CHAPTER I
INTRODUCTION

1.1 Introduction

Increasing awareness of the need to reduce air pollution has led to development of environmental technology. Nitrogen oxides are important pollutants, which contribute largely to variety of environmental problems: the formation of acid rain, smog, and ground-level ozone. Furthermore, they cause lung infection and respiratory system allergy in human. From these reasons, nitrogen oxide emissions have to be controlled.

Nitrogen oxides consist of several compounds: Nitric oxide (NO), Nitrogen dioxide (NO_2), Nitrous oxide (N_2O), Nitrogen sesquioxide (N_2O_3), Nitrogen tetraoxide (N_2O_4) and Nitrogen pentoxide (N_2O_5). The first two gases are known as " **NO_x**". They are significantly concerned because of large amount emission. Especially, NO is released around 95% of all nitrogen oxides emission.

The major source of NO_x emission is fossil fuel combustion such as in power plant and automobile engine. Furthermore, chemical operations such as nitric acid producing plant, and natural producing can also originate NO_x emission.

When the exhaust gas from engine contains a mixture of oxygen and the pollutants, which are hydrocarbons, carbon monoxide and nitrogen oxides, close to the stoichiometric air/fuel ratio (14.6) operating conditions, as in case for most gasoline-fueled cars today, the pollutants can be transformed simultaneously to water, carbon dioxide and dinitrogen using three-way catalysts (TWC).

From viewpoint of environmental protection, there have been increasing demands for cleaner exhaust and better fuel economy. A gasoline lean-burn and diesel engine (air/fuel ratio $\approx$ 22.4) are the effective technologies for suppressing the fuel consumption. Depending on driving conditions, a lean-burn engine can decrease fuel consumption by up to 30% compared with a stoichiometric engine. However, the lean conditions operated at high amount of air or oxygen have been limited because NO_x emitted under oxidizing conditions could not be purified using conventional three-way catalysts.

The most appealing method to remove NO_x is the decomposition of NO because it needs no reducing agent. Theoretically, NO is thermodynamically unstable, so, it should be decomposed to dinitrogen and oxygen as soon as it is formed. Nevertheless, this decomposition reaction is inhibited by high activation energy. Thus, a catalyst must be used to decrease this activation energy. However, despite intensive research on catalytic decomposition of NO, no suitable catalyst with a significant activity in real exhaust gas has been identified. The main problem is the poisoning of catalyst surface by oxygen from the decomposition itself.

The selective catalytic reduction (SCR) of NO_x is an alternative way to reduce NO_x emission. In order to convert NO_x to dinitrogen, reducing agents are used to react with NO_x. Carbon monoxide, hydrogen, ammonia and hydrocarbons can be used as reducing agents. Using hydrocarbons is a reasonable target, because a certain amount (100-1000 ppm) of ethene and propene is usually contained in the exhaust gases of lean-burn gasoline and diesel engine.

$$NO_x + O_2 + C_2H_4 \text{ or } C_3H_6 \longrightarrow N_2 + CO_2 + H_2O$$

Although selective catalytic reduction with hydrocarbons is a promising technique and it has been studied in recent years, there remain several problems to be solved such as catalyst deactivation, narrow operation temperature window, etc. Thus, one of the most interesting approaches presently for NO_x removal in excess oxygen is to use a so-called **NO_x Storage-Reduction (NSR)**. The NSR catalyst is used in an engine, which operated under lean (high air/fuel ratio), and rich (low air/fuel ratio) conditions. During lean operation, the NO_x in exhaust are stored in the catalyst. As the NO_x storage capacity of the catalyst becomes saturated, it is necessary to regenerate the catalyst by switching the engine to rich condition for a short period. During rich period, stored NO_x is released and subsequently reduced to dinitrogen. The Japanese 10-15 mode regulated emission test using a passenger car equipped with a lean-burn engine revealed that the NO_x conversion to dinitogen of NO_x storage-reduction technique operated under the dynamic oxidizing conditions was higher than the conversion under the static oxidizing conditions (Takahashi *et al.*, 1996).

To handle the exhaust gases from lean-burn gasoline engines and diesel engines for practical applications, a catalyst should be operated under an excess of oxygen and the moisture. Therefore, this work will investigate the catalytic activities of gold supported on alumina catalyst and barium-containing in gold/alumina catalyst under oxygen and moisture conditions.

1.2 Research Objectives

The objectives of this study were to test the lean De-NO_x activity of Au/Al_2O_3 and $Au/Ba/Al_2O_3$ catalysts by studying the effect of :

- preparation method
- % metal loading
- water vapor content
- impregnation sequence, and
- pretreatment gas.