DEVELOPMENT OF CATALYTIC CONVERTER FOR UNLEADED GASOLINE PROGRAM IN INDONESIA*

by

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ABSTRACT

Recent economic development in Indonesia, as in other ASEAN countries, has resulted in improved prosperity as reflected by the significant increase in the numbers of motor vehicles, particularly in big cities. This growth in car population and traffic is unfortunately accompanied by increase in not only of the country's energy consumption but also increased air pollution. Almost 70% of atmospheric pollution in big cities is reported to be contributed by motor vehicles.

In view of this situation, the Government of Indonesia has launched the "Blue Sky program" and introduced unleaded gasoline. Indonesian manufacturing industries have responded also by designing "national automobiles" to be fabricated in Indonesia, and to run with unleaded gasoline. Some of these vehicles will be equipped with catalytic converters to reduce exhaust gas emissions.

In support of this program, LEMIGAS R/D Center for Oil and Gas Technology is developing its-own catalytic converters that can be fitted into these cars.

The work carried out toward this end is outlined briefly in this paper.

I. INTRODUCTION

Indonesia, as a developing country, has enjoyed a relatively high economic growth, and this is reflected by the increasing prosperity of the people. One of the effect of this growth is the significant increase in the number of motor vehicles particularly in the big cities. The annual increase of the car population in Indonesia is shown in Table 1. The growth in car population can be expected to increase even faster because of the current program of the Indonesian government to build the car industry through the "national automobiles" program.

The increased prosperity in country has had not only a positive impact but also a negative impact to the people, as evident from the increase in fuel consumption

Table 1
Motor vehicles population in Indonesia (1991 - 1994)

Year	Private cars	Buses	Trucks	Motor cycles	Total
1991	1,494,607	504,720	1,087,940	6,494,871	9,582,138
1992	1,590,750	539,943	1,126,262	6,941,000	10,197,955
1993	1,700,454	568,490	1,160,539	7,355,114	10,784,597
1994	1,870,968	671,240	1,501,932	9,940,230	13,984,370

Source: Central Bureau of Statistics

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and the aggravation of air pollution. In addition, such growth in the number of motor vehicles will aggravate traffic jams which reduce the efficiency of the fuel consumed.

Motor vehicles have been viewed as one of the main sources of air pollution. It is estimated that 70% of air pollution in the big cities is due to motor vehicles exhaust emission. Motor vehicles exhaust emission of carbon monoxide (CO) and unburned hydrocarbon (UHC) in Indonesia from year 1991 to 1994 are shown in Table 2 and 3, respectively.

Since 1993 Indonesian government has issued the clean air policy which is known the Blue Sky program as an expression of its concern over the air pollution. It has decreed Law No. 14/1992 on Road - Traffic to prevent air pollution due to emission of motor vehicles and enforced that they meet the emission standard. Pollution caused by lead which is contained in leaded gasoline can be eliminated by using unleaded fuel. Since August 1995, the government, through its State Oil Enterprise PERTAMINA, has launched unleaded gasoline which called Super TT for automobiles.

Although lead compound can be eliminated, emission of exhaust motor vehicles such as CO, UHC, and NO_X still exists. These emissions can only be reduced by using catalytic converter , which is fitted into car's exhaust pipe, to convert CO and UHC to CO_2 and at the same time reduce NO_X to N_2[7]. Unleaded gasoline and catalytic converter have been used in developed countries such as USA, Europe, Japan and also in some ASEAN countries to overcome exhaust motor vehicle emissions.

