CO2 Methanation over Ru-Ni/CeO2 Catalysts

Authors : Nathalie Elia, Samer Aouad, Jane Estephane, Christophe Poupin, Bilal Nsouli, Edmond Abi Aad Abstract : Carbon dioxide is one of the main contributors to greenhouse effect and hence to climate change. As a result, the methanation reaction CO2(g) + $4H2(g) \rightarrow CH4(g) + 2H2O (\Delta H^{\circ}298 = -165 \text{ kJ/mol})$, also known as Sabatier reaction, has received great interest as a process for the valorization of the greenhouse gas CO2 into methane which is a hydrogen-carrier gas. The methanation of CO2 is an exothermic reaction favored at low temperature and high pressure. However, this reaction requires a high energy input to activate the very stable CO2 molecule, and exhibits serious kinetic limitations. Consequently, the development of active and stable catalysts is essential to overcome these difficulties. Catalytic methanation of CO2 has been studied using catalysts containing Rh, Pd, Ru, Co and Ni on various supports. Among them, the Ni-based catalysts have been extensively investigated under various conditions for their comparable methanation activity with highly improved costefficiency. The addition of promoters are common strategies to increase the performance and stability of Ni catalysts. In this work, a small amount of Ru was used as a promoter for Ni catalysts supported on ceria and tested in the CO2 methanation reaction. The nickel loading was 5 wt. % and ruthenium loading is 0.5wt. %. The catalysts were prepared by successive impregnation method using Ni(NO3)2.6H2O and Ru(NO)(NO3)3 as precursors. The calcined support was impregnated with Ni(NO3)2.6H2O, dried, calcined at 600°C for 4h, and afterward, was impregnated with Ru(NO)(NO3)3. The resulting solid was dried and calcined at 600°C for 4 h. Supported monometallic catalysts were prepared likewise. The prepared solids Ru(0.5%)/CeO2, Ni(5%)/CeO2 and Ru(0.5%)-Ni(5%)/CeO2 were then reduced prior to the catalytic test under a flow of 50% H2/Ar (50 ml/min) for 4h at 500°C. Finally, their catalytic performances were evaluated in the CO2 methanation reaction, in the temperature range of 100-350°C by using a gaseous mixture of CO2 (10%) and H2 (40%) in Ar balanced at a total flow rate of 100 mL/min. The effect of pressure on the CO2 methanation was studied by varying the pressure between 1 and 10 bar. The various catalysts showed negligible CO2 conversion at temperatures lower than 250°C. The conversion of CO2 increases with increasing reaction temperature. The addition of Ru as promoter to Ni/CeO2 improved the CO2 methanation. It was shown that the CO2 conversion increases from 15 to 70% at 350°C and 1 bar. The effect of pressure on CO2 conversion was also studied. Increasing the pressure from 1 to 5 bar increases the CO2 conversion from 70% to 87%, while increasing the pressure from 5 to 10 bar increases the CO2 conversion from 87% to 91%. Ru-Ni catalysts showed excellent catalytic performance in the methanation of carbon dioxide with respect to Ni catalysts. Therefore the addition of Ru onto Ni catalysts improved remarkably the catalytic activity of Ni catalysts. It was also found that the pressure plays an important role in improving the CO2 methanation.

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