

CHAPTER I

INTRODUCTION

A major source of nitric oxide emissions is fuel combustion in engines and power plants. NO_x emission also can be significant in chemical operations such as nitric acid plants. Nitric oxide (NO) is the key starting material for all of the other nitrogen oxides. NO is a major atmospheric pollutant because NO can react with photochemical pollutants such as ozone, formaldehyde, organic hydroperoxides, and to generate more NO_x and organic nitrates. Therefore, if NO is prevented from entering into the atmosphere, most downstream effects can be eliminated.

The control technology of NO is divided into 4 groups.

1. The selective catalytic reduction of NO with ammonia

This method is used in many chemical plants and power plants, but ammonia is an expensive reductant and, within certain limits, is a pollutant itself. Therefore, there is interest in using, cheaper and less polluting reductants.

2. The selective catalytic reduction of NO in the presence of CO and/or H_2

This method is used for transportation vehicles which have gasoline engines. The reduction of NO in the presence of CO and/or H_2 is another intensive research area.

3. The selective catalytic reduction of NO in the presence of hydrocarbons

This method is used for automotive pollution control and in various industrial plants. The main advantage of the corresponding reaction is the use of a gas mixture very similar to that found in exhausts.

4. The direct NO decomposition

This method eliminates the need for reductants, which eliminates the addition pollution associated with the other three methods.

The selective catalytic reduction of NO by hydrocarbons in the presence of large amounts of oxygen has attracted much attention recently because it has the potential ability to remove NO from diesel exhaust gases. This method has not yet reached industrial use but can be applied both for automotive pollution control and in industrial plants. In this project, propylene was used as the reducing agent.

From previous work (Lorpongpaiboon, 1998), the catalytic reduction of NO in an oxidizing atmosphere was investigated at various silver loadings on alumina (Al_2O_3). He found that the sol-gel silver supported on alumina catalyst showed the highest activity compared to the conventional coprecipitation method. The sol-gel process is one of the most useful processes to make catalytic materials because it involves the use of molecular precursors, mainly alkoxides, as the starting materials. The incorporation of an active metal in the sol during the gelation step allows the metal to have a direct interaction with the support, therefore the catalysts made through this method show special catalytic properties.

The lean-DeNO_x process, in which NO_x are reduced by hydrocarbons in oxidizing conditions, has become of great interest in the last 10 years, as it has opened the possibility of controlling the NO_x emissions from diesel engines. Since 1990 when the groups of Held *et al.* (1990) and Iwamoto *et al.* (1990) described the activity of Cu-ZSM5 in this reaction, various other systems have been shown to be active for this process by Amiridis *et al.* (1996).

From the two main groups of materials-zeolites and metal oxides-that have been used as supports, the majority of works that deal with oxides have used alumina based catalysts. Alumina is a major component of these catalytic

materials because it disperses the metals which are used as active centers in a very broad range of temperatures very well. Alumina can resist sintering up to temperatures of 1200°C for catalytic applications. Because alumina is deactivated in the presence of sulfur dioxide by sulfate formation, while titania (TiO₂) is hardly sulfatized (Matsuda *et al.*, 1982). Therefore alumina and titania were used as support in the catalysts presented in this work. These two oxides have been used as support for NO_x reduction in lean burn conditions, as powder.

Recently, several publications have appeared which report the enhancement of NO reduction activity by combination of two active catalytic species, because multi-stage systems are effective to broaden the temperature window for the reduction of NO .

The aim of the present work has been, firstly, to test the lean-DeNO_x activity of Ag/Al₂O₃, Pt/Al₂O₃, Ag/TiO₂, and Pt/TiO₂ catalysts by using propylene as reductant. Secondly, to develop with these catalysts a two-stage system active in a wide range of operating temperatures, studying the influence of the water vapor and operating condition on the performance of the system.