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Sustained concentration and temperature oscillations

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Abstract

A diesel oxidation catalyst (DOC) comprising an alumina-based washcoat of Platinum (Pt) and Palladium (Pd) is used to catalyze the oxidation of a diesel exhaust containing hydrocarbons (HCs) with sufficiently high conversion. A large-pore zeolite such as Beta (BEA) is added to the DOC to trap diesel exhaust hydrocarbons during the cold start. There is a need to gain insight on how BEA affects the performance of the HC light-off (LO) and of the transient HC oxidation. We conducted steady-state and transient trapping experiments involving the n-dodecane in order to enable improved trap design and operating strategy. Simultaneous spatio-temporal measurements of concentration and temperature, respectively by spatially-resolved mass spectrometry and coherent optical frequency domain reflectometer, provide detailed insight into the reactor dynamics. Steady-state experiments reveal unexpected periodic oscillations of CO₂ and temperature in a feed temperature range (142-181 °C). The oscillatory amplitude increases at lower space velocity. Temperature programmed oxidation experiments revealed unusual multipeak CO₂ generation and temperature excursion behavior. A lower ramp-rate increases the number of the additional peaks. In addition, at a low heating/cooling rate of ±1 °C/min, a counter-clockwise hysteresis behavior exists. The oscillations and multi-peak behavior indicates coupling between HC trapping and oxidation. Analysis of the behavior during the hysteresis suggests that HC trapping and oxidation dominate in different regions along the length of the monolith. Since the downstream is much hotter than the upstream, the HC is re-trapped by the upstream BEA and is oxidized at the downstream. This study provides insight into the spatiotemporal features of the transient HC trapping and oxidation of BEA/DOC catalysts. To our knowledge, it is the first report of oscillations in a hydrocarbon trap.

KEYWORDS: hydrocarbon trap; Beta-zeolite, diesel oxidation catalyst, oscillations; dodecane;

oxidation; platinum

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