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Formation and Control of Nitric Oxide from Fuel Nitrogen
(2nd Report, Formation and Decomposition of Fuel-NO in
Diffusion Flames)

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Problems in the formation mechanism of fuel-NO in diffusion flames are researched by adding ammonia or nitric oxide to fuel or combustion air. Conversion ratios of ammonia to nitric oxide in diffusion flames are lower than those in premixed flames because added ammonia is decomposed in fuel-rich region near the inner flame surface. Conversion ratios are strongly affected by the amount of air included in fuel. Formation processes of fuel-NO in hydrogen diffusion flames can be explained by reaction scheme based on extended Zeldovich mechanism as well as in premixed flames if an assumption is made that fuel-N decays finally into N atom. Decomposition reaction of added NO plays a very important role in methane diffusion flames.

1. Introduction

Current concerns over nitrogen oxide emissions to the atmosphere have necessitated a more fundamental examination of the nitrogen oxide formation. Recent studies (1) (2) have indicated that nitrogenous species such as hydrogen cyanide or ammonia are detected in fuel-rich hydrocarbon flames even if nitrogenous species except for nitrogen molecule are not included in fuel. These species can be easily oxidized to NO. Therefore it is more important to investigate the role in the NO formation process of the inner flame region of the diffusion flame where nitrogenous species may be formed.

It seems difficult to discuss the formation process of NO by relating the emission level of NO with the operating parameter (thermal-NO(3) - (7), fuel-NO(8) - (12)) because the mixing process of fuel and air in combustion chamber has delicate effects. The quantitative understanding of NO formation process through detailed measurements(13) - (17) of concentration and temperature is also difficult because of the existence of the velocity distribution, diffusion of NO and fluctuation of flames. Fundamental data, such as the effect of the fuel kind, the relation between thermal-NO and fuel-NO, differences of NO formation process in diffusion flames and in premixed flames, have not been reported. Recognizing this situation on NO research and the results of our first report(18), an experiment was done using cylindrical diffusion flames.

2. Experimental

A known amount of NO or NH_3 is added to fuel or to combustion air and the exhaust concentration of NO is analyzed to get information on the formation mechanism of nitric oxide from fuel nitrogen.

The schematic model of the combustor is shown in Fig. 1. Gaseous fuel is injected into vycor glass tube (5) coaxially through fuel injection nozzle (4) of 2mm in inside diameter. Combustion air is fed uniformly through an air inlet pipe (2) and a porous metal (3). The flow rate of the fuel is set at $152 \text{ Ncm}^3/\text{s}$ for H_2 flames and 38 Ncc/s for CH_4 flames so as to equalize the flow rate of combustion air for both fuels. The combustion gas is sampled at the exit of the combustor where

Fig. 1 Schematic of the diffusion flame burner

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the cross sectional concentration profile of NO is flat. A chemiluminescent type analyzer is used for NO_x measurement and a magnetic type for O₂ measurement. Ammonia is determined by either the neutralization titration method or the indophenol method and hydrogen cyanide is determined by an ion electrode method. The quantity of fuel-N is defined as the quantity when one percent of fuel-N has all converted to NO_x, NO_x* (18) (NO_x* = NO_x measured x combustion gas quantity/stoichiometric combustion gas quantity) amounting to 1340 ppm for CH₄ flames and 1520 ppm for H₂ flames.

3. Results

3.1 Results of the H₂ diffusion flame and discussion

Figure 2 shows the experimental results of the hydrogen diffusion flame in which pure NO gas is added to air (subscript; out) or to fuel (subscript; in) before combustion to know the reaction mechanism of NO. The mark (o) in this figure represents the NO_x* emission with no addition of NO (thermal-NO). The abscissa is the overall equivalence ratio $\bar{\lambda}$ ($\bar{\lambda}$ = air fuel ratio/stoichiometric air fuel ratio). Fairly much thermal-NO_x is exhausted even when no NO is added. When pure NO is added to air, the NO_x* emission is less than the sum of the thermal-NO and the added NO (1520 ppm for 1 % Fuel-N). This implies that decomposition reactions of NO cannot be ignored in comparison with formation reactions of NO. NO added to fuel is not decomposed in combustion process. In any case, the decomposition ratio of added NO is so low that the decomposition reaction of NO seems less important than the formation reaction of NO in the process of thermal-NO formation for the hydrogen flame.

