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Optimization of methane conversion to liquid fuels over W-Cu/ZSM-5 catalysts by response surface methodology

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Abstract: The conversion of methane to liquid fuels is still in the development process. The modified HZSM-5 by loading with Tungsten (W) enhanced its heat resistant performance, and the high reaction temperature (800 °C) did not lead to the loss of W component by sublimation. The loading of ZSM-5 with Tungsten and Copper (Cu) resulted in an increment in the methane conversion, CO_2, and C_{5+} selectivities. The high methane conversion and C_{5+} selectivity, and low H_2O selectivity are obtained by using W/3.0Cu/ZSM-5. The optimization of methane conversion over 3.0 W/3.0Cu/ZSM-5 under different temperature and oxygen concentration using response surface methodology (RSM) are studied. The optimum point for methane conversion is 19% when temperature is 753 °C, and oxygen concentration is 12%. The highest C_{5+} selectivity is 27% when temperature is 751 °C, and oxygen concentration is 11%.

Key words: methane; W-Cu/ZSM-5; liquid hydrocarbons; response surface methodology

1. Introduction

The use of ZSM-5 zeolite as a support of the metal oxide phase is very interesting due to three reasons: its thermal stability, the high surface area that enables high metal oxide loading, and the presence of acid sites that could lead to the formation of certain active metal oxide species [1].

Cu loaded ZSM-5 catalyst *via* acidic ion exchange method has been identified to be the potential catalyst for conversion of methane to liquid fuels [2]. However, the infrared study of metal loaded ZSM-5 catalyst indicated that the catalysts are not resistant to high temperature. Previous studies have indicated that metal loaded ZSM-5 did not exhibit vibration band at 3610 cm^{-1} and 3660 cm^{-1}, except for ZSM-5 which showed a weak vibration band at 3666 cm^{-1}. The result suggested that the framework and nonframework aluminum were either extracted to acidic solution or became silanol defect form when calcined at 800 °C and made the catalysts inactive [3]. Previous studies [4,5] indicated that the Cu loaded W/ZSM-5 catalyst was thermally stable at the reaction temperature (700−800 °C).

In our previous study [5], it was reported that the loading of HZSM-5 with tungsten and copper decreased the crystallinity, surface area, and also total volume of the catalysts. However, the average pore diameter and the acidity of the zeolites increased as a result of the modification with the metals. The modified ZSM-5 by loading with Tungsten enhanced its heat resistant performance and the high reaction temperature (800 °C) did not lead to loss of W component by sublimation. The process of converting methane to liquid hydrocarbons (C_{5+}) is dependent on the metal surface area and the acidity of the zeolite. The W/3.0Cu/ZSM-5 is the potential catalyst, because over this catalyst high methane conversion and C_{5+} selectivity, and low H_2O selectivity are obtained.

Amin and Anggoro [5] studied the optimization of Cu loaded W/ZSM-5 using the Response Surface Methodology. The low, middle, and high levels of all the independent variables were W content in weight doped into the 3.0Cu/W/ZSM-5, O_2 concentration, and flow rate of feed gases. This article reports that the optimizations of W loaded Cu/ZSM-5 with the independent variables were temperature and oxygen concentration.

Response surface methodology (RSM) is a method to determine the optimum condition of a process. RSM has similarity with regression analysis. In regression analysis, empirical mathematical model are derived from the experiment

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