

which is difficult to measure directly due to its sensitivity. The results demonstrate that the symmetrical inlet geometry as well as pressure drop and particle separation play an important role in the cyclone separator. Further investigations including the effects of turbulence characteristics and particle load on the flow pattern are required in order to more accurately reveal the mechanism of pressure drop and particle separation in the symmetrical inlet cyclone.

Received: March 18, 2005 [K 0088]

Symbols used

<i>a</i>	[m]	inlet height
<i>b</i>	[m]	inlet width
<i>B</i>	[m]	particle outlet diameter
<i>D</i>	[m]	cyclone body diameter
<i>D_e</i>	[m]	gas outlet diameter
<i>h</i>	[m]	cyclone cylinder height
<i>H</i>	[m]	cyclone height
<i>L</i>	[m]	natural length of cyclone
<i>n</i>	[–]	outer vortex exponent
<i>r</i>	[m]	radial dimension
<i>R</i>	[m]	cyclone body radius
<i>S</i>	[m]	gas outlet duct length
<i>T</i>	[K]	gas temperature
<i>v</i>	[m/s]	gas velocity

Subscripts

in	cyclone inlet
r	radial coordinate direction
s	static pressure
t	tangential coordinate direction

References

- [1] B. Zhao, H. Shen, Y. Kang, *Powder Technol.* **2004**, *145* (1), 47.
- [2] A. J. ter Linden, *Proc. Inst. Mech. Eng.* **1949**, *160*, 129.
- [3] Y. Wei, Y. Jin, Z. Ji, M. Shi, in *Proc. of INTERPEC*, Beijing, China, **1991**.
- [4] R. Wakelin, *Ph. D. Thesis*, University of Canterbury **1992**.
- [5] D. R. Garwood et al., in *Filtech Conf.*, Horsham, UK, **1993**.
- [6] P. A. Patterson, R. J. Munz, *Can. J. Chem. Eng.* **1996**, *74*, 213.
- [7] W. Peng, A. C. Hoffmann, P. J. A. J. Boot et al., *Powder Technol.* **2002**, *127*, 212.
- [8] L. X. Zhou, S. L. Soo, *Powder Technol.* **1990**, *63*, 45.
- [9] A. C. Hoffmann, M. de Groot, A. Hospers, *Can. J. Chem. Eng.* **1996**, *74*, 464.
- [10] L. Z. Wang, Q. S. Yan, L. L. Liu, *Aerosol Sci. Technol.* **2001**, *35* (5), 909.
- [11] S. Obermair, J. Woisetschläger, G. Staudinger, *Powder Technol.* **2003**, *138*, 239.
- [12] R. M. Alexander, *Proc. Inst. Min. Met.* **1949**, *152*, 202.

Application of a Capillary Microreactor for Selective Hydrogenation of α,β -Unsaturated Aldehydes in Aqueous Multiphase Catalysis*

By *Yücel Önal, Martin Lucas, and Peter Claus**

The direct regioselective hydrogenation of α,β -unsaturated aldehydes in aqueous multiphase catalysis was investigated in a capillary microreactor with channel diameters ranging from 500 to 1000 μm . By connecting two mixers in line, a regular and stable alternating plug flow behavior could be generated, allowing accurate measurement of the plug geometry and hence the calculation of the specific surface area relevant for mass transport. These parameters are essential for reliable kinetic modeling of multiphase reaction systems containing microkinetic as well as transport terms.

1 Introduction

The employment of microreactors instead of conventional reactors for performing kinetic and high throughput experiments [1] has proven very fruitful in the past years. Special emphasis is being placed on high heat and mass transport rates in microreactors due to an enhanced specific surface area [2–4]. This important feature of microreactors is also appreciated when carrying out reactions comprising multiple phases. For example, the direct fluorination of aromatics being a highly exothermic reaction can be carried out isothermally even under relatively harsh reaction conditions. Compared to conventional reactors, operated at relatively mild conditions because of safety reasons, improved space-time yields can be achieved [5–7]. Microreactors even allow examination of heterogeneous catalysts by using microstructured plates coated with the catalyst by an appropriate method. The fluid phase passes through the microchannels and reacts over the catalyst.

In the past years, special fluid dynamic characteristics in microchannels have motivated some research groups to successfully carry out L/L-biphase reactions in capillaries with channel diameters less than 1000 μm [8–11]. By pumping the two immiscible liquids via two separate feed lines and bringing them together in an appropriate mixer (T or Y mixer), an alternating plug flow of the two liquid phases is generated. Since channel walls are very close, resulting Re numbers are small and laminar flow characteristics within the plugs are obtained. The geometry of the plugs depends on

[*] Lecture presented at the DECHEMA/GVC Annual Meeting, Karlsruhe (Germany), Oct. 12–14, 2004.

