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John, Jinu Joseph; Kuhn, Simon; BRAEKEN, Leen & Van Gerven, Tom (2017)

Ultrasound assisted liquid-liquid extraction with a novel interval-contact reactor. In:  
CHEMICAL ENGINEERING AND PROCESSING, 113(SI), p. 35-41.

DOI: 10.1016/j.cep.2016.09.008

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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 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].

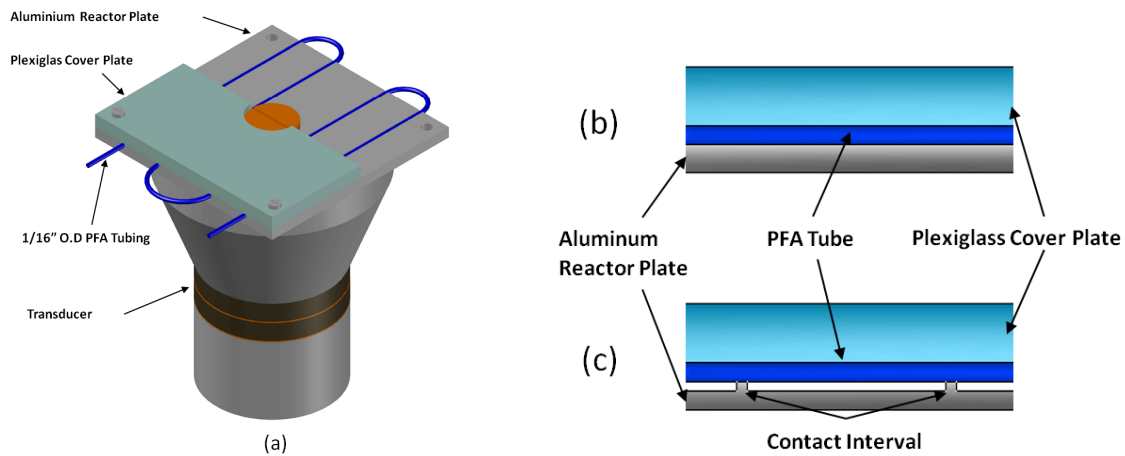
There are many methodologies developed for combining ultrasound with microreactors, the most common of which is having the microstructured device suspended in a liquid medium and ultrasound being applied to the medium for effective transfer [18,19,27]. In most cases this is achieved by making use of 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.**

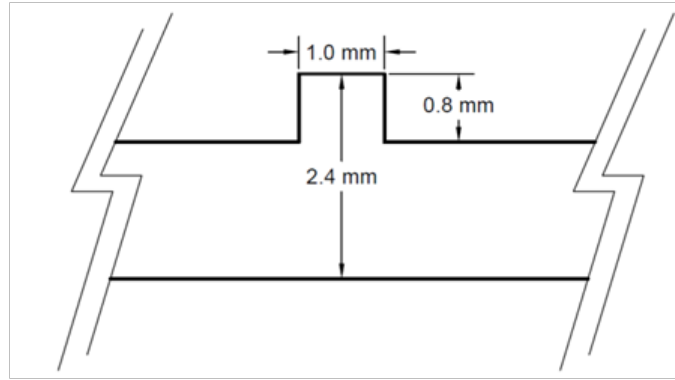
### **2.1 Design of Reactor**

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 80x80 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 has 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 (Figure 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.



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

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 channel of the direct-contact reactor is shown in Figure 1 (b), where it can be seen that the tube is in contact with the reactor plate along the full length of the channel. Based on the pseudo-sonicated zone observed in the direct-contact type it was hypothesized that the contact with the entire length of the channel may not be really required, so that by reducing the contact length it may be possible to have a more focused transmission of sound at specific points along the channel.

