Chemical Engineering and Processing 104 (2016) 190–200

Contents lists available at ScienceDirect

Chemical Engineering and Processing: Process Intensification

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

Comparison of liquid-liquid extraction in batch systems and micro-channels

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ARTICLE INFO

Article history:
Received 23 December 2015
Received in revised form 29 January 2016
Accepted 14 March 2016
Available online 15 March 2016

Keywords:
Micro-channel
Liquid-liquid extraction
Non-linear equilibrium
Stratified flow
Slug flow

ABSTRACT

Liquid-liquid extraction is an important mass transfer operation in the chemical, food processing and pharmaceutical industries. Our work focuses on experimentally quantifying mass transfer performance in a well stirred batch vessel and in stratified flow and slug flow in micro-channels. Extraction of propionic acid from toluene to water is chosen as a test system. The distribution of propionic acid in toluene and water at equilibrium was found to be non-linear. The batch experiments were carried out both with a flat interface and as a well-mixed dispersed system. The continuous experiments were carried out in micro-channels with a square cross-section. A lumped parameter model was used to quantify the mass transfer coefficient in batch mode. A lumped (distributed) model was used to describe the extraction in a slug (stratified) flow in a micro-channel. The overall extraction performance is characterized as a function of residence time of the phases and the hydrodynamics. It was found that the micro-channel gives a superior extraction performance in the slug flow regime as compared to the well mixed batch system although both have comparable sizes of the dispersed phase. This is attributed to the strong internal circulations induced by shear in the slug flow regime.

1. Introduction

For several decades conventional contactors like agitated contactors [1], packed bed columns [2], RTL extractors [3,4] etc. have been used in chemical process industries to carry out liquid-liquid extraction. The disadvantages of conventional contactors such as large space requirements and high energy input motivates us to develop an extraction process which effectively uses raw materials and requires low energy input. These objectives can be met by carrying out extraction in microfluidic devices as they possess the advantages of high surface area to volume ratio, low diffusion path lengths and low inventory requirements. Microfluidics is also a preferred choice in systems where toxic chemicals are manufactured or handled as the volumes processed are low. It also plays an important role in systems where high heat transfer rates are required [5]. Due to the low diffusion path lengths, micro-channels are very effective in analyzing two-phase processes which are limited by mass transfer. These systems are characterized by having a characteristic dimension which is less than 1 mm [6].

Liquid-liquid two phase flows play a vital role in liquid-liquid extraction and two-phase reactions (phase transfer catalysis). Depending on the channel geometry and material, fluid properties and operating conditions, different types of flow patterns are observed in micro-channels. In liquid-liquid two phase systems stratified flow, core-annular flow, slug and droplet flows are commonly observed depending on the interplay of inertial, viscous and surface tension forces. Sarkar et al. [7] and Zhao et al. [8] studied liquid-liquid two-phase flow patterns in a serpentine glass and a PMMA micro-channel respectively. Both observed seven different flow patterns such as slug flow, slug and droplet flow, droplet flow, unstable annular flow, annular flow, annular dispersed flow and fully dispersed flow. The flow pattern affects the mass transfer rate and hence conversion and selectivity when carrying out chemical engineering applications as consecutive-competitive reactions in them.

Stratified flow in which two phases flow side by side offers ease of phase separation at outlet. This flow pattern has applications in the field of liquid-liquid extraction and phase transfer catalysis. Here, the inertial forces dominate the viscous forces. Mixing in each phase occurs primarily by diffusion, as convection is parallel to the interface. An alternate way to enhance mixing and the mass transfer rates in a micro-channel is by operating in the slug flow regime. This is a very stable flow regime and is realized at very low flow rates. In this regime the interfacial forces dominate the

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http://dx.doi.org/10.1016/j.cep.2016.03.010
0255-2701/

Nomenclature

List of symbols

H	Width of channel (m)
C_i	Dimensional concentration of species in the ith phase
c_i	Dimensionless concentration of species in the ith phase
$Q_i's$	Volumetric flow rate per unit depth of the ith phase ($m^2\,s^{-1}$)
L	Length of channel (m)
D_i	Diffusivity of solute in the ith fluid phase ($m^2\,s^{-1}$)
v_i	Velocity of the ith phase in stratified flow
k_{la}	Overall mass transfer coefficient (s^{-1})
Pe_i	Peclet number in the ith phase

Greek letters

$\varepsilon_{aq,f}$	Volume fraction of aqueous phase
$\varepsilon_{org,f}$	Volume fraction of organic phase
α_s	Holdup in stratified flow
μ_i	Viscosity of ith phase
τ_{res}	Residence time (s)

Subscripts

i = 1, 2	Phase-1, phase-2 in stratified flow
i = aq, org	Aqueous phase and organic phase

inertial forces and is characterized by strong internal circulations induced by shear. This gives rise to convection in the individual slugs [9]. This induces mixing in each phase which greatly enhances the extraction.