[**] Dipl.-Ing. Y. Önal, Dipl.-Ing. M. Lucas, Prof. Dr. P. Claus (claus@ct.chemie.tu-darmstadt.de), TU Darmstadt, Fachbereich Chemie, Ernst-Berl-Institut, Technische Chemie II, Petersenstr. 20, D-64287 Darmstadt, Germany.

the channel diameter, the mixer type and the flow rates of the liquid phases. Reaction takes place either at the interphase of the plugs or following mass transport within the bulk of a specific phase comprising, for example, a soluble catalyst. The impact of flow rate and geometry of the plugs on the mass transport rate have been extensively studied for nitrogenation of aromatics which is a mass transport limited reaction [9]. Mass transport rates are commonly faster than in conventional reactors, since diffusion pathways defined by the channel diameter are often shorter than in usual reactors. Additionally, special fluid dynamics within the plugs can enhance mass transport in the capillaries. This regards mainly internal convective circulations, also called Taylor flow, occurring in the plugs due to shear forces at the channel walls opposite to the flow direction. Hence, provided the reaction of interest is limited by mass transport, the replacement of a conventional reactor by a capillary-microreactor results in an improved overall mass transport coefficient and improved reaction rates.

The objective of the present work is to investigate the direct regioselective hydrogenation of α,β -unsaturated aldehydes in aqueous multiphase catalysis (see Fig. 1) in a capillary microreactor. The main advantage of carrying out homogeneously catalyzed reactions in a biphasic or multiphase mode in industry is the possibility of simplifying catalyst recycling to a phase separation without any thermal stress and catalyst deactivation caused, for example, by distillation steps. As a matter of fact, homogeneous catalysis is becoming more attractive for industrial application, since energy consumption and environmental impact can be dramatically lowered by the new process design. The catalyst used for the hydrogenation reactions in the present work is Ru(II)-TPPTS (TPPTS: triphenylphosphine trisulfonate sodium) which is highly soluble in the aqueous phase [12]. The substrate is used as solution in an organic solvent which is immiscible with water. Hydrogen gas serves as reduction agent.

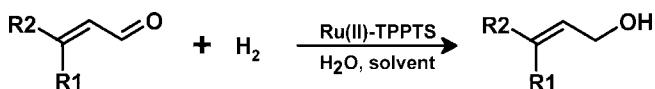


Figure 1. Chemoselective hydrogenation of α,β -unsaturated aldehydes to unsaturated alcohols with Ru(II)-TPPTS by aqueous multiphase catalysis.

Full conversions with complete selectivity to the unsaturated alcohol can be achieved in a batch reactor at moderate temperatures ($50\text{--}60^\circ\text{C}$) and hydrogen partial pressure (2.0 MPa) within a few hours, depending on the substrate. Reaction rates are in the kinetic regime and can be described by a simple rate law according to Eq. (1):

$$r = \frac{dc_{PA,org}}{dt} = -k \left(\frac{P_{H_2}}{K_H} \right)^{1,2} \left(\frac{c_{PA,org}}{K_N} \right)^{0,1} c_{Ru,aq}^{0,9} \quad (1)$$

According to the literature available, the investigation of G/L/L-multiphase catalytic reactions in a capillary micro-

reactor has been performed for the first time. Investigational results will be reported below.

2 Experimental Setup

2.1 Characterization of Aqueous and Organic Phase

The homogeneous water-soluble complex Ru(II)-TPPTS is a highly active and selective catalyst for the regioselective hydrogenation of carbonyl groups in α,β -unsaturated aldehydes [12]. During reaction at elevated hydrogen pressure (> 0.5 MPa) and reaction temperature ($> 50^\circ\text{C}$), the catalytic active species is generated *in situ* by reaction of the metal precursor RuCl₃ and TPPTS within a few minutes. Prior to its use in the capillary-microreactor, RuCl₃ and TPPTS were pretreated in aqueous solution ($c_{Ru} = 0.005$ M, $c_{TPPTS} = 0.05$ M) in a batch reactor at $p_{H_2} = 2.0$ MPa and $T = 50^\circ\text{C}$ for 1 h. The solution was then feeded in a piston pump for further application in the microreactor. The aqueous solution was buffered at pH 7.0 because, as is known from preliminary experiments, the pH value has a dramatic impact on the chemical nature of the complex generated and hence on the selectivity of hydrogenation.

As substrates for selective hydrogenation, prenal ($R_1 = R_2 = \text{CH}_3$, $c_0 = 0.5$ M) and citral ($R_1 = \text{CH}_3$, $R_2 = \text{C}_6\text{H}_{11}$, $c_0 = 0.25$ M) were used as solutions in an organic solvent immiscible with water (n-hexane, toluene). Prior to their use in the microreactor, they were pretreated in an ultrasonic bath for 15 min in order to remove the dissolved oxygen.

