

Catalyst Supports for Selective Catalytic Reactions: Wire Mesh Screens vs. Monoliths

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Introduction

A highly active catalytic material is always required to meet the conversion requirements of a given process but often the reaction rate is limited by heat and mass transfer from the bulk phase to the catalytic sites. This is especially the case for conventional parallel channel catalyzed monoliths currently used worldwide for environmental applications. The use of monoliths is however limited to applications where mechanical stability and low pressure drop make them highly desirable, but due to their limited heat and mass transfer properties as well as poor radial mixing they have not gained an advantage over the widely used pellet beds for industrial applications. Wire mesh screens, representing the wider category of short contact time (SCT) supports, provide an advantage over conventional straight channel monoliths while still granting a pressure drop and mechanical stability advantage over the pebble bed.

This work aims to investigate the selective oxidation of CO in the presence of H_2 , H_2O and CO_2 (PROX process), for wire mesh screens and monoliths with two different catalyst formulations: Pt/Al_2O_3 and $Pt/FeAl_2O_3$. The process conditions for the comparison had to be selected accurately in order to avoid thermally induced state multiplicity. Comparison was performed in terms of CO conversion and CO selectivity, which in this case is defined as the fraction of O_2 that goes towards CO oxidation instead of H_2 oxidation[1].

Materials and Methods

The test rig consisted of a gas delivery manifold, using N_2 , H_2 , CO, CO_2 and air via mass flow controllers mixed to generate the simulated reformat feed. An inlet and exit gas sample port was installed and coupled directly to an on-line gas chromatograph (Agilent MicroGC-3000) and an infrared CO/CO_2 gas analyzer (California Analytical ZRF) for analysis. A syringe pump was used to introduce controlled amounts of water in an in-house steam generator assisted by a N_2 sweep gas for stability. All reactants with (except for N_2) were introduced after the steam generator, and air was introduced with a separate line to a 0.75" ID quartz tube reactor. The reactor was tested for a variety of inlet temperature, and each condition was operated at steady state

Results and Discussion

The performance envelope of the PROX reactor is divided in 3 representative regimes, here listed in order of increasing reaction temperatures. 1. The low temperature kinetically controlled regime, characterized by increasing conversion with increasing temperature, 2. The CO conversion plateau: a temperature window at which the CO conversion levels off, 3. The post-plateau CO conversion drop: in this regime the selectivity can remain constant or drop depending on the reaction conditions. Our work sheds light into the transport-kinetic background for the conversion/selectivity behavior in regimes 2 and 3. The conversion

drop from the kinetic regime to the plateau regime can be attributed to a switch in reaction mechanism (by the onset of the reverse water gas shift (rWGS) reaction which produces CO effectively reversing the conversion trend) or by CO bulk mass transfer. In Figure 1 cases C and D are test data of wire-mesh SCT and monolith and indicate that the SCT reactor reaches ~20% higher conversion for the same selectivity of 55% compared to the monolith reactor. Our in-house reactor model confirms that the mass transfer rate for wire mesh screens is higher than the one for monolith by a factor 3, hence delaying the onset of bulk mass transfer. The selectivity drop during CO bulk mass transfer limit can be attributed to the fact that H_2 oxidation is not yet bulk mass transfer limited, which however is not confirmed by our reactor model.

Comparison of the shape of A, B, and C shows that conversion maxima have different shapes according to how the reactor is operated: Case C at a space velocity of 300 L/gcat s shows that the maximum is shaped as a sharp cusp, indicating that the CO conversion levels off with a constant CO selectivity. The decrease in conversion following the CO conversion plateau is traditionally imputable to the onset of the rWGS reaction however our data contradicts with this notion, as experiments shows that for CO concentrations <1500ppm and $O/C < 1$ for a space velocities 300-900 L/gcat s the rWGS reaction on Pt/Alumina is inactive below 230°C. Under these conditions the CO conversion drop follows a different mechanism, which is under investigation.

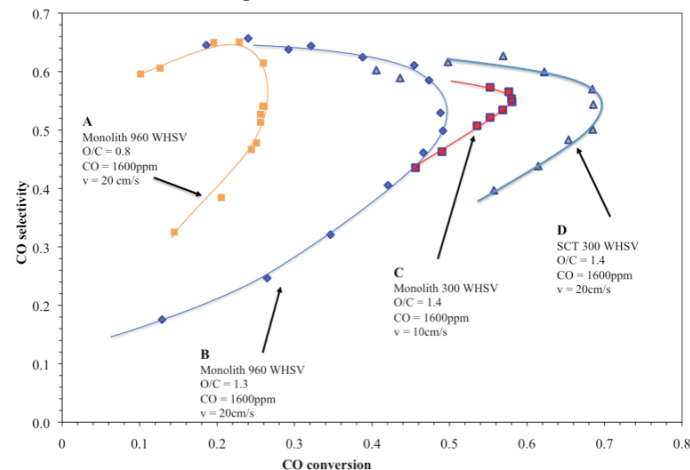


Figure 1: Selectivity-conversion envelope for monolith and wire mesh SCT
Significance

This research provides insight into fundamental aspects of supported catalysis by investigating heat and mass transfer impact on reaction rate and mechanism, and it elucidates aspects of the PROX process that are to date not fully understood.

References

1. M. J. Kahlich, *Journal of Catalysis* **171**, 93-105 (1997).