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Catalysis Today 125 (2007) 48–55

CATALYSIS TODAY

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Experimental studies on hydrogenation of anthraquinone derivative in a microreactor

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Abstract

Microreactors provide a novel means for chemical synthesis in a highly controlled way to obtain better yield and selectivity. In the present work hydrogenation of ethylanthraquinone, which is the first step in the commercial production of hydrogen peroxide by Reidl–Pfleiderer process, is carried out in a microreactor packed with supported palladium catalyst. The effects of reaction temperature, hydrogen to liquid reactant flow rate ratio, reactor pressure, ethylanthraquinone concentration and fluid velocity on conversion and reactor space-time yield were studied. The performance data obtained from this microreactor study were compared with the data from conventional reactors reported in the literature. The data show that the reactor space-time yield is much higher in the microreactor than in conventional reactors.

Keywords: Microreactor; Hydrogen peroxide; Ethylanthraquinone; Supported palladium catalyst

1. Introduction

Hydrogen peroxide is being increasingly used in many green chemistries for its environmentally friendly properties. For example, traditional bleaching of pulp and paper by chlorine or sodium hypochlorite solutions produces a large amount of trihalomethanes, mainly chloroform, which are released to the atmosphere. These trihalomethanes are known to be carcinogenic in nature and our atmosphere is thus polluted by the traditional pulp and paper bleaching process. Bleaching of pulp and paper by hydrogen peroxide produces no pollutants and today about 50% of the hydrogen peroxide produced in the world is used in pulp and paper bleaching [1]. Other uses of hydrogen peroxide include treatment of water and wastewater.

Hydrogen peroxide is commercially produced by a process known as Reidl–Pfleiderer process. In this process 2-ethylanthraquinone (EAQ) in an organic solvent is first hydrogenated in the presence of a supported Pd catalyst to yield 2-ethylanthrahydroquinone ($EAQH_2$). The $EAQH_2$ is then oxidized by molecular oxygen to produce hydrogen peroxide regenerating EAQ in the process. Hydrogen peroxide is then extracted with water to obtain an aqueous solution of hydrogen peroxide. The reaction scheme is presented as Scheme 1 given by Drelinkiewicz et al. [2]. The heat of reaction for the hydrogenation of EAQ is 104 kJ/mol while that of the oxidation step is 85 kJ/mol as given by Kirk–Othmer [3].

The regenerated EAQ is again hydrogenated in the next cycle to obtain another molecule of H_2O_2.

Although starting with EAQ the main reaction is as shown above, some $EAQH_2$ on further hydrogenation is converted to tetrahydro-2 ethylanthrahydroquinone (H_4EAQH_2). When H_4EAQH_2 is oxidized it produces one molecule of H_2O_2 and one molecule of tetrahydroanthraquinone (H_4EAQ). Like EAQ, this H_4EAQ is also used again and again to produce H_2O_2. Some other side reactions involving $EAQH_2$ also occur in which the products do not yield H_2O_2 and are termed degradation products. The degradation products include ethyloxanthrone, octahydro-ethylanthrahydroquinone, ethylanthrone and ethylanthracene. The routes for formation of degradation products are as shown below in reaction Scheme 2 as given by Drelinkiewicz et al. [2].

In the present study, fresh EAQ solution is used in all the runs to avoid the degradation products.

Santacesaria et al. [4] reported that hydrogenation of anthraquinone derivatives on Pd catalyst is a fast reaction, and mass transfer resistance controls the reaction rate in conventional

