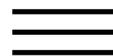


Methane steam reforming over Ni/Ce-ZrO₂ catalyst: Influences of Ce-ZrO₂ support on reactivity, resistance toward carbon formation, and intrinsic reaction kinetics.

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Methane steam reforming over Ni/Ce-ZrO₂ catalyst: Influences of Ce-ZrO₂ support on reactivity, resistance toward carbon formation, and intrinsic reaction kinetics

N. Laosiripojana ^a ... S. Assabumrungrat ^b

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Abstract

Ni/Ce-ZrO₂ showed good methane steam reforming performance in term of stability toward the deactivation by carbon deposition. It was first observed that the catalyst with Ce/Zr ratio of 3/1 showed the best activity among Ni/Ce-ZrO₂ samples with the Ce/Zr ratios of 1/0, 1/1, 1/3, and 3/1. Temperature-programmed oxidation (TPO) experiments indicated the excellent resistance toward carbon formation for this catalyst, compared to conventional Ni/Al₂O₃; the requirement of inlet H₂O/CH₄ to operate without the formation of carbon species is much lower. These benefits are related to the high oxygen storage capacity (OSC) of Ce-ZrO₂. During the steam reforming

process, in addition to the reactions on Ni surface (Ni^0), the redox reactions between the gaseous components present in the system and the lattice oxygen (O_x) on $\text{Ce}^{\text{IV}}\text{-ZrO}_2$ surface also take place. Among these reactions, the redox reactions between the high carbon formation potential compounds (CH_4 , CH_x and CO) and the lattice oxygen (O_x) can prevent the formation of carbon species from the methane decomposition and Boudard reactions, even at low inlet $\text{H}_2\text{O}/\text{CH}_4$ ratio (1.0/1.0).

Regarding the intrinsic kinetic studies in the present work, the reaction order in methane over $\text{Ni}/\text{Ce}^{\text{IV}}\text{-ZrO}_2$ was observed to be approximately 1.0 in all conditions. The dependence of steam on the rate was non-monotonic, whereas addition of oxygen as an autothermal reforming promoted the rate but reduced CO and H_2 production selectivities. The addition of a small amount of hydrogen increased the conversion of methane, however, this positive effect became less pronounced and the methane conversion was eventually inhibited when high hydrogen concentration was added. $\text{Ni}/\text{Ce}^{\text{IV}}\text{-ZrO}_2$ showed significantly stronger negative impact of hydrogen than $\text{Ni}/\text{Al}_2\text{O}_3$. The redox mechanism on ceria proposed by Otsuka et al. [K. Otsuka, T. Ushiyama, I. Yamanaka, Chem. Lett. (1993) 1517; K. Otsuka, M. Hatano, A. Morikawa, J. Catal. 79 (1983) 493; K. Otsuka, M. Hatano, A. Morikawa, Inorg. Chim. Acta 109 (1985) 193] can explain this high inhibition.



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