CATALYTIC OXIDATION OF UNBURNED HYDROCARBON IN SIMULATED EXHAUST GAS OF DME ENGINE

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Abstract

Characteristic of dimethyl ether (DME) oxidation with NO_X reduction catalyst was investigated in comparison with noble metal catalyst. The catalysts used in this research were Co and Sn as NO_X catalyst and Pt as a noble metal catalyst. In addition, pure alumina (denoted by "PA" in the following section) catalyst was also used. Laboratory experiments were conducted by using above catalysts in oxygen rich or Diesel type DME engine simulated exhaust gas. Catalyst temperature and concentration of NO were selected as experimental parameters. The experimental results showed that low temperature DME oxidation was possible with Pt whereas, higher temperature DME oxidation was observed with Co, Sn and PA catalyst. Large amount of formaldehyde (HCHO) was formed as an intermediate species during the use of NO_X reduction catalysts. However, Pt catalyst did not show any formation of formaldehyde. The effect of concentration of reducing agent on NO reduction was also carried out with the NO_X reduction catalyst.

Key Words: *Internal Combustion Engines, Exhaust Gas, NO_X Catalyst, Formaldehyde, And Lean Combustion*

Introduction

At the beginning of 21st century, the energy utilization and environmental protection have become two subjects, which are of most urgency and great importance for the development of the human society. However, with the increase of the traveling automobile and the step rise of the motor fuel demand, moreover with the sharp decrease of worldwide petroleum resource and the heavy pollution by the automobile tail pipe exhaust gas to our living environment, every country in the world is speeding up to seek and develop new alternative automobile fuels.

Today, the hopeful substitute fuel energies are alcohol fuels, natural gas, DME, LPG, H_2 and so on. Physical and chemical properties of these fuels are shown in Table 1. Alcohol fuels are liquid fuels, so we can use them easily by former petroleum systems, the systems may not be changed, and the economic merit is large. Recently, we have kept our eyes on the global environment problem of the global heating by CO_2, etc., and we have directed our

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attention to alternative fuel that has superior environment characteristics of NO_x, particulate matter, etc.

DME and methanol are especially interesting when the energy is needed or desired as a transportation fuel and when it has to be transported over increasingly long distances. Methanol and DME can be industrially produced from coal, natural gas and many kinds of biomass fuels. Several reports were published showing DME as a fuel additive [1, 2] and after that DME was rediscovered as a clean alternative fuel for Diesel engine. Combustion in a compression ignition (CI) engine is possible without any ignition aid due to high cetane number of DME and its high oxygen content introduce smoke free combustion [3] when burned in a proper manner.

Table 1 Properties of some alternative fuels compared to other conventional fuels

	UNIT	DME	DIESEL	PROPANE	CNG	METHANOL	GASOLINE
Liquid Density	Kg/m³	667	831	500.5	-	795	750
Relative Gas Density	Kg/m³	1.59		1.52	0.56	-	-
Cetane Number		>55	40~55	-	-	-	-
Methane Number		-	-	-	75	-	-
Chemical Structure		C_2H_6O	-	C_3H_8	CH_4+rest	CH_3OH	-
Stoich. A/F Ratio	Kg/Kg	9	14.6	15.68	16.86	6.46	14.7
Boiling Point	°C	-25	180/370	-42	-162/-88	65	30/190
RON		-	-	112.1	120	106	98
C	%wt	52.2	86	82	76	37.5	85
H	%wt	13	14	18	24	12.5	15
O	%wt	34.8	0	0	0	50	0
Velocity of Sound	m/s	980	1,330	-	-	-	-
Kinematic Viscosity (liquid)	cSt	<1	3	-	-	-	-
LCV	MJ/Kg	28.8	42.7	46.3	49	19.8	43.2
Ignition Limit		0.34	0.48	0.42/2.0	0.7/2.1	0.34/2.0	0.4/1.4
Vapor Pressure (293 k)	KPa	530	?	830	-	37	45/90
Mol. wt.	g/mol	46.07	170	44.1	17	32.1	98
Min. Ign. Energy (λ=1)	mJ	0.29	?	0.31	0.32	0.22	?
Auto Ign. Temp	°C	235	250	470	650	450	?
Liquid Specific Heat	KJ/Kg-K	?	2.2	2.5	0.63	2.6	2.4
Gaseous Specific Heat	KJ/Kg-K	2.99	1.7	1.67	2.2	1.72	1.7
Heat of Vaporization	KJ/Kg	467.13	300	372	510	1,110	420

∗ Superscript numbers indicate references listed at the end of the paper

Though DME is known as a clean fuel, some emissions, such as NO_X, unburned DME and formaldehyde will create problem. NO_X emission from DME fueled engine is comparable to the conventional Diesel engine [4]. NO_X emission might be improved with three-way catalyst if the engine would operate in fuel-rich condition. However, lean burn or Diesel type engine is quite preferable to improve energy efficiency. When the lean burn or Diesel type engine is used, one of the most promising way to reduce NO_X emission is the use of NO_X reduction catalyst (called as "NO_X catalyst" in the following part) [5 ~ 7] since NO_X catalyst is mainly designed for NO_X reduction with oxygen rich exhaust gas condition. The use of NO_X catalyst will much increase in the future to meet the NO_X emission regulation under the requirements for low fuel consumption and better air quality standard. When the NO_X catalyst is applied to the DME engine, we need to keep our eyes on unburned DME and formaldehyde emissions. Especially, formaldehyde is toxic species for human beings and also has photochemical reactivity. Sometime formaldehyde was formed as an intermediate species during the catalytic oxidation of unburned hydrocarbon [3]. Thus, DME oxidation and formaldehyde formation characteristics are also important feature for NO_X catalyst, when it has to be applied to DME fueled engine.

Main part of the NO_X reduction catalyst researches is, of course, it's NO_X reduction performance in different atmosphere such as, in engine exhaust gas and synthesized gas. Yamamoto et al. [8] reported the performance of alumina catalyst as a NO_X catalyst in exhaust gas from methanol fueled engine. However, since the main focus of the reports until now have been NO_X reduction characteristics, the systematic research on DME oxidation and formaldehyde formation with NO_X catalyst concerning its application to DME fueled engine is quite limited. Also it is important to know the particular effect of NO on catalytic oxidation of unburned hydrocarbon. Simulated exhaust gas technique is an art to understand the species effect on catalyst performance.