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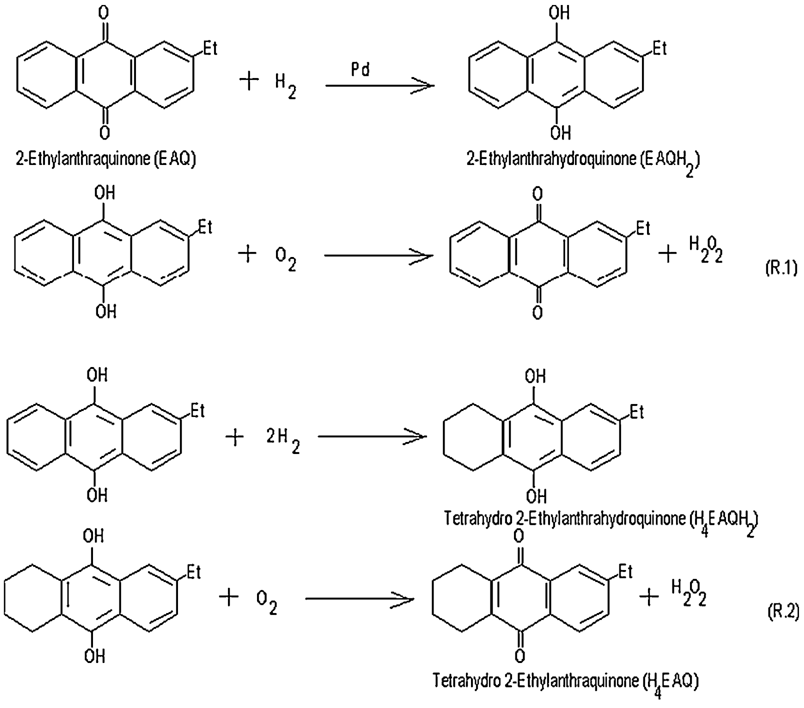
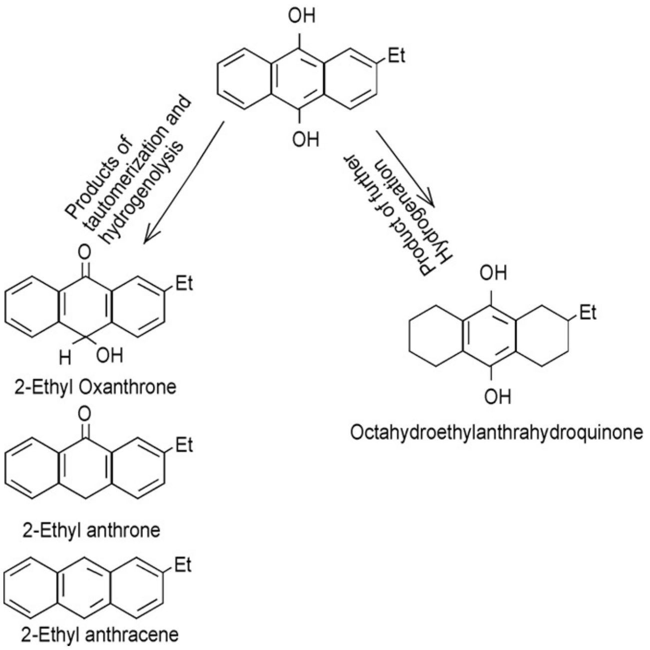
doi:10.1016/j.cattod.2007.03.055



Scheme 1. Production routes for hydrogen peroxide.

reactors. Microreactors are known to have enhanced mass transfer rates compared to conventional reactors for multiphase reactions where the development of Taylor slug flow or pulsing flow has been shown to lead to orders of magnitude improvement in the mass transfer rates across the gas–liquid interface as shown by Losey et al. [5]. Since in this hydrogenation reaction mass transfer rate controls the reaction rate in conventional reactors, the reactor space-time yield can be increased to a great extent by using a microreactor. Also, microreactors have very high surface area to volume ratio thus enabling the control of exothermic reactions such as this hydrogenation reaction very efficiently. In this study, the hydrogenation reaction of anthraquinone derivative is experimentally conducted in a microreactor in order to demonstrate that the reactor space-time yield for this reaction is superior to that in conventional reactors.



Scheme 2. Production routes of degradation products.

2. Experimental

2.1. Chemicals

The solid EAQ is dissolved in a solvent mixture containing a polar and a non-polar solvent. The non-polar solvent is 1,3,5 trimethylbenzene, and the polar solvent is trioctyl phosphate. The EAQ, 1,3,5 trimethylbenzene and trioctyl phosphate (all reagent grade), were purchased from Sigma–Aldrich. The weight ratio of the trioctylphosphate and the trimethyl benzene used in the solvent mixture was 1:2.6. The concentration of the EAQ employed in the solution was in the range 0.22–0.44 mol/l.

Hydrogen gas used in this study was obtained from a cylinder and was of ultra high purity grade (99.9999%). The catalyst used in this work is 1% Pd supported on silica and, supplied by Johnson Matthey. The properties of the catalyst are given below:

- BET surface area = 320 m^2/g.
- Pore volume = 1.6 cm^3/g.
- Average pore diameter = 20 nm.

