AIDIC EFCE
ISCRE25 FLORENCE 2018
BRIDGING SCIENCE & TECHNOLOGY
The 25th International Symposium on Chemical Reaction Engineering
20-23 May 2018
Florence, Italy
www.aidic.it/iscre25

Reaction Engineering of Catalytic Converters for Lean Burn Vehicles: Tailored Catalyst Architectures and Operational Strategies for Enhanced Performance

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Highlights

- The exhaust system of the lean burn vehicle requires several sequential reactors.
- Lean NOx reduction requires multi-functional catalysts and structured reactors.
- Targeted experiments uncover key trends while predictive models provide understanding.
- Dual-layer and zoned catalysts with modified porosity provide enhanced performance.

1. Introduction

The need both to reduce the consumption of transportation fuels and improve urban air quality confronts both developed and developing nations. Furthering the challenge is the ever-tightening emissions rules for nitrogen oxides (NOx), N_2O, and hydrocarbons including methane. Diesel, lean burn gasoline, and natural gas fueled engines are of interest because they are more fuel efficient than conventional stoichiometric engines. However, while the unconverted oxygen in the exhaust is beneficial for oxidation of hydrocarbons and CO, it prevents the use of the conventional three-way catalytic converter to reduce NOx to N_2. This has led to intense research and technology development during the past 15 years with some notable successes utilizing concepts and tools of catalytic reaction engineering. In this plenary, several examples are described that show how the use of multi-functional catalysts and structured reactors can achieve high NOx conversion over a wide range of operating conditions. Multi-functional catalyst architectures that combine two or more active layers or zones can be effective strategies to address cost and/or performance limitations. To be highlighted are dual-layer catalysts for ammonia-based selective catalytic reduction (SCR), ammonia slip catalysts (ASC), and combined NOx Storage and Reduction (NSR) and SCR. A combination of targeted experiments and modeling provide insight that enables a systematic optimization of catalyst architectures and operating strategies for enhanced performance.

2. Results and discussion

SCR is adopted from the stationary source process which utilizes NH_3 as the NOx reductant, and utilizes both Cu- and Fe-exchanged zeolite catalysts. While Cu is particularly active at low to moderate temperatures, Fe is active at high temperature. The combination of both in dual-layer or sequential reactors is shown to be an effective approach to expand the high NOx conversion temperature window [1,2].

The ASC combines an ammonia oxidation catalyst and a metal-exchanged zeolite as a NOx reduction catalyst in a dual-layer architecture to oxidize NH_3 with high selectivity to N_2. This architecture provides an effective combination of ammonia oxidation and selective NOx reduction [3]. Also discussed is the use of sacrificial additives to provide incremental macroporosity in the SCR layer which increases overall activity without compromising N_2 selectivity.

NSR is a promising but complex catalytic process that involves the sequential periodic reactive trapping of NOx and its rapid reduction on multi-functional catalysts containing precious metal and storage components [4]. Operated in the stand-alone lean NOx trap (LNT), NSR has the noted disadvantage of cost (precious metal) and byproducts (NH_3, N_2O). However, the ammonia generating capability of the LNT may be exploited in the combined use of NSR and SCR. To this end, the “NSR + SCR” catalyst combines periodic NOx storage and reduction with NH_3 generation and selective catalytic reduction of NOx. The use of layering and zoning of the two functions is described [5] as well as in situ measurement of reacting species

concentration profiles using spatially-resolved mass spectrometry [6]. The measurements providing insight into NOx reduction pathways with hydrocarbons, including water-gas shift reaction and steam reforming.

Finally, the cyclic operation of NSR poses particular challenges including significant exotherms and cycle time effects. Experiments and modeling quantify the impact of cycle time and nonisothermal effects [7,8]. The mechanism of fast cycling features better utilization of NOx storage sites, resulting in a higher NOx conversion and lower NH3 selectivity. Significant exotherms are detected and modeled under conventional ca. 1 minute cycle, resulting in increased NOx slip. In contrast, reduced NOx slip is encountered during near-isothermal, sub-10 second cycle time operation (fast cycling).The combination of experiments and modeling help to identify the mechanism responsible for the NOx conversion enhancement obtained when using selected hydrocarbon reductants.

3. Conclusions

Reaction engineering analysis and tools are used to elucidate the complex features of advanced catalytic converters for lean NOx reduction. Experiments uncover key trends while predictive models provide understanding, enabling the development and optimization of multi-functional catalysts in structured reactors.

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Keywords
NOx; vehicle emission control; lean NOx trap (LNT); ammonia slip catalyst (ASC); selective catalytic reduction (SCR); monolith reactor;