In the authors' previous works, methanol oxidation and formaldehyde formation characteristics in many kinds of oxidation catalysts as well as NO_X catalysts were investigated [9 ~ 13]. Those results showed that though the methanol oxidation was quite high however, significant amount of formaldehyde formation appeared during the methanol oxidation process. Especially, the amount of formaldehyde formation increased with increase in NO in the reaction gas.

In this report, the systematic research on DME oxidation and formaldehyde formation with NO_X catalyst such as Sn or Co were carried out by the following experimental method of the previous researches [9 ~ 13]. The performances of NO_X catalyst for DME oxidation and formaldehyde formation were compared to Pt and PA catalyst to make the performance of NO_X catalyst clearer. Also the effect of concentration of reducing agent on NO reduction was also performed with NO_X catalyst. Concentration of NO and reaction temperature were selected as experimental parameters based on the information from the previous researches [9 ~ 13].

Experimental

A schematic diagram of the experimental system is shown in Fig.1. Synthesized (reaction) gas used in this experiment is shown in Table 2. Synthesized gas of N_2, O_2, and DME was blended with NO and fed into a quartz glass flow reactor where catalytic oxidation of DME was carried out. The reaction gases were supplied from the gas cylinder at an atmospheric pressure, and made to pass through the reactor and temperature controlling section. The simulated exhaust gas used in this research was a general one where CO, CO_2 and H_2O were not considered. Since the co-existence effect of CO, CO_2 and H_2O are much lower than the effect of NO when noble metal catalyst was used [14]. Also the main concern of this research was to find out the effect of NO on DME oxidation and formation of intermediate species therefore, the species with less effect was not considered here.
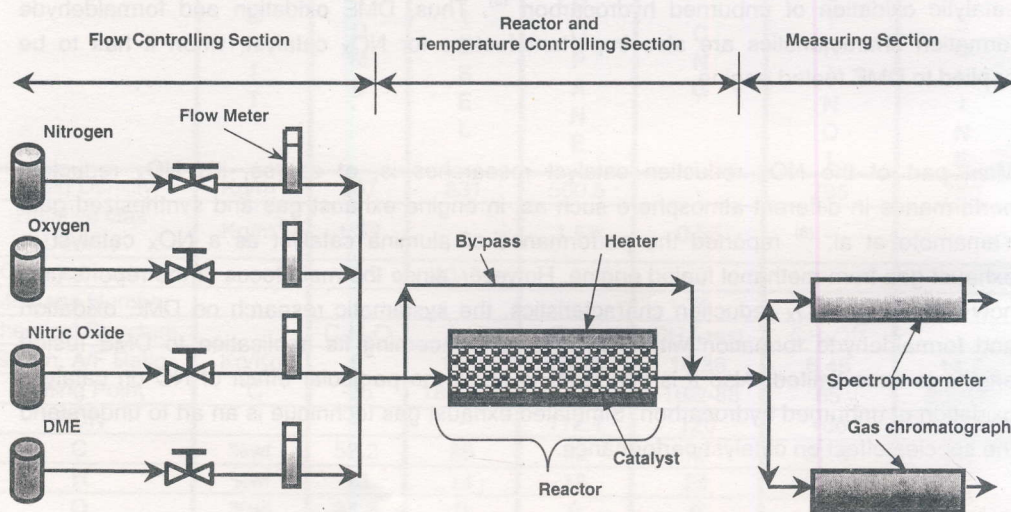

Fig 1 Schematic diagram of the experimental system

Table 2 Composition of simulated exhaust gas

Species	Concentration (vol %)
O_2	5 %
DME	0.15 %
NO	0, 500 and 1000 ppm
N_2	Balance

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The catalytic reactor was heated electrically to keep the temperature profile uniform within the catalyst. The gas temperature was monitored at the middle of the reactor. Any temperature between 0 ~ 400 $^{\circ}$C can be attained by this temperature controlling section.

The tested catalysts are shown in Table 3. The noble metal catalyst was manufactured by Japan Engelhard Co. Ltd. Riken Corporation manufactured the NO_X and PA catalyst. PA catalyst was used for the comparison of the catalyst activity among the catalysts. All the catalysts used in this research were loaded on gamma alumina. Noble metal catalyst was cylindrical in shape with diameter 2.5mm and 3.5mm long. NO_X and PA catalyst were spherical shape with 2 ~ 3 mm in diameter. All the catalysts used in this experiment were pellets.

Simulated exhaust gas entered into the gas analyzing section after passing through the reactor. A second-order derivative spectrophotometer [15] was used for the measurement of NO, NO_2 and HCHO. A gas chromatograph equipped with flame ionization detector (FID) was used for DME and methanol determination.

Results

NO Reduction Characteristics: NO_X catalyst is mainly designed for NO_X reduction however, how it works when it has to be applied for the purification of unburned species and at the same time formation behavior of the intermediate species are the main focus of this research. Figure 2 represents NO reduction ratio [= {1 - (NO_{out} / NO_{in})} where, NO_{out} and NO_{in} are the concentration of NO at the outlet and inlet of the catalytic reactor, respectively] with 500ppm of NO concentration of the simulated exhaust gas. NO reduction ratio with Co, Sn and PA was plotted as a function of catalyst temperature.
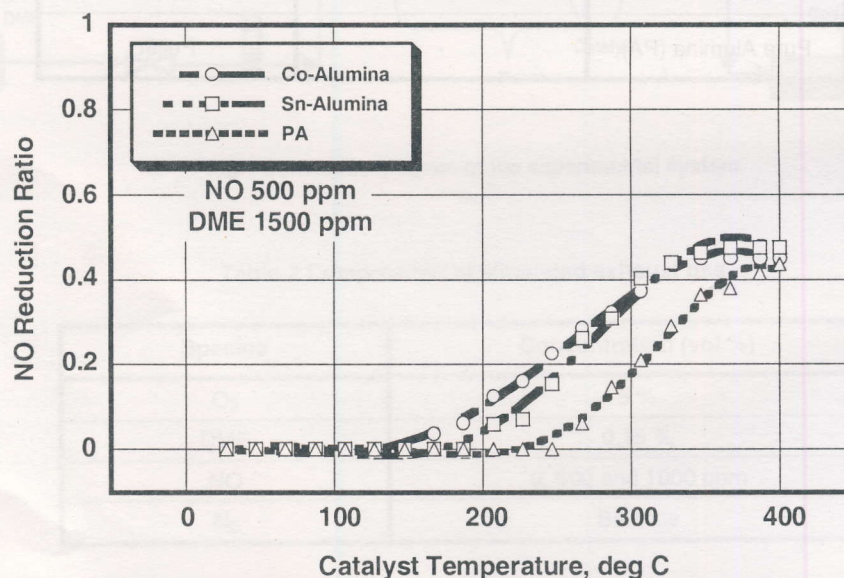