2.2. Experimental set-up and procedure

A sketch of the experimental set-up used for the present study is shown in Fig. 1. The EAQ solution was pumped at a

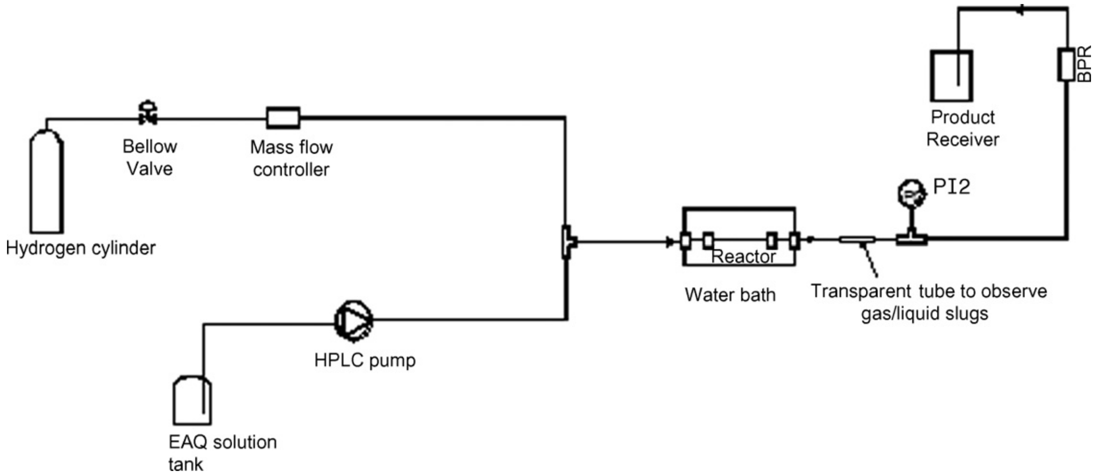


Fig. 1. Lab set-up for EAQ hydrogenation.

predetermined flow rate by the HPLC pump. Hydrogen gas flowed from a cylinder through a mass flow controller, which accurately measured and controlled the hydrogen gas flow rate. The hydrogen gas was mixed with the EAQ solution in a 1/16 in. Swagelok tee. A pneumatically actuated bellow valve was used to shut down or turn on the gas flow.

All the fittings used downstream of this mixing tee are of size 1/16 in. and the tubing used has an outer diameter of 1/16 in. (1.588 mm) and an inner diameter of 0.775 mm. The tube and all the metallic fittings are made of 316L stainless steel. After mixing the two reactants, the reaction mixture was passed through a tubular microreactor packed with the silica supported palladium catalyst. The reactor was made of SS316L stainless steel tube having an inner diameter of 775 μm. The total length of the reactor was 9.5 cm of which 7.5 cm was packed with the catalyst. Downstream of the catalyst the remaining length of the reactor was filled with inert glass beads. The catalyst particles used were 75–150 μm size range. The glass beads were spherical and were in the 63–75 μm size range. The glass beads were used to prevent any fine catalyst particles from clogging the filter placed at the end of the reactor. In each of the experiments reported here, except in the experiments of Fig. 9, 13 mg of supported catalyst containing 1% palladium and about 15 mg of glass beads were used. The reactor was connected to the set-up with the help of PEEK fittings and then immersed in a constant temperature water bath to obtain uniform temperature. The pressure at the outlet of the reactor was measured by a pressure transducer, and the associated meter as shown in the sketch (PI2) while the inlet pressure was measured by a pressure transducer installed in the HPLC pump. Downstream of the reactor, a small length of 1/16 in. ID transparent PTFE tube was installed to observe visually the gas and liquid slugs as they exit the reactor. The reactor pressure was adjusted by a back-pressure regulator (BPR). From the back pressure regulator, the product mixture was collected in a product receiver, and the un-reacted hydrogen was vented to the atmosphere. After product concentration attained steady state, the liquid product was collected for analysis. With a fresh catalyst, the reactor was run for a maximum period of 6–8 h to ensure that the catalyst activity remained close to its original activity and the concentrations of the degradation products, if any, could be neglected. It has been shown that appreciable quantities of degradation products are formed when the reaction solution is recycled numerous times [2]. After this 6–8 h run, the reactor and the catalyst are discarded and new run is started with fresh catalyst in a new reactor.

2.3. Analytical procedure

About 10 ml of the collected product was oxidized completely by injecting oxygen gas at 138–207 kPa(g) while stirring the solution with a magnetic stirrer in an oxidation glass tube for about 15 min. The color of the solution indicates whether complete oxidization has occurred or not. After the complete oxidation, 5.0 ml of the oxidized solution was extracted twice with 15% (w/w) sulfuric acid solution using 15 ml of the sulfuric acid solution each time. The hydrogen peroxide amount in the aqueous extract was then determined by titration with standard potassium permanganate solution. From this amount, the concentration of total hydrogenated anthraquinones was determined.