Figure 3 shows the NO_x* emissions for the H₂ flame. Ammonia is added to the H₂ fuel in order to examine the formation process of fuel-NO. Hitherto, nitrogenous

Fig. 2 NO_x* emissions vs. $\bar{\lambda}$ (H₂ + NO)

compounds contained in fuel have been considered more or less to be a NO_x formation factor (8) - (12). This experiment, however, gives an interesting result especially when 1 % of fuel-N is added to fuel. The NO_x emission is less than the level of thermal-NO. The tendency is remarkable under fuel-rich flames added with 5 % fuel-N and so cannot be accounted for by the reduction of thermal-NO or the variations of the flame structure. Therefore it seems that added NH₃ reacts with thermal-NO and decomposes the latter. On the other hand, NH₃ added to air is a formation factor of NO_x. This fact indicates that the yield of NO from fuel-N differs according to the combustion condition.

Figure 4 shows the result of an experiment for H₂ fuel with 1.13 times N₂ gas. As the thermal-NO formed is less than 30 ppm, it will be easy to separate the quantity of the fuel-NO from the thermal-NO. Ammonia added to fuel increases the NO_x* emissions. But the conversion ratio of NH₃ to NO is much lower than that of premixed flame (18). It is supposed to explain this phenomenon that in a diffusion flame the fuel-N may be decomposed inside of the flame where oxygen concentration is low. Then, some amount of air is added to fuel to verify the above supposition.

Fig. 3 NO_x* emissions vs. $\bar{\lambda}$ (H₂ + NH₃)

Fig. 4 NO_x* emissions vs. $\bar{\lambda}$ (H₂ + N₂ + NH₃)

3.2 Modeling of fuel-NO formation process

By allowing a part of combustion air to flow into fuel pipe ① through a bypass line, the premixed air fuel ratio λ_1 , is made changeable keeping the total air ratio $\bar{\lambda}$ the same. In this examination, a fuel nozzle of 4 mm in inner diameter is used lest the velocity of the premixed fuel should become too large. Figure 5 shows NO_x^* emissions as a function of λ_1 . With an increasing λ_1 , the flame structure changes from a diffusion flame to a double construction flame with an inner flame, and further to a premixed flame. With these variations of the flame structure, the NO_x^* emissions decreases remarkably in case of no addition of NH_3 . On the other hand, in case of $\bar{\lambda} = 1.3$ (Fig. 5-(a)), the NO_x^* emission with NH_3 addition begins to increase immediately after the λ_1 exceeds 0.3. And as λ_1 approaches unity, the flame turns to a premixed one and the conversion ratio of fuel-N to NO runs up to about 100 % (arrow mark on the right ordinate indicates the concentration of fuel- NO_x^* with 100 % conversion).

Figure 5(b) shows the results of $\bar{\lambda} = 0.8$. It is observed that in the range of small λ_1 the NO_x^* emission decreases with

Fig. 5 Effect of premixed air on NO_x^* emissions

an increasing fuel-N %. But as λ_1 increases, the influence of the amount of fuel-N diminishes a little later after NO_x^* exceeds the thermal- NO_x . And when λ_1 becomes larger, the NO_x^* emission increases with an increasing fuel-N and the effect of $\bar{\lambda}$ disappears [comparing Fig. (a) with Fig. (b), NO_x^* is nearly equal for the same λ_1]. From these experiments, it was found that the formation of the fuel-NO depends strongly on the quantity of air included inside of the flame.

The flame of an operating utility burner is rather a slightly premixed flame than a pure diffusion flame. Then, referring to the results of the hydrogen flame and the first report (18), a modeling of the formation process of fuel-NO is tried for the simplest hydrogen flame. Besides, from a standpoint of making an overall formation model, all of the fuel-N are assumed to become nitrogen atoms in the process of the combustion reaction.

