SELECTIVE REDUCTION OF NO_X IN OXYGEN-RICH ENVIRONMENTS WITH PLASMA-ASSISTED CATALYSIS: CATALYST DEVELOPMENT AND MECHANISTIC STUDIES

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ABSTRACT

The control of NO_x (NO and NO_2) emissions from so-called 'lean-burn' vehicle en-gines remains a challenge. In recent years, there have been a number of reports that show that a plasma device combined with a catalyst can reduce as high as 90 percent or more of NO_x in simulated diesel and other 'lean-burn' exhaust.

Non-thermal plasma (NTP)-catalysis treatment of a gas stream consists of the sequential (or possibly simultaneous) action of NTP and heterogeneous catalysis. The most common reported strategy for lean NO_x treatment is to separate the plasma from the catalyst, as it is difficult to create robust plasma inside a catalyst bed. In the case of propylene-containing simulated diesel exhaust, the beneficial role of a plasma treatment is now thought to be due to oxidation of NO to NO_2 and the formation of partially oxidized hydrocarbons that are more active for the catalytic reduction of NO_2 than propylene. For plasma-assisted catalysis, both zeolite- and alumina-based materials have shown high activity, albeit in somewhat different temperature ranges, when preceded by an NTP reactor.

This presentation will describe our research efforts aimed at optimizing the catalyst materials for NTP-catalysis devices based, in part, on our continuing studies of the NTP- and catalytic-reaction mechanisms. Various alkali- and alkaline earth-cation-exchanged Y zeolites have been prepared and their material properties characterized. They have been tested as catalytic materials for NO_x reduction in laboratory NTP-catalysis reactors. Interestingly, NO_2 formed in the plasma, and not subsequently removed over these catalysts, will back-convert to NO, albeit to varying extents depending upon the nature of the cation.

Besides this comparative reactivity, we will also discuss selected synthesis strategies for enhancing the performance of these zeolite-based catalyst materials. A particularly important result from our mechanistic studies is the observation that aldehydes, formed during the plasma treatment of simulated diesel exhaust, are the important species for the reduction of NO_x to N_2. Indeed, acetaldehyde has been found to be especially effective in the *thermal* reduction of both NO and NO_2 over Ba- and Na-zeolite-Y catalysts.

Finally, we will discuss experiments aimed at understanding and controlling deactivation of these zeolite-based catalysts due to 'coking.' We believe that this catalyst deactivation mechanism explains a loss of catalyst efficiency with time that is observed in engine dynamometer tests.