3. Results and discussion

3.1. Effect of reaction temperature on conversion and space-time yield (STY)

Fig. 2 shows the effect of reaction temperature on the conversion of the limiting reactant and reactor space-time yield. In this study the STY was calculated as follows: from the titration results, the moles of $EAQH_2$ per liter of the product were calculated. The change of the liquid flow rate from the inlet of the reactor to the outlet was assumed negligible. The mol/l of $EAQH_2$ formed was multiplied by the liquid flow rate (l/h) to calculate the actual moles of $EAQH_2$ formed in an hour, which is equal to moles of H_2O_2 formed in an hour. The gram of H_2O_2 produced per hour was divided by the amount of palladium in the catalyst bed to obtain the STY. In all the runs the EAQ was the limiting reactant and hydrogen was in excess. The experimental runs, the data of which are shown in Fig. 2

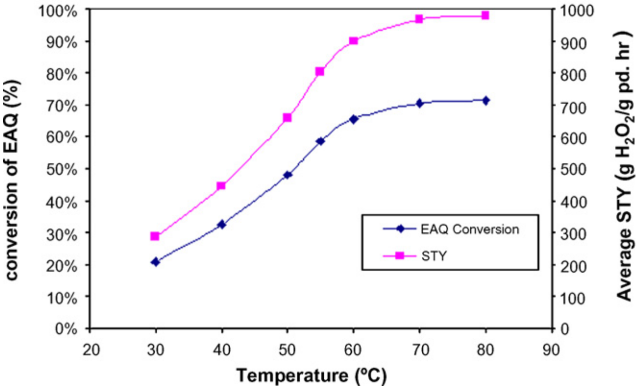


Fig. 2. Effect of reaction temperature on conversion and reactor STY (689 kPa(g) reactor outlet pressure, 0.22 M EAQ solution flow = 0.40 ml/min, H_2 feed flow = 4.0 sccm).

were conducted at 689 kPa(g) reactor outlet pressure. As expected, the conversion and reactor STY increase with increase in temperature. At low temperature levels (30–50 °C) the temperature has a stronger effect on conversion and STY than at high temperature levels. It appears that even at the low temperature levels, mass transfer has some influence on the reaction rate because for a kinetically controlled reaction one would expect higher increase in conversion with the increase in temperature than that obtained in these runs. This has been confirmed by conducting experiments on the effect of flow velocity on STY (see Section 3.6 below). At high temperature levels, the increase in STY, and conversion with increase in temperature becomes smaller suggesting that influence of mass transfer rate on the reaction rate becomes dominant. The reason is that at high temperature levels the intrinsic kinetics of the reaction becomes faster resulting in the mass transfer resistance becoming more dominant.

3.2. Effect of reaction pressure on conversion and STY

The effect of hydrogen pressure on conversion, and reactor STY is shown in Fig. 3. Although the data show that the conversion and STY increase slightly with increase in pressure, the increase is very marginal. Since under the operating conditions given in Fig. 3 the concentration of hydrogen in the liquid can be assumed to be proportional to the pressure, the data show that hydrogen concentration in the liquid has negligible effect on conversion or STY. For a differential reactor, the plot of log(STY) versus log(pressure) would give the dependency of reaction rate on hydrogen concentration. The maximum conversion for the data shown in Fig. 3 is about 50% for which the differential reactor analysis is not acceptable. Nevertheless, a plot of log(STY) versus log(outlet hydrogen pressure) from the present data should give an approximate value of the dependence of reaction rate on the hydrogen concentration. Fig. 4 shows the plot of log(STY) versus log(outlet hydrogen pressure) for the data shown in Fig. 3. From this plot it appears that the dependency of rate on hydrogen concentration is almost zero order. Santacesaria et al. [4,6,7] conducted the EAQ hydrogenation reaction in conventional reactors, and also found the reaction to be zero order in hydrogen concentration.

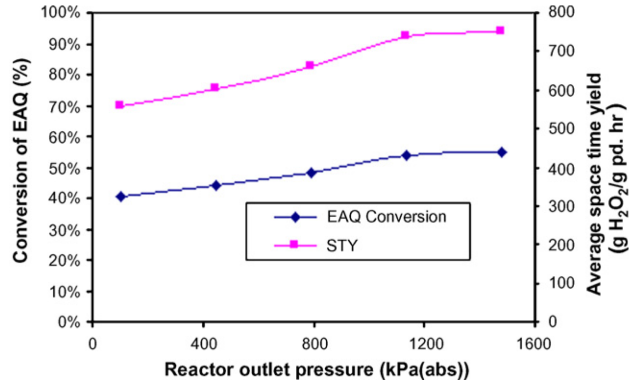


Fig. 3. Effect of reactor pressure on conversion and STY (0.22 M EAQ solution flow = 0.40 ml/min, H_2 feed flow = 4.0 sccm, reactor temperature = 50 °C).