Figure 6 shows a fuel-NO formation model which corresponds to Fig. 5. It is ascertained in the investigation by Ogasawara et al. (14) that the thermal-NO is formed mainly in the flame zone which is shown in Fig. 6 marked N . In the case of (a) where $\bar{\lambda} = 1.3$, $\lambda_1 \neq 0$, as the oxygen concentration is fairly low inside of the zone N (14), most of nitrogen atoms (NH_m ?) from NH_3 becomes N_2 through the next reaction.

(The following explanation will be also done according to the extended Zeldovich mechanism). Some part of the thermal-NO formed in the flame zone marked N may be decomposed in the inner flame zone through the reaction $(1)_f$. On the other hand, in case of $\bar{\lambda} = 0.8$, (a), NH_m remaining with-

Fig. 6 Fuel NO formation model

out passing through the flame zone $\bar{\lambda}$ will decompose thermal-NO in the downstream region of the flame. Therefore, the NO_x^* emission decreases with an increasing amount of NH_3 addition.

Fig. (c) and (c)' show models for large λ_1 . Premixed inner flames are formed and next reactions $(2)_f$ and $(3)_f$ take place more rapidly than the reaction $(1)_f$. So much fuel-NO may be formed.

From the results of the premixed flame (18) and Fig. 2, it follows that the fuel-NO would be formed rapidly near the inner flame and that most of them would persist in the exhaust gas. Flames (b) and (b') are in the middle of the above two conditions. NH_3 is apt to remain near the inner flame for little λ_1 and NO is apt to be formed for large λ_1 . The formation process of NO from nitrogen in fuel seems to become clear from the above modeling for the hydrogen flames.

3.3 Results of the CH_4 diffusion flames and discussion

Figure 7 show the NO_x^* emission in case of small addition of pure NO to fuel or combustion air. The decomposition of NO added to fuel is significant under air-rich combustion and the decomposition of NO added to air is remarkable under fuel-rich combustion. So it may be concluded that the added NO is decomposed by passing through not the fuel-rich region but the flame zone where main combustion reactions proceed. In view of the fact that such decomposition does not occur in H_2 flames, this phenomenon of NO decomposition may also be as peculiar to the hydrocarbon flame as the formation of 'Prompt NO'. The fact that some of the added NO is decomposed, means that the decomposition reaction plays a very important role in the formation process of thermal-NO as well as the formation reaction.

Figure 8 shows the result of the CH_4 flame with added NH_3 . The NO_x^* emission is the larger for the higher $\bar{\lambda}$ and the more fuel-N. The behavior is similar to

Fig. 7 NO_x^* emissions vs. $\bar{\lambda}$ ($\text{CH}_4 + \text{NO}$)

the results of H_2 flame with little thermal-NO (Fig. 4). Fig. 9 shows the influence of the amount of fuel-N at $\bar{\lambda} = 1.3$. The decomposition ratio of added NO increase with an increasing NO addition. In case of small addition of NH_3 , the NO_x^* emission is almost the same as that of the NO addition. But for large NH_3 addition, the conversion ratio of NH_3 to NO_x is fairly low.

4. Discussion of NO reaction mechanism

From the experiments of diffusion flames, it was found that the decomposition reaction of NO played an important role in methane flames. To discuss the reaction mechanism of this decomposition phenomenon, some experimental studies using the simplest premixed flame were carried out and the results were compared with those of reaction model calculation. Experimental works (19)(20) with NO added premixed flames or analytical research (21) in high temperature region related to engines have been reported, and the behavior of NO is well explained by only the extended Zeldovich reaction mechanism. Otherwise, in the well-stirred reactor (22) or flat flames (23) (24) with rather lower flame temperature a considerable part of added NO is found decomposed in the fuel-rich condition. In this field, no discussion including chemical reaction models nor studies on the effect of fuel type seem to have been done.

Fig. 8 NO_x^* emissions vs. $\bar{\lambda}$ ($\text{CH}_4 + \text{NH}_3$)

Fig. 9 Effect of the quantity of NH_3 or NO on NO_x^* emissions

4.1 Experimental results and discussion with a premixed flat flame.

The experimental apparatus and the procedure of the test are the same as the case of the 1st report⁽¹⁸⁾, except use of pure NO instead of NH_3 as an additive. Figure 10 represents the effects of flame temperature on the NO_x retention ratio (NO_x mass flow rate ratio before and after combustions) in CH_4 premixed flames. In case of $\lambda = 1.3$ almost all amount of added NO comes out through the flame, and there seems no temperature effect under the experimental condition. On the contrary, in case of $\lambda = 0.8$ some amount of added NO is converted to other species and the amount of decomposition increases with a drop in flame temperature. The decomposition of added NO is also found to occur in and near the flame zone.

