



Highly selective NO_x reduction for diesel engine exhaust via an electrochemical system



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ABSTRACT

It is challenging to reduce the nitrogen oxides (NO_x) in diesel engine exhaust due to the inhibiting effect of excess oxygen. In this study, a novel electrochemical deNO_x system was developed, which eliminated the need for additional reducing materials or a sophisticated controlling system as used in current diesel after-treatment techniques. The electrochemical system consisted of an electrochemical cell modified with NO_x adsorbents and a diesel oxidation catalyst placed upstream of the cell. The system offers highly selective NO_x reduction and a strong resistance to oxygen interference with almost zero emission of secondary pollutants.

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1. Introduction

Diesel engines offer superior fuel economy and potential for a great reduction in the emission of greenhouse gases [1]. However, the high-temperature combustion in diesel engines generates significant amounts of nitrogen oxides (NO_x) [2] that are harmful for the environment [3,4] and humans [5]. Currently, the most mature deNO_x technologies for diesel engine exhaust are selective catalytic reduction with ammonia (NH₃-SCR) and NO_x storage and reduction catalysts (NSR) [6]. The NH₃-SCR method uses ammonia or urea to reduce NO_x selectively and requires a secondary fluid tank with an injection system for the reducing agents. This results in added system complexity and problems related to storage, spill management, refilling network infrastructure, and pollution from side products. The NSR system achieves NO_x reduction by selectively storing NO_x under lean conditions and subsequently reducing the stored NO_x upon switching to rich conditions. However, a sophisticated adaptive control system is necessary for the implementation of this approach in mobile applications. Consequently there is substantial interest in the development of alternative NO_x removal techniques to overcome these problems.

In this study, we report an electrochemical deNO_x system that exhibits high activity and selectivity toward NO_x reduction while simultaneously eliminating the need to add reducing agents or change the

operational state of the engine. The new system also minimizes the release of damaging secondary pollutants. Electrochemical NO_x reduction has been extensively studied as an alternative technology for diesel exhaust purification since the concept was proposed by Pancharatnam et al. in 1975 [7]. Here, a solid state cell is used to reduce NO_x to N₂ on the cathode under negative polarization, requiring no additional reducing materials other than electrons and showing the potential for the formation of N₂ and O₂ only. Owing to its potential as a simple, clean and compact deNO_x technology, this approach has attracted much research interest [8–11].

To reduce NO_x in an oxygen-rich environment, in addition to a high activity, the cathode of the electrochemical cell must simultaneously exhibit a high selectivity toward NO_x reduction in order to suppress the competitive O₂ reduction reaction [12]. Despite an extensive research effort, only a small number of materials (e.g., Ni [13,14], Ir [15], and (doped-)LaCoO₃ [16,17]) have been observed to be more electrochemically active toward NO_x than O₂, and even these materials alone did not provide satisfactory deNO_x selectivity (less than 5%) in the presence of excess O₂. It was suggested that adding NO_x adsorption materials (alkali metal oxides or alkaline earth metal oxides) to the cathode could effectively improve its selectivity [8,18,19]. However, the best selectivity/current efficiency reported in the literature was below 20% [18].

In the present study, we show that the issue of selectivity in electrochemical NO_x reduction can be solved by integrating a diesel oxidation catalyst (DOC) upstream of an electrochemical cell modified with NO_x adsorbents (Fig. 1a). A current efficiency of 50% to 65% is achieved with an NO_x conversion of 50% to 70% in 8% O₂ and 1000 ppm NO at 400 °C. A nearly complete reduction of NO_x to N₂ was realized with an N₂ selectivity above 90%. The principle of the system is to first oxidize