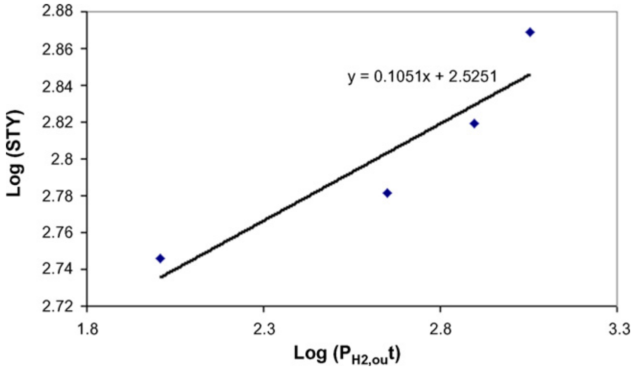


Fig. 4. Plot of log(STY) vs. log($P_{H_2,out}$).

3.3. Effect of EAQ concentration

Experiments were conducted to study the effect of EAQ concentration on the STY and EAQ conversion, and the data are shown in Fig. 5. With increase in EAQ concentration, the conversion is nearly constant while the STY increases. The increase in STY with increase in EAQ concentration indicates that the reaction rate is dependent on the EAQ concentration. Fig. 6 shows a plot of log(STY) versus log(C_{EAQ}) for the data shown in Fig. 5. From the slope of this plot, it appears that the

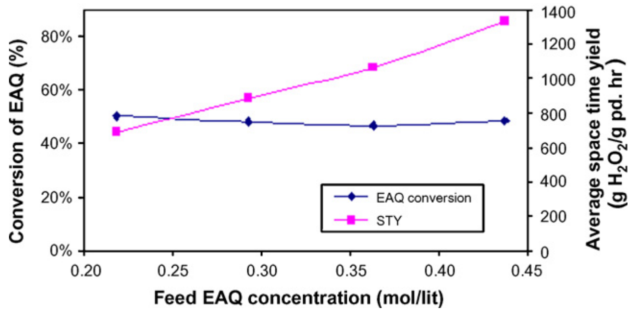


Fig. 5. Effect of EAQ concentration on conversion and STY (liquid feed flow = 0.40 ml/min, reaction temperature = 50 °C, reactor outlet pressure = 689 kPa(g)).

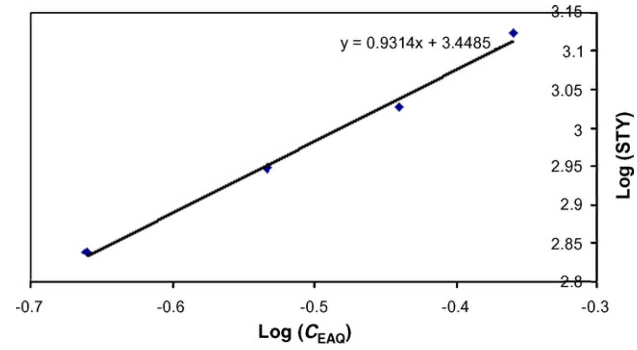


Fig. 6. Plot of log(STY) vs. log(C_{EAQ}).

reaction rate has approximately a first-order dependence on the EAQ concentration. Santacesaria et al. [4] also found that the rate reaction rate has a first-order dependency on the EAQ concentration in a conventional reactor.

3.4. *Effect of hydrogen gas to liquid flow ratio*

We found that with increase in hydrogen gas to liquid feed flow ratio, the conversion of the limiting reactant and the reactor STY increase. Fig. 7 shows the data obtained in the present study with EAQ as the limiting reactant. As observed from Fig. 7, with increase in hydrogen flow rate the conversion and STY increase slightly. The increase in conversion with H_2 flow rate is possibly due to better dispersion of hydrogen into the liquid and high mass transfer rate at high linear velocities.

3.5. *Effect of residence time*

The effect of residence time on the reactor STY, and EAQ conversion is shown in Fig. 8. The experiments were conducted at 50 °C with EAQ as the limiting reactant. Both the gas and liquid flow rates were varied, but the liquid to gas flow ratio was kept constant in all the runs with a fixed amount of catalyst of 13 mg in the reactor. Fig. 8 shows that under the experimental conditions, the residence time has a negligible effect on the conversion. For a kinetically controlled reaction, the conversion is expected to increase with increase in residence time. On the other hand for a mass transfer controlled reaction the mass transfer rate increases with increase in velocity (i.e., with decrease in residence time). The net effect could lead to negligible effect on conversion and an increase in STY with increase in flow velocity. So, this observation again suggests that mass transfer is still playing a role in the reaction rate even in the microreactor. Since, conversion is nearly the same at different flow velocities, the STY is found to decrease with increase in residence time.