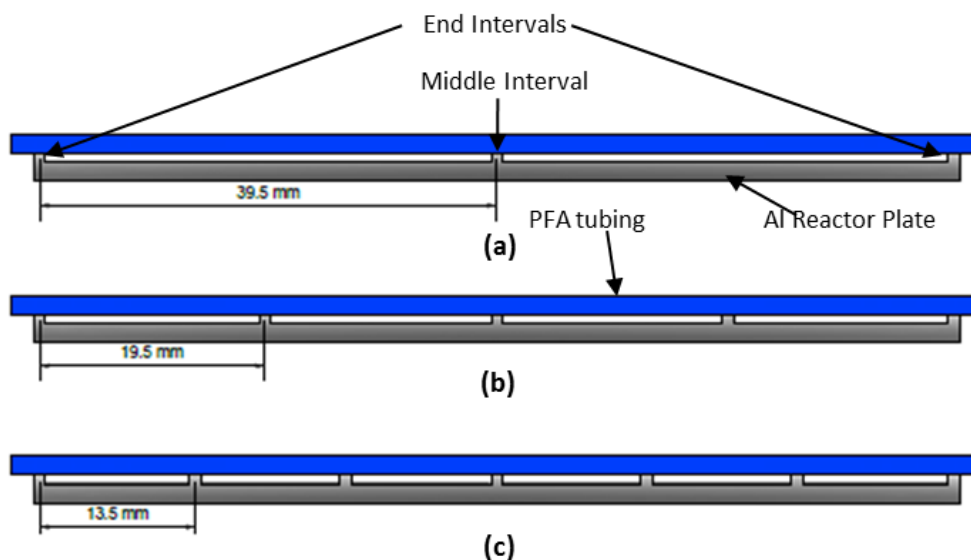


**Figure 2. Design details of a single interval**

Therefore, a new design was developed where the tubing is not in contact with the plate along the entire length as shown in Figure 1 (c), but only at specific points. For reference purpose we will term these points of contacts as intervals. Each interval (Figure 2) is 1 mm wide and has a height of 2.4 mm from the bottom of the plate. The space between the intervals is at a height of 1.6 mm from the bottom of the plate, so the tubing is in contact with the bottom part of the channel only at the intervals. Thus only the intervals are at the same height at which the tubing was in contact in the direct-contact type. We assume with this configuration that, instead of the entire length of channel transmitting the sound waves in the form of one single element, the individual intervals act as different focused transmission points along the length of the channel. The parts of the tubing that are not in contact with the intervals will still be under the influence of the ultrasound as a result of the pseudo-sonicated zone created by the adjacent intervals.

The number of interval elements was varied to determine the ideal number required along each channel for an effective transfer of the ultrasound. There are different ways in which the intervals could be arranged on the channel, but the following design criteria were used to ensure stability and comparability of the designs. (a) There are always end intervals for each channel as absence of them can cause the tubing to sag and no longer provide the intermittent contact required. (b) An interval is to be placed at the centre of the channel as they ensure at least one interval will always coincide with the position of the transducer as a circular transducer is used on a square plate.



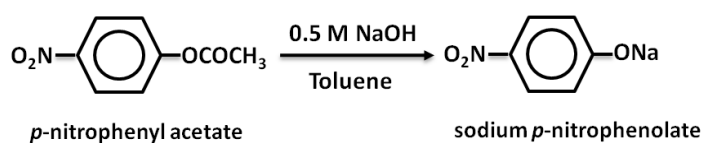


**Figure 3: The arrangement of intervals (cross section of one channel) (a) Three Intervals arrangement (b) Five Intervals arrangement (c) Seven Interval arrangement.**

Hence, three variations of three, five and seven intervals along each channel were selected and studied. They were arranged along the channel as shown in Figure 3. With this arrangement three plates were manufactured to perform the required experiments. The thermal profile of the plates for different variations in the design and process parameters were also studied using a FLIR T335 thermal camera with a spatial resolution of 1.36 mrad. The qualitative behaviours of the two phase system is studied with a high speed camera Photron Fastcam Mini UX-100.

## 2.2 Reaction

The reaction of interest is the hydrolysis of *p*-nitrophenyl acetate, as this has been well described in terms of solvent extraction and acoustic enhancement [13,18,26].



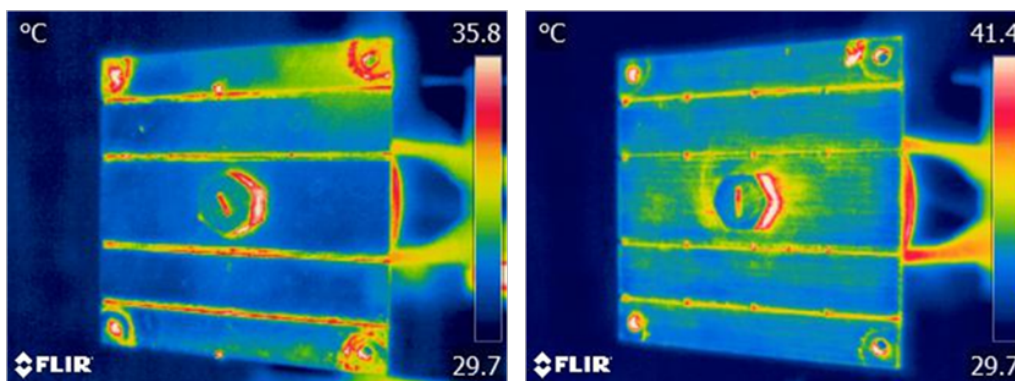
**Figure 4: Hydrolysis of *p*-nitrophenylacetate**

The hydrolysis of *p*-nitrophenyl acetate is a reactive extraction process (Figure 4). The organic input stream is *p*-nitrophenyl acetate dissolved in toluene at a concentration of 0.05 M, while the aqueous phase consists of a 0.5 M NaOH solution. This is an instantaneous reaction and hence it is mass transfer controlled. As an excess of NaOH is used, this reaction can be considered as a pseudo-first order reaction. When the sodium *p*-nitrophenolate is dissolved in water, the solution turns yellow, which can be utilized to quantify its concentration in a UV-1601 Shimadzu spectrophotometer at a wavelength of 400 nm.

