Chemical Engineering and Processing 77 (2014) 13–21

Contents lists available at ScienceDirect

Chemical Engineering and Processing: Process Intensification

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

Effect of high frequency ultrasound on micromixing efficiency in microchannels

Mahboubeh Faryadi, Masoud Rahimi*, Sahar Safari, Negin Moradi

CFD Research Center, Chemical Engineering Department, Razi University, Kermanshah, Iran

ARTICLE INFO

Article history:
Received 8 October 2013
Received in revised form 9 December 2013
Accepted 4 January 2014
Available online 9 January 2014

Keywords:
Micromixing
Microchannels
Ultrasound
Mixing
Mixing enhancement

ABSTRACT

In the present work, an investigation on the effect of high frequency ultrasound wave on micromixing in the studied microchannels was carried out. Three types of microchannels with different shapes are examined. A 1.7 MHz piezoelectric transducer (PZT) was employed to induce the vibration in these microchannels through an indirect contact. A method based on the Villermaux–Dushman reaction was employed to study the micromixing in these microchannels. The segregation intensity was determined for layouts with and in the absence of ultrasound irradiation. Further, the effect of ultrasound waves, in various flow rates and initial concentrations of acid, on the segregation index (X_S) and micromixing time (t_m) was investigated. The experimental results showed that the ultrasound waves have a significant influence on product distribution and segregation index at various flow rate ratios. The data obtained in all cases showed that the segregation index was reduced when the flow rate ratios were increased. Also the results demonstrate that in spite of a low energy consumption of PZT, the relative segregation index improved up to 18–36% at various flow rate ratios.

1. Introduction

The application of microstructured reactors in the chemical process industry has caught significant attention in recent years and microtechnology is currently an area of rapid growth in many areas of application. Microstructured reactors (or microchannels) are generally three-dimensional structures with inner dimensions below millimeter, and more specifically between 10 and 100 μm and reaction volumes in the nanoliter to microliter range [1,2]. Microchannels provide high specific surface area allowing an effective mass and heat transfer. The miniaturization of chemical reactors has many other benefits such as minimal environmental hazards and enhanced safety because of smaller volume (reagents holdup) besides process cost reduction [2–5].

Mixing is a basic unit operation for chemical reactions and at the molecular scale (micromixing) can extremely influence the selectivity, yield and quality of final products in various chemical processes. The mixing in the microchannel takes place through diffusion and convection depending on the flow geometry and the operating condition [6–8].

Depending on demand, more microchannel units could be connected in parallel (numbering-up) so that the required amounts of products can be produced [9–11]. Microchannel geometry can greatly influence the micromixing performance and some investigations [5,12,13] have revealed that the mixing efficiency strongly depends on channel geometry.

On the other hand, the ultrasound wave irradiation, as a well known technique, can improve a wide variety of processes. Acoustic cavitation and acoustic streaming are two phenomena caused by the ultrasound wave propagation through a liquid. During the cavity collapse, shock waves and microjets are produced. Moreover, ultrasound energy dissipation in the liquid leads to the formation of a rather intense macroscopic liquid flow called the acoustic streaming. These phenomena result in interesting mechanical effects [14–17]. Mechanical effects of ultrasound can also improve the mixing during the homogeneous liquid phase reactions [18].

In many studies [19–27], the effect of ultrasound wave on mixing efficiency has been investigated. In addition, some efforts were undertaken to combine the ultrasound wave irradiation and microchannels [28,29].

Although the low frequency reactors give dominant physical effects and hence mixing efficiencies, but ultrasound waves with higher frequencies, (more than 1 MHz), produce a stable cavitation with small bubbles [26], which leads to the generation of acoustic streams in the bulk of liquid with mixing effect. High frequency ultrasound, in the range of MHz, induces convective flows and micro-streams, simultaneously. Therefore, it is possible to reach the more efficient macro and micromixing [30]. The high frequency wave gives significant micromixing efficiencies due to

* Corresponding author at: Chemical Engineering Department, Razi University, Taghe Bostan, Kermanshah, Iran. Tel.: +98 8314274530; fax: +98 8314274542.
E-mail addresses: m.rahimi@razi.ac.ir, masoudrahimi@yahoo.com (M. Rahimi).

0255-2701/$ – see front matter © 2014 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.cep.2014.01.001

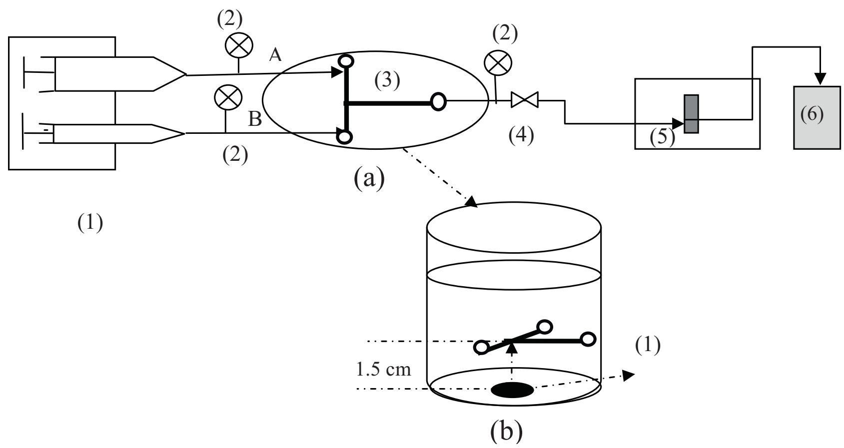

Fig. 1. A schematic diagram of the experimental setup: (a) – (1) syringe pump, (2) pressure sensor, (3) microchannel, (4) valve, (5) spectrophotometer, (6) product container and (b) container ultrasonic – (1) piezoelectric transducer 1.7 MHz.

