CHAPTER I

INTRODUCTION

One cause of air pollution is exhaust gases from automobile having serious harmful effects for human health, the world's atmosphere etc. A number of damages are attributed to tailpipe emissions. The toxic pollutants cause health impairments, plant damage and damages to materials. [1] In the case of gasoline engines such pollutants are carbon monoxide (CO), oxides of nitrogen (NO_X), and volatile hydrocarbons (VOC). Emissions of these pollutants are a function of five parameters :

- the quantity of fuel consumed ;
- the systems of combustion used ;
- the conditions under which combustion takes place ;
- fuel composition ;
- any abatement technologies applied.

The last four parameters are, to variable degrees, currently subject to regulations and can define the emission factors. Each of pollutants have the negative effects of which are mainly local (urban areas) and regional. The first pollutant to concern was carbon monoxide because it was emitted in so high quantities that ambient air concentrations were acutely toxic. Black fumes from cars disturb visibility and are also responsible for soot formation. But particulates which have a smaller diameter white smoke have risen concern to cause cancer because they contain on their very porous surface all kinds of carcinogenic compounds such as heterocyclic hydrocarbons. Particulates have such a small diameter that they pass right into the lung alveoles. Nitrogen oxides are affecting in particular elderly people, asthmatics and people suffering from heart and lung disease. The limit value for air quality was fixed at 200 $\mu g/m^3$, but that seems to be too high, especially for this group and in particular when NO_X occur together with other air pollutants.

Environmentalists realize in this problem, it is necessary to reduce CO, HC, and NO_X by applying the best available technology. One of these technologies, we can use "the catalytic converter". Thus, catalytic converter were introduced to meet stringent emissions requirements legislated for the mineties. Table 1.1 summarises the decision reached on the emission limit values and the corresponding dated of implementation.

Table 1.1 The limit values and the corresponding dates of implementation. [2]

CAR CATEGORY	CO G/TEST	HC+NOx G/TEST	NOx G/TEST	DATE IMPLETATION
Gasoline Engines > 2000 cc	25	6.5	3.6	NM* 1/10/89 AT 1/10/89
1400-2000 cc	30	8	-	NM 1/10/91 AT 1/10/93
<1400 cc STEP 1	45	15	5	NM 1/10/90 AT 1/10/91
STEP 2	TO BE DEFINED			NM 1/10/92 AT 1/10/93
Diesel Engines** >1400 cc	30	8	-	NM 1/10/90 AT 1/10/91
<1400 cc STEP 1	45	15	6	NM 1/10/90 AT 1/10/91
STEP 2	TO BE DEFINED			

* NM.- NEW MODELS AT - ALL TYPES

** FOR DIRECT INJECTION DIESEL ENGINES. 200 CC., THE DATES OF IMPLANTATION HAVE BEEN FIXED AT 1/10/96 FOR ALL TYPES.

The history of catalytic converters was reviewed recently by Ebel. Some of the early patents and publications on catalytic treatment of exhaust gases dated from 1925 [3]. The catalytic converter can control hydrocarbon (HC) and carbon monoxide (CO) in oxidizing condition where oxygen is added to the exhaust constituents. In addition catalytic converter can also control nitrogen oxides in reducing conditions in the exhaust where oxygen is removed from the harmful compound, too. Thus it can control all three emissions (HC, CO, and NO_X) simultaneously. The catalytic converter which can control the three pollutants is called "Three-way Catalyst" (Figure 1.1) [4].