In support of the effort to solve the pollution caused by motor vehicles exhaust emissions in Indonesia,

Table 2
Emission of CO from motor vehicles source in Indonesia for year 1991 to 1994

CO, kg/Year

Year	Private cars	Buses	Trucks	Motor cycles	Total
1991	28,845.90	1,261.82	2,719.87	125,350.95	158,178.54
1992	30,701.46	1,349.86	2,815.67	133,961.23	168,828.22
1993	32,818.74	1,421.23	2,901.36	141,953.63	179,094.96
1994	36,109.51	1,678.10	3,754.83	150,303.00	191,845.45

Source: Central Bureau of Statistics

Table 3
Emission of UHC from motor vehicles source in Indonesia for year 1991 to 1994

UHC, kg/Year

Year	Private cars	Buses	Trucks	Motor cycles	Total
1991	2,750.07	600.61	1,294.64	11,950.52	16,595.83
1992	2,926.97	642.53	1,340.24	12,771.39	17,681.13
1993	3,128.82	676.50	1,381.03	13,533.36	18,719.71
1994	3,442.56	619.22	1,421.20	14,329.40	19,812.39

Source: Central Bureau of Statistics

LEMIGAS R/D Center for Oil and Gas Technology has carried out catalyst research for catalytic converter.

II. MOTOR VEHICLES EXHAUST EMISSIONS

A. *Origin of motor vehicle exhaust emission*

Emissions are, basically, the direct consequences of incomplete combustion in the engine. Complete oxidation of the hydrocarbons presents in the fuel leads to the harmless reaction products such as CO_2 and H_2O vapor. Since all material conversion processes, much like energy conversion processes, are incomplete, these processes originate a number of pollutants [1,2;].

- Uncombusted hydrocarbon such as paraffins, olefins, and aromatics
- Only partly combusted hydrocarbons such as carbon monoxide, aldehydes, and ketones
- Numerous products of thermal cracking such as soot and polycyclic aromatic hydrocarbons
- By products such as nitrogen oxides and lead oxide (if using leaded fuel).

Most of those emissions detrimentally affect human health.

Generally, the emissions of motor vehicle are strongly influenced by the following factors [1]:

- The motor vehicle class, i.e., cars, trucks, busses, two or four-cycle engines, gasoline or diesel engines.
- The model year of the vehicle, due to changing design affecting emissions.
- The condition of the vehicle.
- The conditions of operation of the vehicle, i.e., starting, continuous versus discontinuous operation, stop and go traffic, and individual attributes of the driver.

B. *Alternative fuel*

Various efforts have been carried out to reduce exhaust motor vehicle emissions, i.e. using clean fuel or alternative fuel such as gaseous fuel (natural gas) or reformulated gasoline [3,6,7]. Many believed that, to meet the low emissions levels specified, alternative fuels would be required. However, such fuels show that the certain emission will be increased while another decreased. For example, according to the work of Shell Research, which used variation in gasoline composition (MTBE, aromatics, olefins, and using paraffins to balance), emissions will be affected by the variables of fuel composition [6]. The results show that:

- Inclusion of MTBE (and removal of paraffin) yields HC and CO reduction benefits in both non-catalyst- and catalyst-equipped cars.
- MTBE effects on NO_x are small and no clear trend is apparent.
- The apparent effect of reducing aromatics content is to reduce tail pipe HC and CO emissions further, but the accompanying volatility changes in the fuels make the reasons for this effect uncertain.
- Reducing aromatics content increases NO_x emissions.

Similar results were also reported by Koehl *et. al* in their work with Auto/Oil Air Quality Improvement Research Program, AQIRP [11]. Adding oxygenates to the fuel decreased benzene but increased aldehydes. Oxygenate effects have been found to vary among control technology systems.

In another work, Schoonveld and Marshall reported that the effect of reformulated gasoline on vehicle emissions showed significant reduction of CO, UHC, NO_X, and benzene but increases formaldehyde emission [13].

According to US 1990 Clean Air Act Amendment there are five specific toxic compounds to be considered in evaluation of reformulated gasoline, namely: formaldehyde, acetaldehyde, 1,3-butadiene, benzene, and polycyclic organic matter.

Comparison of the emission between natural gas and conventional gasoline is shown in Table 4 [3].

III. CATALYTIC CONVERTER

Catalysts have been widely used to lower carbon monoxide (CO) and hydrocarbons (HC) in the exhaust of motor vehicles since 1975 in the United States [14].

