

Preface

Current and upcoming environmental policy throughout most of the world is imposing significant decreases in tailpipe NO_x , hydrocarbon, CO and particulate emissions from diesel engines. Past regulations have been addressed by the engine manufacturing community with engine adaptation through a variety of technologies, but future reductions, and even some current, will be addressed with aftertreatment technologies that include catalyst systems. Current, “standard” three-way catalyst technology for gasoline-using vehicles does not result in the needed reductions, which is not surprising as it was designed for emissions control in exhaust conditions that are net stoichiometric and with no consideration for particulate. Therefore, significant amounts of research and development over the last several years in the catalyst area have been directed at diesel and lean-burn exhaust aftertreatment.

Removal of the hydrocarbon and CO emissions in the lean exhaust conditions is currently accomplished using oxidation catalysts. Although there are now niche applications with particulate control and an introduction of new NO_x control technologies for spark-ignited engines on the market, the control technologies and solutions of NO_x and particulate from diesel engines are still being developed. The major challenges still include controlled oxidation of soot under nominal operating conditions and reduction of NO_x to N_2 under the lean exhaust gas conditions. Particulate filters are considered the technology that will be implemented for particulate removal. Periodic oxidation of the trapped soot to CO_2 can occur whether via interaction with NO_2 , termed passive oxidation, or O_2 , termed active oxidation. The reactions associated with these two occur in different temperature regimes, NO_2 at lower temperatures. However, with ever-decreasing engine-out NO_x emissions, interestingly due to increasingly stringent regulations, the amounts of NO_2 are not always sufficient to react with the accumulating soot on the filters. In many duty cycles there is therefore a need for active oxidation by O_2 . The temperatures necessary for oxidation by O_2 , with no catalytic help, are higher than the normal exhaust temperatures. A primary challenge is therefore achieving the temperature necessary for soot oxidation in the exhaust or developing a catalyst coating for the filter that lowers the oxidation temperature. The coincident challenge is controlling the reaction once it begins. The C oxidation reaction is highly

exothermic and ensuring no runaway reaction dictates a precise understanding of the kinetics as well as flow, heat and mass transfer characteristics of the catalyst/filter system.

Two pathways are under current development for NO_x reduction to N_2 . First is the selective reduction of NO_x to N_2 using ammonia, or urea as a precursor, or hydrocarbons already available in the exhaust gas. Europe appears to have adopted the former as a solution with some concern directed at NH_3 slip in lieu of NO_x and therefore control strategies for transient operation become critical. Hydrocarbon as the reductant has been plagued by low conversions. In each case however, the understanding of the reaction pathways has been increasing and the coincident selectivity and conversions have increased as well. The alternate solution is the periodic non-selective reduction of NO_x via NO_x storage/reduction (NSR) or NO_x trap catalysis. One step of this process includes the trapping of NO_x on a surface containing a basic component. Unfortunately, SO_x species are trapped as well and the surface S species are quite stable. Periodic removal of S to regain activity is necessary, but difficult as this reaction requires high temperature and a coincident reducing environment. Both the periodic NO_x to N_2 and S removal steps have an associated fuel penalty, which needs to be minimized. As this is a relatively new technology for mobile applications, a significant amount of research is continuing and directed at understanding the reduction pathways as well as removing S as quickly and infrequently as possible.

Associated with these challenges are the quickly approaching regulation time frames in Europe and North America. Reducing NO_x and particulate with such significant challenges have therefore become areas of very significant topical interest. At the most recent North American Meeting of the North American Catalysis Society, the 19th NAM, four half-day sessions were dedicated to diesel emissions reduction. One session was dedicated to catalyst-based particulate control, one to lean- NO_x catalysis via selective catalytic reduction (SCR) with ammonia or hydrocarbons and two were dedicated to NSR catalyst research and development. Three keynote speakers and one plenary speaker were invited and over 70 abstracts were submitted, with 24 chosen for oral presentations. Interestingly, each session contained presentations from industry, academia and national laboratories.

This edition of Catalysis Today contains manuscripts solicited from 13 groups working in these areas and are based on the oral or poster presentations given at the meeting. Soot oxidation, SCR with ammonia and hydrocarbons, NSR catalysis and NO oxidation are all addressed in the papers presented. Some highlights include novel catalyst formulations for soot oxidation over catalyst-coated particulate filters, SCR with hydrocarbons, and increased NSR catalyst performance. The first-published N_2 evolution profiles during NSR catalyst regeneration are presented, along with several papers addressing the reaction mechanism of NSR catalyst operation. During the SCR session, there was an interesting discussion concerning the “fast” mechanism in NH_3 -based SCR. Both sides realized they agreed on the mechanism, but arrived at their conclusions via differing methods. The details of this mechanism and one of the approaches are presented in this edition.

The guest editors thank all of the catalyst scientists and engineers who presented at the 19th NAM, the authors who submitted manuscripts with such a short time frame, and the conference organizers, especially Bob Farrauto for putting our team together. Finally, the three junior partners would like to recognize Barry Cooper for his leadership through the last year

in bringing together such successful sessions in diesel emissions reduction.

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