CHAPTER V
THE EFFECT OF COPPER CONTENT ION-EXCHANGED ON MFI TYPE ZEOLITE ON NITRIC OXIDE CONVERSION

5.1 Introduction

The abatement of nitric oxide emissions from lean exhaust streams produced by stationary and mobile sources has been of interest. It is well known that Cu/MFI zeolites have been promoted as potential catalysts for both direct decomposition and selective catalytic reduction of nitric oxide. These kinds of catalysts exhibit high NO conversion in the presence of oxygen using a little hydrocarbon amount as reducing agents. The purpose of this chapter is to investigate the copper loading on NO SCR using n-octane as reducing agent in a wide range of reaction temperatures.

5.2 Experimental

5.2.1 Catalyst preparation

Experiments on several Cu/MFI catalysts with different copper levels (0-200% ion-exchange of copper) were carried out in order to analyze the effect of copper content on NO conversion as a function of temperature. The amount of metal compositions on Cu/MFI catalysts are shown in table 5.1. In this study we set the lower ion-exchange of copper on MFI zeolite as 50% ion-exchange of copper on MFI, 100% ion-exchange of copper on MFI and over ion-exchange of copper on MFI as 200% ion-exchange of copper on MFI by using the assumption that one Cu^{2+} is exchanged with two H^+ or Na^+ ions.

5.2.2 Reaction test

The catalyst as powder form was tabletted, crushed, and sieved into 12 - 22 mesh to reduce the pressure drop. In order to investigate the activity for NO removal, catalyst was heated in a He stream from room temperature to 600°C and maintained at 600°C for 30 min. After that, a feed gas composed of 1,000 ppm NO, 1,000 ppm n-octane, 2mol% O_2, and 0-10mol% H_2O balanced with He was introduced to flow with a GHSV of 30,000 h^{-1}. Every 10 min after the feed gas was introduced to ensure the steady state of catalytic activity, the reactants and products were analyzed by gas chromatographs (Chrompack, Micro GC CP 2002 with MS-5A and porapak Q column) equipped with integrators. The reaction test was undertaken every 50°C

Table 5.1 Main characteristics of copper ion-exchanged MFI catalysts

Catalyst	Abridged notation	Si/Al	Cu content (%wt)	Cu/Al	Cu exchange level (%)
50%ion-exchanged copper on MFI	50%Cu/MFI	50	0.57	0.25	50
100%ion-exchanged copper on MFI	100%Cu/MFI	50	1.40	0.44	100
200%ion-exchanged copper on MFI	200%Cu/MFI	50	2.89	1.03	205

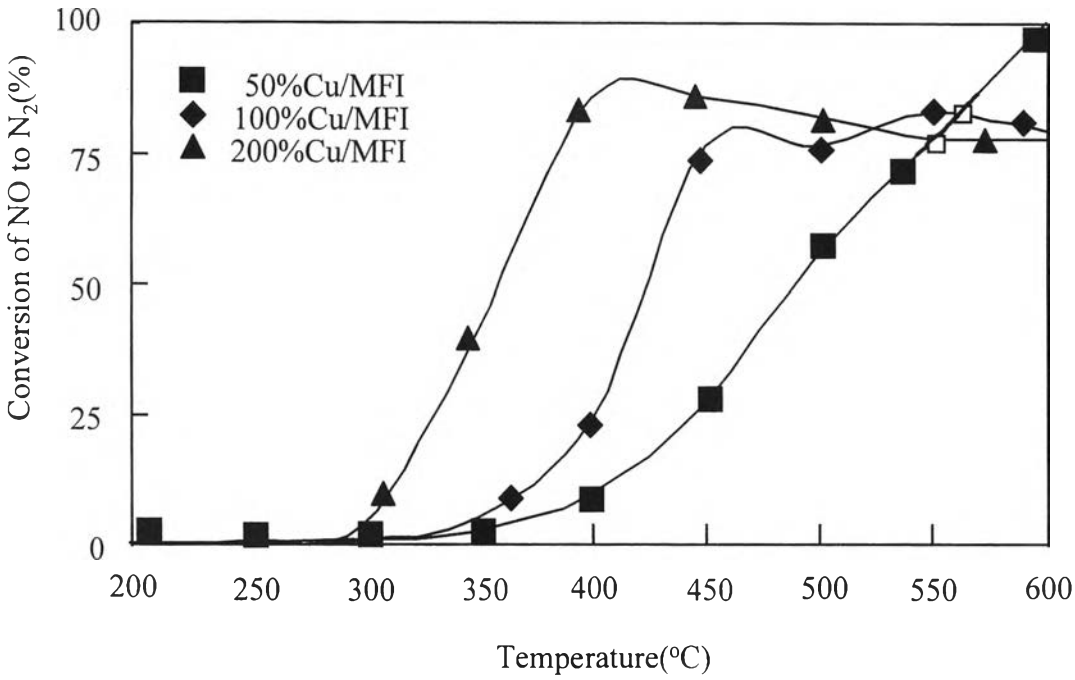

Figure 5.1 Conversion of NO of Cu/MFIs at various ion-exchange level of copper
Feed gas: NO 1,000 ppm, n-octane 1,000 ppm, O_2 2mol%, He balance, GHSV 30,000 h^{-1}

