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Ultrasound assisted liquid–liquid extraction with a novel interval-contact reactor

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ABSTRACT

A novel reactor was developed for ultrasound-assisted liquid–liquid extraction. This reactor design entails introducing short contact intervals for the microchannel tubing along the reactor plate channel to have a more focused transmission of the ultrasound. The non-contacted parts of the tubing are still under the influence of the ultrasound as a result of the pseudo-sonicated zone created by the adjacent intervals. The effect of introduction of these elements was first studied by comparing the thermal profiles with and without the presence of intervals and it was found that the maximum intensities along the channel become focused at these intervals. The influence of the intervals on a sonicated two-phase flow was also studied and revealed a repetitive splitting (at the intervals) and coalescence (downstream from the interval) of the emulsified aqueous phase. This dynamic change in the size of the emulsified aqueous phase introduces additional interfacial area and improves the mass transfer between the phases. The number of intervals was varied between three, five and seven. The five intervals showed the best performance. On comparing the five-interval design with a direct-contact design it was shown that the interval design gave the best improvement in yield for the process conditions studied.

1. Introduction

Miniaturisation of chemical reactors is a promising field of reactor design to improve mass and energy transfer. An important unit operation in chemical processes is liquid–liquid (or solvent) extraction, for which microstructured devices are also studied. Liquid-liquid extractions consist of heterogeneous systems of immiscible liquids. Microreactors can provide intensified effects for such systems owing to the different flow patterns that can be generated as a result of the mixing elements used and the flow rate applied [1–7]. The various flow patterns generated can provide very high interfacial areas, shorter diffusion lengths and internal mixing effects, even under laminar flow conditions [5,8–13]. To accommodate for sufficient throughput in order to match the industrial production capabilities, methodologies such as numbering up [14] have been developed, which involves introducing identical parallel flow channels. However, numbering up may lead to an excessive amount of parallel channels in order to reach the desired throughput. To reduce the need for numbering up, increase of the reaction, separation or transport rates may be a better approach. In extraction processes, intensified mixing of the immiscible phases or of the liquid volumes close to the interfaces of the immiscible phases is appropriate to increase the extraction rate. There are many methods developed for improving the mixing effects, which are broadly classified as passive and active [15].

The passive methodology involves using innovative structures, splits, curvatures and obstructions in the microstructure to intensify mixing [6,16,17]. A two-phase system that has been reported is the hydrolysis of *p*-nitrophenyl acetate. This is an instantaneous pseudo-first order reaction which is mass-transfer controlled. Plouffe et al. [6] compared the performance of this reaction with several of the passive elements available and saw that these were not really effective in improving the performance of this system, which was quantified in terms of the conversion and the volumetric mass transfer coefficient. Plouffe et al. [6] then proceeded to improve the performance of the system by changing the solvent to one that has an increased solubility with the aqueous phase and ensuring parallel flow in the two phase system. As this may not always be feasible for different process, an alternative method is to utilize external forms of energy to improve the mixing effects. This approach is called an active methodology.

There are many active methods available [15,18,19], with ultrasound being one of them. When ultrasound in the frequency

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Nomenclature

Latin
$K_l a$ Volumetric mass transfer coefficient (s^{-1})

Greek
α Fraction of the reactant consumed (−)
τ Residence time (s)

range of 20 kHz to 2 MHz is applied to a liquid it is known to create cavitation bubbles, which in turn cause various chemical and physical effects within the liquid medium [20]. In the case of solvent extraction the physical effects are of more importance. The physical effects are a result of cavitation bubbles formation, vibration and collapse, which cause improved internal circulation that enhances the mass transfer resulting in faster and increased conversion [19,21–24]. When ultrasound is applied to a liquid–liquid extraction system, an emulsion of one of the phases in the other is created. This formation of emulsion leads to increased interfacial areas between the two immiscible phases resulting in an enhanced mass transfer flux between the phases. The mechanism of this emulsion formation by ultrasound has already been discussed by different researchers [18,25,26].

There are many methodologies developed for combining ultrasound with microreactors, the most common of which is having the microstructured device suspended in a liquid medium and ultrasound being applied to the medium for effective transfer [18,19,27]. In most cases this is achieved by making use of an ultrasonic bath. These methods were proved to be effective in improving the performance of the hydrolysis reaction. Such methods are, however, accompanied by different disadvantages such as dissipation of power in the liquid medium, inhomogeneous ultrasonic field, non-reproducibility and dependence of the performance on the dimensions and type of the ultrasonic bath used. A different approach is the direct application of ultrasound to the microreactor without using a liquid medium for energy transfer.