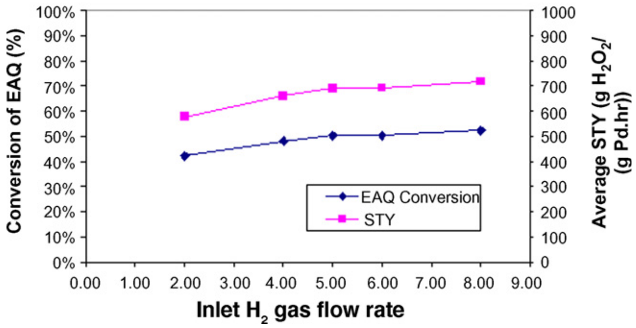


Fig. 7. Effect of hydrogen flow rate on EAQ conversion and reactor STY (0.22 M EAQ solution flow = 0.40 ml/min, reactor temperature = 50 °C, reactor outlet pressure = 689 kPa(g)).

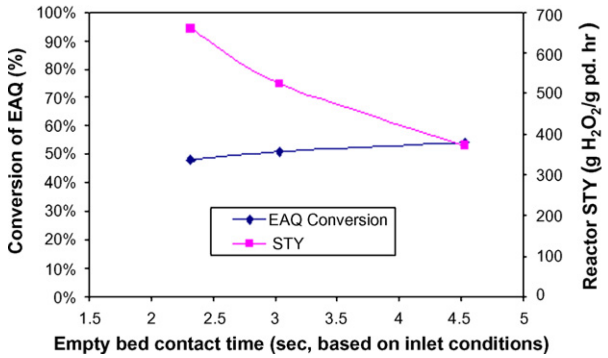


Fig. 8. Effect of residence time on conversion and reactor STY (0.22 M EAQ solution, reactor temperature = 50 °C, reactor outlet pressure = 689 kPa(g)).

3.6. *Effect of external mass transfer resistance*

The influence of external mass transfer rate on the reaction can be neglected if the overall external mass transfer rate given by, $K_L a C_A$, is 10 times or more than the observed reaction rate as given by Mills and Chaudhari [8], where $K_L a$ is the overall mass transfer coeffiecient times interfacial area per unit liquid volume, which is defined by the following equation and C_A is the concentration of the diffusing reactant in the liquid.

$$\frac{1}{K_L a} = \frac{1}{k_l a_i} + \frac{1}{k_c a_s} \qquad (1)$$

where $1/k_l a_i$ is the mass transfer resistance for gas absorption into liquid and $1/k_c a_s$ is the mass transfer resistance for liquid to catalyst surface.

For the system used here, the mass transfer coefficient cannot be directly calculated because all the known correlations of mass transfer coefficients, such as the ones for trickle or bubble flow in a packed bed given by Satterfield [9] or Taylor flow in an empty tube as given by Bercic and Pintar [10], cannot be applied to Taylor flow in a packed bed microreactor. However, Losey et al. [5] determined the value of overall mass transfer coefficient in a packed bed microreactor and found that the overall mass transfer coeffiecient in a packed bed microreactor is orders of magnitude higher than the mass transfer coefficient generally found in a conventional reactor. The values of $K_L a$ found in a microreactor by Losey et al. was in the range 5–15 s^{-1} whereas the values in a laboratory trickle bed reactor is 0.01–0.08 s^{-1}. Work done in our lab by Tadepalli et al. [11] for a different hydrogenation reaction also showed that the mass transfer coefficient, $K_L a$ value in a packed bed microreactor is similar to that obtained by Losey et al. in the range of parameters studied in this work. The flow velocity used in the prersent work is higher than than that used by Losey et al. and hence the mass transfer coefficient in our reactor would even be higher than that obtained by Losey et al. Also, in the microreactor periodic drying of the catalyst bed might happen. In a dried catalyst bed, mass transfer rate of hydrogen would be higher than the mass transfer through liquid because mass transfer would take place directly from gas to catalyst surface. The adsorbed hydrogen would react with dissolved ethylanthraquinone when the liquid comes into contact with the catalyst.

So, the mass transfer rate in a microreactor is much higher compared to a conventionsl packed bed reactor. The overall superficial velocity in the reactor was varied to see the effect of external mass transfer rate on the observed reaction rate. If the mass transfer rate is very high (more than 10 times) compared to the intrinsic kinetic rate, then the superficial velocity will not influence the STY, and the STY will be fully controlled by intrinsic kinetics. If the mass transfer rate has an influence on the STY, then with increase in velocity while keeping all other parameters constant, the STY will increase. In our study we had kept the amount of catalyst in the bed proportional to the overall linear velocity in order to keep the contact time constant.

