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Gas phase hydroxylation of benzene to phenol by using CO_2 as an oxidant over Cu-exchanged ZSM-5 catalysts

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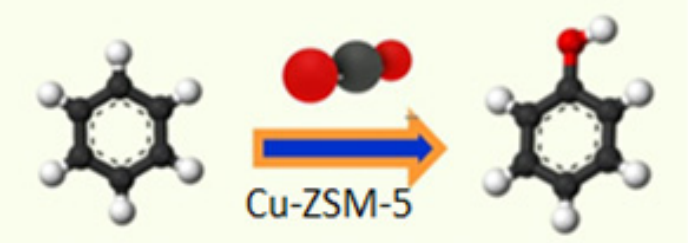
Direct hydroxylation of benzene is an assuring strategy to synthesize phenol. Formation of phenol was achieved under the continuous flow micro-reactor system by the concurrent feed of benzene and carbon dioxide over Cu loaded M+-ZSM-5 catalysts (M = Na^+, Cs^+, Mg^{2+} Ca^{2+} and Cr^{3+}). The formation of phenol was observed (9.5% at 450 ^{0}C) in the temperature range of 4250C–5250C with a co-feed of CO_2 (15 mL/min) and C6H6 (0.8 mL/min). When the catalyst was calcined at 450 ^{0}C, the activity peaked in the first hour and then slowly deactivated, but when the calcination temperature for the synthesized catalysts was increased to 7500C, the activity slowly increased until it reached a plateau. At high (750 ^{0}C) calcination temperatures, only a small fraction of the Cu undergoes the two-step reduction and most of the Cu remains in the +2 state. Weaker Bronsted acidity is helpful to obtain a higher phenol selectivity and less by-products. The obtained results indicated that the M+ assists in enriching surface active CO_2 in the form of carbonates and subsequently reacted with the homolytic activated C-H over Cu0-H-ZSM-5. The isolated square pyramidal Cu2+ species was suggested to be the active species for the phenol formation. The increase in the square-pyramidal Cu2+ species ensured when the CuH-ZSM-5 was calcined at higher temperatures and when HZSM-5 with lower Si/Al atomic ratios was used as a support.

Key words: CO_2 utilization, Cu-ZSM-5, hydroxylation, Nanoparticles



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