### 3. Results

#### 3.1 Effect of the interval

The effect of introducing the intervals along the length of the channel was first studied in terms of the thermal profile of the reactor plate when sonicated for a certain amount of time. Both the direct-contact type and the interval-contact type with 5 intervals were subjected to a frequency of 20 kHz, at an amplitude of 590 mV and net applied electrical power of 20 W. The thermal profile was then observed after a period of 30 min with the thermal camera. The images obtained are shown in Figure 5.

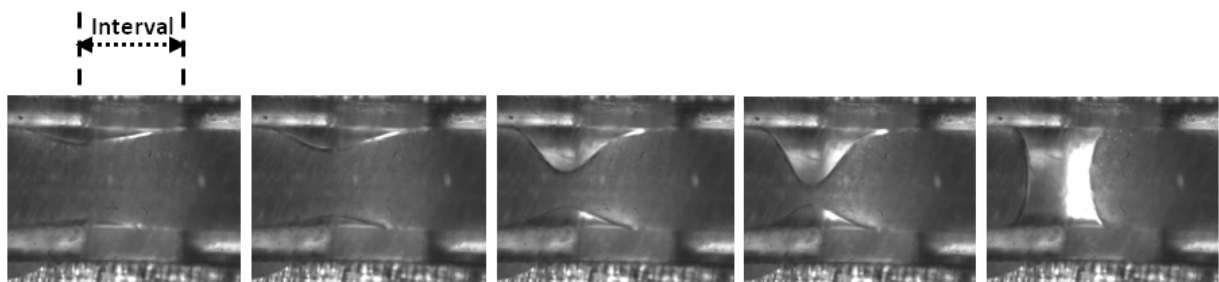


*Figure 5: Thermal imaging temperature for (a) the direct contact type and (b) the interval contact type*

From the images obtained it can be seen that for the direct-contact type the high temperature area in the channel is spread in a non-uniform way along the length of the channel, whereas the introduction

of the intervals in the interval-type reactor seems to concentrate the high temperature points at the intervals. Also it is to be noted from the images that the high intensity points of the interval-contact type are at a higher temperature (average of 37 °C - 41 °C at the intervals) than the direct-contact type (average of 32 °C – 35 °C along the channels) . Considering that the temperature variation is approximately proportional to the ultrasound intensity variation along the plate, we perceive that the objective of focusing the ultrasound at defined points along the channel is met with this design.

In addition to the thermal profile of the reactor plate, the behaviour of the two-phase system was also observed with a high speed camera. The organic phase is the continuous phase and the aqueous phase is the dispersed phase. There was emulsification of the aqueous phase by the organic phase upon sonication with a similar initiation mechanism as observed in the direct-contact type [26] , which made use of the vibration of the bubble clusters and large bubbles formed in the continuous phase to disturb the interface and cause the emulsion formation. In addition to the emulsification, a specific behaviour was observed along the interval points of especially the third and the fourth pass of the tubing, where the emulsified aqueous phase appeared to split, as shown in Figure 6.