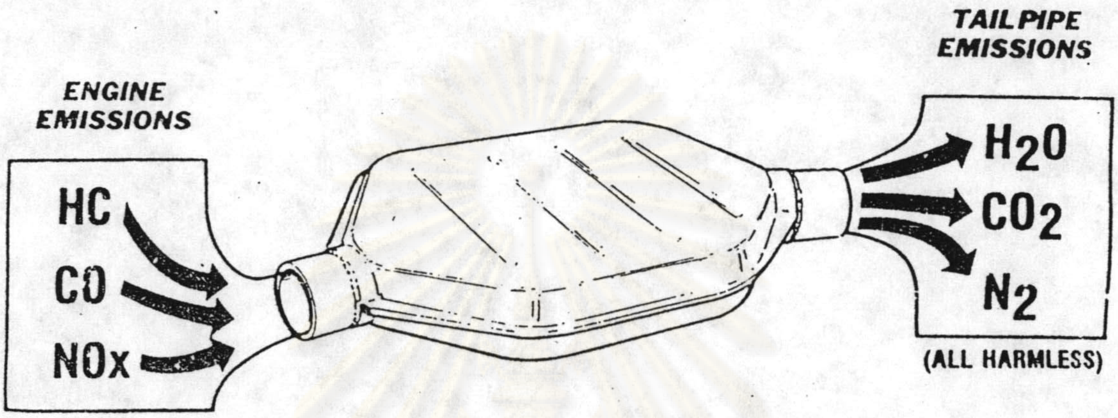

Figure 1.1 Three-way Catalyst System.

The three-way catalytic converter is able to both oxidize and reduce exhaust gas constituents. [5] Inside of the converter, there are five major components : substrate, support, stabilisers, base metal promoters and platinum group metals. The most commonly used substrate materials are multicellular ceramaic monoliths which have a high open area and exert little back pressure in the exhaust system. Metallic monoliths are also used, especially in situations where the greater open area and even lower back pressure of these systems is an advantage. The properties and uses of ceramic and metallic monoliths are reviewed in depth elsewhere and will not be discussed further here. In order to increase the surface area of the monolith, a coating of a highly porous material, usually alumina, is applied. This is known as the washcoat. Stabilisers are often added to the washcoat to maintain the high surface area at the elevated temperatures which are encountered under operating conditions. Promoters are included to improve the activity or selectivity of the catalyst and can have a strong influence on performance. The most widely applied promoters in three-way catalysts are nickel and cerium. The primary catalytic components of current car exhaust catalysts are platinum group metals, which combine the benefits of high activity, particularly at low temperatures, with stability and resistance to poisoning. The three-way catalyst system requires two very stringent conditions, both difficult to realize in practice : nearly stoichiometric air/fuel mixture preparation and a nearly ideal equilibration catalyst. The achievement of the first necessary condition, based on sensing the exhaust composition and continually adjusting the air/fuel ratio, is being pursued vigorously. The narrow ranges which automotive catalytic converter operate well are called windows. Much progress has been ade in widening the operating windows [6, 7]. Research on improved versions of such catalysts is continuing and one can anticipate better conversion, wider operating range and other advances in the future.

The aims of this work were to investigate the preparation of three-way catalysts first. The effects of the addition of promoters were examined on catalytic activities. Finally, modifications of the prepared catalysts were studied to develop their activities.

1.1. The Objectives of This Study.

1.1.1 To develop the preparation method of three-way catalysts for control carbon monoxide, hydrocarbon, and nitric oxide.

1.1.2 To characterize the catalysts prepared in this research.

1.1.3 To study the effect of the reaction conditions (calcination temperatures and calcination conditions) on the redox reaction.

1.2 The Scope and Methodology of This Study.

1.2.1 Studying the preparation of catalysts by

- varying the weight percent of cerium, rhodium, and platium.
- varying the ratio of platium to rhodium.
- varying the order of metal loading.

1.2.1.1 Investigation of the activities of the above-mentioned prepared catalysts on the conversion of carbon monoxide, hydrocarbon, and nitric oxide.

1.2.1.2 Characterizing the prepared catalyts by CO Adsorption Capacity Measurements, and BET Surface Area Measurement

1.2.2 Studying the effect of calcination temperatures in a reducing atmosphere on the prepared catalysts by varying the calcination temperatures ranging from 400 °C to 700 °C :

- The prepared calcined under different temperatures were tested for their activities on the conversion of carbon monoxide, hydrocarbon, and nitric oxide as mentioned in 1.2.1.1.

- The catalysts then were characterized with the same method as mentioned previously in 1.2.1.2.

1.2.3 Evaluating the effects of times on stream and the variation of air/fuel ratio on catalytic performances of catalysts from sections 1.2.1 and 1.2.2.

1.2.4 Modifying the catalysts from section 1.2.1 by fresh catalyst pretreatment :

- The activities of the modified catalysts were tested according to 1.2.1.1.