

Effect of Catalyst Size on Hydrodynamics and Mass Transfer during Hydrogenation of Styrene in a Trickle Bed Reactor

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Highlights

- Trickle to bubble transition maps are dependent on temperature, pressure and catalyst size.
- Higher temperature shifts transition to higher liquid velocities, pressure shifts to lower.
- Transition map trend with catalyst size is dependent on temperature.
- Hydrogenation rates are fastest in conditions offering highest gas-liquid mass transfer.

1. Introduction

While the trickle-pulsing transition for trickle bed reactors (TBR) has been extensively studied, the literature focusing on the trickle-bubble transition is rather limited [1, 2]. Given the propensity for the prior work to use air and water, we designed this effort to include (1) characterization of that flow regime transition and (2) measurement of mass transfer-limited reaction rates with a catalyst and fluids that more closely mimic conditions found in industrial trickle bed hydrogenators. We evaluated the effect of temperature, pressure, and catalyst size for a model system, the styrene hydrogenation over a supported palladium (Pd) catalyst.

2. Methods

The batch recycle TBR is a 2" jacketed stainless steel pipe with a gas-liquid distributor on top and a 10 L tank at the bottom that serves as a liquid reservoir. The liquid from the tank and the gases enter the reactor concurrently from the top through the distributor. A pump continuously recirculates the liquid, while the gases flow once through. After the reactor, the gas flows through a condenser before being vented. The reactor is heated using a water bath, the liquid tank is heated using electrical heating tapes, and a back pressure regulator is used to pressurize the system.

The Pd catalyst is supported on 1/8" and 1/16" spheres. We developed transition maps for the two catalyst sizes using a toluene/N₂ system at temperatures ranging from 20 to 65°C and pressures of 100 and 195 psig. Pressure transducers monitor the transition from the trickle to the bubble flow regime, which is marked by a sudden significant increase in the differential pressure across the bed. We selected two liquid velocities, one in each regime, for the hydrogenation experiments, using a 3.6% (by weight) styrene solution balanced with toluene and 30% (by moles) H₂ balanced with N₂. We used gas chromatography to analyze samples collected intermittently during the hydrogenation experiments.

3. Results and Discussion

Figure 1 shows the locus of the transition from the trickle to the bubble regime as a function of the superficial gas and liquid velocities, reactor temperature, pressure, and catalyst size. As shown in the figure, at the same superficial gas velocity, a higher temperature causes the transition to shift to a higher liquid velocity. However, the trickle to bubble transition shifts to lower liquid velocities with increasing pressure. Effect of catalyst size on the transition maps differs depending on reactor temperature. At 65°C, the smaller 1/16" catalyst transitioned at a higher liquid velocity than the 1/8" catalyst. However, at 20°C, the same catalyst shifted the liquid transition velocity to a lower value compared to the 1/8" catalyst.

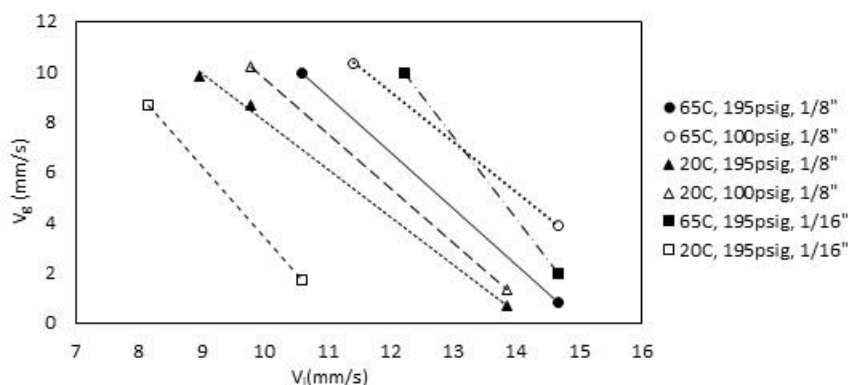


Figure 1. Transition map as a function of superficial gas and liquid velocities, temperature, pressure, and catalyst size.

Figure 2 shows the styrene conversion over time for the two different catalyst sizes under both the trickle and bubble flow regimes. As seen in the figure, under the same flow regime, the smaller catalyst size exhibits faster hydrogenation rates. Also, for both catalyst sizes, the hydrogenation rates in the bubble regime are significantly higher than those observed in the trickle regime. This hydrogenation reaction system exhibits a strong gas-liquid mass transfer dependence, which is reflected in the trends observed in the hydrogenation rates. The bubble regime is the high interaction regime, where enhanced gas-liquid mass transfer can be achieved. The smaller catalyst size also enhances mass transfer. Consequently, the smaller catalyst size in the bubble regime gives the fastest hydrogenation rate.

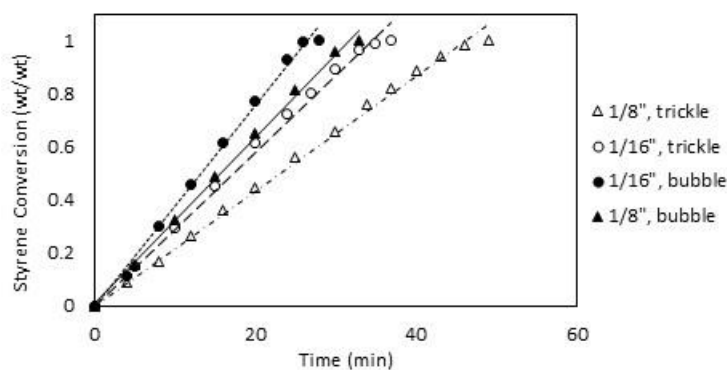


Figure 2. Styrene conversion over time with different catalyst sizes and under different flow regimes.

4. Conclusions

We investigated the effects of temperature, pressure, and catalyst size on the trickle to bubble flow regime transition. While increasing temperature shifted the transition to higher liquid velocities, an opposite trend occurred with increasing pressure. The effect of change in catalyst size on the transition maps is found to be dependent on temperature. One hypothesis to explain this opposing trend is that the combination of higher surface tension at lower temperature and the smaller catalyst size exacerbate bubble coalescence, causing transition to occur at lower liquid velocities. Understanding the location of the trickle to bubble transition becomes important in the case of reactive systems controlled by mass transfer effects. This is corroborated using styrene hydrogenation as an example, where operation in the bubble regime and a smaller catalyst size enhanced the gas-liquid mass transfer, resulting in significantly enhanced rates.

References

- [1] W. J. A. Wammes, S. J. Mechielsen, K. R. Westerterp, Chem. Eng. Sci. 45 (1990) 3149-3158.
- [2] G. S. Honda, J. H. Pazmino, E. Lehmann, D. A. Hickman, A. Varma, Chem. Eng. J. 285 (2016) 402-408.

Keywords

Trickle bed reactor; transition map; temperature and pressure effect; hydrogenation.