Figure 11 shows the effects of air-fuel equivalence ratio on the NO_x retention ratio. In the CH_4 flame, the retention ratio is nearly 100% on the air-rich side and goes down on the fuel-rich side approaching zero at $\lambda = 0.6$. The result with H_2 flame is also plotted and the behavior is a little different from that with CH_4 flames. This significant decomposition of added NO in CH_4 flames seems to correspond to the decomposition phenomenon in CH_4 diffusion flames (Fig. 7) and so a detailed discussion considering the elementary reaction rate will be done next.

Just as in the 1st report, 52 forward and backward reactions which include CH_4 decomposition reactions are taken into consideration. The initial gas is given as $\text{CH}_4 + \text{NO} + \text{Air}$, and the following gas composition is determined numerically. The results are shown in Table 1.

Fig. 10 Effect of flame temperature on the NO_x retention ratio

Fig. 11 NO_x retention ratio as a function of λ , fuel n% and kinds of fuel

The remarkable decomposition of added NO which occurred in the experiment of fuel-rich methane flame ($\lambda \approx 0.6$) does not appear in this calculation. This suggests that some other reactions which include NO must be taken into account.

4.2 A technical model of prompt NO

According to the above calculation under fuel-rich condition, NO decomposition reactions are mainly

and the amount of reaction $(1)_f$ is nearly the same of reaction $(3)_b$. In other words, if there exist some reactions which supply N atoms, NO added would decompose in large amounts through reaction $(1)_f$. Then, reaction models which will be able to explain our experimental results are discussed next. When a certain amount of Fuel N (1%), for example, has changed into N atoms in 0.1 ms, NO which is initially given changes as shown in Fig. 12 through reactions $(1) - (3)$. Thus, the phenomenon is well explained that NO is formed rapidly when initial concentration is low and it is decomposed rapidly when initial concentration is high. But in this model the amount of decomposed NO can't exceed the amount of released N atom, so the assumption that a certain amount of N atom is released must be abandoned. Figure 12 will correspond to the phenomenon

Table 1 Decomposition of NO (calculated)

Initial conditions			Results	
λ	Temperature °K	(NO) in ppm	$(\text{NO})_t = \frac{1}{\text{ms}} \times \text{ppm}$	Net NO increased ppm
0.6	1784	1000	931.7	- 1.6
0.8	2097	1000	950.1	- 2.7
1.3	1949	1000	997.9	+ 2.6

Fig. 12 Formation and decomposition model of fuel-NO

which was explained for the H_2 diffusion flames that fuel-N could be either formation or decomposition factor of NO depending on the level of thermal-NO. From this figure it can be pointed out that the concentration of NO decomposed by fuel-N doesn't decrease to the level of equilibrium concentration $(NO)_{eq}$.

Now, we reform the model on the assumption that the concentration of N atom is constant independently of initial NO concentration. Taking into account the forward reactions of extended Zeldovich mechanism (1) - (3) yields the following kinetic equation.

$$\frac{d(NO)}{dt} = k_{2f}(N)(O_2) + k_{3f}(N)(OH) - k_{1f}(N)(NO)$$

Under the initial condition $(NO) = (NO)_{in}$ at $t = 0$ and the assumption that (N) is constant concentration in duration τ and that other species except for NO and N take equilibrium concentration, above equation is solved as

$$(NO) - X = e^{-k_{1f}(N)\tau} \{ (NO)_{in} - X \} \quad \text{---(1)}$$

$$\text{where } X = \{ k_{2f}(O_2) + k_{3f}(OH) \} / k_{1f} \quad \text{---(2)}$$