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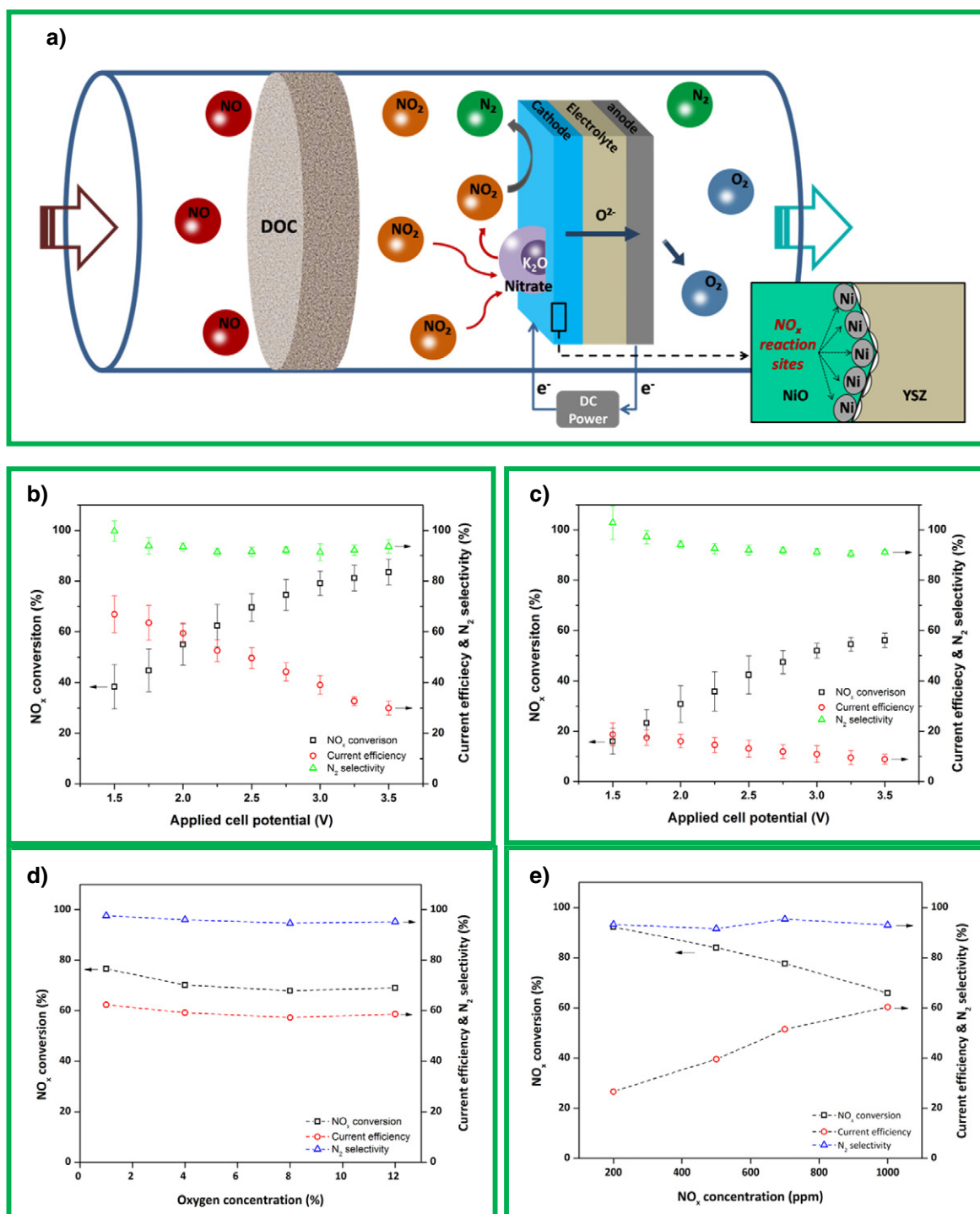


Fig. 1. a) Schematic diagram of the electrochemical deNO_x system; NO_x reduction properties of b) the electrochemical system, and c) the modified cell without the DOC in 1000 ppm NO and 8% O₂ in a balance of Ar at 400 °C under various voltages; effects of d) O₂ concentration (with 1000 ppm NO) and e) NO_x concentration (with 8% O₂) on the NO_x removal properties of the electrochemical system at 400 °C at 2.5 V.

NO to NO₂ using the DOC, then selectively adsorb and reduce NO₂ to N₂ in the electrochemical cell under polarization.

2. Experimental

We used a DOC catalyst consisting of 2 wt% Pt/γ-Al₂O₃ supported on cordierite monolith. The electrochemical cell was composed of a YSZ (8% Y₂O₃ doped ZrO₂) electrolyte substrate, a NiO|YSZ (55 mol%/45 mol%) cathode, a mesh-patterned Pt current collector,

an Al₂O₃-supported K₂O adsorption layer, and a Pt|YSZ anode. The cell was fabricated by tape casting the electrolyte, screen printing the electrodes, and drop casting the adsorption layer. After preparation, the large cell (5 cm × 5 cm) was laser-cut into several small round cells with a diameter of 15 mm. The active area of the cell was approximately 1.77 cm². The DOC catalyst and one cell were placed in a quartz tube heated by a furnace. The cell was placed perpendicular to the gas flow and connected to a Zahner Zennium electrochemical workstation. To simulate the gas atmosphere in the diesel engine exhaust, gas with

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