CHAPTER I

INTRODUCTION

Air pollution is a very important social problem that must be solved as soon as possible. The problems arising from vehicle air pollution vary widely throughout the world, depending on vehicle population density, climatic and geographical conditions. Therefore, different emission standards are imposed in various countries. The notorious air conditions in California and Japan have prompted very strict controls. However, the legislation on vehicles in Japan is closely following the lead set by the USA in the early 1970s and will probably develop in the same manner [1]. Currently, the environmental crisis announced in the 1970s has returned more forcefully. Increasing concerns about air quality and the environment have led to tougher standards in many countries throughout world in the control of exhaust emission on mobile and stationary engines, and even more stringent regulations are expected to be introduced in the near furture [2].

The exhaust gases from engines of vehicles contain mainly carbon oxides (CO and CO_2), nitrogen oxides (NO_X), hydrocarbons, sulfur dioxide, particles and soot. At present, one of the highly significant problems is removal of NO_X, which is produced during high temperature combustion [3]. In particular, the removal of nitrogen monoxide (NO) is a dominant target to be achieved because NO is the major component of NO_X in the exhaust gases [4]. Since NO is thermodynamically unstable relative to N_2 and O_2 at low temperature, its catalytic decomposition is the simplest method for NO removal from exhaust

streams. Two main catalytic technologies are currently available to decrease the problem. One is the three-way catalyst system [5] and the other is the selective catalytic reduction process with ammonia [6]. Of the two processes, only the latter can be used to remove NO in oxygen-rich atmospheres. However, the use of ammonia makes this process unsuitable for small scale oxygen-rich NO emissions such as diesel engine exhausts [7] and inherently has many problems, e.g. ammonia slip (another air pollutant), equipment corrosion, and high equipment costs. Therefore, an effective NO reduction process with an affordable, non-ammonia reductant is desirable.

Recently, selective catalytic reduction of nitric oxide with hydrocarbons has received great attention both from an academic and an industrial point of view because this may offer an effective way of removing nitric oxides even in the presence of a large excess of oxygen, whereas present automotive catalysts do not work effectively under these conditions. The use of hydrocarbons as reducing agents for NO has been extensively studied on Al_2O_3 [8], SiO_2-Al_2O_3 [9,10], metallosilicate [11], silicoaluminophosphate (SAPO) [12], and metal ion-exchanged zeolite [13-18].

ZSM-5 zeolites having MFI type structure (IUPAC name) are useful catalysts in chemical reactions, because of their high activity and shape selectivity. They have two kinds of very fine micropores inside their crystallites: straight and zig-zag pores. Acid sites are different depending on the kind of chemical reaction. For the selective reduction of NO, Held et al. [19] and Iwamoto [20] were the first who reported that the catalytic activity of Cu-ZSM-5 could be greatly enhanced with the presence of small amounts of hydrocarbon in an excess oxygen.

1.1 The Objectives of This Study

1.1.1 To study the method of preparation of ion-exchanged zeolite and copper silicate catalysts.

1.1.2 To study the characterization of 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 copper ion-exchanged zeolite for the selective reduction of nitric oxide by different hydrocarbons in oxygen-rich atmospheres.

1.2 The Scope of This Study

1.2.1 Preparing of metallosilicates by rapid crystallization method. Atomic ratio of Si/metal are set at Si/Al = 20, 50, 3200, and ∞ and Si/Cu = 50, respectively. In addition, H-ZSM-5 (H-Al-silicate) and Cu/Na-ZSM-5 were prepared by using ion exchange method.

1.2.2 Characterizing of the prepared catalysts by the following methods.

- Identifying the synthesis 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 shape and size of the crystals by Scanning Electron Microscope (SEM).

- Analyzing the structure details of catalysts by FT-IR spectroscope.

- 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 ^{o}C

- Space velocity : 4000 h^{-1}

1.2.4 Comparing the catalytic activities for the selective reduction of NO by various hydrocarbons in oxidizing atmosphere on Cu/Na-ZSM-5.