

CHAPTER I

INTRODUCTION



1.1 Introduction

Public concern with environmental pollution has grown phenomenally in recent years. The increase in atmospheric pollution experienced in industrialized countries is inevitably associated with the Industrial Revolution and the associated growth of cities. Especially, Bangkok is one of the cities that encounters serious pollution problems from exhaust gases nowadays.

The exhaust gases from automobile engines and other industrial sources contain mainly carbon oxides (CO and CO₂), nitrogen oxides (NO_x), unburned hydrocarbons, sulfur oxides (SO_x), particles, and soot (Iwamoto and Hamada, 1991).

At present, one of the most significant problems is removal of NO_x, which are produced during high-temperature combustion and are an important group of air contaminants.

Nitrogen oxides (NO_x) include six known gaseous compounds: Nitric oxide (NO), Nitrogen dioxide (NO₂), Nitrous oxide (N₂O), Nitrogen sesquioxide (N₂O₃), Nitrogen tetroxide (N₂O₄), and Nitrogen pentoxide (N₂O₅) (Howard *et al.*, 1985). The first two oxides of nitrogen are the primary concern in air pollution because they are emitted in large quantities into the atmosphere, especially NO which is emitted in much larger quantities than NO₂. It accounts for 95% of all nitrogen oxide emissions.

NO is the key starting material for all of the other oxides of nitrogen. Once produced, it is rapidly oxidized by ozone and by reaction with radicals such as OH^* and HO_2 is transformed into NO_2 , HNO_2 , and HO_2NO_2 (Armor, 1994). NO plays a major role in the photochemistry of the troposphere and the stratosphere. It reacts with photochemical pollutants such as formaldehyde, organic hydroperoxide, and peroxyacyl nitrates that all are very reactive and have a very short lifetime. These are very fast reaction, which generate more nitrogen oxides and organic nitrates (Parvulescu *et al.*, 1998). Thus, if NO is prevented from entering to atmosphere, most of down stream effects can be eliminated.

NO is formed in combustion process by a reaction between the nitrogen and oxygen present in the air. At temperatures greater than $1,500^\circ\text{C}$, this reaction proceeds at appreciable rates. The major source of nitrogen oxides is the combustion of fossil fuels such as coke in electrical power plants or petroleum in the engines of vehicles and aeroplanes. NO_x emissions also can be significant in chemical operations such as nitric acid plants. More recently, the emissions of nitrous oxides from fiber production plants has received attention because of its global warming effects. The nitric oxide and sulfur oxides emissions are components of acid rain since, when mixed with water vapor in the clouds, they form nitric and sulfuric acid, respectively. Furthermore, NO_x particles in photochemical ozone or smog can be generated by reaction with hydrocarbons (Ronald and Farrauto, 1995).

Regarding the effects on human health, nitric oxide is a relatively inert gas and only moderately toxic. Like CO, NO can combine with hemoglobin to reduce the oxygen carrying capacity of blood (Howard *et al.*, 1985). In the presence of air NO is rapidly oxidized to NO_2 . This reaction is further accelerated in the presence of sunlight and hydrocarbons from unburned gasoline. This oxide is poisonous for the respiratory system, provoking both

lung infection and respiration allergies since it is able to diffuse through the alveolar cells and capillary vessels of the lungs and damage their structure through their propensity towards oxidation. There are studies that reveals the concentrations of nitrogen oxides having hazardous effects for people in good health are above 0.05 ppm for an exposure of over 24 hour (Pitchon and Fritz, 1997). Consequently, NO_x have to be reduced to nitrogen to render them harmless.

Due to the severe regulations, the motor car manufacturers have had to decrease considerably the NO_x emissions from their vehicles. Even though diesel engines have the advantage of lower consumption of fuel and of lower emissions of carbon dioxide, carbon monoxide, and hydrocarbons than usual gasoline engines (Niwa *et al.*, 1998) they emit more NO_x than cars equipped with three way catalysts because three way catalysts do not work with diesel engines.

Some improvements have been made in cleaning up the emissions of diesel engines through modification of the combustion chamber and cleaning up of the diesel fuel. More recently, these measures have proved less than sufficient. So, to prevent pollution, an exhaust after treatment for diesels has become necessary. By the end of this century, the governments of the US, Europe, Japan, and also Thailand will strengthen the legislation to reduce the amount of emitted NO_x , hydrocarbons, and CO from the car. Therefore, NO_x elimination technology applicable to the diesel and lean burned gasoline engines has to be developed.

As prescribed, NO is thermodynamically unstable relative to N_2 and O_2 at low temperature. Theoretically, it should decompose as soon as it is formed. Therefore its decomposition is the simplest and cheapest method for the removal of NO from the exhaust streams. However, the decomposition reaction is inhibited by high activation energy of 364 kJ/mol (Pitchon and

Fritz, 1997). So a catalyst is necessary to lower this activation energy in order to facilitate this decomposition. Nevertheless, no suitable catalyst of consistently high activity has been found in the presence of oxygen. Thus, catalytic reduction processes are being looked upon as the second best method for removing NO, although catalytic decomposition is essentially the best approach (Iwamoto and Hamada, 1991).

The selective catalytic reduction of nitrogen oxides by hydrocarbons has attracted much attention recently because it provides the potential ability to remove nitrogen oxides from diesel exhaust and other oxygen rich flue gases. From the previous work (Lorpongpaiboon, 1998), the catalytic reduction of NO with propene in an oxidizing atmosphere was investigated at various silver and gold loadings on alumina (Al_2O_3). The sol-gel silver supported on alumina catalyst showed the highest activity compared to the conventional coprecipitation method. Therefore, it is necessary to investigate further for understanding the catalytic activity of these sol-gel catalysts.

1.2 Research Objectives

The objectives of this study were:

- i) To clarify the effect of silver loading on the catalytic activity of the sol-gel Ag/Al₂O₃ catalysts.
- ii) To study the combination of the Ag/Al₂O₃ catalyst mixed physically with manganese oxide catalyst.
- iii) To study the promoting effect of oxygen on the Ag/Al₂O₃ catalyst.