CHAPTER I

INTRODUCTION

The present technologies for minimizing the toxic gases in automotive exhaust are based on closed-loop control configuration and the three-way catalyst concept. With a combination of the computer control and the three-way catalyst, over 90 percent removal of the three major toxic gases: carbon monoxide, oxides of nitrogen and hydrocarbon can be achieved. The resulting exhaust satisfies all the known emission standards used today.

However, the closed-loop system, which maintains the air/fuel(A/F) ratio very close to the stoichiometric value, always gives small A/F oscillations around its set point. An addition of easily reducible metal oxide can help the catalyst keep its high efficiency. When A/F ratio is too low, the metal oxide can give oxygen to oxidize carbon monoxide and hydrocarbons, taking oxygen back when air is in excess (A/F ratio is too high). Cerium oxide (Ceria) is a well-known additive that acts as an oxygen storage component added to a present-day catalytic converter. It is believed that ceria can interchange between Ce₂O₃ and CeO₂ depending on oxygen partial pressure in the gas-phase. This is why ceria is useful. However, the mechanism of oxygen transfer inside the catalyst is still unclear.

This project aims to identify the source of the transferable oxygen atoms. The useful oxygen atoms must be only those which can be taken off

and restored back within 1-2 Hz of A/F ratio oscillation. Therefore, in this project, the hypothesis we want to prove is that "the useful oxygen atoms come from the surfaces and the boundaries between crystallites of ceria. They are not the lattice oxygen of ceria in bulk".

This study used a model Pt/CeO₂/Al₂O₃ (platinum on ceria dispersed on alumina support) as the catalyst. The difference in crystallite sizes of ceria was established by varying the calcination temperatures. The amount of oxygen supplied by ceria was measured from the extent of oxidation of carbon monoxide to carbon dioxide in absence of oxygen in the gas phase. Because the measured catalyst oxygen storage capacity(OSC) needs to be representative of the useful oxygen only, the contact time of carbon monoxide on each point in a catalyst bed was kept to about 1 second which corresponds to the 1 Hz oscillation of A/F ratio in the practical automotive system.