2.2 Capillary Microreactor

A schematic presentation of the capillary microreactor is shown in Fig. 2. The main device of the microplant is a PTFE capillary which was used for better visualizing the fluid dynamic behavior and which can be operated at a maximum pressure limit of 3.0 MPa. The fluid characteristics of the plug flow within the capillary regarding the length of the fluid plugs and the specific surface area between them can be further analyzed by photographic imaging. Optionally, three different capillaries with internal diameters of 500, 750, and 1000 μm were used. The lengths of the capillaries were fitted to the internal diameters in a way that the total reaction volume was kept constant (3, 6, and 12 m).

The organic and the aqueous fluid are fed via two separate lines and brought together in a T-mixer (Valco, Mixer I). At this point, hydrogen is not mixed with the liquid phases. The organic phase is pumped by a HPLC pump with a volumetric flow rate of 250 $\mu\text{L min}^{-1}$ whereas for the supply of the aqueous phase a piston pump with a filling volume of 100 mL is used, enabling flow rates smaller than 600 $\mu\text{L min}^{-1}$. The resulting alternating L/L-plug flow consisting of the two liquid phases is then mixed with hydrogen in a second T-mixer (Valco, Mixer II). The volume rate of hydrogen was controlled by a MFC in the range of 0 to 10 000 $\mu\text{L min}^{-1}$.

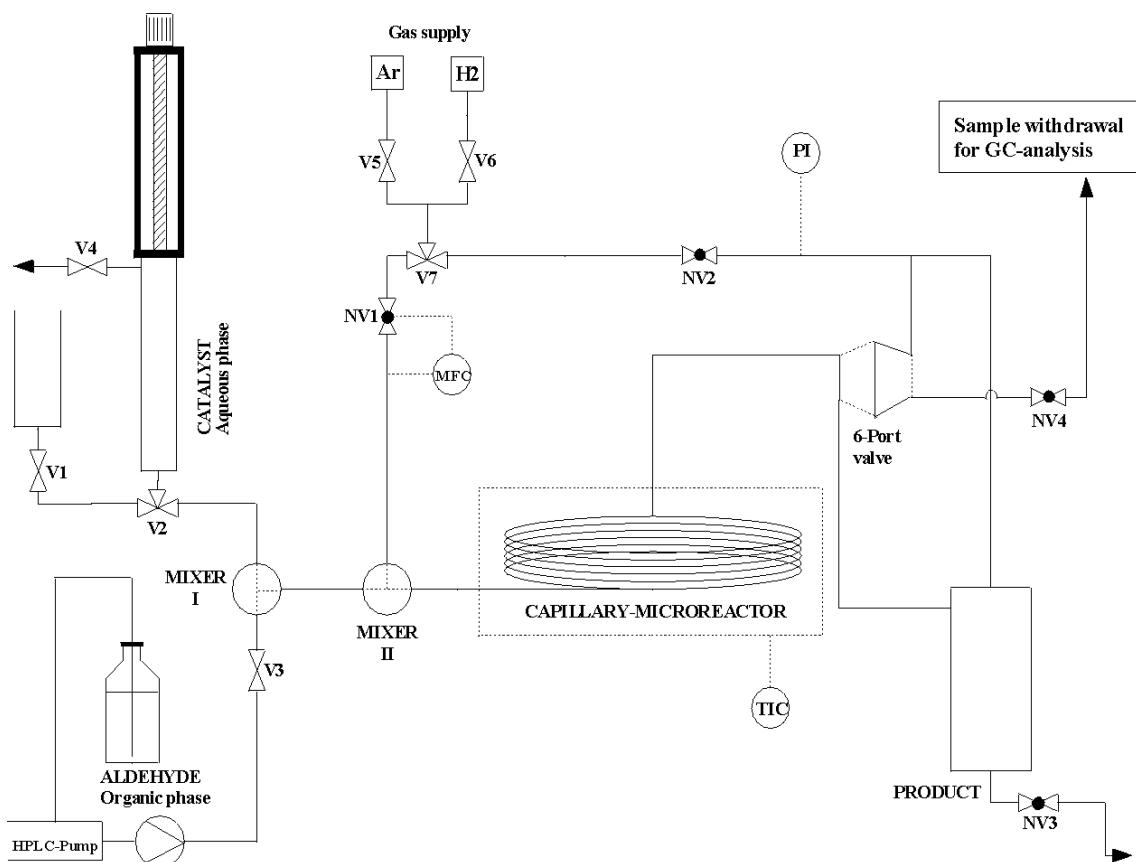


Figure 2. Flow chart of microplant with capillary microreactor.

