Applied Catalysis A: General

Volume 290, Issues 1â€“2, 18 August 2005, Pages 200-211

Methane steam reforming over Ni/Ceâ€“ZrO_2 catalyst: Influences of Ceâ€“ZrO_2 support on reactivity, resistance toward carbon formation, and intrinsic reaction kinetics

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https://doi.org/10.1016/j.apcata.2005.05.026

Abstract

Ni/Ceâ€“ZrO_2 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/Ceâ€“ZrO_2 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/Al_2O_3; the requirement of inlet H_2O/CH_4 to operate without the formation of carbon species is much lower. These benefits are related to the high oxygen storage capacity (OSC) of Ceâ€“ZrO_2. During the steam reforming

process, in addition to the reactions on Ni surface (*), the redox reactions between the gaseous components present in the system and the lattice oxygen (O_x) on Ceâ€“ZrO_2 surface also take place. Among these reactions, the redox reactions between the high carbon formation potential compounds (CH_4, CH_x-*n 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 H_2O/CH_4 ratio (1.0/1.0).

Regarding the intrinsic kinetic studies in the present work, the reaction order in methane over Ni/Ceâ€“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. Ni/Ceâ€“ZrO_2 showed significantly stronger negative impact of hydrogen than Ni/Al_2O_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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