Enhanced Redox Property in Ni-Nb-O/$Ce_xZr_{1-x}O_2$ for Selective Production of Ethylene from Ethane

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Ethylene is an important primary building block for preparing value-added intermediates [1]. One of the currently highlighted reactions to produce ethylene is a catalytic conversion of ethane using oxygen, and this is now referred to as the oxidative dehydrogenation of ethane (ODHE) [2]. Niobium-doped NiO (Ni-Nb-O) is one of the promising catalysts for ODHE reaction at low reaction temperature (≤420 °C) [3]. However, low level of ethane conversion (30~35%) caused by low reaction temperature limits further increase in ethylene productivity [4]. The presence of this limitation would be due to the difficulties in maintaining high ethylene selectivity at high reaction temperature, with producing C1 chemicals such as CO_x and/or CH_4.

The ODHE reaction pathway on Ni-Nb-O is believed to follow Mars-van Krevelen mechanism. From this description, lattice oxygen within the catalyst directly participates in the production of ethylene [5]. It has generally been accepted that the byproducts are produced by the surface oxygen generated by surface positive holes (h^+) when ethylene over-oxidation is a bystander [6]. Therefore, suppressed ethylene selectivity at relatively high reaction temperature can be explained by this theoretical approach, which might be ascribed to the insufficient reactive lattice oxygen capability within the Ni-Nb-O. On the basis of this reaction mechanism, enabling of a rapid uptake and release of lattice oxygen can be an effective strategy to improve the catalytic performance of Ni-Nb-O and increases ethylene production via the ODHE process.

In this study, we report on a development of Ni-Nb-O/$Ce_xZr_{1-x}O_2$ catalyst for selective production of ethylene via ODHE reaction. Compared with the conventional manner of using Ni-Nb-O alone, the introduction of $Ce_xZr_{1-x}O_2$ to the Ni-Nb-O active catalyst contributes on suppressing the formation of byproducts (CO, CO_2, and CH_4) at relatively high reaction temperature (450 °C). The increased reaction temperature also leads to an enhancement of ethane conversion (~55%) and subsequent increase in the production of ethylene. Electrochemical tests and control tests demonstrate that the promotion effect is attributed to the better redox property of Ni-Nb-O with the introduction of $Ce_xZr_{1-x}O_2$.

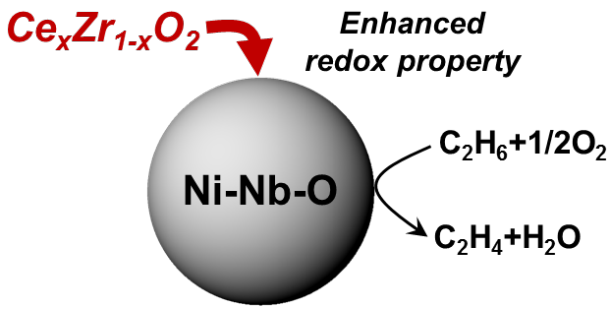

Fig. 1 Schematic illustration of enhanced redox property in Ni-Nb-O/$Ce_xZr_{1-x}O_2$ composite for selective production of ethylene *via* oxidative dehydrogenation of ethane.

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