Table 4
Comparison of natural gas and conventional gasoline emissions [4]

Pollutant	Gasoline	Natural gas
THC + NOx	0.50	0.24
CO	1.54	0.90

Then catalyst research for NO_x emission has been carried out more intensively and as the result, the three-way catalyst which simultaneously reduce HC, CO and NO_x emissions have been founded. Such catalyst system, called catalytic converter, is installed in the tail pipe (exhaust systems) and controls all three pollutants together.

The principle of the catalytic converter is to convert the pollutants to be harmless emissions. The most important catalytic reactions in the catalytic converter [14] are :

- Oxidation of the unburned hydrocarbons (HC):

  $H_nC_m + (m+\frac{1}{4}n)\,O_2 \rightarrow$ [illegible] $+ \frac{1}{2}\,H_2O$

- Oxidation of the carbon monoxide (CO):

  $CO + \frac{1}{2}\,O_2 \rightarrow CO_2$

  $CO + H_2O \rightarrow CO_2 + H_2$

- Reduction of the nitrogen oxides (NO_x):

  $NO + CO \rightarrow N_2 + CO_2$

  $NO + H2 \rightarrow N_2 + H_2O$

The desired products of the reactions are N_2, CO_2, and H_2O.

Although the catalyst is the key ingredient, a vital element in converter design is the manner in which the catalyst is supported. Three different physical forms of catalyst have been introduced, *i.e.* pellets, ceramic monolith, and metal monolith [9,15].

The "pelleted" catalysts are in the form of extrudates, spherical particles, or cylindrical pellets of about 1/8 to 1/10 of an inch in diameter. It is well suited for a large volume, low speed engines with relatively low exhaust gas temperature. Unfortunately, the "pelleted" catalysts have encountered a mechanical design problem after a period of time in customer use which has resulted in loss of power and drivability of the vehicles.

The ceramic monolith is presently the substrate of choice in the world market because its cellular design provides the following benefits: a high degree of geometric surface area, fast catalyst light off, low exhaust gas back pressure, compatibility with the catalysts and coatings, low cost, and high temperature resistance

Monolithic ceramic substrate have almost entirely been produced from cordierite. Ceramic monolith have been produced in various cell densities and geometries.

Monolithic catalyst supports of metallic alloys have been under development for a long period. This catalyst offers a number of potential advantages for various reasons as reported by Harkonen *et al.*[10]. They are basically the cellular design of ceramic monolith but the honey comb is formed normally by spirally wound alternate sheets of flat and corrugated metal.

The composition of three-way catalyst are precious metals or base transition metals and their oxides [7,8]. Pt and Pd are the only active ingredients for oxidation catalysts that have proved durable for application in vehicle exhaust. Rh is used for reduction catalysts. Each precious metal is combined with one or two others to exhibit a synergism. Other elements (La, Ce) are added as promoter and/or stabilizer. All these ingredients (active metals, promoter) are impregnated into alumina support [8,14].

Vehicle exhaust catalysts or catalytic converter plays an important role in environmental pollution controls [5,12,15]. However, such catalysts can only be effective when vehicle fuel is unleaded gasoline whereas the catalysts are poisoned by lead, which causes the catalysts to be deactivated and their performance will decreased [7,8,14].

IV. STUDY OF VEHICLE EXHAUST CATALYST

In support of the program to reduce of air pollution due to vehicle exhaust emission, "LEMIGAS" R/D Center for Oil and Gas Technology participated by carrying out a study of catalyst formulation for catalytic converter. To produce catalyst which is relatively good for controlling vehicle emission, several requirements needed such as: high activity and selectivity, resistant to poison, chemically stable, high strength, thermal shock resistance, low pressure drop, harmless, and long life.

A. *Objective of the study*

The objective of the present study is to investigate the influence of preparation condition and addition of other components to improve the characteristic of the vehicle exhaust catalysts.

This study is conducted to obtain the catalyst formulation with high activity to reduce the vehicle exhaust emissions.

