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Effective additives to improve hydrothermal aging of DME reforming catalyst in terms of hydrogen yield and de-NOx performance of RC + LNT combined system

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ABSTRACT

The purpose of this study is to observe the effects of H_2 generation by a reforming catalyst ($Cu30/\gamma\text{-}Al_2O_3$) with various additives, so as to improve the durability of the combined system of reforming catalyst (RC) + LNT. Under both fresh and hydrothermal aging conditions, the H_2 yields of the $Cu29CeO_21/\gamma\text{-}Al_2O_3$ catalyst were the highest among the reforming catalysts prepared by the sol–gel method. Even though the H_2 yield obtained using the DME reforming reaction of the exhaust gases of DME vehicles has the advantage of using the heat source and water in the exhaust gas, the existence of NO, CO_2 and O_2 in the exhaust gases is disadvantageous to the DME reforming reaction. The NOx conversion of the combined system of RC ($Cu29CeO_21/\gamma\text{-}Al_2O_3$) + LNT was higher than that of the LNT alone. This shows that the H_2 generated by the reforming reaction using the exhaust gases of the DME engine is a more effective reductant. Therefore, in order to effectively improve the NOx conversion, the combined system of RC + LNT is needed as an after-treatment system for DME vehicles.

1. Introduction

Dimethyl ether (DME) is being actively studied as an alternative fuel to diesel because it has a high cetane number and produces no sulfur or particle matter in its exhaust [1,2]. Engines using DME as a fuel produce less carbon dioxide (CO_2) and nitrogen oxides (NOx) than diesel engines [3,4], but further decrease in the NOx emission remains an important issue for DME engines due to stringent exhaust emission regulations [5,6]. Currently, a number of NOx reduction catalysts are known, such as the Lean NOx trap (LNT) and selective catalytic reduction (SCR, urea or HC-SCR) [7,8]. LNT, which is commercialized for light vehicles, can effectively reduce NOx by using the reductants (hydrogen (H_2), carbon monoxide (CO), and hydrocarbon (HC)) which are generated through precise engine control and the post injection of fuel without additional devices [9]. To effectively reduce the NOx exhausted from DME engines, we recently not only developed a combined system consisting of a DME reforming catalyst (RC) and LNT using the H_2 generated from the DME RC, but also demonstrated these improved performance of the de-NOx in DME engines [10,11]. However, no reforming catalyst has been reported that can improve the generation of H_2 from the DME steam reforming (SR) or oxidative steam reforming (OSR) reactions using the exhaust gases of a DME engine. The reforming catalyst used as an after-treatment system for DME engines is confronted with three critical problems, which are very different from those of the reforming catalyst used in fuel cells [12]. Firstly, CO_2, NO, and O_2 coexist in the exhaust gases of DME engines [13,14], which makes the DME reforming process more difficult and decreases the amount of H_2 generated. Secondly, the space velocity (SV) of the RC changes frequently according to the load conditions of the DME engine. When the DME engine drives under high load conditions, the SV increases and the H_2 generated by the DME reforming reaction decreases. Thirdly, the heat source and water vapor for DME reforming can be supplied by the hot exhaust gases of the DME engine, but if they are used for a long time, the reaction activity of the RC and LNT decreases due to hydrothermal aging [15,16]. As the catalysts come to suffer from hydrothermal aging after long term use, it is difficult not only to maintain the surface area, size and dispersion of the metal particles, and the surface area of the supports such as alumina, but also to prevent the phase transition depending on the interactions between the supports and metals, and the growth and cohesion of the active sites over the supports. To address these problems, many studies have been performed recently on the maintenance of the surface area, size and dispersion of metal particles by mixing various additives with DME RC [17–19]. However, no studies have been demonstrated on the effects of the hydrothermal aging of RC in DME vehicles on H_2 generation and de-NOx performance of the LNT combined system.

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