

METHANE AROMATIZATION OVER 3 PERCENT Mo/HZSM-5 CATALYST INVESTIGATED BY TOF-MS UNDER SUPERSONIC JET EXPANSION CONDITION

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Methane dehydroaromatization over the 3%Mo/HZSM-5 catalyst under supersonic jet expansion condition was investigated via direct monitoring of the reactant and products using time-of-flight mass spectrometry. We found that unlike the results observed over the same catalyst at atmospheric pressure, naphthalene was the major product. The results collected during the induction period of CH₄ or ¹³CH₄ interaction with the catalyst at 700 °C revealed that the CH_x radicals produced from CH₄ decomposition reacted with the surface oxygen to form CHO radicals which dissociated into CO and H atoms. With the conversion of MoO₃ to Mo₂C, aromatic compounds such as naphthalene and toluene were generated at temperature as low as 380 °C. The formation of the C₄-C₁₀ compounds was related to the aromatization of C₃H₇ radicals on the Bronsted acid sites. The fresh and used catalysts were characterized by means of X-ray photoelectron spectroscopy and high-resolution transmission electron microscopy. Based on the results, a reaction mechanism of methane dehydroaromatization under supersonic jet expansion condition was proposed. The results confirmed that the step of methane dehydrogenation over Mo₂C/ZSM-5 is the only step that requires a temperature as high as 700 °C. The later steps of the aromatization of alkyl entities should be conducted at much lower temperatures so as to avoid the deep oxidation of the desired products. For such an aim, a condition similar to that of supersonic jet expansion should be adopted. The work described here was supported by a grant from the Research Grants Council of the Hong Kong Special Administrative Region, China (Project No. HKU 701507P).