The steps of study are as follows: preparation of catalysts, characterisation, and activity testing with gas CO.

B. *Result and discussion*

The first step of the study is to evaluate the potential of addition cerium (Ce), lanthanum (La), active metals palladium (Pd) and rhodium (Rh) on platinum (Pt)-contained catalysts with alumina supported in reduction of model exhausts emission. Five catalysts formulation have been prepared, their characteristics are shown in Figure 1, 2 and 3 for surface area, pore volume and pore size, respectively. The activity of such catalysts is shown in Figure 4 and 5 for catalysts that were calcined at 400°C and 700°C, respectively.

Figure 1 reflects the effect of calcination temperature in the preparation catalysts, high surface areas are produced at 400°C calcination temperature. Meanwhile the effect of addition ingredients La, Ce, La, Pd, and Rh and calcination temperature on the magnitude of surface area indicates that the surface area decreased by the addition of the ingredients except for Ce addition which resulted the small increase.

In contrast, calcination temperature and addition of La, Ce, Pd, and Rh have indicated insignificant difference on pore volume for those catalysts (Figure 2). The

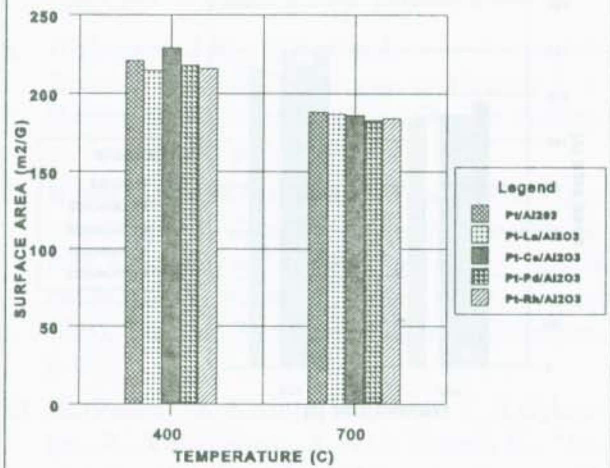

Figure 1
The effects of calcination temperature and addition of La, Ce, Pd , and Rh on surface area of Pt/Al_2O_3 catalyst

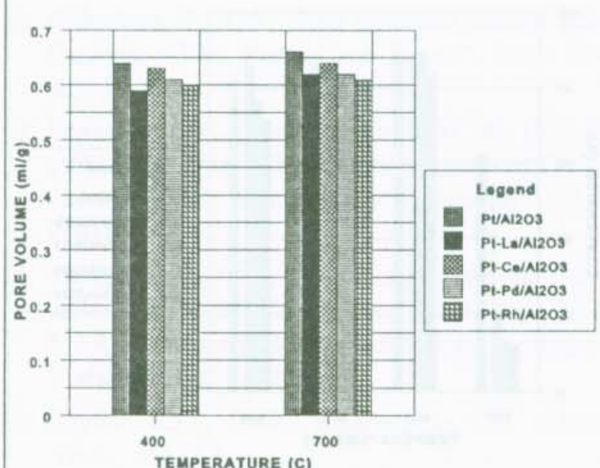

Figure 2
The effects of calcination temperature and addition of La, Ce, Pd , and Rh on pore volume of Pt/Al_2O_3 catalyst

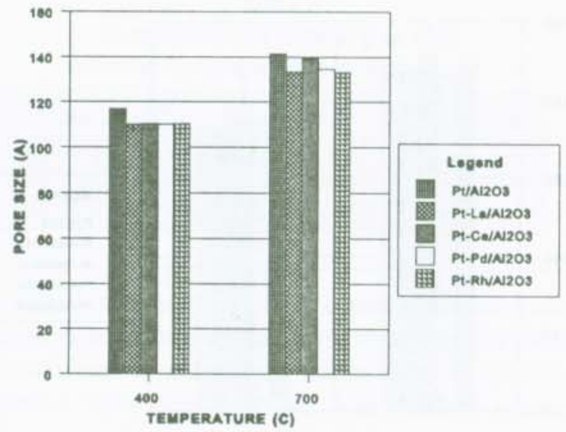

Figure 3
The effects of calcination temperature and addition of La, Ce, Pd , and Rh on pore size of Pt/Al_2O_3 catalyst

pore size of catalysts is only affected by the calcination temperature whereas the pore size increased at 700°C (Figure 3), since at this temperature sintering or agglomeration of catalysts particles can be expected.