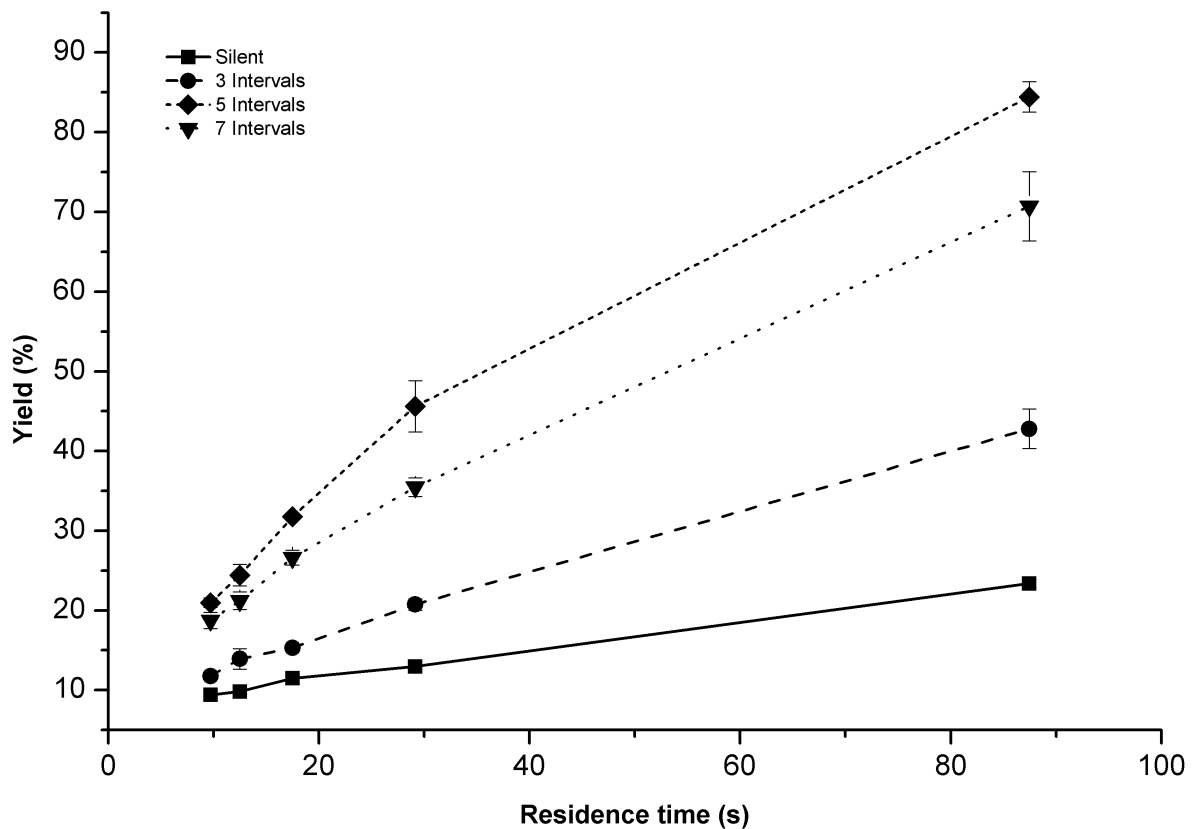
**Figure 6: Behaviour of emulsified slug at an interval (start to end 0.031S)**

Sonication of the two-phase flow causes the regularity of the segmented phases to be disturbed by alternately causing coalescence and subsequent splitting up again. The increase in interfacial area by emulsification, reported earlier [26], is combined in this case with the increase in mixing by the repetitive cycles of coalescence and phase splitting, thereby increasing the mass transfer between the

phases, and can further improving the yield of the process. Hence, this design seems to be beneficial for the desired fluids interaction.

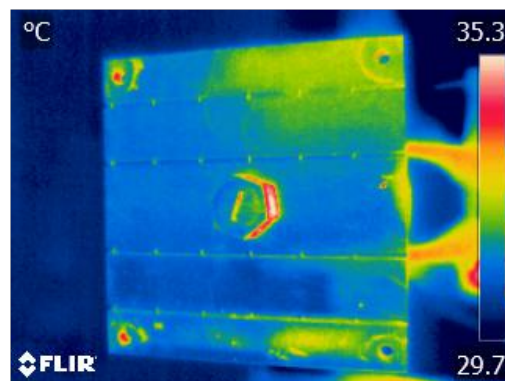
### 3.2 Variation of the number of intervals

Having studied the effect of the presence of the interval on the design and fluid flow characteristics, the next step is to obtain the ideal number of intervals along each channel. The sonicated reactive extraction of *p*-nitrophenyl acetate experiment was carried out in each of these plates at a frequency of 20.3 kHz, amplitude of 590 m.V and the net applied power of 20 W. In all the experiments both the aqueous and the organic streams are pumped at the same flow rate (i.e. phase ratio 1:1). The extraction yields obtained at different residence times are plotted in Figure 7.



**Figure 7: Variation of yield with number of intervals per channel of the reactor plate. Error bars calculated on the basis of three replicates. (If no error bars are shown they are smaller than the symbol).**

From the graph it is very evident that irrespective of the number of intervals there is always an improvement in the yield by sonication. Comparing the three interval designs, the lowest yield at all residence times was obtained for the 3 interval arrangement, which is possibly related to insufficient ultrasound contact length (i.e. three times 1 mm). In the three-interval design the contact length per channel is reduced by a factor of 26 in comparison to the direct-contact type. Comparing the five-intervals and the seven-interval designs, it is seen that the highest yield was obtained for the five-interval type, even though the contact length for the 7 intervals is greater (7 mm compared to 5 mm for the five-interval type). To better understand this, the thermal profile of both reactor plates was captured with the thermal camera after sonication at the same operating conditions for a period of 30 minutes. The infrared image for the five-interval type was already shown in Figure 5(b), while that for the seven-intervals design is shown in Figure 8.



