CHAPTER I
INTRODUCTION

Carbon monoxide is a gas produced from incomplete combustion of fossil fuels as in automobile engines and from atmospheric chemical processes, mostly the oxidation of hydrocarbons. At sufficiently high concentrations, it is deadly. At lower concentrations, it can be health hazard and causes air pollution. The major carbon monoxide source is automobiles when ignited in fuel rich conditions. From the perspective of environmental concern, there have been increasing demands for cleaner exhaust. Many techniques are applied for carbon monoxide removal. Among these techniques, catalytic oxidation is mostly used. The highest conversion of carbon monoxide is, indeed, attained over a three-way-catalyst (TWC) which converts exhaust to water, carbon dioxide, and dinitrogen.

CeO_2 is widely used as automotive catalysts because the oxygen storage capacity (OSC) of CeO_2 results in its ability to undergo rapid reduction and oxidation cycles. The OSC of added CeO_2 acts as an oxygen partial pressure regulator. In pure CeO_2 system, the redox process is essentially limited to the reaction temperature. One way to improve its property is the addition of other oxides, and ZrO_2 was discovered to be the best doping material.

Cerium (IV) ammonium nitrate, cerium acetate, zirconium nitrate, zirconium oxychloride, cerium alkoxides and zirconium alkoxides can be used as starting materials to prepare CeO_2-ZrO_2 mixed oxide catalysts. However, commercial cerium alkoxides and zirconium alkoxides are expensive and more sensitive to water than other precursors. There is a new route to synthesize zirconium and cerium glycolate complexes, which can be used as alkoxide precursors, by the Oxide One Pot Synthesis (OOPS) process (Ksapabutr et al., to be submitted). Not only starting materials are diversed, but also preparation methods are varied. There are many methods for preparing CeO_2-ZrO_2 mixed oxide catalysts. Among these techniques, sol-gel technique is considered to be a very practical method, since it produces high purity, homogeneity, well-controlled properties and low temperature processing of the product.

Since many factors affect the catalytic properties, that means a large numbers of catalyst formulations need to be tested. If conventional testing methods are applied, it will take long time to discover desired catalysts. Consequently, the development of a system allowing the activity determination of a large number of catalyst formulations at the same time is required. Combinatorial catalysis and high throughput methods become more and more applicable nowadays.

Senken (2001) defined combinatorial catalysis as a methodology or a set of tools where large diversities of solid state material libraries are prepared, processed and tested for activity and selectivity in high throughput fashion.

The essential components of combinatorial catalyst are:

- Rapid library synthesis,
- High throughput testing, and
- Large-scale information management.

For examples, high throughput synthesis and screening of mixed metal oxide libraries for ethane oxidative dehydrogenation to ethylene on V-Al-Nb and Cr-Al-Nb oxide catalysts have been developed (Liu *et al.*, 2000). A 144-member catalyst library can be screened in slightly more than 2 hours. Furthermore, researchers attempted to develop the low-temperature catalysts for the oxidation of low-concentration propane (Buyevskaya *et. al.*, 2001). A 15-sample can be tested simultaneously. Most of the final materials converted propane to CO_2 at 150°C and the best ones oxidized propane even at as low as 50°C.

Some examples of successful catalyst discoveries from using high throughput screening techniques are the evaluation of Mo-V-Nb catalysts for oxidative dehydrogenation of ethane (Cong *et al.*, 1999), and the discovery of Pt-Ru-Os-Ir for reforming of methanol (Reddington *et al.*, 1998).

For testing the activity of a large number of catalytic formulations, optical techniques are mostly employed. Among optical techniques, IR thermography is interesting and used to investigate for its potential.

Infrared technique was firstly developed for military purposes. The ability of infrared is the detection the heat generated and illustration of temperature differences from objects. This technique can distinguish the differences in temperature of objects by the color differences of thermogram, images viewed under infrared ray. Consequently, infrared technique can be applied for the research aim in order to observe the temperature changes during reaction, especially for the exothermic reactions. The most active catalyst will lead to the highest yield in the reaction and therefore to the largest temperature change. Since, this work examined carbon monoxide oxidation on CeO_2-ZrO_2 mixed oxide catalysts as a model reaction. Infrared technique was applied to detect surface temperature of catalyst samples during reaction to determine the catalytic activities. This technique could be suitably applied with such a system, because carbon monoxide oxidation is a reaction with 100% selectivity. A large number of catalyst samples were tested at the same time with infrared technique to find the active catalysts. After that, the lead formulations were studied further for more accurate activity by conventional testing.