The internal diameter of the channels within the mixers was fitted to the internal diameter of the capillaries used, so that a constant and stable plug flow could be generated. Six mixers, two each with the same channel diameter as the used capillary, were applied. The capillary was connected directly to mixer II, so that the fluid dynamic behavior of the G/L/L plug flow could be analyzed at the exit of the mixer. The temperature of the capillary was normally kept at 60 °C. Hydrogen partial pressure corresponding to the overall pressure in the capillary ranged between 1.0 and 2.0 MPa. Samples of the reaction mixture were drawn at the exit of the capillary via a 6-port valve. After phase separation, the organic phase of the samples was analyzed by gas chromatography.

3 Results

3.1 Fluid Dynamic Characterization

With the above combination of two mixers (channel diameter < 1.0 mm) connected in line it is possible to generate a regular and stable fluid dynamic behavior of the G/L/L multiphase system. The flow presents a plug flow of alternating fluid elements consisting of the aqueous and the organic phase. Since the organic phase has a higher affinity for

PTFE, the inner wall of the capillary is completely covered with the organic phase, so that the aqueous phase is never in direct contact with the capillary wall. Therefore, hydrogen fed to the L/L-mixture at mixer II can be located only in the organic phase (see Fig. 3).

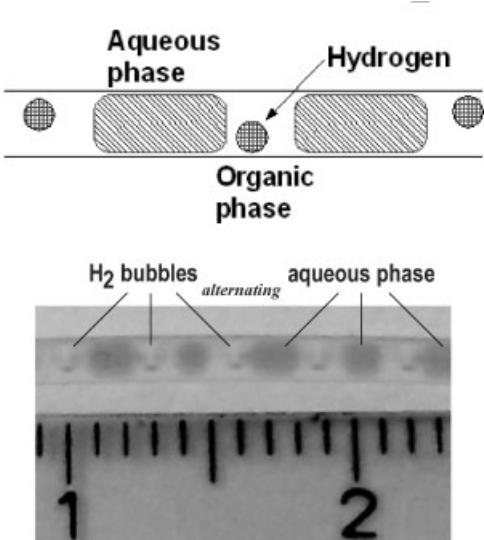


Figure 3. G/L/L-flow scheme in capillary reactor (left: schematic, right: image).

Provided the mean length of the liquid plugs has a value of 1.3 mm as it is the case for the G/L/L-plug flow in a capillary with an internal diameter of 750 μm ($T = 60^\circ\text{C}$, $p_{\text{H}_2} = 1.0\text{--}2.0 \text{ MPa}$, $\dot{V}_a q = 420 \mu\text{L min}^{-1}$, $\dot{V}_{\text{org}} = 250 \mu\text{L min}^{-1}$, $\dot{V}_{\text{H}_2} = 2800 \mu\text{L min}^{-1}$), the specific surface area adds up to 769 m^{-1} , on the assumption that the area relevant for mass transport is composed of the base and top face of each cylindrical plug.

There is an essential need for extremely precise pumping devices providing stable flow rates of the aqueous and organic phase at relatively small flow rates in order to generate a regular G/L/L-plug flow behavior, since that is the most important parameter influencing the geometry of the plugs.

3.2 Volume Rate of Catalyst Phase

Prenal hydrogenation was examined at various volume rates of the aqueous phase which contains the homogeneous catalyst and in which the reaction is supposed to take place. The volume rates of the organic phase ($250 \mu\text{L min}^{-1}$) and hydrogen ($2800 \mu\text{L min}^{-1}$) were kept constant. Reaction temperature was 50°C and hydrogen pressure within the capillary was 2.0 MPa . Conversions as well as observed reaction rates clearly increase as the volume rate of the aqueous phase increases in the range of 0.19 to 0.51 mL min^{-1} (see Fig. 4).

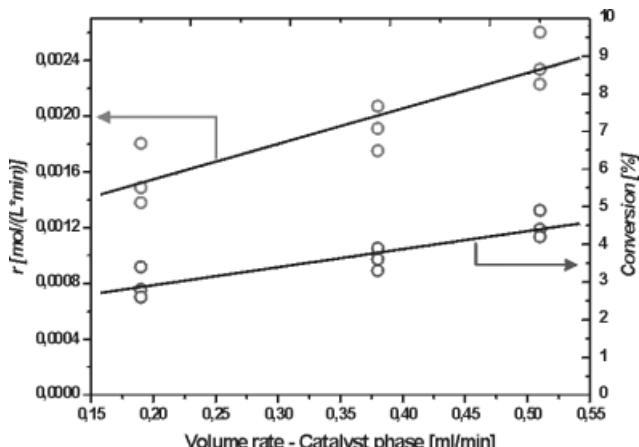


Figure 4. Influence of catalyst phase volume rate on conversion and observed reaction rate.

The increase in volume rate of the aqueous phase results in an increase in the characteristic Re number of the fluid flow and the overall mass transport coefficient k which directly correlates with the Re number. As a matter of fact, the mass transport rate at the L/L-phase boundary is accelerated which is confirmed by the enhanced reaction rate determined experimentally. Although an increasing trend can be observed for the conversion rates, the values still remain at relatively small values, since the mean residence time within the capillary is only 2 to 3 min, depending on the overall flow rate. On the other hand, as changes in the substrate

concentration are very marginal, the reaction rate over the whole length of the capillary can be assumed to be constant.

