A NOVEL PARAMETRIC ESTIMATION METHOD FOR REACTOR OPTIMIZATION IN PLUG FLOW REACTORS

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Introduction

- Parametric optimization is a feasible route towards efficient and cost-effective operation of reactors.
- Here we report, an easy, fast and cost-effective approach through direct utilization of experimental data to model and simulate the direct conversion of CO2 and H2 to methanol.
- Monolithic reactors, showing lesser mass transfer resistance than the conventional packed bed reactors, were chosen, specifically in this study.
- This novel technique allows for translation of lab-scale experimentally acquired data into kinetic, equilibrium and adsorption parameters for the methanol synthesis reaction.

Method

- To obtain the kinetic factors, driving force factors and adsorption factors, the parametric estimation was carried out using a modified version of Nelder-Mead Simplex Algorithm in MATLAB.
- Experimental data (input for parametric fitting) was obtained using a home-made Cu/ZnO/Al2O3 catalyst on cordierite honeycomb monoliths.
- Fitted parameters were used in COMSOL Multiphysics 5.6 to simulate a monolithic reactor for CO2 hydrogenation to methanol.
- Transport of diluted species, free and porous media flow module, and chemistry module were adopted to study the effect of kinetics, external and internal mass transfer.

Results

- Direct hydrogenation of CO2 to methanol is a kinetically slow reaction and internal diffusion is not a limiting factor within the temperature window of interest (150-350°C).
- Some of the prominent calculated parameters are as under:

<table>
<thead>
<tr>
<th>Variable</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dg (Diffusion coefficient)</td>
<td>0.59 cm²/s</td>
</tr>
<tr>
<td>( \rho_m ) (Mixture density)</td>
<td>0.597 kg/m³</td>
</tr>
<tr>
<td>( \Delta H_{\text{f,chem}} ) (Mixture enthalpy)</td>
<td>110.34 kJ/kg</td>
</tr>
<tr>
<td>( L_e ) (Entrance length)</td>
<td>0.2 mm</td>
</tr>
<tr>
<td>Re (Reynolds no.)</td>
<td>3.95</td>
</tr>
<tr>
<td>Sc (Schmidt no.)</td>
<td>0.298</td>
</tr>
<tr>
<td>( \delta ) (Diffusion layer coefficient)</td>
<td>0.8</td>
</tr>
<tr>
<td>( \beta_{\text{eff}} ) (External mass transfer (coefficient))</td>
<td>36.68 mm/s</td>
</tr>
</tbody>
</table>

- Figure 4 (a) depicts the change in surface area of monolithic reactor based on the channel shape while Figure 4 (b) shows that the triangular channel shows the highest methanol yield due to relatively high surface area.

Conclusion

- Monolithic reactors offer lower external and internal mass transport resistance in comparison to packed bed reactors.
- Parametric estimation with the combination of simulated models offer a fast and cost effective route to test thousand of reactor parameters in limited time.
- It was found that triangular channel shape with fixed catalyst loading is the optimum as triangular shape exhibits the highest surface area for a given fixed volume among the different shapes tested.
- Similarly, a channel hydraulic diameter of 0.4 mm results in the highest methanol yield without incurring high pressure drop. Moreover, a channel length of 5 cm is ideal for a given space velocity, catalyst loading at 300°C.
- Furthermore, reducing the space velocity favors methanol production, provides the reaction gases higher residence time within the reactor.
- These estimated results can then be used as a reference point for experimental validation.

Reactions

Three reactions are involved in the synthesis of methanol via CO2 hydrogenation [13]:

1. \( \text{CO} + 2\text{H}_2 \rightleftharpoons \text{CH}_3\text{OH} (\Delta H_{\text{f,chem}} = -90.77 \text{ kJ/mol}) \)
2. \( \text{CO}_2 + \text{H}_2 \rightleftharpoons \text{CO} + \text{H}_2\text{O} (\Delta H_{\text{f,chem}} = 41.21 \text{ kJ/mol}) \)
3. \( \text{CO} + \text{H}_2 \rightleftharpoons \text{CH}_3\text{OH} + \text{H}_2 (\Delta H_{\text{f,chem}} = 48.18 \text{ kJ/mol}) \)

The rate equations for these three reactions listed above can be generalized in the form below:

\[ \text{Rate} = k \cdot \text{Ad} \cdot (\text{Kinetic factor}) \cdot (\text{Driving force expression}) \]

\( \text{Ad} \) = Adsorption term

References