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Improved Benzene Selctivity for Methane Dehydroaromatization via Modifying the Zeolitic Pores by Dual Templating Approach

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Abstract : Catalytic transformation of simplest hydrocarbon methane into benzene and valuable chemicals over Mo/HZSM-5 has a great economic potential, however, it suffers serious hurdles due to the blockage in the micropores because of extensive coking at high temperature during methane dehydroaromatization (MDA). Under such conditions, it necessitates the design of micro/mesoporous ZSM-5, which has the advantages viz. uniform dispersibility of MoOx species, consequently the formation of active Mo sites in the micro/mesoporous channel and lower carbon deposition because of improved mass transfer rate within the hierarchical pores. In this study, we report a unique strategy to control the porous structures of ZSM-5 through a dual templating approach, utilizing C6 and C12 -surfactants as porogen. DFT studies were carried out to correlate the ZSM-5 framework development using the C6 and C12 surfactants with structure directing agent. The structural and morphological parameters of the synthesized ZSM-5 were explored in detail to determine the crystallinity, porosity, Si/Al ratio, particle shape, size, and acidic strength, which were further correlated with the physicochemical and catalytic properties of Mo modified HZSM-5 catalysts. After Mo incorporation, all the catalysts were tested for MDA reaction. From the activity test, it was observed that C6 surfactant-modified hierarchically porous Mo/HZSM-5(H) showed the highest benzene formation rate (1.5 µmol/gcat. s) and longer catalytic stability up to 270 min of reaction as compared to the conventional microporous Mo/HZSM-5(C). In contrary, C12 surfactant modified Mo/HZSM-5(D) is inferior towards MDA reaction (benzene formation rate: 0.5 µmol/gcat. s). We ascribed that the difference in MDA activity could be due to the hierarchically interconnected meso/microporous feature of Mo/HZSM-5(H) that precludes secondary reaction of coking from benzene and hence contributing substantial stability towards MDA reaction.

Keywords : hierarchical pores, Mo/HZSM-5, methane dehydroaromatization, coke deposition
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