3.3 Hydrogen Flow Rate

One of the main factors limiting the observed reaction rate in direct hydrogenations carried out in a batch reactor is the mass transport rate at the G/L-phase boundary. The rate of hydrogen uptake within the reactor can therefore be essential in optimizing the overall reaction rate. The capillary microreactor offers the advantage that this effect can be examined at a micro-dimensioned level. To show this, hydrogen flow rate was slowly increased ($0\text{--}10\,000 \mu\text{L min}^{-1}$) at constant volume rates of the aqueous phase ($510 \mu\text{L min}^{-1}$) and the organic phase ($250 \mu\text{L min}^{-1}$), and the observed reaction rates for preenal hydrogenation were analyzed (see Fig. 5).

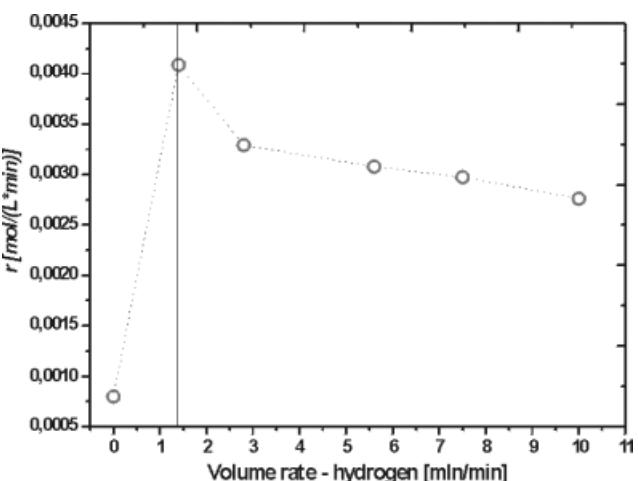


Figure 5. Influence of hydrogen flow rate on overall reaction rate in preenal hydrogenation, $T = 50^\circ\text{C}$, $p_{\text{H}_2} = 2.0 \text{ MPa}$.

Additionally, the phase behavior of the G/L/L-plug flow system was characterized at various volume hold-ups of the gas phase and correlated with the observed reaction rates. It could be clearly shown that up to a hydrogen flow rate of 1.4 mL min^{-1} the observed reaction rate directly correlates with the gas volume hold-up. Increasing the hydrogen flow rate results in a slight decrease in the observed reaction rate. It could be demonstrated by visual analysis of the fluid phase behavior that at low hydrogen flow rates ($< 1.4 \text{ mL min}^{-1}$) hydrogen bubbles are not present in every organic plug. In these domains hydrogenation reaction is supposed to be slower. At a hydrogen flow rate of 1.4 mL min^{-1} hydrogen bubbles are present in every organic plug and reaction takes place in all domains at a comparable level. Further increase in hydrogen flow rate results in a slight decrease in the observed reaction rate because now there are more gas bubbles in the organic plugs, which reduces the effective reaction volume of the capillary and the mean residence time inside the capillary.

3.4 Reaction Temperature

The effect of reaction temperature on the overall reaction rate was examined for citral hydrogenation in a temperature range from 60 to 90 °C at constant hydrogen partial pressure (1.0 MPa) and constant volumetric flow rates of the aqueous, the organic and the gas phase. The observed reaction rates could be analyzed according to an Arrhenius plot which resulted in a theoretic activation energy of $32.3 \pm 1.3 \text{ kJ mol}^{-1}$ (see Fig. 6).

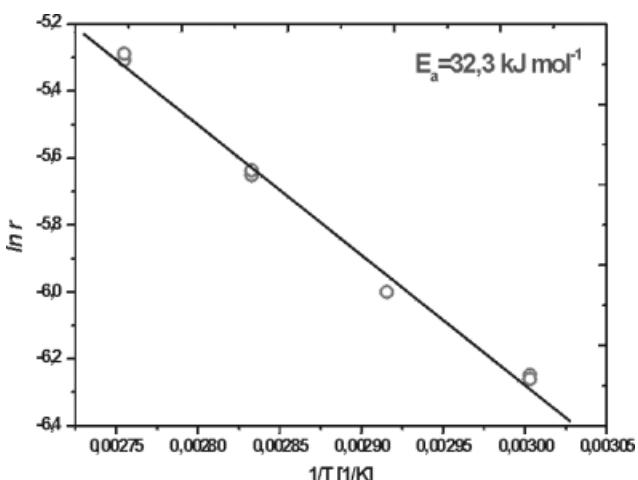


Figure 6. Temperature dependency of citral hydrogenation in capillary-micro-reactor.