Kashid et al. [10] studied extraction using a non-reacting water-acetone-toluene system for six different flow regimes such as slug, slug-drop, deformed interface, parallel/annular, slug-dispersed and dispersed flow. Mass transfer coefficient was calculated for these regimes as a function of channel cross-section, flow rates and surface area. Fries et al. [11] studied the extraction of vanillin dissolved in water to the organic phase toluene. They showed that with a decrease in channel cross-section, the mass transfer coefficient increased. These two studies are primarily experimental in nature. Vir et al [12] developed a semi-analytical solution for extraction in core-annular and stratified flow. They compared their model prediction with experimental results from the literature. Malengier et al. [13] compared extraction under co-current and counter-current stratified flows theoretically and showed that counter-current flow resulted in a superior performance over co-current flow. Ghaini et al. [14] analyzed mass transfer to determine the effective interfacial area in slug flow using a physical and chemical method. Tamagawa and Muto [15] studied extraction of cesium (Cs^+) from aqueous cesium nitrate using a solvent (phosphoric acid di(2-ethyhexyl) ester in cyclohexane). In this study the extraction in a micro-channel was compared with batch extraction. They observed that in slug flow a high extraction efficiency could be obtained at a low residence time. Naleini et al. [16] compared the extraction of oleuropein from ethyl acetate into aqueous phase in a micro-channel with a batch system. They found that the extraction yield using a micro-channel with an extraction time of 0.13 min was 18% more than in the batch mode at 45 min. Nandagopal et al. [17] studied the extraction of phenol from dodecane into distilled water. They compared the slug flow based extraction performance with conventional extraction techniques such as batch extraction, microwave assisted extraction and ultrasound assisted extraction. They realized 94.18% extraction in the slug flow regime at a residence time of 3 min and 43.15% extraction in the batch mode in 16 min. Asano et al. [18] compared the extraction of sunflower oil from aqueous phase to hexane in batch system and in a microfluidic device with slug flow. They have shown that the microfluidic device is effective for improving extraction efficiency.

All these studies have focused on the improvement in extraction performance in a micro-channel compared to the batch process. Some studies have considered the hydrodynamics of the system [15,17] while others have focused on the improvement in extraction performance without giving emphasis to the flow regime [16]. Besides most of the models in the literature are based on the assumption of a linear equilibrium relationship of the solute between the two phases. This may not be valid for most systems. In this work we focus on improvement of extraction performance by considering hydrodynamics of system and describe how the models can be extended to the case when the equilibrium relationship is non-linear.

Several studies exist in the literature which describe the superior performance of the microfluidic system to a batch system. However in most of these studies it is not clear whether the comparison is fair. For example were the flow regimes in the two systems “equivalent”. For instance were the drop size of the batch system and the slugs in a micro-channel comparable? If so why is there an improvement in the micro-channel performance? At low stirring speeds the interface between the two fluids in batch mode would be flat and this would result in a poor performance of the batch system. At higher stirring speeds the contents are well mixed and a fine dispersion of one phase in another is formed. The size of the dispersed phase here is comparable to that existing in the micro-channels. The comparison of the extraction performance under these conditions would then be “fair”. To obtain a clear understanding of the physico-chemical processes involved, in this work we systematically study the extraction behavior in a batch mode as well as a micro-channel. The batch system is analysed for two cases i) when the interface is flat between the fluids and ii) when there is a well dispersed mixture of one fluid in another. The micro-channel is analysed under both stratified flow and slug flow. The study focuses both on experiments and modeling. Our objective is to analyse extraction keeping the conditions constant so that we can establish if indeed the performance of a micro-channel is superior. Specifically the batch reactor is studied in the well-mixed mode and the micro-channel in the slug flow mode to keep the dispersed phase size comparable. The slug and stratified flow experiments are performed in two channels with the same cross-section and varying lengths such that the mean residence times are comparable. Besides in all experiments both the phases are analysed and this is used to verify the accuracy of the experiments.

The paper is organized in the following manner. Section 2 discusses the material and methods employed during experiments. In Section 3, the experimental procedure and the lumped parameter model for the batch system is explained. Experiments in micro-channel and the mathematical model to analyse the extraction in a micro-channel are described in Section 4. Section 5 introduces various terms that are used to characterize extraction performance. In Section 6, the results of extraction in the different modes of operation are discussed and compared. Finally we summarize the results in Section 7.

2. Materials and methods

All the chemicals used in the experiments were analytical reagent (AR) grade. Propionic acid (>99%), toluene (>99%), 0.1 N sodium hydroxide (NaOH), and phenolphthalein indicator were purchased from Merck India Ltd. The aqueous phase used for the extraction of propionic acid was milli Q water. Two different