Fig. 9 shows the experimental data obtained at two different temperatures of 40 and 70 °C. The overall velocity was calculated based on the liquid flow and the calculated hydrogen flow under the reactor conditions. We have calculated the flow velocities by assuming 40% external void in the catalyst bed and the velocities shown in the figure are the actual average velocities calculated based on the inlet and outlet flow conditions. The runs were conducted with 0.22 mol/l of inlet EAQ solution and at 689 kPa(g) reactor outlet pressure. The empty bed contact time under inlet conditions was approximately 2.0 s in these runs while the empty bed contact times based on outlet velocities were in the range 1.85–1.88 s depending on the conditions. As seen in Fig. 9, at both these temperature levels with increase in velocity, the STY increases indicating that the mass transfer rate has influence on the STY. However, as shown in the figure, the temperature also has a strong influence on the reaction rate under the conversions studied. At conversions below 40% the reaction rate increased by approximately two and half times when the temperature is increased from 40 to 70 °C. Although, to obtain precise kinetics information on the dependency of reaction rate on temperature the conversion must be very low, nevertheless the data indicate that the temperature has much higher influence on the reaction rate than the change in mass transfer rate. Hence, it appears that under our operating conditions, both the intrinsic kinetics and the mass transfer resistance are influencing the reaction rate. However, as seen from Fig. 9, at low conversions, the slope of the plot of STY versus flow rate is higher at 70 °C than at 40 °C, which indicates that influence of mass transfer resistance on the reaction rate is higher at higher temperature. Intrinsic kinetic rate increases much more rapidly with increase in temperature than the mass transfer rate, thus mass transfer resistance becomes more predominant at higher temperature.

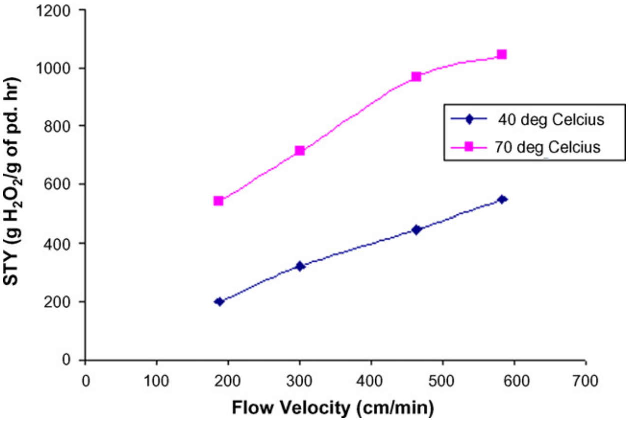


Fig. 9. Effect of flow velocity on STY (0.22 M EAQ solution, H_2/EAQ inlet molar flow ratio = 2.0, reactor outlet pressure = 689 kPa(g)).

As mentioned previously in this work, we found that the reaction rate is almost independent of hydrogen pressure. With increase in pressure, the saturation concentration of hydrogen in the liquid should increase which in turn should lead to an increase in the liquid-solid mass transfer rate at the same velocity. However, with increase in pressure the actual volumetric flow rate of the gas decreases with same mass flow rate resulting in decrease in overall velocity through the bed. Thus, even if the hydrogen mass transfer rate has an influence on the reaction rate in the microreactor, the increase in higher hydrogen mass transfer rate from liquid to catalyst due to higher concentration in the liquid could be offset by the decrease in mass transfer rate due to decrease in velocity, resulting in the reaction rate to be relatively independent of hydrogen pressure. With the increase in velocity the mass transfer rate of EAQ from liquid to solid also increases which might have also influenced the rate of reaction.

3.7. Effect of internal mass transfer resistance

To determine the effect of internal pore diffusion resistance on the reaction rate the Thiele modulus was calculated by assuming spherical catalyst particles for some experimentally obtained high reaction rates, i.e., 800 g H_2O_2/g of Pd h (0.00392 mol H_2O_2/g of catalyst min) obtained at 55 °C. The Thiele modulus, as given by Smith [12] for spherical catalyst particles is defined as:

