# Carbon dioxide hydrogenation to methanol in a tubular packed-bed chemical reactor: an unsteady particle-resolved CFD simulation in 3D

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***Highlights***

- Turbulence can affect catalytic reaction rate even in surface reaction limited conditions
- Unsteady numerical solutions are advantageous in presence of non-linear surface reaction kinetics
- Impact of a chemical reaction on fluid flow path through packed bed is significant

## 1. Introduction

Particle Resolved Computational Fluid Dynamics (PR-CFD) modelling using Direct Numerical Simulation (DNS) deepens the understanding of chemical reaction dynamics in low packed bed reactors of low ratio of reactor tube to particle diameter (d/D) This is possible by going beyond the assumption of steady state reaction into a more realistic representation of turbulent, unsteady fields of partial pressures and temperature in the inter-particle space of packed bed reactors [1].

Even in reactions limited by surface reaction kinetics, the intensity of turbulence can significantly impact the dynamics of local conditions at catalyst active sites. This can cause a divergence between results obtained from experiments and simulations, where a steady state environment was assumed. This is particularly true when it comes to modelling chemical kinetics using pseudo-empirical models. These models determine local reaction rates and are often highly non-linear in relation to their parameters, such as local species' concentrations and temperature. This non-linearity causes error propagation whenever fields adjacent to reacting sites are assumed to be steady state.

Turbulence is a chaotic instability of fields of species and temperature in the fluid domain, caused by a high ratio of flow inertia to fluid viscosity. Turbulence is present in the inter-particle volume of the packed bed during the operation of industrial catalytic reactors, causing unsteadiness in these local fields. Such unsteadiness affects the conversion rate, unless internal diffusion in the catalyst pellet is so large that it is the limiting factor of the reaction.

DNS, unlike computationally cheaper and much more efficient RANS method, guarantees that local variations of species and temperature fields are captured accurately, which depending on the case, could have a significant impact on modelling result [2], especially when chemical kinetics are highly non-linear.

A tubular packed bed reactor with low d/D is a non-uniform reactor domain, where PR-CFD is needed to resolve the fluid flow without the assumption of homogeneous bed space. One chemical reaction which would typically take place in such a reactor due to its exothermicity is carbon dioxide hydrogenation to methanol. Previous studies have revealed discrepancies of results between modelling this reaction in 1D (homogeneous) and 2D (particle-resolved) conditions [3].

## 2. Methods

The computational technique used for solving this fluid flow problem is DNS of Navier-Stokes equations discretised along the domain using Finite Volume Method (FVM). The solution is fully resolved in the whole range of spatial and temporal scales, without using any turbulence model [2]. This

approach is very demanding in terms of computational resources [4]. However, the solution it offers is closer to reality than that obtained through RANS due to the inclusion of unsteady effects. The computational environment of choice is OpenFOAM, specifically reactingFoam solver adjusted to the chemical reactions being researched.

The simulation domain consists of a 3D cutaway of a packed bed of a tubular reactor in a scale magnitude of 20x20x20 mm. The domain is big enough to capture multiple layers of cylindrical catalyst particles [6]. Available computational resources limit both the simulation domain size as well as maximum flowrate for which the study can be performed to Re=800. Such flowrate is relatively low in comparison to industrial conditions, partially thanks to recycling rates of 3-8, which corresponds to Re=12000-32000.

## 3. Expected results and discussion

Multiple simulations are performed to find out how different reaction conditions, such as Reynolds Number (Re), affect local reaction rates and resulting conversion. Due to the influence of unsteady local species and temperature fields at the reacting catalytic sites, turbulence effects are expected to have an impact even if the reaction is not limited by external mass transfer.

A comparison is made between non-reacting conditions (cold gas flow) against reacting conditions to investigate the bulk influence of the chemical reaction on the flow through the packed bed. A significant discrepancy between these cases is expected, predominantly due to the decreasing number of molecules in this reaction.

Key results will include catalyst particle effectiveness factors, residence time distributions, axial and radial effective dispersion of mass and heat and plots of temperature and species fields' variations over time.

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***Keywords***

CFD, packed bed, reactor, turbulence