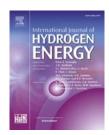
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Kinetics and mechanistic studies of CO-rich hydrogen production by CH₄/CO₂ reforming over Praseodymia supported cobalt catalysts



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ABSTRACT

The production of hydrogen and syngas by catalytic methane dry reforming is often accompanied by carbon deposition. In order to mitigate the effect of carbon deposition, it is important to have a full understanding of the elementary steps involve in the methane dry reforming over a given catalyst. This will enable the proper design and optimization of such catalyst to minimize catalyst deactivation by carbon deposition. In this study, the kinetic and mechanistic features of 20 wt%Co/Pr₂O₃ catalyst in the methane dry reforming reaction has been investigated as a function of CH₄ and CO₂ partial pressures and reaction temperature. The 20 wt%Co/Pr₂O₃ catalyst was synthesized by wet impregnation method and characterized for its physicochemical properties by TGA, XRD, N₂-physisorption analysis, TEM, FESEM, EDX, H₂-TPR, NH₃ and CO₂. The excellent physicochemical properties of the 20 wt%Co/Pr₂O₃ catalysts resulted in a high rate of CH_4 (r_{CH_4}) and CO_2 (r_{CO_2}) consumption. The highest values of 3.6 mmol gcat $^{-1}$ min $^{-1}$ and 3.2 mmol gcat $^{-1}$ min $^{-1}$ were obtained for r_{CH_4} and $r_{CO_2},$ respectively. tively at 50 kPa and 1023 K. The kinetic behavior of the as-synthesized 20 wt%Co/Pr $_2O_3$ catalyst in the methane dry reforming reaction was measured in a fixed bed stainless steel reactor at CH₄/CO₂ partial pressure range of 5–50 kPa and temperature range of 923–1023 K. The data obtained from the kinetic measurement were fitted into seven Langmuir-Hinshelwood (LH) Models. The Model were statistically discriminated using root mean square deviation (rmsd) and coefficient of determination (\mathbb{R}^2). The statistical analysis revealed that LH kinetic Model 7 (2-step dual site rate determining steps (RDS) involving CH4 activation by metal Co and C gasification by adsorbed CO_2 on support site) fits very well the experimental data. The R^2 values of 0.962, 0.982, and 0.989 as well as, rmsd values of 0.095 0.038, and 0.035 were obtained at 923, 973, and 1023 K respectively. Activation energies of 61.67 and 32.52 kJ/mol were obtained for the rate of consumptions of CH₄ and CO₂, an indication that lower energy barrier is required for the activation of CO₂ compared to CH₄. Based on Model 7, the mechanism of the methane dry reforming reaction over the 20 wt%Co/ Pr_2O_3 catalyst can best be described by 2step dual site rate determining steps whereby the activation of CH₄ by the metal Co resulted in hydrogen and carbon formation. The carbon formed was subsequently gasified by the lattice oxygen released from the activation of the CO_2 by the support site.

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