Figure 2 NO reduction ratio with the variation of catalyst temperature

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The Co, Sn and PA catalysts showed high performance to reduce NO in presence of DME and O_2. The reduction ratio with NO_X catalyst increased with increase in catalyst temperature and reached above 0.70 at around at 400 $^{\circ}$C. The reduction temperature of NO was lower with NO_X catalyst compared to PA catalyst. Most active temperature zone with NO_X catalyst was in between 200 ~ 400 $^{\circ}$C and which is almost identical to an actual DME engine exhaust gas temperature [4]. NO_X reduction with Co and Sn catalyst might not be a problem however purification of unburned species from the exhaust gas of DME engine is another concern for Co and Sn catalyst.

NO reduction ratio with Pt catalyst is shown in Fig. 3 where concentration of NO and catalyst temperature was considered as an experimental parameters. During the catalytic reduction of NO, there is a possibility of conversion of NO into NO_2 since, NO is less stable compared to NO_2. Therefore, NO_2 formation was also checked with the derivative spectrophotometer. No such conversion was observed With NO_X and PA catalyst or in other words formation of NO_2 was too small for detection. However, NO_2 formation was observed with Pt catalyst (Fig. 4) during the reduction of NO in the simulated exhaust gas. Formation of NO_2 started after 200 $^{\circ}$C and increased with increase in NO concentration of the reaction gas. Formation of NO_2 increased up to a certain limit with increase in reaction temperature, after that formation decreased with increase in reaction temperature. Possibly, at higher temperature reducing agent (DME, HCHO) is consumed due to the higher reactivity of Pt. Therefore, in presence of enough oxygen NO is oxidized and NO_2 formation occurs.
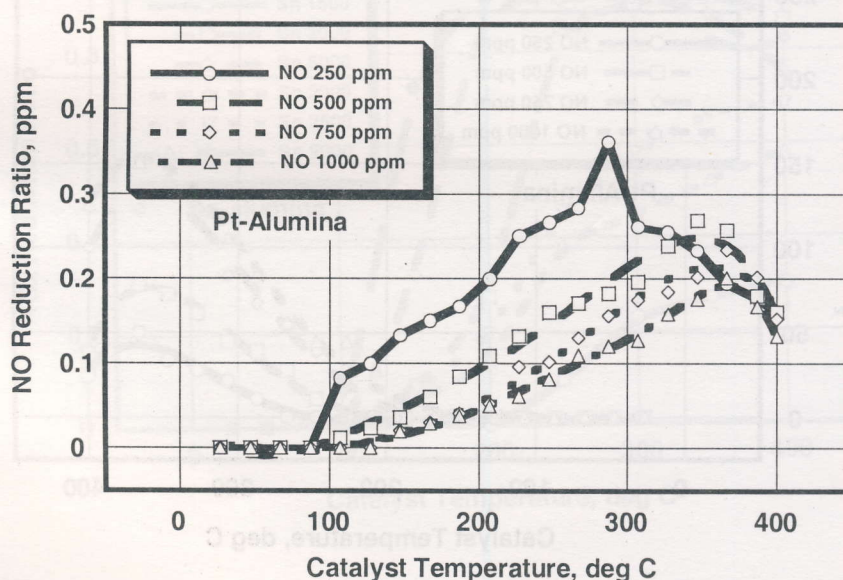

Figure 3 No reduction with Noble metal catalyst

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Effect of Concentration of Reducing Agent on NO Reduction: Every reducing agent has some effect on catalytic reduction of NO. Some of them are effective to reduce NO_X even at lower temperature. Some of them perform well at higher temperature. It depends on the catalyst, reaction temperature, species of exhaust gas, concentration of NO_X and so on. Also the concentration of reducing agent in the exhaust gas will play an important role for the selective catalytic reduction of NO_X.

It is considered that the selective catalytic reduction of NO_X will increase with the increase of reducing agent. However, report is not available for the catalytic reduction of NO_X with the change of DME concentrations in the exhaust gas. In this experiment NO_X catalyst Co and Sn were used to check their performances during the variation of concentration of reducing agent. All through the experiment 500 ppm of NO was mixed with the simulated exhaust gas. Figures 5 and 6 represent the NO reduction ratio with the variation of catalyst temperature for Co and Sn respectively, where concentration of reducing agent (DME) was considered as an experimental parameter. In these figures several different curves are presented for NO reduction and each curve represents NO reduction ratio for a particular concentration of reducing agent such as, Co 9000 indicates reduction ratio with Co catalyst where simulated exhaust gas contains 9000ppm of DME.
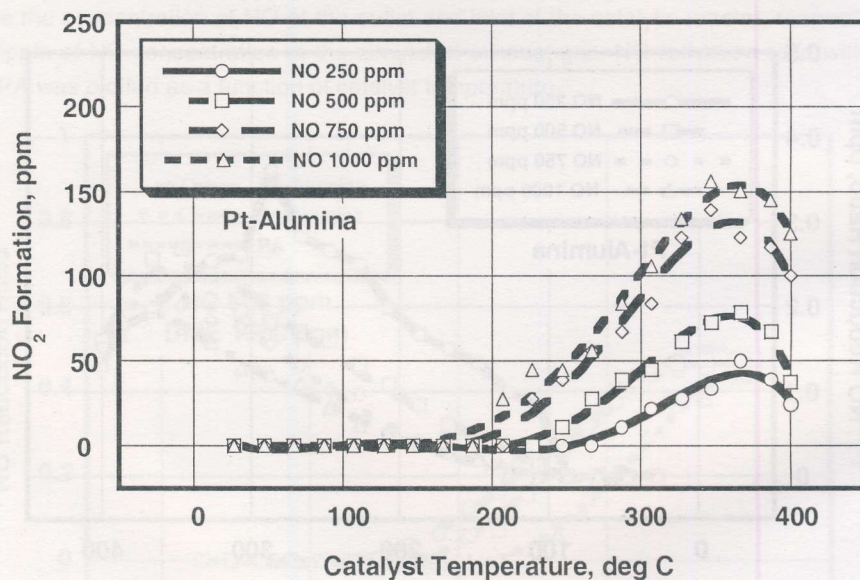