the microstreams and micro jets created in the liquid. In our previous works [25,26], there have been investigations on using high frequency for improving the micromixing level in both plug and batch reactor and it was demonstrated that high frequency ultrasound wave (1.7 MHz) has a great effect on micromixing efficiency. Our previous works also showed that ultrasound with a frequency of 1.7 MHz can generate micromixing with a low energy consumption which is desirable [25].

In the present study, the influence of high frequency ultrasound wave propagation on reaction efficiency in three-types of microchannels, was investigated. For this purpose, the Villermaux/Dushman reactions consisting of a neutralization reaction coupled with the iodide–iodate reaction were used. The key issue in this work is employing the high frequency ultrasound wave to enhance micromixing in the studied microchannels. The microchannels were placed in an ultrasonic container equipped with a high frequency (1.7 MHz) piezoelectric transducer in order to evaluate the effects of ultrasonic waves on mixing in the microchannels. The effect of ultrasound irradiation on the micromixing performance at various operational conditions including flow rate ratio and acid concentration were investigated.

2. Experimental work

2.1. Experimental setup

A schematic diagram of the experimental setup in this work is shown in Fig. 1(a). Dushman reaction was carried out in the microchannels to show the performance of mixing in continuous condition. The microchannels were fabricated from glass tubes with outer diameter, inner diameter and length of 1 mm, 800 μm and 75 mm, respectively. The inlet streams, A and B, are pumped at selected flow rates into the microchannel using syringe pumps. The pressure drop across the channel was measured. In order to study the effect of microchannel shape on the micromixing, three microchannels were used (Fig. 2). Detail of the microchannels is listed in Table 1. The microchannels are placed in a container, equipped with an ultrasound wave transducer, to evaluate the effects of ultrasonic waves on the mixing performance inside the microchannels. The high frequency piezoelectric transducer (1.7 MHz, Model ANN-2517GRL, Annon Piezo Technology Co. Ltd., China) with a diameter of 1.5 cm was installed at the bottom of the vessel, in a way that the wave has a proper contact with the inlet channels junction point (Fig. 1(b)).

2.2. Micromixing characterization

The competitive parallel reactions are affected by the micromixing level since their product distribution depends on the degree of segregation. Thus the micromixing performance can be evaluated by using these reactions. In this work, the micromixing level estimate, generated by microchannel, was tested using the well-known parallel competing reactions that have been presented by Dushman [31–35]. The Dushman reaction equations are as follows:

$$H_2BO_3{}^- + H^+ \rightarrow H_3BO_3 \quad \text{quasi-instantaneous} \tag{1}$$

$$5I^- + IO_3{}^- + 6H^+ \rightarrow 3I_2 + 3H2O \quad \text{veryfast} \tag{2}$$

$$I_2 + I^- \leftrightarrow I_3{}^- \tag{3}$$

The reaction rates are reported as follows [33,35,36]:

$$r_1 = k_1[H^+][H_2BO_3{}^-] \tag{4}$$

$$r_2 = k_2[H^+][I^-][IO_3{}^-] \tag{5}$$

$$r_3 = r_3{}^+ - r_3{}^- = k_3{}^+[I^-][I_2] - k_3{}^-[I_3{}^-] \tag{6}$$

In which k_1, k_2 and k_3 values were reported in our previous works [19,20].

The concentration of $I_3{}^-$ can be estimated by using a UV spectrophotometer at a wavelength of 353 nm and based on the Beer–Lambert's law [25,33,34,37]:

$$C_{I_3^-} = \frac{A}{\varepsilon_{353} l} \tag{7}$$

$C_{I_3{}^-}$ is the concentration of $I_3{}^-$, l is quartz cell thickness (0.01 m) and A is light absorption. The term of ε_{353} corresponds to the molar extinction coefficient of $I_3{}^-$ at a wavelength of 353 nm, that $1/(\varepsilon_{353} l)$ has been obtained 0.06 experimentally.

The main point in this method is adding a small amount of sulfuric acid to the mixture solution of borate, iodide and iodate ions. The segregation index (X_S) which has been defined as the relative amount of consumed acid for iodine production, is a criterion for indicating micromixing quality. The X_S is defined [34,35] by following equation:

$$X_S = \frac{Y}{Y_{TS}} \tag{8}$$

$$Y = \frac{2F_{I_2+I_3^-}}{F_{H_0^+}} \approx 2\frac{F_{I_3^-}}{F_{H_0^+}} = 2\frac{Q_{I_3^-}[I_3^-]}{Q_{H_0^+}[H_0^+]} \tag{9}$$