CHAPTER I

INTRODUCTION

The increasing volumes of air pollutants such as carbon monoxide (CO), hydrocarbons (HC), nitrogen oxides (NO_x), sulphur dioxide (SO_2), particles, and soot have become a serious global problem that must be solved as soon as possible. Beside these emissions, the increasing carbon dioxide (CO_2) concentration in the atmosphere and its contribution to the greenhouse effect is also of general concern. The main contribution to the man-made CO_2 emission is due to combustion process in power plants, industries, and transportation systems.

The world demands greater efficiences in transportation but without the harmful effect of increased pollution. The gasoline fueled automobile is a great luxury for the average citizen of the world, but it is a large fossil fuel consumer, and it directly generates enormous amount of emissions. Therefore, the development of internal combustion engines with improved fuel economy is of social interest. The three-way catalytic converter (TWC) [1] is efficient in the simultaneous reduction of CO, HC, and NO_x. However, it requires an exhaust gas composition operating very close to the stoichiometric point, i.e. the point that there is sufficient but not excess O_2 for reacting with CO and HC. The operation of the gasoline engine with a lean air-to-fuel ratio results in a decrease in fuel consumption, and less generation of CO_2, but the TWC cannot reduce NO_x in this condition.

The most attractive method for the removal of NO_x is the catalytic decomposition of NO [2-4], because no reducing agent is required. Because NO is thermodynamically unstable relative to N_2 and O_2 [5], this method is feasible. However, no suitable catalyst with a significant activity in real exhaust gas has been identified. It is widely accepted that the poisoning of the catalyst surface by oxygen from the exhaust gas or from the decomposition itself is the main problem. Therefore, up to now the only way to reduce NO_x emission in lean exhaust gas is the injection of reducing agents into the exhaust gas stream [6, 9-12].

For a power plant or a stationary diesel engine, the selective catalytic reduction of NO by injection of ammonia into the exhaust gas is well known [6-8]. However, the use of NH_3 brings disadvantages such as high costs of facilities, an offensive odour during running, and leakage of unreacted dangerous ammonia. For these reasons, the use of ammonia in this process is unsuitable for small scale oxygen-rich NO emissions such as diesel engine exhaust [9]. Thus, an effective NO reduction process with an affordable, non-ammonia reductant is desirable.

Recently, the reduction of nitric oxide with hydrocarbons has also drawn attention as a process for the catalytic removal NO in the exhaust gas of diesel or lean-burn gasoline engines [10-17]. Some studies on the NO decomposition in the presence of excess oxygen and a low concentration of hydrocarbons were reported using microporous crystalline metallosilicate such as H-ZSM-5 [10], Cu/ZSM-5 [11], Cu-incorporated zeolite [12], some metallosilicate [13], and metal ion-exchanged zeolite [14-18]. Among the

catalyts which have been reported, Cu-ion exchanged ZSM-5 (Cu/ZSM-5) is potentially the most effective catalyst for the reduction of NO with hydrocarbons [19-21]. However, Cu/ZSM-5 showed decrease NO conversion when the operating temperature was more than 300 °C. Thus, there is a need to develop a new catalyst which can show a wider temperature window for nitric oxide reduction than Cu/ZSM-5 by hydrocarbon in oxidizing atmosphere.

1.1 The Objectives of This Study

1.1.1 To study the method of preparation of Copper Aluminosilicate (Cu,Al-silicate) and Copper ion-exchanged Copper Aluminosilicate (Cu/Cu,Al-silicate) Catalysts.

1.1.2 To characterize the prepared catalysts.

1.1.3 To study the catalytic reduction of nitric oxide in the presence of excess oxygen over the prepared catalysts with low concentration of propane.

1.1.4 To compare the catalytic activities of the prepared catalysts with Cu/Na-ZSM-5 for the selective reduction of nitric oxide by propane in excess oxygen atmosphere.

1.2 The Scope of This Study

1.2.1 Prepare of bimetallosilicate (Cu,Al-silicate) by adding the metal sources at the stage of gel formation and synthesizing the bimetallosilicate according to the rapid crystallization method. The source of metals are aluminium chloride ($AlCl_3$), and copper nitrate ($Cu(NO_3)_2$). The metal elements were incorporated in the structure. Cu/Cu,Al-silicate were prepared by using an

ion exchange method. In addition, coated Cu/Cu,Al-silicate on monolith structure were prepared.

1.2.2 Characterize the prepared catalysts by the following methods :

- Identifying the synthesized crystals by X-ray diffractometer.

- Analyzing the surface areas and pore size distribution of the catalysts by the techniques of BET Surface Areas Measurement and N_2 adsorption.

- Analyzing the percentage of metal loading of the catalysts by Atomic Absorption Spectrometer (AAS).

1.2.3 Studying the catalytic activities of the prepared catalysts for the selective reduction of NO in the presence of excess oxygen with low concentration of propane under the following conditions:

- Reactant : NO 1000 ppm, C_3H_8 3000 ppm, O_2 10%, and He as a balance gas

- Reaction temperatures : 150-600 $^{\circ}$C

- Space velocity : 4000 h^{-1}