Oxidative Coupling of Methane using Modified Na/W/Mn Catalysts

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Oxidative coupling of methane (OCM) is a useful method to produce methane, a major component of natural and shale gas, to more valuable chemical building blocks of coupled compounds including C_{2+} compounds of ethane, ethylene, propane, and propylene [1]. Because the C-H bond of methane is very stable, its activation using oxygen molecues to produce C_{2+} compounds avoiding combusted products of CO and CO_2 requires high reaction temperature with selective catalysts. While a lot of catalysts including alkali, alkali earth mtals, and their mixed oxides have been suggested for the OCM, supported Na_2WO_4/Mn catalysts exhibited the highly active and stable activity and frequently selected for the scaled-up reaction systems [2,3].

Although the activity of Na_2WO_4/Mn catalysts has been reported in the literature, its catalytic performance is not fully understood and the design of improved catalysts based on Na_2WO_4/Mn catalysts is complex [4]. In this study, we develop the modified Na_2WO_4/Mn catalysts and investigate the origin of its adjusted activity. Structures of catalysts were observed using XRD, XPS, FT-IR, Raman, and electron microscopy and the relations between structure and activity were discussed. The reaction conditions of catalysis were also observed and their effects on the product compositions were discussed. By adjusting process conditions, the catalytic activity was modified, which was further discussed to understand the descriptors of catalysis.

With the modification of catalysts, the highest C_2 yield of 26.0% was obtained for the nanosheet-supported Na_2WO_4/Mn catalyst (Fig. 1), which is higher than that of the conventional fumed-silica supported catalyst (17 - 18%) prepared in this study. In addition to the C_2 yield, selectivity to oxygen-free coupled hydrocarbons and conversion of methane were further discussed for the future development of more efficient OCM catalysts.

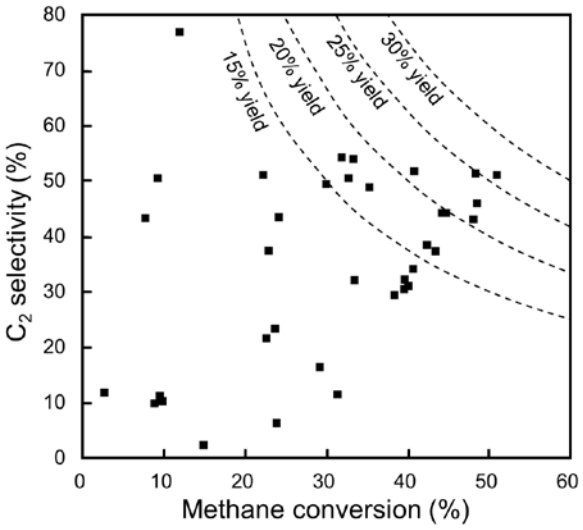

Fig. 1 OCM activity of catalysts at 800 °C. (GHSV = 10,000 h^{-1}, $(CH_4)/(O_2)$ = 3 (mol/mol))

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