Figure 4 NO_2 formation characteristics with Pt catalyst
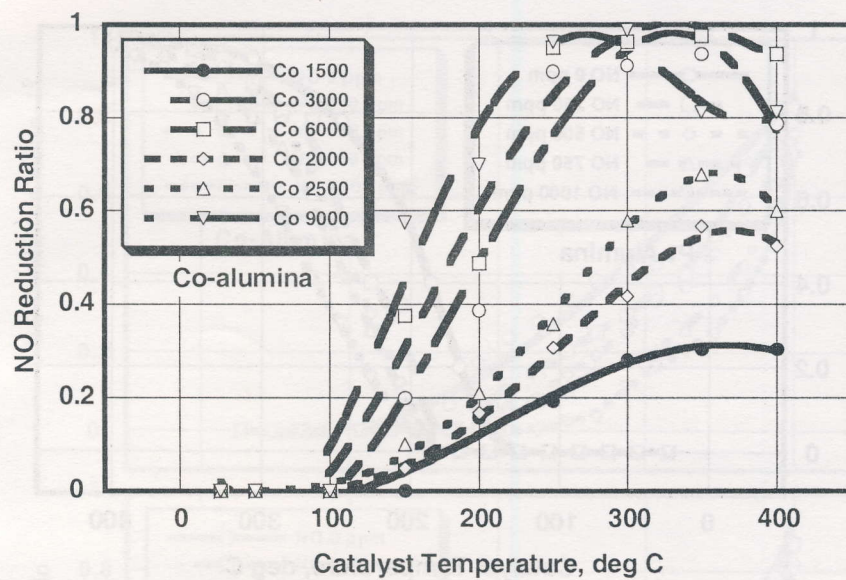
Figure 5 NO reduction with the variation of reducing agent
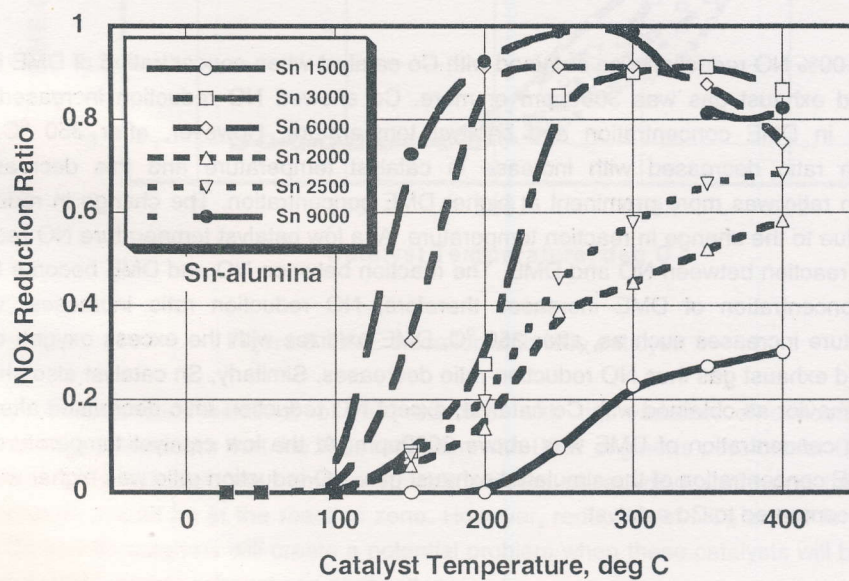

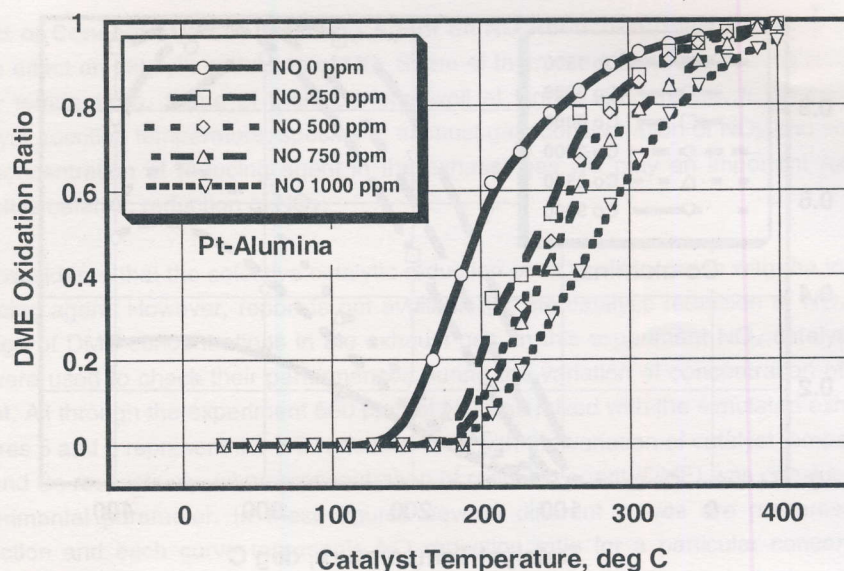Figure 6 NO reduction with the variation of reducing agent

