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APPENDIX A

DERIVATION OF EQUATION 4-24

This appendix uses the same notation as Chapter 4. For a sphere with symmetrical boundary conditions, the diffusion-reaction equations for an elementary $A + B \rightarrow C$ reaction are:

$$\begin{aligned} \frac{d^2 a}{d\rho^2} + \frac{2}{\rho} \frac{da}{d\rho} - \frac{k_r r_p^2 C_{B,bulk}}{D_A} a \cdot b &= 0 \\ \frac{d^2 b}{d\rho^2} + \frac{2}{\rho} \frac{db}{d\rho} - \frac{k_r r_p^2 C_{A,bulk}}{D_B} a \cdot b &= 0 \\ \text{If } \gamma &= \frac{\phi_A^2}{\phi_B^2} \\ \frac{d^2 a}{d\rho^2} + \frac{2}{\rho} \frac{da}{d\rho} - \gamma \phi_B^2 a \cdot b &= 0 \\ \frac{d^2 b}{d\rho^2} + \frac{2}{\rho} \frac{db}{d\rho} - \phi_B^2 a \cdot b &= 0 \\ \therefore \frac{d^2 a}{d\rho^2} + \frac{2}{\rho} \frac{da}{d\rho} &= \gamma \left(\frac{d^2 b}{d\rho^2} + \frac{2}{\rho} \frac{db}{d\rho} \right) \end{aligned} \quad (\text{A.1})$$

$$\text{And } \frac{d^2}{d\rho^2} (\gamma \cdot b - a) = \frac{2}{\rho} \frac{d}{d\rho} (a - \gamma \cdot b)$$

$$\begin{aligned} \text{Let } \Theta &= a - \gamma \cdot b \\ \text{Then: } \frac{d^2 \Theta}{d\rho^2} + \frac{2}{\rho} \frac{d\Theta}{d\rho} &= 0 \end{aligned}$$

(A.2)

Let $y = \Theta\rho$

It can be shown that: $\frac{d^2y}{d\rho} = 0$

$$\therefore y(\rho) = k_1\rho + k_2$$

$$\Theta(\rho) = k_1 + \frac{k_2}{\rho}$$

(A.3)

Since Θ is finite at $\rho = 0$:

$$\Theta(\rho) = k_1$$

At $\rho = 1$, $a = 1$ and $b = 1$:

$$\therefore k_1 = 1 - \gamma$$

$$\therefore a - \gamma b = 1 - \gamma$$

$$\text{And } b = \frac{a - 1 + \gamma}{\gamma}$$

(A.4)

The same relationship can be derived for an infinite slab.

APPENDIX B

HYDROGENATION OF LINEAR OCTENE UNDER GAS-LIMITED CONDITIONS

Many trickle-bed reactor studies under gas-limited conditions are reported in the literature. In terms of wetting efficiency, many of these are based on the additive procedure as shown in Chapter 2. In Chapter 4, it is shown that, especially for eggshell catalysts, gas-limited reactions may be subject to limitations of the liquid reagent even when a reaction is generally considered as gas-limited. A preliminary study on the hydrogenation of linear octenes under gas-limited conditions is reported here.

Reaction system and operating conditions. The experimental procedure followed is exactly the same as that described in Chapter 5, except for the reaction system and operating conditions. In order to operate under gas-limited conditions, reactor operating pressures were kept low. For all liquid flow rates, a gas feed mixture of 85% N_2 and 15% H_2 was fed to the reactor, operating at a pressure of 6 bar and a temperature of 60 °C. At these conditions, the liquid reagent can still be regarded as non-volatile. Relevant properties other than those reported in Chapter 5 are listed in Table B.1. The total gas flow rate was 10 nL/min for all liquid flow rates, corresponding to a superficial gas velocity $v_{SG} = 17 \text{ mm/s}$. The liquid feed contained 8% (v/v) linear octenes and no isooctenes. Under these conditions, $\gamma \approx 40$.

The reactor was packed with 110 g catalyst, corresponding to a bed height of approximately 9 cm, between two layers of inert support. Startup and operation were exactly the same as in the previous chapter. Experiments were conducted for $v_{SL} = 1.9, 2.7, 3.8$ and 4.7 mm/s . Only two repeat runs were performed.

Table B.1: Hydrogen property estimations

Property	Estimated value	Estimation method
Saturated H ₂ concentration in solvent	3.25 mol/m ³	Florusse et al. (2003)
Hydrogen molecular diffusivity in solvent	4.5 × 10 ⁻⁹ m ² /s	Wilke-Chang

Experimental results. Overall reaction rates are reported in Figure B.1. Since all conversions were less than 10%, the reactor was regarded as a differential reactor. Reaction rates are unexpectedly high, and upflow reaction rates can not be explained with the liquid-solid mass transfer results from Chapter 5, and especially not if considered that overall hydrogen-to-catalyst surface mass transfer is also affected by gas-liquid mass transfer. Therefore, it was not possible to use upflow rate data as a basis for the treatment of trickle-flow data, as was done in Chapter 5, and only the general trends will be discussed. The trends reported in Figure B.1 also seem counterintuitive to some extent.

