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Single-stage micro-scale extraction: Studies with single microbore tubes and scale-up

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ABSTRACT

The study focuses on single-stage solvent extraction of HNO_3 from 3 M HNO_3 using 30% TBP in dodecane in microbore tubes. The objective of the study is to identify a configuration that ensures high stage efficiency ($\geqslant$90%), quick settling and low pressure drop ($\leqslant$2 bar) and to demonstrate this configuration at a total throughput of 1 LPH using parallel paths. To find out the right configuration, experiments with single microbore tubes are conducted to observe the effects of flow rate, diameter and length of microbore tube on stage efficiency, pressure drop and settling behavior. The experiments show that the configuration consisting of a Y-junction of 750 μm diameter having a single microbore tube of 800 μm diameter and 300 cm length on its downstream side is capable of handling total flow rate of 0.5 LPH with high stage efficiency, quick settling and low pressure drop. Two such configurations are used in parallel to achieve total flow rate of 1 LPH. Specific extraction rate achieved in the contactor based on microbore tubes is compared with the specific extraction rate estimated for a mixer-settler. The comparison shows that the use of microbore tube contactor leads to process intensification.

1. Introduction

Chemical processing in microchannels has several distinct advantages. High surface to volume ratio in a microchannel ensures intensified heat transfer and is very useful for carrying out highly exothermic reactions [1–4]. Numbering up approach followed for scale-up makes the scale-up easy and certain [5–7]. Small volumes of microchannels lead to low inventory of chemicals in the setups based on microchannels. This makes such setups very useful for carrying out reactions involving toxic and hazardous chemicals [8–13].

There are several advantages of carrying out extraction in microchannels. Unlike conventional contactors, quality of dispersion can be controlled in a microchannel to ensure fast settling [14]. Fast settling can reduce the size of phase disengagement section of the contactor eventually resulting in reduction of solvent inventory and footprint area of the contactor. Unlike conventional contactors, dispersion generated in a microchannel can be tailored to have very narrow drop size distributions [15,16]. This can avoid the problems of generation of fine droplets and resulting entrainment losses which is often a problem in conventional contactors [17]. Due to smaller flow cross-sections, the size of the dispersed phase is restricted leading to high specific interfacial area and high overall volumetric mass transfer coefficient [18–24]. Scale-up is very easy and confident as throughput is increased by numbering up approach [14].

Driven by the advantages of using microchannels for solvent extraction, there have been many studies on liquid–liquid two-phase flow and mass transfer in microchannels. While there are studies on understanding flow regime maps of liquid–liquid two-phase flow [25,26], studies focused on a specific flow regime e.g. slug flow are also reported [27,28]. Both commercially available microchannels [21–23,29,30] as well as microchannels fabricated in-house [24,31] have been used for carrying out solvent extraction. While there are studies with phase systems of interest of the authors [32,33] there are several studies in which standard phase systems recommended by European Federation of Chemical Engineering (EFCE) [21,29,34,35] have been used. While most of the studies are with single microchannels [32,33,36,37], there are few studies focusing on achieving higher flow rates using parallel microchannels or micromixer [29,38] or specially designed microchannels [31].

One of the attractive features of solvent extraction in microchannels is that overall volumetric mass transfer coefficient increases with reduction in contact time. High stage efficiencies in contact times of the order of few seconds have been reported

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Nomenclature

a	specific interfacial area (m^{-1})
C_A^{in}	solute concentration in the aqueous phase at the inlet (mol l^{-1})
$C_{A,Eqb}^{in}$	solute concentration in the aqueous phase in equilibrium with the solute concentration of the organic phase at the inlet (mol l^{-1})
C_A^{out}	solute concentration in the aqueous phase at the outlet (mol l^{-1})
$C_{A,Eqb}^{out}$	solute concentration in the aqueous phase in equilibrium with the solute concentration in the organic phase at the outlet (mol l^{-1})
C_O^{out}	solute concentration in the extract (mol l^{-1})
D_i	diameter of i^{th} microbore tube (μm)
D_{max}	maximum diameter among available microbore tubes (μm)
D_T	diameter of selected microbore tube (μm)
K_D	distribution coefficient (–)
K_L	overall mass transfer coefficient (m s^{-1})
K_La	overall volumetric mass transfer coefficient (s^{-1})
L_T	tube length (cm)
L_{min}	minimum tube length (cm)
ΔL	change in tube length (cm)
N_T	numbers of tubes (–)
O/A	ratio of organic flow rate to aqueous flow rate (–)
P	power input (W)
ΔP	pressure drop (bar)
ΔP_{max}	maximum permissible pressure drop (bar)
Q	flow rate in a single microbore tube (LPH)
Q_A	aqueous flow rate (LPH)
Q_T	Total flow rate (LPH)
V_{Total}	total volume of the contactor (l)
V_{mixer}	mixer volume (l)
$V_{settler}$	settler volume (l)

