

Novel Reactors (Single Particle Temporal Analysis of Products Reactor)

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Introduction

In order to successfully model the transient-response curves obtained from a typical TAP reactor system using a one-dimensional Knudsen diffusion model, it is necessary to assume that temperature and concentration gradients are minimized. Historically, several transitions have occurred from a fully-packed catalyst bed to a three-zone model to a thin zone model have allowed for increasingly smaller temperature and concentration gradients through the active catalyst zone [1,2,3]. In continuation of this tendency, we submit the development of a single particle or pellet source reactor. In this reactor, a single active catalyst particle is placed within a sea of inert quartz particles (fig 1). Having such a configuration, now we are able to move the active catalyst particle in both axial and radial directions. It creates an experimental basis for extracting the corresponding profiles of gas concentrations and, in the future, a map of gas composition.

Materials and Methods

A TAP-2 multipulse reactor fitted with a high-throughput ultra-high vacuum system coupled to an UTI quadrupole mass spectrometer and data acquisition computer running version 2 of the TAPsoft software was the test bed for these experiments. The cylindrical micro reactor measured 2.54 cm in length and 0.95 cm in diameter and was fitted with a resistance heating system capable of 0.1° accuracy. Typical TAP pulse intensity was 1×10^{14} molecules per pulse. Single particles of platinum foil and powder served as feedstock to this work. A 250 μm diameter high-purity (99.9%) platinum particle was used. CO oxidation was studied both under transient pulse experimentation (UHV) and high-pressure flow conditions (FLOW). In both cases the catalyst was pretreated by oxidation at elevated temperatures. In UHV experiments, catalyst is titrated by CO-Ar pulses. Conversion was calculated as a function of axial position, and surface area. FLOW experiments performed at high flow rate (~40ml/min) of stoichiometric O₂ and CO flowed over the catalyst. Temperature ramping allowed the direct comparison of FLOW and UHV experiments over a wide range of operating conditions. Our estimates show that in both experiments, the temperature gradients within one particle are not significant.

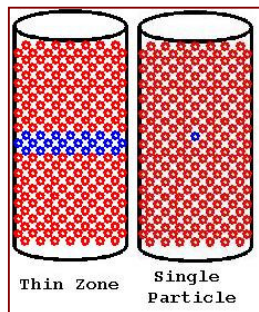


Fig 1. Progression from Thin zone to Single Particle configuration.

Results and Discussion

Under FLOW conditions, the conversion is lower because it is governed by the small cross sectional surface area of the catalytic pellet. In contrast, under UHV conditions,

the back diffusional process is the dominating factor as proved previously in thin zone TAP studies[4]. Comparison of FLOW and UHV experiments have shown that under flow experiments, CO conversion exhibits hysteresis due to the competitive adsorption mechanism. According to this mechanism, catalyst surface coverage may dramatically change from a preliminarily CO-covered surface at low temperatures to a O₂ covered surface at higher temperatures. By contrast, UHV experiments do not exhibit hysteresis because they do not significantly affect the catalyst surface (i.e. they are “state defining”)[2]. Rigorous theoretical studies of these phenomena will be studied in the future.

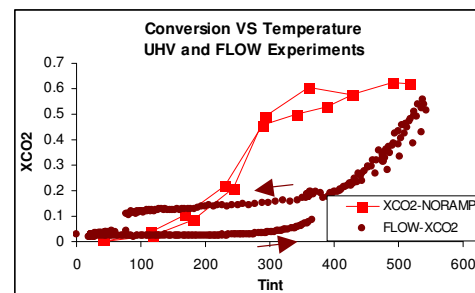


Fig. 2. Under FLOW conditions, a sizable Hysteresis effect is noticeable due to the change of catalyst surface loading from CO at low temperatures to O₂ at high temperatures. Under UHV conditions, the hysteresis effect is not visible.

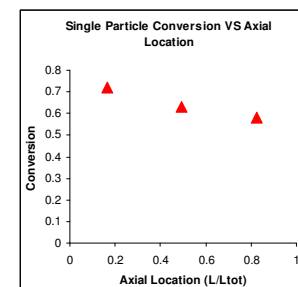


Fig. 3. Catalyst conversion as a function of axial position within the TAP reactor vessel under UHV using platinum foil.

Significance

The sequence of TAP zone models has been expanded by the addition of a single particle model in which the temperature and concentration gradients can truly be neglected.

References

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