerical setup

The fluid flow and thermal analysis was performed using a CFD code *ANSYS CFX12.0*. The numerical study will allow examination of the flow behavior, mixing and heat transfer processes in the channel. The flow involved in current study is laminar. The governing equations are the incompressible Navier–Stokes equations.

$$\nabla \cdot \mathbf{u} = 0 \tag{1a}$$

$$\frac{D\mathbf{u}}{Dt} = f - \frac{1}{\rho}\nabla p + \nu\nabla^2\mathbf{u} \tag{1b}$$

The finite volume method is used to discretize the spatial domain. The governing equations are integrated over each control volume, and the relevant quantity is conserved over each volume to satisfy strict global conservation. The convergence criterion is set as RMS residual $\leq 10^{-5}$.

In heat transfer simulations, the variable-density effects are neglected. The thermal energy equation is solved.

$$\frac{\partial(\rho e)}{\partial t} + \nabla \cdot (Ue) = \nabla \cdot (\lambda \nabla T) - P\nabla \cdot U + \tau : \nabla U + S_E \tag{2}$$

where e is the internal energy, $\tau : \nabla U + S_E$ is the dissipation item.

To check thermal performance of the channel, the Nusselt number (Nu) for H2 boundary conditions, i.e. constant wall heat flux and constant fluid axial heat flux, is calculated. Nu is defined as

$$Nu = \frac{Q}{T_{wall} - T_m}\frac{D_h}{\lambda} \tag{3}$$

where Q is the constant wall heat flux; T_{wall} is the channel wall temperature; T_m is the bulk mean temperature of the fluid over the cross-sectional area of the channel; λ is the thermal conductivity of the fluid, for water it is 0.607 W/(m·K). The temperature uniformity is measured by the standard temperature deviation, which is calculated as

$$\sigma_T = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(T_i - \bar{T})^2} \tag{4}$$