

Spatially-Resolved Calorimetry - Using IR Thermography to Evaluate Axial Distribution of Sorbed Species Across Integral Catalyst Beds

William Epling^{*1}, Khurram Aftab¹, Fan Kit Cheuk¹, Aleksey Yezerets² and Neal W. Currier²
¹ Dept of Chemical Engineering, University of Waterloo, Waterloo, ON, N2L 3G1 (Canada)
² Cummins Inc., 1900 McKinley Ave, Columbus, IN 47201
^{*}wepling@uwaterloo.ca

Introduction

In catalytic systems, such as engine exhaust emissions control devices, large gradients of gas and surface species inevitably exist along the gas flow direction due to the integral nature of reactor operation. Understanding spatial distributions of adsorbed species is critical for developing physically relevant, predictive models of such catalysts. In this presentation, we will demonstrate that IR thermography can be used to infer the distribution of a reactive species based on the heat released during reaction. The method was validated by measuring the distribution of nitrate species on a model diesel NO_x adsorber catalyst using different degrees of surface saturation created by different exposure times and different NO/NO₂ ratios, and at two different temperatures.

Materials and Methods

The temperature changes and gradients formed during simulated operation of a Pt/Ba/Al₂O₃ NO_x adsorber catalyst (NAC) for diesel exhaust applications were monitored using IR thermography and standard thermocouples. The temperature gradient formed during nitrate reduction was used as a probe reaction, to evaluate the amount and position of NO_x deposited on the catalyst surface. The temperature gradient and distribution measurements are further being used to characterize the reactions and as input into models.

The experimental apparatus consists of a reactor designed to facilitate the use of IR thermography in measuring the temperature patterns that evolve on a catalyst during simulated operation. This unique reactor was designed and built to ensure a square-wave of gas composition and flow enters a monolith-supported catalyst which is 3" wide, 3" long and approximately 0.10" thick (two channels). An IR transparent window makes the upper seal of the reactor. The temperatures evaluated were 300 and 400°C. A fast-acting valve was used to switch from the lean, NO_x-containing stream to the reductant-containing stream. The exit gases were analyzed using an MKS MultiGas 2030 FTIR gas analyzer.

A FLIR Merlin Mid-Wave IR camera acquired images of the catalyst surface with a spatial resolution of 0.01" and a temporal resolution of 4 Hz. In our analysis, 20 equidistant temperature points are selected from inlet to outlet of the catalyst surface, along the radial center of the sample surface. These are labeled pos1 through pos20 in Figure 1, from inlet to outlet respectively.

Results and Discussion

Different levels of NO_x were trapped on a Pt/Ba/Al₂O₃ catalyst surface by varying the NO_x-exposure time intervals. Two limiting case experiments included (1) no NO_x in the lean stream and (2) complete saturation of the surface by exposing the catalyst to the lean, NO_x-containing stream until the outlet NO_x concentration was equal to the inlet level. The data were analyzed beginning with the onset of the reductant-containing or rich phase. The temperature rise as a function of time was integrated over the time period where an exotherm

associated with nitrate reduction was clearly observed. A small temperature rise was observed when no NO_x had been added, likely from H₂ reacting with PtO, or any other sources of oxygen in the reactor system. This rise was significantly lower than that observed when NO_x was added. The calculated, integrated area at each position (of temperature and time at each position) is plotted as a function of amount of NO_x deposited in Figure 1. The integrated areas clearly demonstrate axial gradients in sorbed NO_x species on the catalyst surface.

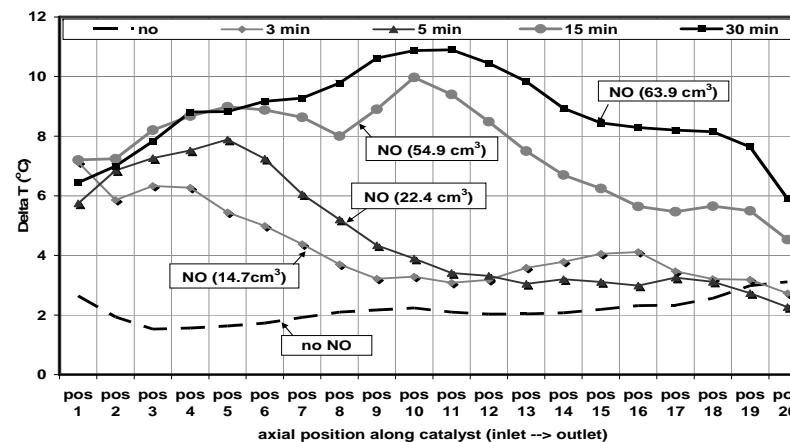


Figure 1. Integrated temperature/time as a function of axial position and amount of NO_x trapped at 400°C (units are °C-seconds).

The data show that NO_x trapping starts at the front of the catalyst and propagates along the catalyst bed with time, as expected [1,2]. But the data also show that with higher levels of NO_x trapped, more accumulates downstream of position 4, indicating it is not a simple saturation wave. Data obtained at the 300°C and with NO₂ will also be discussed in relation to the axial distributions and gradients observed.

Significance

IR thermography can be used to spatially resolve species distributions on a catalyst when a reaction can be induced with the target sorbate that is exothermic or endothermic. This form of calorimetry can be used to monitor changes in such distributions as a function of key reaction variables and furthermore can be used to evaluate effects of catalyst poisoning or aging.

References

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2. J.S. Choi, W.P. Partridge W.S. Epling, N.W. Currier, T.M. Yonushonis, *Catal. Tod.* 114, 102(2006).