Figure 7 DME oxidation with Pt catalyst

Almost 100% NO reduction was obtained with Co catalyst when concentration of DME in the simulated exhaust gas was 3000ppm or more. Co showed NO reduction increased with increase in DME concentration and catalyst temperature. However, after 350 $^{\circ}$C, NO reduction ratio decreased with increase in catalyst temperature and this decrease in reduction ratio was more prominent at higher DME concentration. The change in reduction ratio is due to the change in reaction temperature. At a low catalyst temperature NO reduces with the reaction between NO and DME. The reaction between NO and DME become faster when concentration of DME increases therefore, NO reduction ratio increases. When temperature increases such as, after 350 $^{\circ}$C, DME oxidizes with the excess oxygen of the simulated exhaust gas thus NO reduction ratio decreases. Similarly, Sn catalyst also showed same behavior as obtained with Co catalyst, except NO reduction ratio decreased after 300 $^{\circ}$C when concentration of DME was above 3000ppm. At the low catalyst temperature and high DME concentration of the simulated exhaust gas, NO reduction ratio was higher with Sn catalyst compared to Co catalyst.
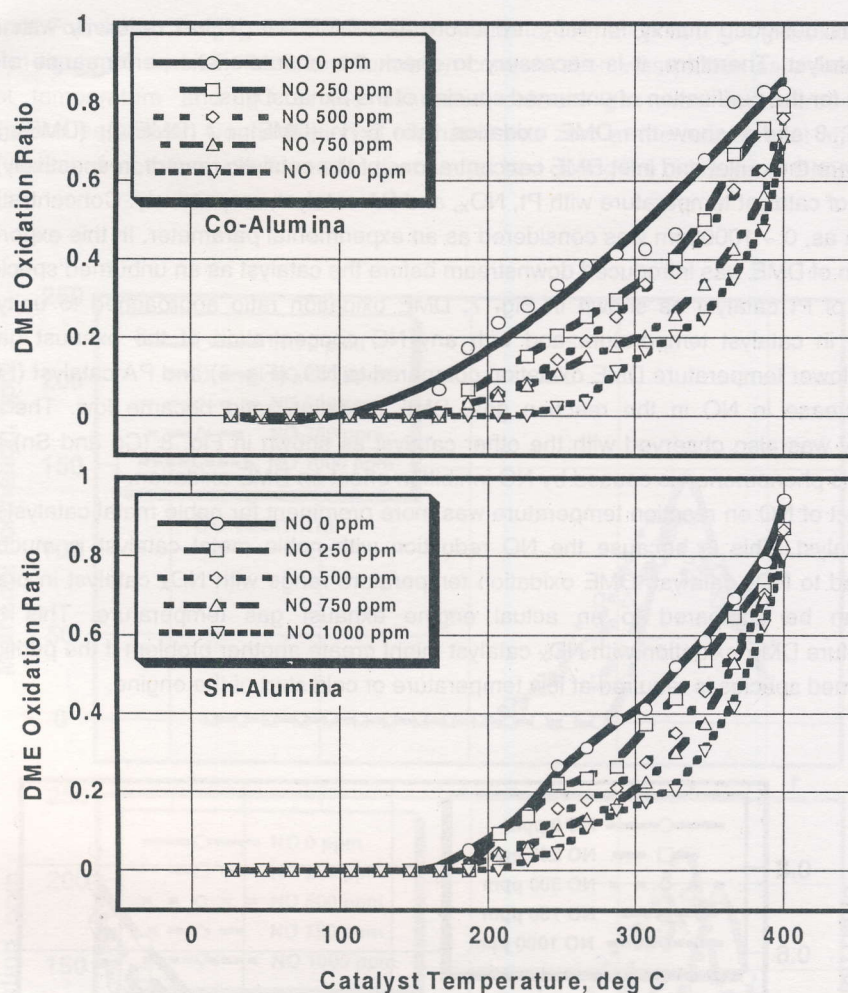
Figure 8 DME oxidation with NO$_X$ catalyst

Above characteristics behavior of NO reduction with the variation of concentration of reducing agent indicates that the NO$_X$ reduction with DME engine is possible for Co and Sn catalysts if the exhaust gas contains suitable amount of reducing agent and the catalyst temperature should be in the reaction zone. However, reduction of NO$_X$ at low temperature with Co and Sn catalysts will create a potential problem when these catalysts will be applied to actual DME engine exhaust gas purification.

DME Oxidation Characteristics: Generally, oxidation ratio of unburned emission of DME with NO$_X$ catalysts is expected to be lower than that of noble metal catalyst. Because NO$_X$

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catalyst is designed mainly for NO_X reduction, also DME has higher reactivity with noble metal catalyst. Therefore, it is necessary to check the characteristic performance of NO_X catalysts for the purification of unburned species of the exhaust gas.

Figure 7, 8 and 9 show the DME oxidation ratio [$=\{1-(DME)_{out}/(DME)_{in}\}$; $(DME)_{out}$ and $(DME)_{in}$ are the outlet and inlet DME concentrations of the catalytic reactor, respectively] as a function of catalyst temperature with Pt, NO_X, and PA catalyst, respectively. Concentration of NO such as, 0 ~ 1000ppm was considered as an experimental parameter. In this experiment 1500ppm of DME was introduced downstream before the catalyst as an unburned species.

In case of Pt catalyst as shown in Fig. 7, DME oxidation ratio approached to unity with increase in catalyst temperature and with any NO concentration of the exhaust gas. Pt showed lower temperature DME oxidation compared to NO_X (Fig. 8) and PA catalyst (Fig. 9). With increase in NO in the reaction gas, DME oxidation ratio became low. The same tendency was also observed with the other catalyst as shown in Fig. 8 (Co and Sn) and 9 (PA). This phenomenon is caused by NO inhibition effect on DME oxidation.