The relatively low value of the activation energy indicates that the observed reaction rate is predominantly limited by mass transport particularly at the L/L-phase boundary because of the low solubility of citral in water ($\approx 1.0 \text{ g L}^{-1}$). As shown in section 3.2, the overall mass transport coefficient and hence the observed reaction rates can be accelerated by increasing the flow rate of the catalyst phase. As a matter of fact, a regular and stable G/L/L-plug flow, as shown in section 3.1, can only be generated at elevated flow rates by precise pumps that can be operated at a wide range.

The observed reaction rates can further be optimized by a decrease in droplet size which is accompanied by an increase in specific surface area. This can be achieved, for example, by decreasing the internal diameter of the capillary. Results concerning this issue will be presented in the next section.

3.5 Influence of Internal Capillary Diameter

Fluid characteristics in capillaries with internal diameters smaller than 1000 μm as used for investigations in this study, differ from the usual fluid dynamic behavior in conventional plug-flow reactors. This issue was already discussed in the literature [8–11]. Due to the relatively small diameter of the flow profile, shear forces opposite to the flow direction at the walls, caused by friction, must not be neglected. The smaller the capillary diameter the more this effect has to be

taken into account which generates internal convective circulations, also called Taylor flow, within the plugs. These convective streams can dramatically improve mass transport rates in a way which cannot be explained by classical reaction engineering concepts (see Fig. 7).

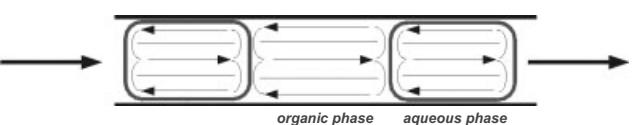


Figure 7. Internal convective circulations within plugs caused by shear forces (Taylor-flow).

In order to check whether this extraordinary fluid dynamic behavior can improve mass transport rates at the L/L-phase boundary and hence the overall reaction rate, citral hydrogenation which is supposed to be a mass transport limited reaction was examined in detail. Different capillaries with internal diameters of 500, 750, and 1000 μm, but with the same reaction volume were tested (see Fig. 8). By changing the capillaries droplet size and hence the specific surface area relevant for mass transport also changed.

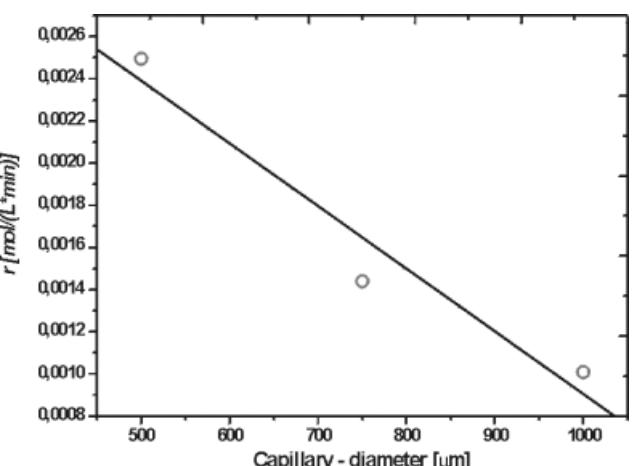


Figure 8. Influence of capillary diameter on overall reaction rate, $T = 60^\circ\text{C}$, $p_{\text{H}_2} = 2.0 \text{ MPa}$.

Overall reaction rates could be dramatically increased by decreasing the internal diameter of the capillaries. This behavior again confirms that mass transport at the L/L-phase boundary limits the overall reaction rate especially for citral hydrogenation, and that this step can be enhanced by using capillaries with smaller internal diameters. An improved specific surface area expected by applying smaller-scaled capillaries can certainly contribute to a faster overall reaction rate, however, of major importance are the internal convective circulations within the plugs accelerating mass transport rates at the L/L-phase boundary. To underline this thesis, theoretic activation energies should be measured again with smaller-scaled capillaries, which should yield higher values because microkinetics gets more involved in the overall reaction rate.

As already shown by Roberts [14], mass transport limitation can also strongly influence the selectivity in parallel reactions provided that the reaction order of the substrate is different. Unfortunately, this effect could not be observed in the presented model reaction systems. This may be due to the very high intrinsic selectivity of the catalyst system used for C=O hydrogenation or to the fact that the reaction order of the substrate is the same for both hydrogenation reactions. Nevertheless, the capillary microreactor is a very interesting reactor design for studying parallel reactions in multiphase systems because mass transport rates can be tuned in a very simple way, as shown above.