Greek letters

Δ_{LMC}	log mean concentration difference (mol m^{-3})
τ	residence time (s)
η	stage efficiency (–)

Abbreviations

EFCE	European Federation of Chemical Engineering
LMCD	log mean concentration difference
LPH	liter per hour
MS	mixer-settler
MTC	microbore tube contactor
PTFE	polytetrafluoroethylene
SER	specific extraction rate
SER_M	specific extraction rate in mixer
SER_{MS}	specific extraction rate in mixer and settler
TBP	tributyl phosphate

[32,33,41]. This makes them particularly attractive for deployment in the back-end of the nuclear fuel cycle where short contact times are required to avoid solvent degradation [42–47]. Criticality is one of the possible hazards in solvent extraction in the back-end of nuclear fuel cycle [47,48]. High surface to volume ratios in microchannels can keep criticality hazard at bay. Reduction in inventory resulting in compact setup can reduce shielding requirement. There are several studies reported on liquid–liquid extraction in microchannels using phase systems relevant to nuclear fuel cycle [22,32,33,40]. The aqueous phase used in this study is 3 M HNO_3. The organic phase used is 30% (v/v) TBP (tributyl phosphate) in dodecane. This phase system is relevant to nuclear fuel cycle in which extraction of nuclear materials such as uranium is often carried out from nitrate medium of about 3 M acidity using 30% (v/v) TBP in dodecane as the extractant [22,32,49,50].

The objective of the paper is to propose a methodology to be followed for finalizing a setup for micro-scale extraction at higher throughput. The target higher throughput in this study is 1 LPH. Table 1 summarizes throughputs used in some of the studies on solvent extraction in microchannels reported earlier. As can be seen, the throughputs in most of the studies are small. There are some studies in which throughputs of the order of 1 LPH or higher are reported but specially made microchannels or devices have been used to realize such throughputs [29,31]. Also in most of the studies on solvent extraction in microchannels the focus is on intensification of mass transfer. In real life application, besides mass transfer phase disengagement or settling is equally important. If the phase disengagement of the dispersion generated in the contactor is not quick, significant inventory will be locked in the settling device or settler. This will defeat the very idea of miniaturization. Though there are many studies on liquid–liquid extraction in microchannels, studies with a holistic view considering both mass transfer and settling are very few. The requirement of fast mass transfer and quick settling often conflicts. When throughput in a single microbore tube is increased, the dispersion tends to become finer and film coefficients also increase. Thus an increase in throughput in a microbore tube leads to intensification of mass transfer. But as the dispersion becomes finer, settling tends to become slow. Thus considering both mass transfer and settling, there exists a limit on the throughput a single microbore tube can handle.

In this study we take the holistic view of the solvent extraction process and propose a simple methodology which can be used to decide an optimum configuration that ensures fast mass transfer along with quick phase separation and low pressure drop. The study consists of two parts. The first part is focused on experiments with single microbore tubes to identify a configuration that gives high stage efficiency in short contact time with quick settling of resulting dispersion and low pressure drop. In the second part of the study, two such configurations are used in parallel to demonstrate solvent extraction at 1 LPH total throughput. The maximum allowable pressure drop considered in this study is 2 bar. This relatively lower limit on allowable pressure drop is to minimize the possibility of pressure driven leakages.

2. Experimental

2.1. Experimental setup

Fig. 1 shows the image and Fig. 2 shows the schematic diagram of the experimental setup used for the first set of experiments i.e. the experiments with single microbore tubes. This setup consists of a Y-junction of 750 μm diameter drilled in a polytetrafluoroethylene (PTFE) disk. The included angle between the two inlets of the Y-junction is 120°. A replaceable microbore tube of PTFE is connected to the disk using a threaded connector. Dispersion generated at the Y-junction passes through the microbore tube and collects in a glass sample bottle which allows visual observation of settling behavior of the dispersion. The aqueous and organic phases are pumped at the desired flow rates by a dual channel syringe pump (Model ASP-3.0, Syrris Ltd. UK). The aqueous samples are analyzed for nitric acid concentration using acid base titration using 1 N NaOH solution.

Fig. 3 shows the image and Fig. 4 shows the schematic diagram of the experimental setup used for the second set of experiments