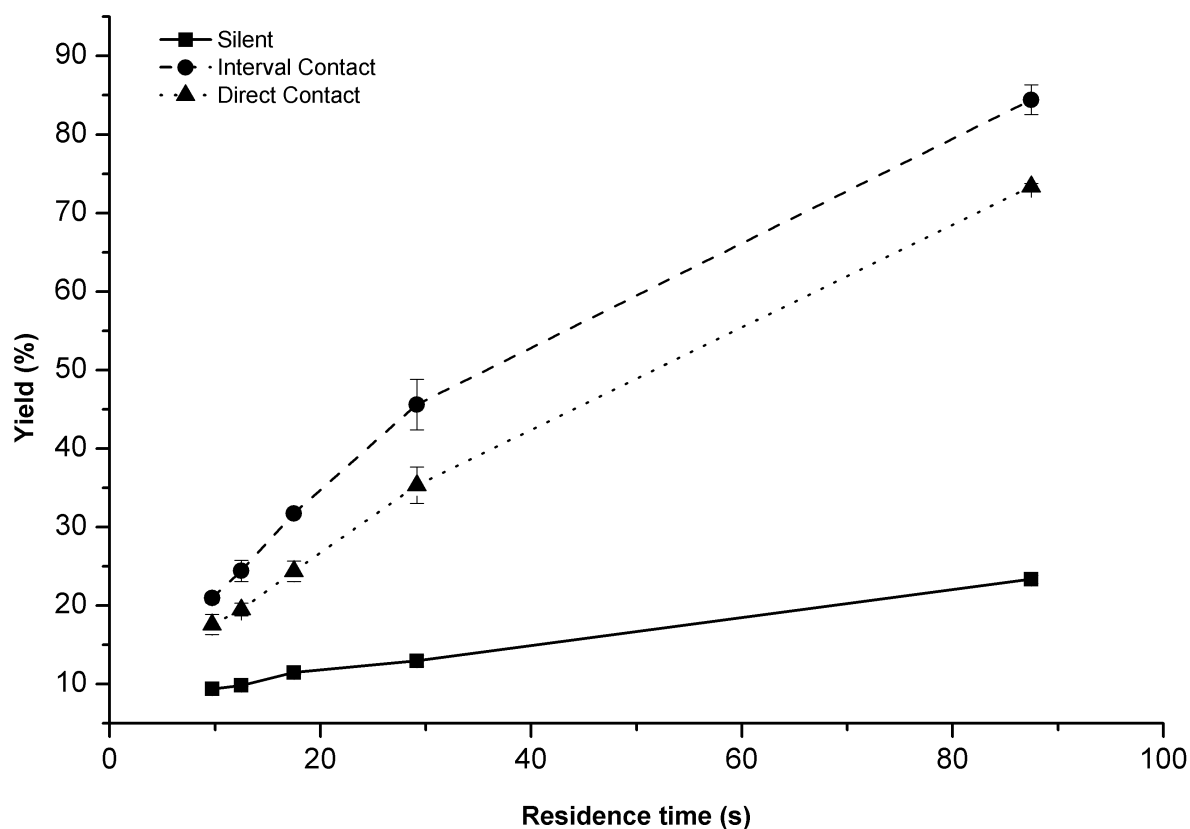
**Figure 8: The thermal profile of plate for seven intervals per channel of the reactor plate.**

Form the thermal profile it is seen that there is a clear difference in the temperature distribution along the surface of the plate depending on the number of intervals. The five-interval design shows a higher temperature in comparison to the seven-interval design, both overall (five interval average of 35.85 °C, seven intervals average of 32.5°C and at the intervals (five interval average of 37 °C - 41 °C, seven intervals average of 32°C - 33°C) . Higher temperature is considered to represent better power transmission along the channel, and hence the five-interval design provides better transmission of power, resulting in better yield of the extraction process. It is hypothesized the intervals being placed

at halfway and quarter way points of the channels may be contributing to the improved performance, but the exact reason for the better performance by the five-interval reactor will be investigated in further research.