4 Discussion

As a matter of fact, the generation of a strongly ordered G/L/L-plug flow is of special importance for accurate determination of the geometries of the aqueous and organic plugs and hence of the specific surface area. These parameters are very important for reliable kinetic modeling of the multiphase reaction system containing microkinetic as well as mass transport terms. Therefore, the most important challenge in realizing G/L/L-multiphase reactions in a capillary microreactor for kinetic investigations is the generation of a regular fluid flow behavior (see Fig. 3). This can be achieved by connecting two mixers in line and mixing the fluids consecutively rather than mixing them all in one mixer unit. Further parameters which have to be optimized are the ratio of the volume rates of the aqueous and the organic phase, the internal diameters of the capillary as well as of the channels inside the mixers.

According to Ramshaw et al. [10], multiphase flow characteristics in capillaries with channel diameters less than 1000 µm are often laminar because of the low Re numbers (1–25), and mass transport at the phase boundaries is limited by diffusion. Investigational results obtained in this study confirm that the overall reaction rates of the model reaction systems examined in the capillary microreactor are limited by mass transport. This is proven by the results obtained from experimental investigations of capillary diameters, volume rates of the catalyst phase and the influence of reaction temperature on the overall reaction rate ($E_A = 32.3 \text{ kJ mol}^{-1}$).

The rate of diffusion is directly correlated with the overall mass transport coefficient k which, in turn, depends on the diffusion pathway according to the film theory. In capillary microreactors with very narrow internal diameters the diffusion pathway has approximately the same length, so that mass transport rates and hence overall reaction rates are supposed to become faster with decreasing internal diameter of the capillaries. This could be confirmed by testing the influence of differently scaled capillaries on the overall reaction rate (see Fig. 8). Another phenomenon characterizing plug-flow behavior in micro-scaled channels is the appearance of relatively strong internal convective circulations

within the plugs at sufficient flow rates caused by shear forces (see Fig. 7). Consequently, the pathway for diffusion becomes even shorter than expected, enhancing the mass transport rate [8]. Taylor flow is expected for capillaries with channel diameters in the range from 100 to 400 µm already at an overall flow rate of $6 \mu\text{L min}^{-1}$ [8].

Wörtz et al. [9] have already examined the nitrogenation of aromatics in biphasic L/L-reaction systems with a special emphasis on the influence of flow rate on conversion. The increase in reaction rate with increasing flow rate was attributed to an enhanced overall mass transport coefficient which was explained by the Taylor flow characteristics of the plug flow observed with increasing flow rate. Additionally, this fluid flow behavior could be successfully simulated by a CFD software. The internal diameters of the capillary and the mixer channel were in the range of 500 to 1000 µm as in the experiments performed here. The reaction system was examined at flow rates between 0.15 and 0.6 mL min^{-1} which are lower than those achieved in this study ($< 1.0 \text{ mL min}^{-1}$), so that convective circulations within the plugs are assumed to play a major role with regard to mass transport rates also in the reactions investigated in this work.

5 Conclusions

The selective hydrogenation of α,β -unsaturated aldehydes in an aqueous G/L/L-multiphase catalytic system was investigated in different capillary microreactors with channel diameters ranging from 500 to 1000 µm. By connecting two mixers in line, a regular and stable alternating plug flow behavior could be generated, allowing accurate measurement of the plug geometry and hence the calculation of the specific surface area relevant for mass transport. The influence of the catalyst phase volume rate on the overall reaction rate as well as the theoretic activation energy which has a relatively low value count for a mass transport limited reaction. In order to enhance the mass transport rate, the influence of the internal capillary diameter on the overall reaction rate was tested. An increase in the overall reaction rate with decreasing channel diameter was explained by a shortened diffusion pathway at the L/L-phase boundary, accelerating the mass transport rate. This effect is not only caused by an obvious increase in specific surface area, but also by the Taylor flow characteristics of the alternating plug flow, enhancing mass transport rates with decreasing channel diameters and increasing flow rates.

Very often overall reaction rates in multiphase reaction systems carried out in capillary reactors are limited by mass transport, as in the case of hydrogenation reactions investigated in here. As flow rates and droplet diameters regulated by the channel diameter can be adjusted very accurately, the capillary microreactor presents an excellent tool to investigate and model the influence of mass transport on the overall reaction rate and on the selectivity of the reaction.

Received: April 30, 2005 [CET 0147]

References

- [1] P. Claus, D. Hönicke, T. Zech, *Catal. Today* **2001**, *67*, 319.
- [2] K. Jähnisch, H. Löwe, V. Hessel, M. Baerns, *Angew. Chem.* **2004**, *116*, 410.
- [3] O. Wörz, *Chem. Unserer Zeit* **2003**, *34* (1), 24.
- [4] O. Wörz, K. P. Jäckel, T. Richter, A. Wolf, *Chem. Eng. Sci.* **2001**, *56*, 1029.
- [5] R. D. Chambers, R. C. H. Spink, *Chem. Commun.* **1999**, *10*, 883.
- [6] R. D. Chambers, D. Holling, R. C. H. Spink, G. Sanford, *LabChip* **2001**, *1*–*2*, 132.
- [7] K. Jähnisch et al., *J. Fluorine Chem.* **2000**, *105*, 117.
- [8] J. R. Burns, C. Ramshaw, *LabChip* **2001**, *1*, 10.
- [9] G. Dummann et al., *Catal. Today* **2003**, *79*–*80*, 433.
- [10] J. R. Burns, C. Ramshaw, *Trans IChemE* **1999**, *77/A*, 206.
- [11] J. R. Burns, *WO 01/64332 A1*, **2001**.
- [12] J. M. Grosselin, C. Mercier, G. Allmang, F. Grass, *Organometallics* **1991**, *10*, 2126.
- [13] Y. Önal, M. Lucas, P. Claus, *Patent Application*, Oct. 11, **2004**.
- [14] G. W. Roberts, *Chem. Eng. Sci.* **1972**, *27*, 1409.