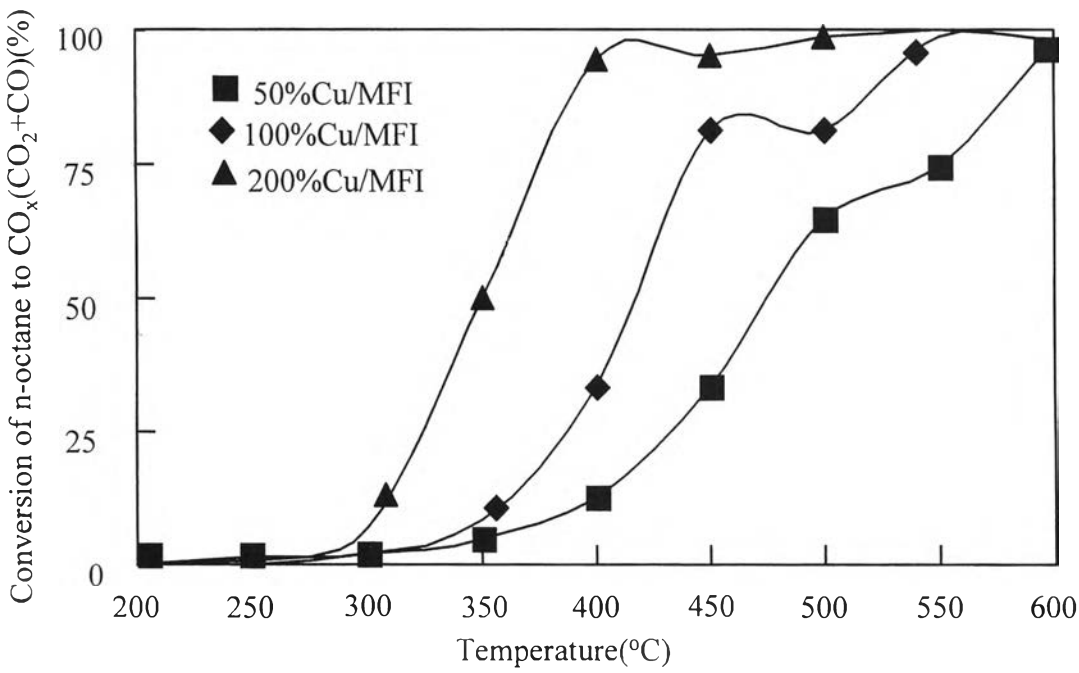

Figure 5.2 Conversion of n-octane Cu/MFIs at various ion-exchange level of copper
Feed gas: NO 1,000 ppm, n-octane 1,000 ppm, O_2 2mol%, He balance, GHSV 30,000 h^{-1}

diminishing from 600°C to 200°C followed the same procedure as mentioned above. The catalytic activity of nitric oxide reduction and n-octane combustion were investigated as the amount of N_2 and carbon oxides(CO_x; CO_2+CO) produced, respectively.

5.3 Results and discussion

The activities of Cu/MFI catalysts for NO and n-octane conversions are depicted in Figures 5.1 and 5.2, respectively. As shown in Figures 5.1 and 5.2, it was found that the copper content on MFI catalyst affects the activities of both NO and n-octane conversions. The more copper content on MFI catalysts, the lower the temperature of maximum NO and n-octane conversions;

50%Cu/MFI (Tmax for NO conversion is about 600°C, Tmax for n-octane conversion is 600°C) > 100%Cu/MFI(550°C, 550°C) > 200%Cu/MFI(400°C, 400°C).

Therefore, it can be concluded that the catalyst with higher copper content exhibited the higher catalytic activity. This appearance is similar to the study of Abreu et al. [115]. Using a H_2 TPR study, the authors suggested that the higher the copper loading promoted easier reducibility of copper species shifting the maximum conversion of NO. Cheung et al. [73] also supported the idea of advantage of overexchange Cu/MFI catalyst. Due to the higher activity for NO conversion 200%Cu/MFI exhibited, it is worth to take into account the activity of 200%Cu/MFI catalyst in the further study.

5.4 Conclusion

The copper loading on MFI catalyst has an influence on nitric oxide removal. The larger amount of copper content loaded, the higher activity for NO removal of Cu/MFIs. The light-off temperatures for both conversions of NO and n-octane of higher amount copper ion-exchanged on MFI catalyst were shifted to lower reaction temperature condition.