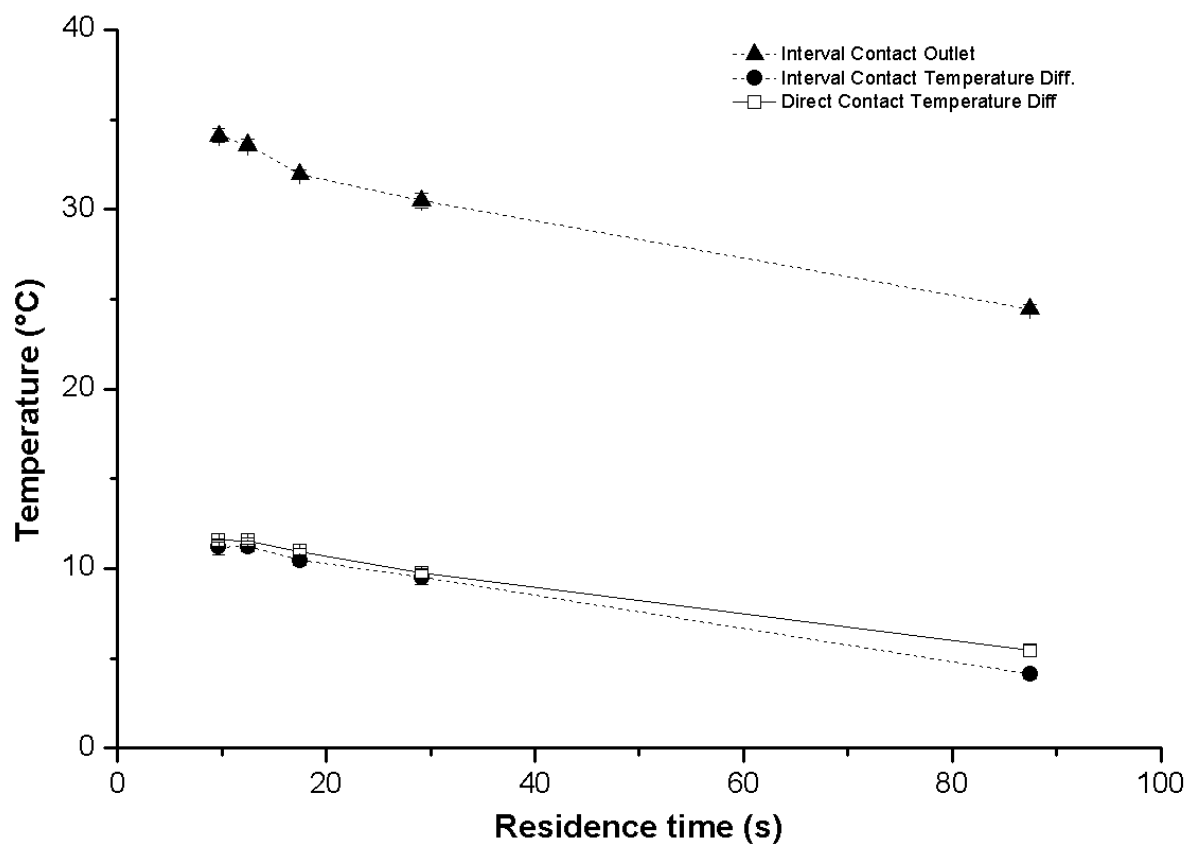
### 3.3 Comparison with direct-contact type reactor

The five-interval design was compared to the direct-contact type at a frequency of 20.3 kHz, amplitude of 590 mV and net applied power of 20 W at the same residence times in terms of the yield obtained for the sonicated hydrolysis reaction of *p*-nitrophenyl acetate. The results obtained are shown in Figure 9.



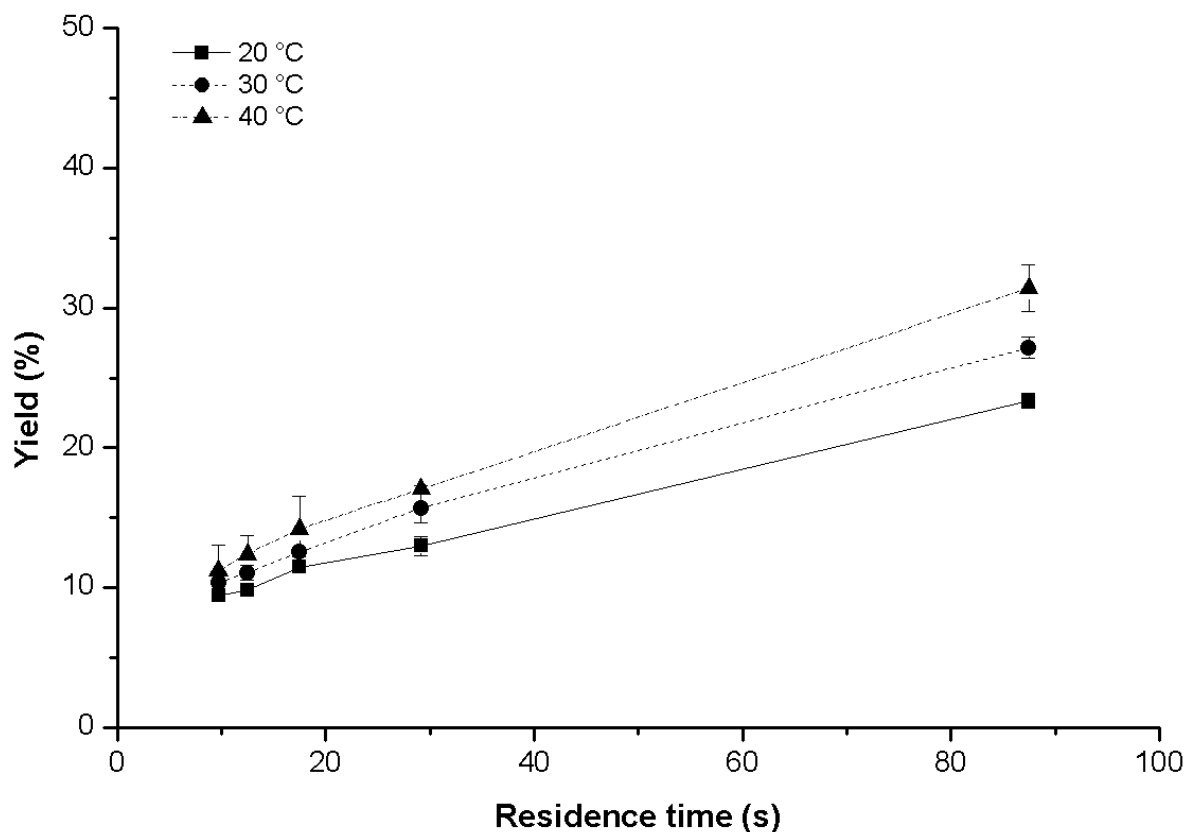
*Figure 9: Variation of extraction yield with reactor design type. Error bars calculated on the basis of three replicates. (If no error bars are shown they are smaller than the symbol).*

