

Catalytic CH₄ reforming with CO₂ over activated carbon based catalysts

The objective of this study is to compare the performance of several activated carbon (AC) based catalysts for CH₄ reforming with CO₂, including raw AC and AC modified with HNO₃ and NaNO₃, designated as AC-HNO₃ and AC-NaNO₃, respectively. CH₄-CO₂ 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, CO₂ chemisorption, temperature programmed oxidation-CO₂ (TPO-CO₂), 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 CH₄ and CO₂ over AC-NaNO₃ 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 CH₄ and CO₂ exceed 90% over these three catalysts. In addition, the mole ratio of H₂/CO increases with temperature for all the catalysts. The mole ratio of H₂/CO over AC-HNO₃, 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, NaNO₃ is more instrumental to the formation of mesopores, surface oxygenated groups, and the reduction of deposited carbon. Hydroxyl group plays an important role in CH₄-CO₂ reforming. According to TPO-CO₂ observations, two types of deposited carbon are formed during reforming reaction. The apparent activation energies of the CH₄-CO₂ reforming reactions catalyzed by AC, AC-HNO₃, and AC-NaNO₃ are 202 kJ/mol, 227 kJ/mol, and 123 kJ/mol.

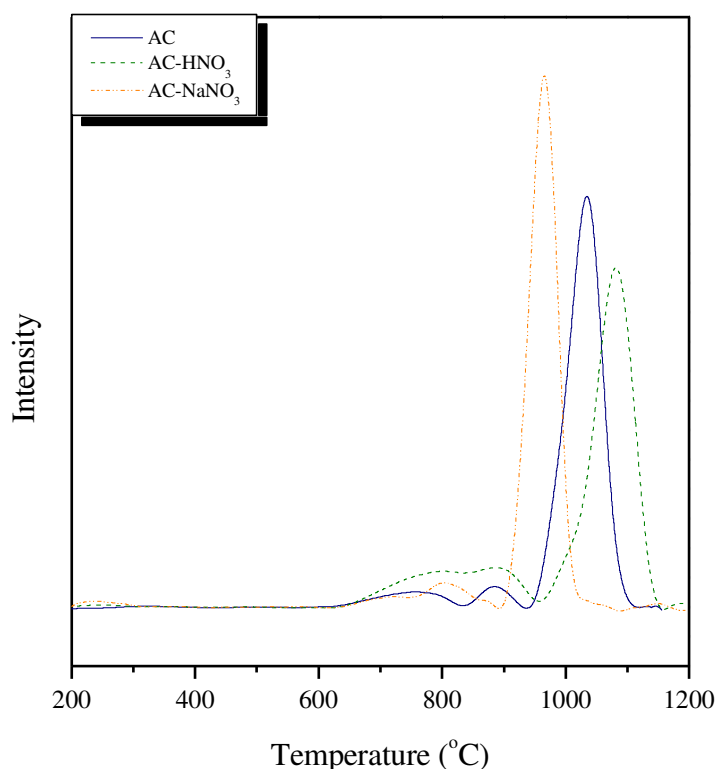


Figure 8. TPO-CO₂ profiles of the used catalysts [Catalyst weight: 0.3 g; Inlet flow rate: 20 ml/min; Inlet CH₄:CO₂ mole ratio: 1; Temperature: 900°C; Reaction time: 240 min]

TPO-CO₂ of the used catalyst was done using the same TA along with a mass spectrometer (Hidden, HPR-20 QIC TMS). The used catalyst was heated in 100 ml/min CO₂ 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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