This paper was also published in *German in Chem. Ing. Tech.* **2005**, *77* (1–2), 101.
DOI: 10.1002/cite.200407067

Power Consumption in Agitated Vessels with Dual Pitched Blade Turbines: Baffle Length and Impeller Spacing Effects

By Joannis Markopoulos*, Evangelia Babalona,
Evangelia Tsiliopoulou, and Katerina Tasopoulou

The power consumed in mechanically agitated vessels is an important parameter, especially for the energy requirements and the heat and mass transfer efficiencies of these systems. Power consumption does not only depend on the impeller type, the fluid properties and the stirring speed, but also on the geometry of the agitated system, including the impeller spacing and the length of the baffles in the mixing vessel. Information is presented on the combined effects of baffle length and impeller spacing on the total power consumption in vessels agitated by dual pitched blade impellers.

1 Introduction

In spite of the often use of dual and multiple impeller systems in chemical and biochemical processes, information available in the literature on power consumption in these systems cannot be considered as complete. Especially problems concerning the effects of baffle length and impeller spacing on the power consumed by these impeller configurations are not completely solved.

Unbaffled agitated vessels or weakly baffled systems consume less power than those with full baffles. This fact, connected also with the energy saving requirements in the process industry, as well as with the better performance reported in the literature for some cases of heat transfer, mass transfer and gas dispersion in unbaffled or weakly baffled systems, shows the importance of the baffle length parameter for an optimal design of these mixing equipments [1–4].

There are only few reports in the literature on the effect of baffle length L on power consumption in agitated vessels with different single impeller types [5,6]. In the turbulent flow regime the Newton number, Ne , was strongly depended on the geometrical parameter p/H , with H as the liquid height in the vessel. For the baffle length, L , and the distance between the lower edge of the baffles and the vessel bottom, p , the following condition is given¹⁾:

$$\frac{p}{H} + \frac{L}{H} = 1 \quad (1)$$

The greatest effect of p/H on Ne was observed for the single Rushton turbine, while no effect could be detected for the single pitched blade turbine, for $p/H \in [0, 0.5]$; on the other hand, a strong effect could be observed for $p/H \in [0.5, 1]$ also for the pitched blade turbine system.

In a previous work, data have been published on the effects of baffle length and impeller spacing on power consumption in vessels agitated by dual Rushton turbines [4]. A sharp decrease of power consumption could be detected for baffle lengths $L < 0.3H$. The Newton number was not significantly affected by L in the range of $0.3H < L < 0.5H$. For $L > 0.5H$ a sharp increase of Ne with increasing L could be observed. The two Rushton turbines act independently at $\Delta H > 1.65d$, with d as the impeller diameter. As the baffle length decreases, an increased mutual interaction between the two impellers could be observed for a large regime of $\Delta H/d$ values; Ne was not affected by ΔH for the unbaffled agitated systems studied.

If the impeller spacing is greater than a critical value, the total power consumed by dual Rushton turbines (RTs) is approximately equal to the sum of the power inputs of the individual impellers, which in this case are acting independently [7–10]. On the other hand, little information is available on the power consumption in systems agitated by dual and multiple pitched blade turbines (PBTs). Bates et al. found that for $\Delta H \geq 4$, a dual 6-PBT system consumes less than twice the power of a single turbine and that for $\Delta H/d < 1$ the total power input decreases with impeller spacing. Similar results are also reported in the literature for small off-bottom clearances of the lower impeller ($1/48 < H_R/D < 1/8$), in a dual 6-PBT system, showing different power numbers for the lower and the upper impeller and a total power consumption, which was smaller than twice the power consumed by the single impeller [11].

[*] Asst. Prof. Dr. J. Markopoulos (imarkopo@auth.gr), Dipl.-Ing. E. Babalona, Dipl.-Ing. E. Tsiliopoulou, Dipl.-Ing. K. Tasopoulou, Department of Chemical Engineering, Aristotle University of Thessaloniki, Univ. Box. 453, GR-54124 Thessaloniki, Greece.

1) List of symbols at the end of the paper.