From the results it is evident that the five-interval design performs better at all the residence times compared to the direct-contact type. The causes of this improvement were investigated. Firstly, It was investigated whether the increase in yield was a result of the temperature increase by the supply of ultrasound to the two-phase system. The temperatures at the inlet & outlet of the reactor plate were measured at the operating ultrasound parameters and residence times for the interval reactor setup with a thermocouple placed in line with the tubing at the entry & exist of the reactor plate. The results obtained are plotted in Figure 10.



**Figure 10: Variation of outlet temperature and the difference between outlet and inlet with residence time. Error bars calculated on the basis of three replicates. (If no error bars are shown they are smaller than the symbol).**

From Figure 10 it is observed that the maximum increase in temperature that was obtained at the outlet was 35 °C. Experiments were conducted at silent conditions at 20 °C, 30 °C and 40 °C to see the extent of the yield change with temperature (Figure 11).



**Figure 11: Variation of yield with change in temperature at silent conditions. Error bars calculated on the basis of three replicates. (If no error bars are shown they are smaller than the symbol).**

It is evident that even at 40 °C the yields obtained from silent processing were not a match for the yields upon sonication. Hence, rather than thermal effects the physical effects of ultrasound contribute considerably to the increase in yield of the hydrolysis reaction.

The temperature differences between the inlet and outlet of both the reactor plate designs were measured at the same operating conditions and are plotted in Figure 10. From these temperature differences it might seem that the behaviour of the system is contradicting the heat transfer principles where for a constant heat source the temperature difference between outlet and inlet should increase with residence time. However, in this reactor design there is no insulation of the reactor tubes, leading to larger thermal dissipation from the reaction liquid at longer residence time, causing in turn a lower outlet-inlet temperature difference. From Figure 10 it is observed that there are no large differences in temperature between the interval and direct-contact designs, leading to the conclusion that in