$$M_T = \left(\frac{r_s}{3}\right)\left(\frac{k_1\rho_p}{D_e}\right)^{1/2}$$

where M_T is the Thiele modulus, k_1 the first-order rate constant, ρ_p the density of the catalyst pellet and D_e is the effective diffusivity of hydrogen in the pellets. As shown previously in this work and also by Santacesaria et al. [4], the rate of reaction is independent of hydrogen concentration. The value of k_1 can be estimated by the average reaction rate and the average concentration of EAQ in the solution. The diffusivity of hydrogen in the solution is estimated under the experimental conditions (from Wilke–Chang equation) [13] to be 9.55×10^{-5} cm^2/s. The effective diffusivity is defined as

$$D_e = D_m\frac{\delta\theta}{\tau}$$

where D_m is the diffusivity of hydrogen in the solution and θ, δ and τ are the catalyst porosity, constriction factor and tortuosity factor, respectively. For these parameters, the ranges are: 0.4–0.6 for θ, 0.7–0.8 for δ and 2–8 for τ. Taking the average values of these parameters, the Thiele modulus was calculated to be 0.35, which indicates that the pore diffusion resistance can be neglected.

We have also made calculations to see if the pore diffusion of EAQ has any effect on the rate of reaction. The diffusivity of EAQ in the solution was calculated using the correlation of King et al. [14]. The diffusivity in the solution comes to be 1.6×10^{-5} cm^2/s which is much less than hydrogen diffusivity. The effective diffusivity is calculated to be 1.2×10^{-6} cm^2/s. The Thiele modulus was calculated to be 0.88 which would give a value of effectiveness factor of 0.8 implying that very little pore diffusion resistance exists. However, the pore diffusion resistance would be significantly lower than that of a commercial packed bed reactor where larger catalyst particles are used.

4. Microreactor STY versus STY of conventional reactors

We have collected data from literature describing the reactor STY of the EAQ hydrogenation on supported palladium catalysts using conventional reactors. Drelinkiewicz et al. [2] conducted the reaction in a tubular reactor of 5.4 cm in diameter charged with 25 cm^3 (19 g) of supported palladium catalyst with 2% palladium. The feed solution had an EAQ concentration of 60 g/l (0.254 mol/l), reactor pressure of 517 kPa(g) and reaction temperature of 50 °C. After 1 h of circulation, they obtained a reactor productivity of 0.282 mol of H_2O_2/h. Under these conditions the reactor STY is calculated to be 25 g H_2O_2/g of Pd h. The results obtained in the present study show that under similar conditions the STY is about 800 g H_2O_2/g of Pd h. Thus, the microreactor STY appears to be more than 30 times that of a conventional tubular reactor. Edvinsson et al. [15] reported a reactor STY of 16 g H_2O_2/g of Pd h for a commercial slurry reactor at 55 °C and 2 bar (200 kPa) pressure. Thus, the STY obtained in the microreactor in the present study appears to be more than 50 times that of the slurry reactors as obtained by Edvinsson et al. Edvinsson et al. also conducted experiments in a packed bed reactor and obtained a STY of 105 g H_2O_2/g of Pd h. As mentioned in this work, pressure has negligible effect on the STY, so the STY in the microreactor is approximately eight times more than that obtained by Edvinsson et al. in their packed bed reactor. Although, the catalyst used in the present study was not the same as that of Drelinkiewicz et al. or Edvinsson et al., an order of magnitude increase of STY in the microreactor might be due to the increase of mass transfer rate in the microreactor as discussed above.

In macroreactors, the reaction is controlled by the mass transfer rates as demonstrated by Santacesaria et al. [4]. In a conventional trickle bed reactor the overall mass transfer coefficient could be 0.01–0.08 s^{-1} as reported by Al Dahhan et al. [16], whereas Losey et al. [5] measured mass transfer coefficients in a packed bed microreactor which were found to be more than two orders of magnitude higher than these values. The higher STY obtained in the microreactor could hence be explained by the high mass transfer coefficients in microreactors.

5. Conclusions

The results obtained in this study and the data collected from the literature suggest that in the multiphase solid catalyzed gas–liquid hydrogenation of ethylanthraquinone, the mass transfer rate controls the rate of reaction in a conventional reactor. In a microreactor both mass transfer and intrinsic kinetics probably influence the reaction rate at lower temperature levels, and the reaction rate is more influenced by the mass transfer at higher temperature levels. Thus, for this reaction much higher reaction rate, in orders of magnitude, could be obtained in a microreactor than in conventional reactor while at the same time preventing any temperature runaway due to the enhanced heat transfer in the microreactor. The data obtained in the present study show that the microreactor technology can be used very successfully to produce very high reactor space-time yield in comparison to conventional reactor for a mass transfer controlled reaction such as hydrogenation of anthraquinone.

Acknowledgement

The authors thank Dr. Robert J. McNair of Johnson Matthey Catalysts for supplying the catalysts.

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