Activity test, using carbon monoxide (CO) as a model, as shown in the Figure 4 and Figure 5, shows that relatively no difference in the conversion of CO is observed for reaction temperatures of 400°C to 600°C. However, addition of La, Ce, Pd, and Rh gives a significant effect on the conversion of CO for the various reaction temperatures. The conversion of CO increased for all catalysts compared with Pt/Al_2O_3 catalyst. In another words, those elements or ingredients have positive effect on the reduction of CO emission. Addition of Ce resulted in high reduction of emission of CO due to its ability to storage the oxygen [10].

This study is expected to be continued with other gas models such hydrocarbon and NO_x and/or mixture thereof for their activity test. Further study will be directed for the modification of the catalysts formulation.

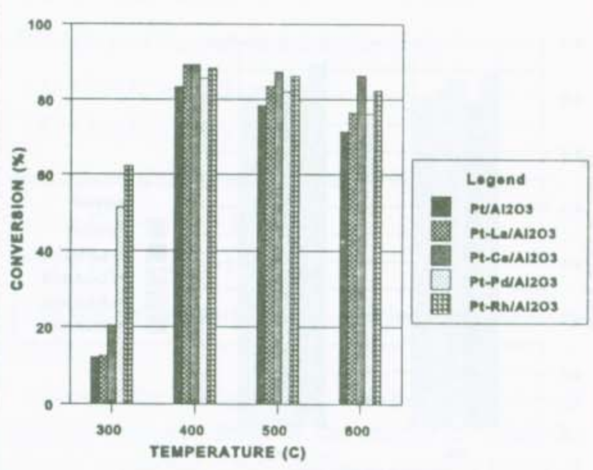

Figure 4
The effect of reaction temperature and addition of La, Ce, Pd, and Rh on Pt/Al_2O_3 catalysts for CO conversion. Catalysts are calcined at 400°C

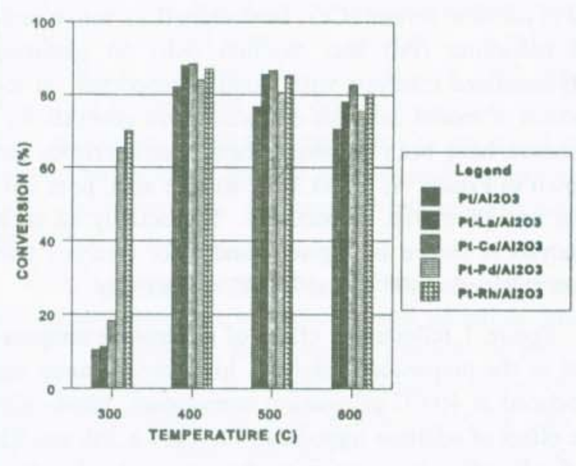

Figure 5
The effect of reaction temperature and addition of La, Ce, Pd, and Rh on Pt/Al_2O_3 catalysts for CO conversion. Catalysts are calcined at 700°C

V. CONCLUSION

Participation of the community and the institutions are expected to achieve of prevention of vehicle exhaust emissions. The use of catalytic converter will aid in the achievement of the goal particularly until the introduction of unleaded gasoline and to the ABlue Sky@ program by the government. Catalytic converter can be fitted with such fuel. Our study is conducted to support the program.

Results of this study indicated that preparation catalyst and addition of La, Ce, Pd, and Rh affected the performance of catalysts. Role of Pd and Rh exhibit a synergism with Pt to reduce CO emission.

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