The effect of NO on reaction temperature was more prominent for noble metal catalysts than NO_X catalyst. This is because the NO reduction with noble metal catalyst is much less compared to NO_X catalyst. DME oxidation temperature range with NO_X catalyst in practical level can be compared to an actual engine exhaust gas temperature. This higher temperature DME oxidation with NO_X catalyst might create another problem if the purification of unburned species is required at low temperature or cold start of the engine.
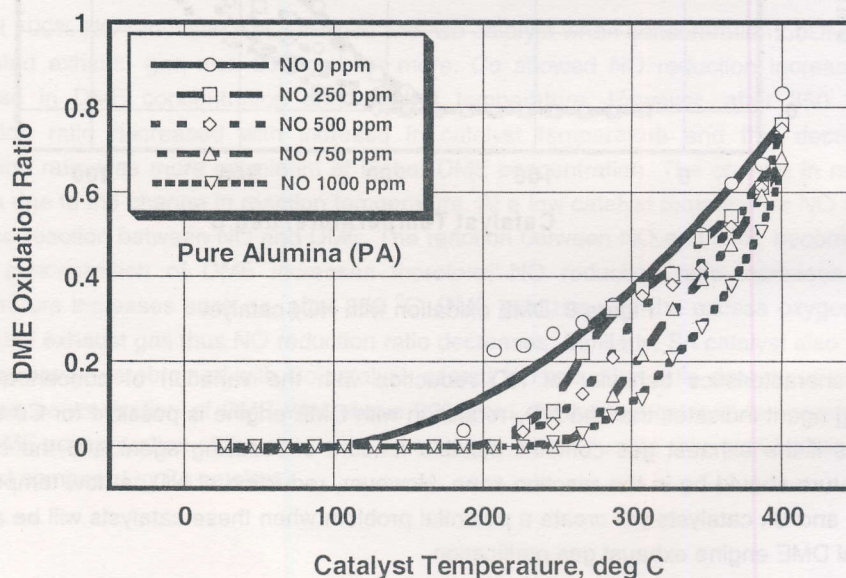

Figure 9 DME oxidation with pure alumina catalyst

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Methanol Formation During DME Oxidation: Methanol formation during DME oxidation is shown in figure 10 and 11. Methanol formation concentration was plotted as a function of catalyst temperature and NO in the simulated exhaust gas was considered as an experimental parameter. Concentration of methanol was zero at the reactor inlet and outlet concentration of methanol was formed in the catalyst bed during the oxidation of DME.
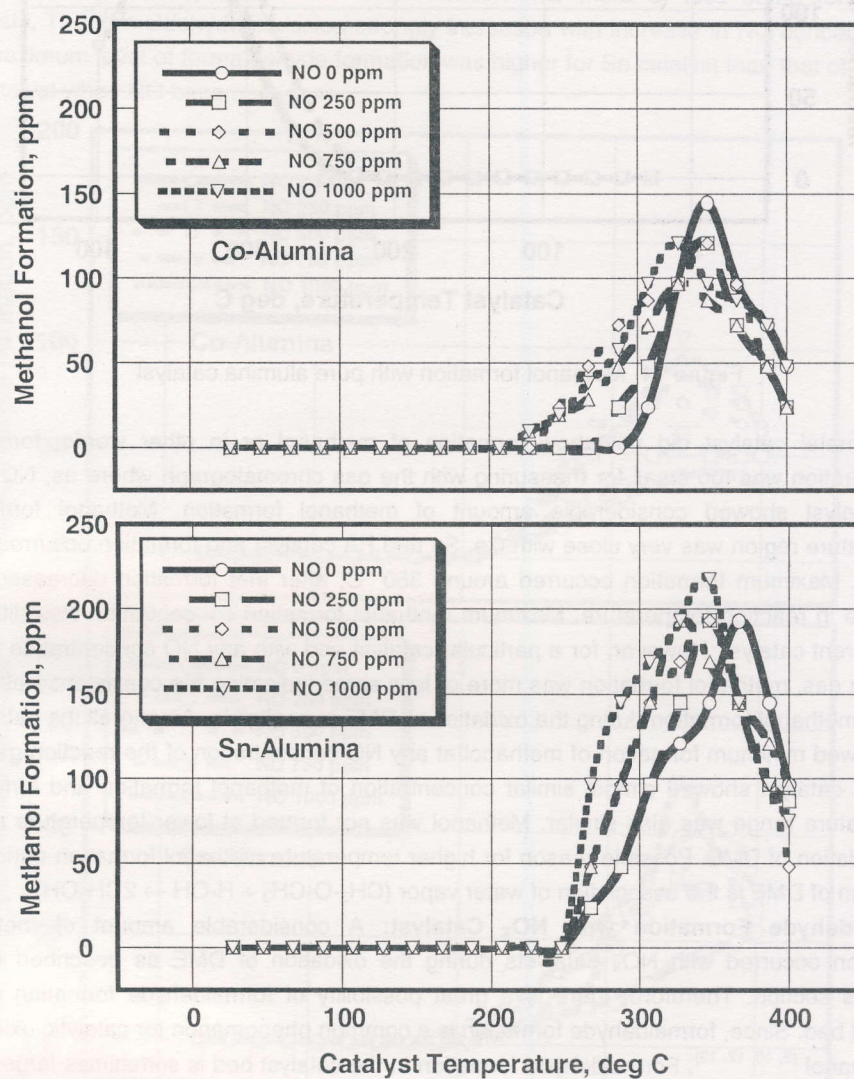

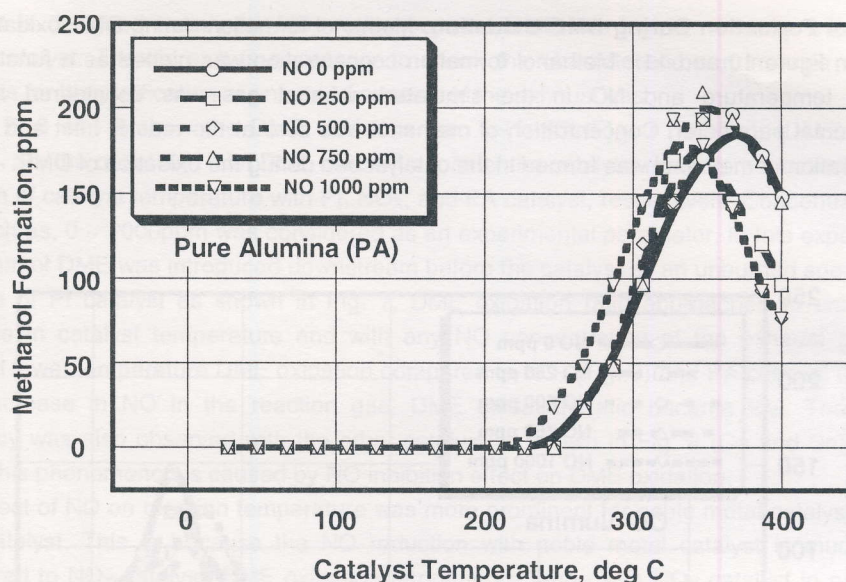Figure 10 Methanol formation with NO_X catalyst