A design for the direct application of ultrasound to a microreactor was presented in previous work [26], which made use of an aluminium plate having channels cut through it to transfer the ultrasound directly to the microchannel tubing. This design was shown to be effective in transferring the ultrasound to the two-phase segmented flow in a microchannel with the yield of the *p*-nitrophenyl acetate hydrolysis reaction increasing by 2.5 times in comparison to the non-sonicated condition for tubing of 0.8 mm at a residence time of 87 s. This work aims at further improving the performance of this design by focusing the transfer of the ultrasound along each channel.

2. Materials and methods

2.1. Design of reactor

The reactor assembly is very similar to the direct-contact type reactor described in a previous publication [26]. The new interval-type reactor, used in this study, consists of an aluminium plate of 4 mm thickness and dimensions of 80 × 80 mm with 4 square channels cut through it. These channels have the same dimensions as the outer diameter of the tubing to be placed in them. The four channels account for four passes through the sonicated zone. The PFA (perfluoroalkoxy) tubing with internal diameter of 0.8 mm and outer diameter of 1.6 mm is held in place in the square channels with a plexiglas cover plate of 5 mm thickness bolted at the four corners on top of the aluminium plate. PFA is used for the tubing even though it has low acoustic impedance as it provides good resistance to different organic solvents and also because of its transparency, which is helpful in studying the effects of ultrasound on a two-phase flow. The plexiglass is used for the cover plate as it is a sound-soft material and hence it is less prone to breakage on applying ultrasound. The entire assembly is bolted onto the transducer whose input parameters are then controlled by a wave form generator and amplifier combination (Fig. 1(a)). The experimental setup was discussed in detail in a previous publication [25] and the reader is referred to this publication for further information.

An observation in the previous publication [26] on the direct-contact type reactor was the presence of a pseudo-sonicated zone, which showed sonication activity in a region of the tube that was not embedded in the reactor plate. This region existed for a distance of up to 8 cm away from the reactor plate, both at the inlet and outlet of the reactor. This behaviour was also observed in the tubing bends in between the sonicated passes, which are also outside the reactor plate. The cross-section of a part of a single

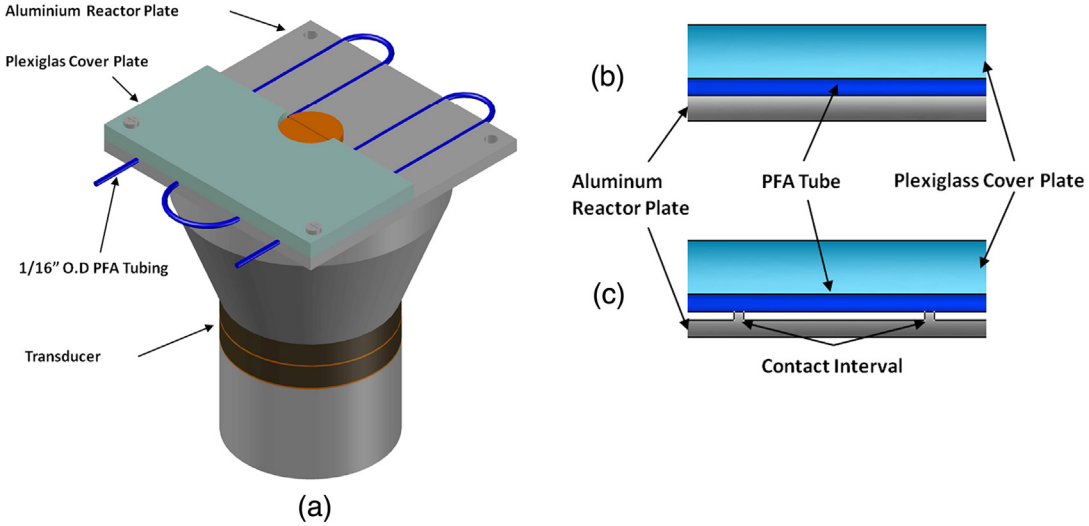

Fig. 1. (a) Schematic representation of the reactor (single column), (b) Cross section of single channel direct-contact type reactor [26], (c) Cross section of single channel for the interval-contact type reactor.

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