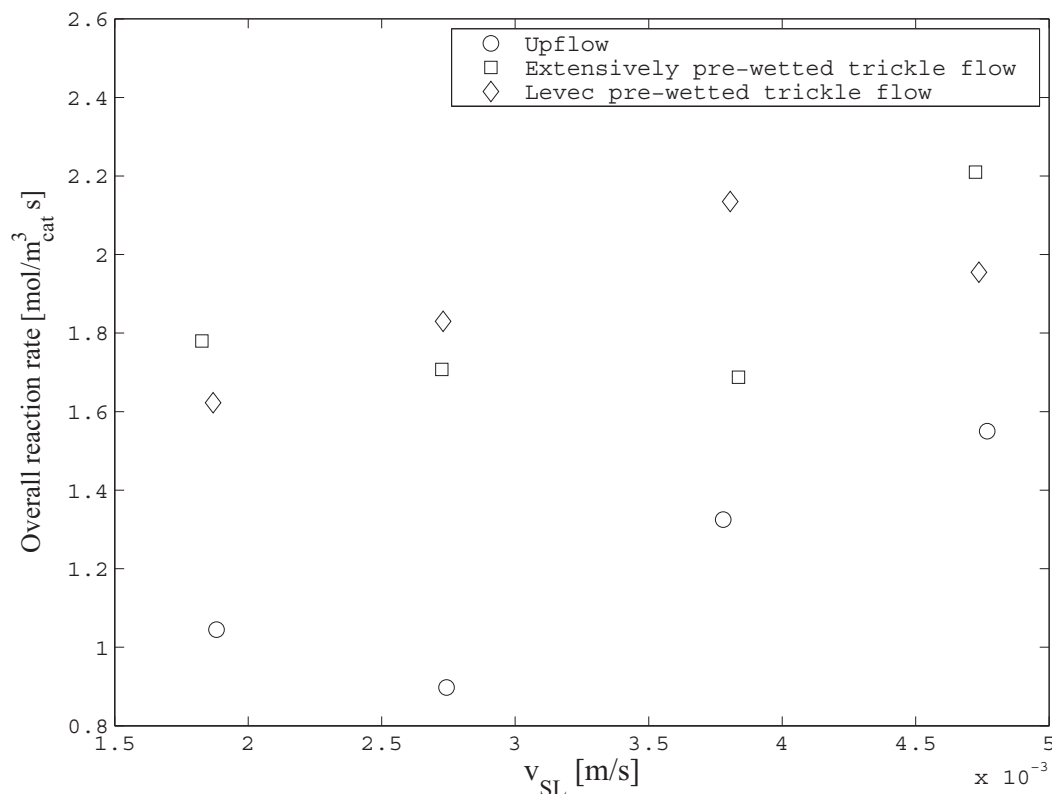


Figure B.1: Overall reaction rates of linear octene hydrogenation under gas-limited conditions.

As expected, downflow outperforms upflow, but any other enhancement effects due to partial wetting seem to be low, or even non-existent. The reaction rates for Levec and extensively pre-wetted flow are more or less the same and reaction rate does not increase (notably) with a decrease in liquid flow rate as is reported for so many of the reactor

studies discussed in the literature review¹. A possible explanation might be the combination of liquid-limited effects in eggshell particles and the distribution of particle wetting due to flow morphology. Figure B.2 shows a rough approximation of overall trickle-bed efficiency (based on particle wetting distribution) for the best and least wetted cases in the colorimetric study as a function of ϕ_A when $\gamma = 40$, using the unified model for eggshell particles. Biot numbers are roughly based on liquid-solid mass transfer results from Chapter 5: experimental k_{LS} -values for octene at the relevant flow rate and pre-wetting conditions were adapted for hydrogen and then halved to allow for gas-liquid mass transfer resistance. It is clear from the figure that it is definitely possible that Levec pre-wetted beds do not outperform extensively pre-wetted beds, or even exhibit lower overall rates of reaction.

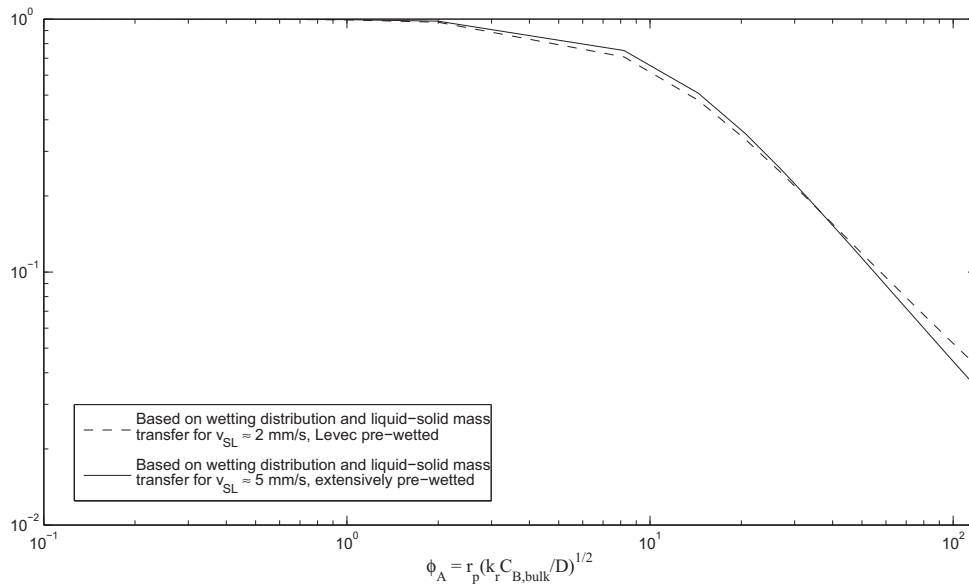


Figure B.2: Approximations of overall efficiency at boundaries of the colorimetric and liquid-limited reactor investigations for the current system, as a function of the Thiele modulus of the gaseous reagent.

¹Extensively pre-wetted results agree to some extent with the study of Mata and Smith (1981)