Figure 11 Methanol formation with pure alumina catalyst

Noble metal catalyst did not show formation of methanol or in other words, formation concentration was too small for measuring with the gas chromatograph where as, NO_X and PA catalyst showed considerable amount of methanol formation. Methanol formation temperature region was very close with Co, Sn and PA catalyst and formation occurred after 200 $^{\circ}$C. Maximum formation occurred around 350 $^{\circ}$C, after that formation decreased with increase in reaction temperature. Maximum methanol formation concentration was different for different catalyst. However, for a particular catalyst and with any NO concentration of the reaction gas, methanol formation was more or less same indicating the coexistence effect of NO on methanol formation during the oxidation of DME is negligible. Among all the catalysts, Co showed minimum formation of methanol at any NO concentration of the reaction gas. Sn and PA catalyst showed almost similar concentration of methanol formation and formation temperature range was also similar. Methanol was not formed at lower temperature during the oxidation of DME. Possible reason for higher temperature methanol formation during the oxidation of DME is the association of water vapor ($CH_3\text{-}O\text{-}CH_3 + H\text{-}OH \rightarrow 2CH_3OH$).

Formaldehyde Formation with NO_X Catalyst: A considerable amount of methanol formation occurred with NO_X catalysts during the oxidation of DME as described in the previous section. Therefore, there is a great possibility of formaldehyde formation in the catalyst bed. Since, formaldehyde formation is a common phenomenon for catalytic oxidation of methanol [9, 10, 12, 13]. Formaldehyde formation in the catalyst bed is sometimes larger than the formaldehyde on the engine out exhaust gas and it poses serious problem for practical use. In this research the inlet concentration of formaldehyde was zero and outlet concentration of formaldehyde was formed in the catalyst bed. Formaldehyde formation for all the catalysts used in this research was investigated carefully.

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During the DME oxidation with noble metal catalyst, formaldehyde formation was hardly detectable. On the other hand, NO_X (Fig. 12) and PA (Fig. 13) catalyst showed considerable amount of formaldehyde formation. This is due to the methanol formation during DME oxidation with NO_X and PA catalyst. Figure 12 and 13 show the formaldehyde concentration at the reactor outlet for various NO concentrations in the simulated exhaust gas. Figure 14 shows formaldehyde formation with all the catalysts tested in the present work where concentration of NO in the simulated exhaust gas was 1000ppm.

Without NO formaldehyde formation concentration was more or less same for all the catalysts. The formaldehyde emission strongly increased with increase in NO concentration. The maximum level of formaldehyde formation was higher for Sn catalyst than that of Co and PA catalyst when NO became higher.
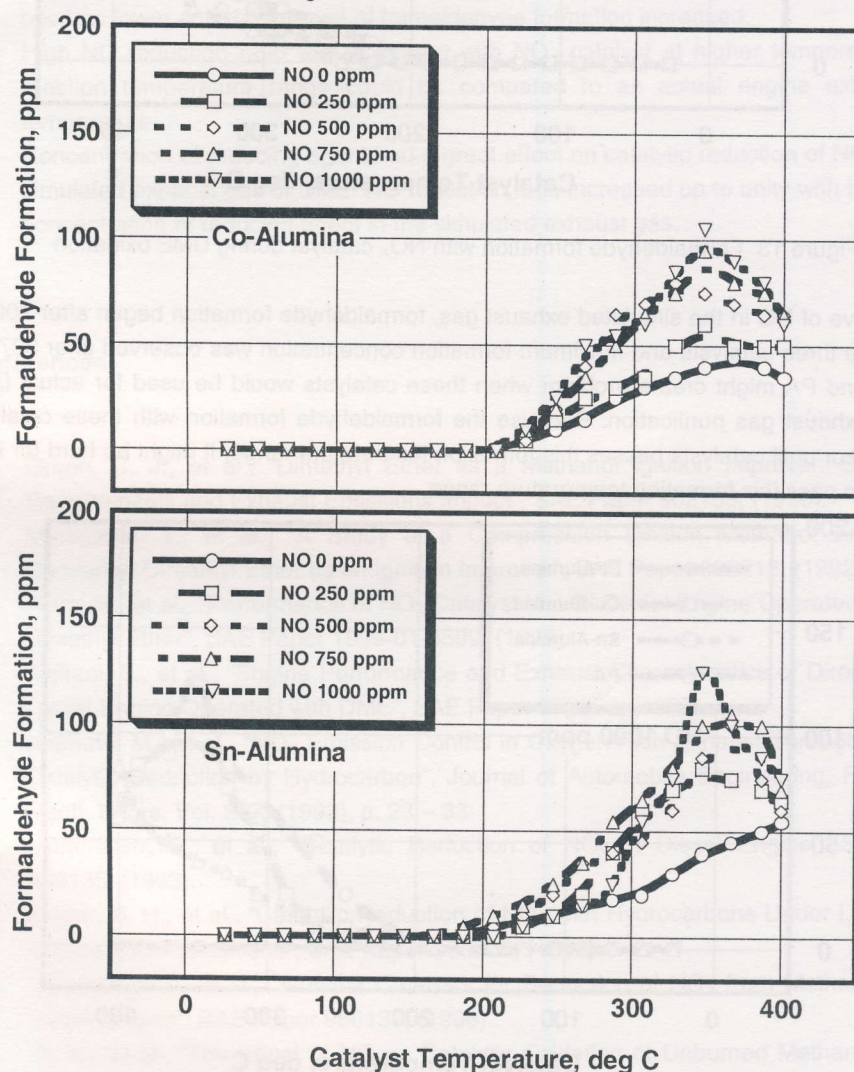

