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Electrochemical Reduction of NO_2

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Abstract Text:

The use of diesel engines is becoming more widespread because their fuel economy is superior to and their emission levels of carbon monoxide and carbon dioxide are considerably lower than those of gasoline engines. However, the high-temperature combustion in diesel engines generates significant amounts of nitrogen oxides (NO_x), which have harmful effects on the environment and human beings, and are limited by increasingly stringent government regulations worldwide. The O_2-rich environment of diesel engine exhaust will deactivate the traditional three-way catalysts that work effectively in gasoline engine exhaust. Therefore, there is a great demand for new technology to control NO_x emissions in diesel engine exhaust. The most extensively researched technologies in this area are currently selective catalytic reduction with ammonia (NH_3-SCR) and NO_x storage and reduction catalysts (NSR), both of which require a reducing agent, either from a secondary supply system or by switching the operation state of the engine between lean and rich conditions. One attractive alternative to these approaches is electrochemical NO_x reduction using a solid state cell. Using this approach, NO_x is reduced to nitrogen at the polarized cathode, thereby eliminating the need for the addition of reducing agents or changes in the operational state of the engine.

At present, the main obstacle to the practical application of this technology is the achievement of high selectivity for NO_x reduction in the presence of excess O_2. Because competitive O_2 reduction consumes substantial amount of current, the current efficiency or selectivity of the cell towards NO_x reduction is genetally a few percent relative to the total current supplied to the cell. Extensive research effort has been put on finding suitable cathode materials or optimizing the cell structure to increase its selectivity. However, the highest current efficiency reported in the literature was below 20% in an oxidizing atmosphere using a multilayered electrochemical cell with Pt cathode [1].

In this study, we proposed a novel concept for the electochemical NO_x reduction, which significantly improve the selectivity towards NO_x reduction. According to our previous research, it is found that the electrochemical reduction of NO_x is probably limited by the formation of intermediate NO_2. Besides, NO_2 is also reported to be an necessary precursor for the NO_x trapping process over the NO_x aborbents added in the electrochemical cell. NO is usually the dominant species of NO_x (90-95%) in exhaust gases, but can be converted to NO_2 efficiently by the catalysis of DOC. As the high efficiency of DOC for NO converstion to NO_2 has already been well demonstrated, in this study, we focus on the reduction of NO_2 on the electrochemical cell. We developed and characterised two typed of cells: one is a modify mulilayered cells with Pt and Ni/YSZ cathode with a K-Pt-Al_2O_3 adsorption layer, the other is a LSM/CGO symmetric cell infiltrated with BaO. We compare the NO_x removal properties of the cells in the atmosphere of mainly NO_2 with O_2 with that in mainly NO with O_2 and investigated the effect of temperature, voltage, and polarization method on the performance of cells. Under O_2-rich conditions, a current efficiency as high as 50-65% is achieved with a NO_x converstion of 50-70% on a modified multilayer cell with Pt and a 10% current efficiency with more than 30% NO_x converstion is realized on a fully ceramic cell free of noble metal. The contribution will briefly outline, how the concept can be further implemented in automotive exhaust cleaning systems.

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