Because the relation between $(NO)_{in}$ and (NO) becomes a linear line passing through (x, x) with a slope of $e^{-k_{1f}(N)\tau}$ (< 1) in co-ordinates with $(NO)_{in}$ as the abscissa and (NO) as the ordinate, decomposition ratio of added NO increase a little with an increasing $(NO)_{in}$. This model seems to be more suitable than the previous model (Fig. 12). In order to explain the experimental fact that the retention ratio of added NO decreases as fuel-richer side, (N) must take higher concentration on fuel-richer side. As the slope changes from 1 to 0, the concentration of NO approaches X as is seen from Eq. (1). Although X is expected to be dependent on the non-equilibrium concentration of the combustion gas, substituting the equilibrium concentration into the equation (2) for convenience sake, its value turns out to make a solid line in Fig. 13 and becomes larger than the equilibrium NO concentration shown as a dotted line. The curve ① shows $(X)_{eq}$ of methane adiabatic flame and it takes smaller value as the flame becomes richer. The curves ② and ③ show the exhaust NO concentration schematically for $(NO)_{in}$ which is larger than $(X)_{eq}$ and zero, respectively. The peak of curve ③ is expected to correspond to the prompt NO formation. The decrease of prompt NO on the fuel richer side may be caused by the lowering of X and its decrease on the fuel-lean side may be caused by the lowering of N atom concentration. As described above, the assumption that the N atom in the flame zone takes higher concentration as the flame becomes fuel-richer, can nearly account for not only the rapid formation of NO but also the rapid decomposition of added NO through the well-known extended Zeldovich mechanism. And so both of them seem to be a similar phenomenon. There have been two opinions in relation to the formation of prompt NO; one is that prompt NO is caused mainly by the nitrogenous intermediates such as HCN or CN(1)(2)(25)(26)(27), and the other is that it is caused by the overshoot of oxygen atom(28)(29). If the formation of prompt NO is caused by a similar mechanism to that of rapid NO decomposition, the former opinion seems better as a systematic explanation of the phenomenon. In this paper, a qualitative explanation could be done by introducing N atom, but in the actual reactions NO formation or decomposition may occur through other nitrogenous species such as HCN, NH_2 , NH , ... So, it seems important to investigate further the reactions involving nitrogenous species.

Fig. 13 Predictions of (NO) and (X)

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Fig. 14 Radial distributions of species in CH_4 laminar diffusion flame

4.3 Concentration distributions in CH_4 diffusion flame

Although the inner flame region of the diffusion flame seems to correspond to the premixed fuel-rich flame, it is impossible to apply the knowledge on the premixed flame directly to the diffusion flame because the flame mechanisms of the both flames are completely different. Therefore, it is necessary to measure the detailed concentration distributions of some kind of chemical species in the diffusion flame. Figure 14 is an illustration for CH_4 flame. A pretty concentration of HCN is detected to be formed from added NH_3 a little inside of the flame and a complexity of the reactions including nitrogenous species is suggested. Also it is shown that fuel-NO is formed rapidly in the vicinity of the flame surface (the given flame is a stable laminar one). A detailed discussion will be done in future research.

5. Conclusions

Formation and decomposition processes of fuel-NO in cylindrical diffusion flames were studied and the following results were obtained.

- (1) Conversion ratios of ammonia to nitric oxide in diffusion flames are lower than those in premixed flames, because added ammonia is decomposed in fuel-rich region near the inner flame surface. Conversion ratios are strongly affected by the amount of air included in fuel.
- (2) Effects of fuel-N on the yield of NO vary widely from formation factor to decomposition factor according to the condition because thermal-NO reacts directly with fuel-N.
- (3) Formation processes of fuel-NO in hydrogen diffusion flames can be explained by reaction scheme based on extended Zeldovich mechanism as well as in premixed flames if an assumption is made that fuel-N decays finally into N atom.
- (4) Decomposition reaction of added NO plays a very important role in methane diffusion flames, but does not in hydrogen flames.
- (5) The assumption that concentration of N atom in premixed methane flame zone is higher on fuel-rich side, may account for not only the rapid decomposition of added NO but also the formation of "prompt NO".
- (6) The formation of NO from the nitrogen compounds in fuel occurs rapidly in the vicinity of the flame surface, where HCN is also detected.

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