Figure 12 Formaldehyde formation with NO_X catalyst during DME oxidation
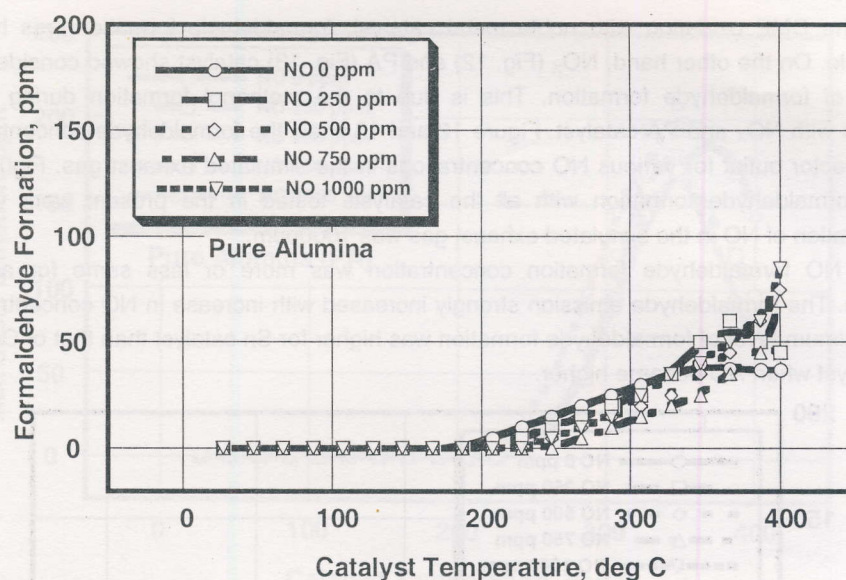
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Figure 13 Formaldehyde formation with NO_X catalyst during DME oxidation

Irrespective of NO in the simulated exhaust gas, formaldehyde formation began after 200 °C with these three catalysts and maximum formation concentration was observed after 327 °C. Co, Sn and PA might create problem when these catalysts would be used for actual DME engine exhaust gas purification. Because the formaldehyde formation with these catalysts would occur until catalysts passes this formation temperature range. It might be hard for NO_X catalyst to pass this formation temperature range.
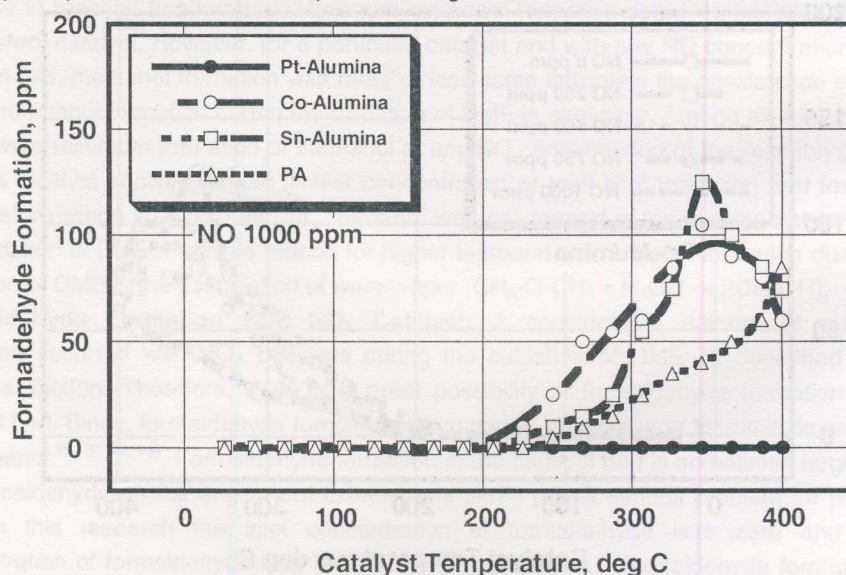

Figure 14 Comparison of formaldehyde formation among the catalysts

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Discussions

In this research catalytic purification of DME, formaldehyde and NO were investigated with NO_X catalyst (Co and Sn) in comparison with noble metal and PA catalyst. The following results were obtained from this research:

1. Low temperature DME oxidation was observed with Pt whereas Co, Sn and PA showed higher temperature DME oxidation. This might be a problem if low temperature DME oxidation is necessary with NO_X catalyst.

2. Considerable amount of methanol and formaldehyde were formed during the use of Co, Sn and PA catalyst at a certain temperature range. However, noble metal catalysts did not show any formation of formaldehyde. With increase in the NO, DME oxidation ratio became lower and the amount of formaldehyde formation increased.

3. High NO reduction ratio was observed with NO_X catalyst at higher temperature. This reaction temperature range could be compared to an actual engine exhaust gas temperature.

4. Concentration of reducing agent had a great effect on catalytic reduction of NO from the simulated exhaust gas of DME. NO reduction ratio increased up to unity with increase in concentration of reducing agent in the simulated exhaust gas.

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AN IMPROVED ALGORITHM FOR FINDING OUT THE PEG STATUS AT ANY MOVEMENT OF M-TH DISK OF THE TOWER OF HANOI

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Abstract

Tower of Hanoi is a familiar puzzle used to show the principle of mathematical induction, the power of recursive algorithms etc. Its general solution is based on recursion method that can be defined generally as the method of solving a problem by dividing it into two or more sub-problems, each of them are same like the original problem in nature, but smaller in size. This paper shows the status of all three pegs after each movement of any disk. It also computes the moves of a particular disk mathematically without solving all disks' movement. Based on this mathematical analysis a new algorithm is devised here.

I. Introduction

The Tower of Hanoi, a popular puzzle of the late nineteenth century consists of three pegs mounted on a board together with disks of different sizes, with the largest on the bottom [1,2,4,5,12]. No existing algorithm is able to describe the status of pegs at any movement of a particular disk without solving all disks movement. Its general solution is based on recursion method that can be defined generally as the method of solving a problem by dividing it into two or more sub-problems, each of them are same like the original problem in nature, but smaller in size [1,2,4,5,12].

Many researchers have tried to state good solutions of the tower of Hanoi problem in different solution [3,6-11]. In [6] a non-recursive solution of multi-peg tower of Hanoi has been given, where intermediate peg is more than one. If we apply this analysis in our single temporary peg tower of Hanoi problem it will compute all disks movement to tell the description of each peg after each movement. Also in [3] that computes all disks to solve a single disk's movement.

The rules of the tower of Hanoi can be summarized as follows: If n disks are arranged on the first peg largest at the bottom and each disk is sitting on a large disk then transferring of the n disks to the third peg will characterize the problem according to the following five conditions:

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