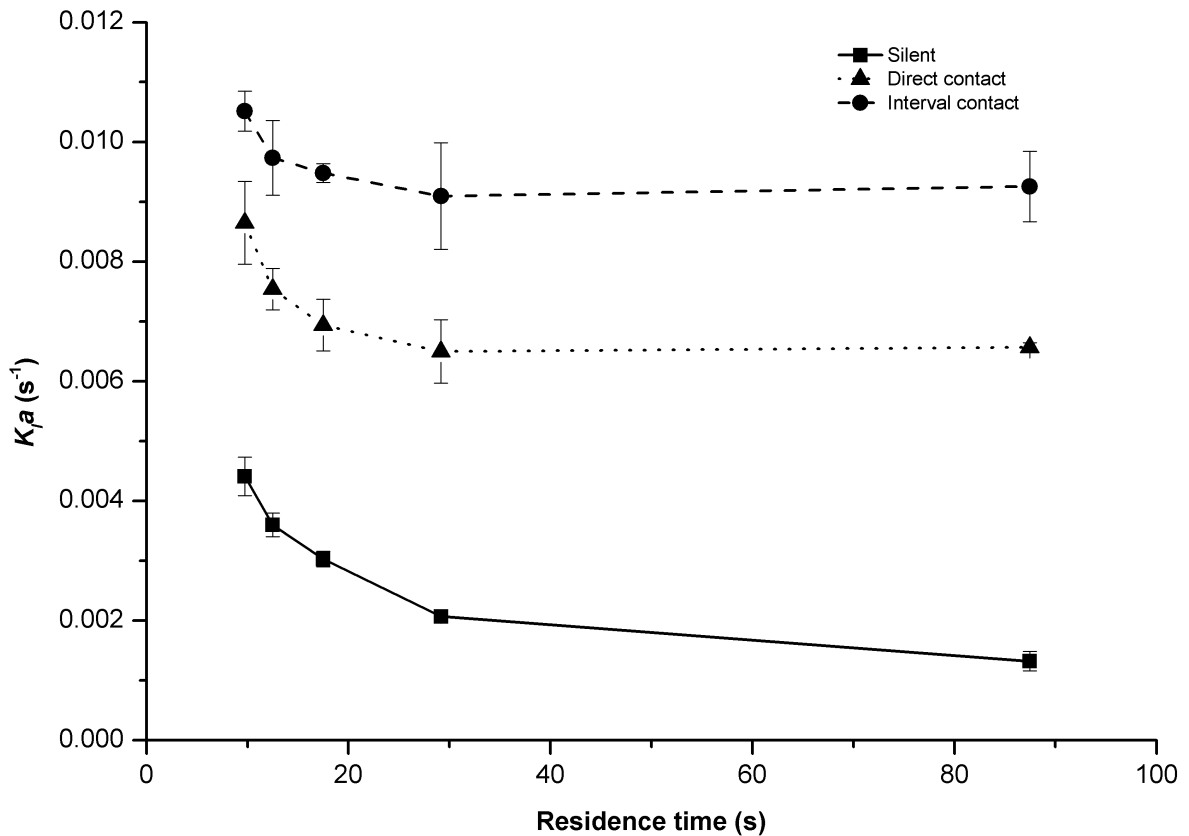


terms of the calorimetric power delivered to the reaction liquid, there is no real difference. This is unexpected because the contact length of the interval type is reduced by a factor of 16 in comparison to the direct-contact type. The only other parameter that can really influence the yield is the interfacial area between organic and aqueous phase. The fluctuating size of the dispersed phase by sequential coalescence and splitting due to the presence of the interval as described in section 3.1 seems to be providing the additional improvement in mass transfer, resulting in higher yields for the interval design.

The improvement in mass transfer was quantified by the estimation of the volumetric mass transfer coefficient ( $K_L a$ ).  $K_L a$  was calculated for this reaction utilizing the method proposed by Ramshaw & Burns [11]. The procedure has been described earlier [25]. The equation used is as follows:

$$K_L a = -\frac{1}{\tau} \ln (1 - \alpha)$$

Where  $\tau$  is the residence time and  $\alpha$  the ratio of the *p*-nitrophenyl acetate consumed at time  $\tau$ . The variation of  $K_L a$  with residence time for the two design types is shown in Figure 12.



**Figure 12: Variation of volumetric mass transfer coefficient with reactor design type. Error bars calculated on the basis of three replicates. (If no error bars are shown they are smaller than the symbol).**

The results obtained for the silent condition show that as a result of the nature of the equation used the  $K_L a$  value seems to exhibit an asymptotic behaviour at higher residence time. With sonication the entire curve shifts upward with the asymptote reached faster. Comparing the interval design to the silent condition, the highest values of  $K_L a$  are observed for the interval-contact type which shows an improvement in the range of 58 to 85 % at residence times of 87 s to 9.7 s respectively.

Comparing the interval-contact design to the direct-contact design, as the temperature difference between the inlet and the outlet streams of both the interval and direct-contact reactors are almost similar (Figure 10) it can be assumed that the  $K_L$  values are similar and that the increase in the  $K_L a$  value can be attributed to the increase in the interfacial area available for mass transfer due to the splitting and combining of the emulsified aqueous slugs. Hence, the improvement in the interfacial area is

calculated to be in the range of 17 to 29 % from the increases in the  $K_L a$  values of the interval contact design with respect to the direct contact design.

## 4. Conclusion

The introduction of intervals along the channels for direct-contact transfer of ultrasound to the microchannels was found to be effective in improving the performance of a reactive extraction process. The intervals brought about two major improvements to the design. Firstly, the points of high temperature became concentrated at the intervals and, secondly, the intervals caused a repetitive change in the size of the emulsified aqueous phase which contributed to additional improvement in mass transfer between the two immiscible phases. The best configuration in terms of number of intervals was found to be five intervals per channel for this particular reactor size and geometry. The improvement in yield of the hydrolysis reaction was found to be the highest for the five-intervals design compared to the direct-contact design at all residence times for the operating conditions studied. When studying the variation in residence time the improvement in the extraction yield under sonication conditions for the interval design, compared to the direct-contact design was between 13 to 23 % and in terms of the volumetric mass transfer coefficient between 17 to 29%.

## 5. Acknowledgements

The research leading to these results has received funding from the European Community's Seventh Framework Programme (FP7/2007-2013) under grant agreement n°NMP2-SL-2012-309874 (ALTEREGO).

## 6. Notation

Latin

$K_L a$  Volumetric mass transfer coefficient  $s^{-1}$

Greek

$\alpha$	Fraction of the reactant consumed	-
$\tau$	Residence time	s

## 7. Reference

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