Catalytic CH_4 reforming with CO_2 over activated carbon based catalysts

The objective of this study is to compare the performance of several activated carbon (AC) based catalysts for CH_4 reforming with CO_2, including raw AC and AC modified with HNO_3 and $NaNO_3$, designated as AC-HNO_3 and AC-$NaNO_3$, respectively. CH_4-CO_2 reforming experiments were conducted in a fixed-bed reactor at temperatures from 700 to 1,000 °C. The catalysts were characterized by various techniques, including Fourier Transform Infrared Spectroscopy (FTIR), Brunauer-Emmett-Teller (BET) analysis, CO_2 chemisorption, temperature programmed oxidation-CO_2 (TPO-CO_2), and scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM-EDX). The catalytic activities of the three catalysts are considerably different at low temperature, although they are similar at high temperature. At 700 °C, the average conversions of CH_4 and CO_2 over AC-$NaNO_3$ are 17.7% and 29.7%, respectively, which are 2.4 and 3.2 times higher than those over AC. At 1,000 °C, the conversions of CH_4 and CO_2 exceed 90% over these three catalysts. In addition, the mole ratio of H_2/CO increases with temperature for all the catalysts. The mole ratio of H_2/CO over AC-HNO_3, which is from 0.76 to 0.94 at different temperatures, is closer to 1 than that over the other two catalysts. Based on the characterization results, $NaNO_3$ is more instrumental to the formation of mesopores, surface oxygenated groups, and the reduction of deposited carbon. Hydroxyl group plays an important role in CH_4-CO_2 reforming. According to TPO-CO_2 observations, two types of deposited carbon are formed during reforming reaction. The apparent activation energies of the CH_4-CO_2 reforming reactions catalyzed by AC, AC-HNO_3, and AC-$NaNO_3$ are 202 kJ/mol, 227 kJ/mol, and 123 kJ/mol.

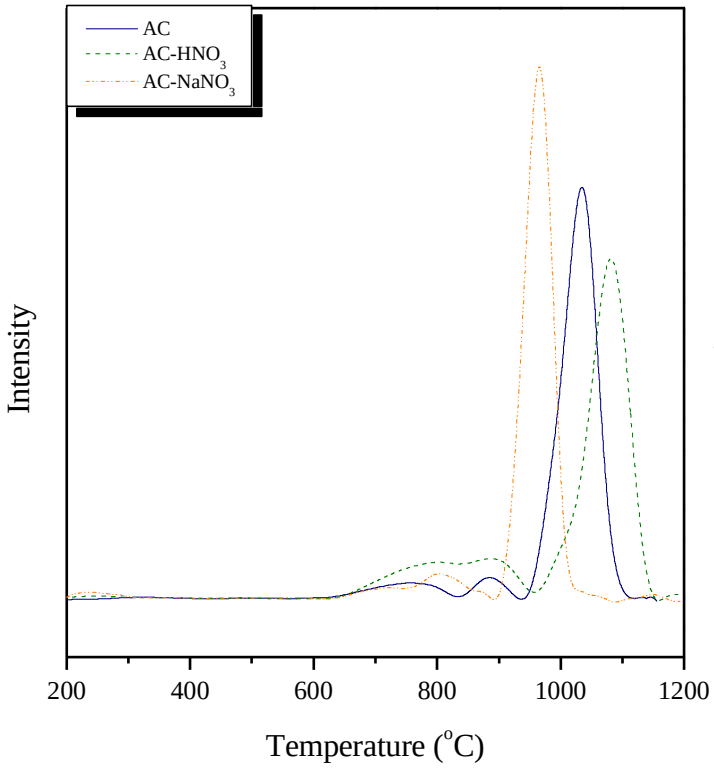

Figure 8. TPO-CO_2 profiles of the used catalysts [Catalyst weight: 0.3 g; Inlet flow rate: 20 ml/min; Inlet CH_4:CO_2 mole ratio: 1; Temperature: 900°C; Reaction time: 240 min]

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TPO-CO_2 of the used catalyst was done using the same TA along with a mass spectrometer (Hiden, HPR-20 QIC TMS). The used catalyst was heated in 100 ml/min CO_2 from room temperature to 1,200°C at the rate of 5°C/min, while the generation of CO was measured by the online MS. The electron ionization voltage of the MS was 0-220 eV.

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