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Electrochemical Reduction of NO₂

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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 (NOₓ), which have harmful effects on the environment and human beings, and are limited by increasingly stringent government regulations worldwide. The O₂-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 NOₓ emissions in diesel engine exhaust. The most extensively researched technologies in this area are currently selective catalytic reduction with ammonia (NH₃-SCR) and NOₓ 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 NOₓ reduction using a solid state cell. Using this approach, NOₓ 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 NOₓ reduction in the presence of excess O₂. Because competitive O₂ reduction consumes substantial amount of current, the current efficiency or selectivity of the cell towards NOₓ reduction is generally 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 electrochemical NOₓ reduction, which significantly improve the selectivity towards NOₓ reduction. According to our previous research, it is found that the electrochemical reduction of NOₓ is probably limited by the formation of intermediate NO₂. Besides, NO₂ is also reported to be an necessary precursor for the NOₓ trapping process over the NOₓ absorbents added in the electrochemical cell. NO is usually the dominant species of NOₓ (90-95%) in exhaust gases, but can be converted to NO₂ efficiently by the catalysis of DOC. As the high efficiency of DOC for NO conversion to NO₂ has already been well demonstrated, in this study, we focus on the reduction of NO₂ 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-Al₂O₃ adsorption layer, the other is a LSM/CGO symmetric cell infiltrated with BaO. We compare the NOₓ removal properties of the cells in the atmosphere of mainly NO₂ with O₂ with that in mainly NO with O₂ and investigated the effect of temperature, voltage, and polarization method on the performance of cells. Under O₂-rich conditions, a current efficiency as high as 50-65% is achieved with a NOₓ conversion of 50-70% on a modified multilayer cell with Pt and a 10% current efficiency with more than 30% NOₓ conversion 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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