

FORMATION OF NITROGEN DIOXIDE IN COMBUSTION PROCESSES

FORMATION DU DIOXYDE D'AZOTE DANS LES PROCESSUS DE COMBUSTION

Jurong Zhuang and Wolfgang Leuckel
Engler-Bunte-Institute, University Karlsruhe, Germany

ABSTRACT

NO₂ detected experimentally in many combustion processes has been considered as a result of a 'quenching effect', not as being formed in combustion processes themselves. With a radiative burner generating a laminar one-dimensional premixed combustion flame, a remarkable NO₂-peak was measured directly above the burner surface. Extensive theoretical investigations were performed based on this phenomenon by means of a Chemkin program Premix as well as the Glarborg-Hadvig or Miller-Baumen reaction mechanism, also considering measuring errors of the sampling system including 'quenching effect'. Results show that the four NO₂-related reactions in Chemkin code and their kinetic data are not adequate for quantitative simulation of the NO₂-profile in the combustion process; and that the reaction responsible for the 'quenching effect' is not sufficient to explain the measured NO₂-peak at the radiative burner. Through those discussions and further experimental results it can be concluded that there must be other mechanisms of NO₂ formation in the flame which are until now not understood.

RESUME

Le NO₂ détecté expérimentalement dans plusieurs processus de combustion a été considéré comme un résultat d'un effet du "quenching" et non comme formé dans le processus de combustion. Avec un brûleur générant une flamme de prémélange laminaire et monodimensionnelle, un remarquable pic de NO₂ était mesuré directement au dessus de la surface du brûleur. De vastes investigations théoriques sont accordées à ce phénomène en utilisant le programme Chemkin Premix aussi bien avec le mécanisme réactionnel de Glarborg-Hadvig que celui de Miller-Bowmen en considérant les erreurs de mesure du système, incluant l'effet du quenching. Les résultats montrent que le NO₂ lié aux réactions et à leurs données cinétiques n'est pas adéquat pour la simulation quantitative du profil de NO₂ dans le processus de combustion et que la réaction responsable du quenching est insuffisante pour expliquer le pic de NO₂ mesuré dans le brûleur. Par ces discussions et avec plus de résultats expérimentaux on peut conclure qu'il doit y être d'autres mécanismes inexplicables de formation de NO₂ dans la flamme.

INTRODUCTION

Because of increasingly strong emission control regulations, one major focus in combustion technology refers to the formation of nitrogen oxides (NO , NO_2 and N_2O) in combustion processes. The relatively low NO_2 concentration compared to NO was often neglected. Although some authors have discussed the formation of NO_2 in combustion processes, one concluded that the NO_2 -concentration measured in experiments was caused by a so-called 'quenching effect'[1-4] during probe sampling.

One of the methods to reduce the formation of nitrogen oxides is to reduce the combustion temperature, which lowers effectively the formation of thermal nitrogen monoxide(NO). But as the thermal NO was decreased, higher NO_2/NO ratios were measured. This challenges researchers to pay more attention to NO_2 -formation. The NO_2 -formation in the combustion processes cannot be neglected anymore.

In this paper, measured NO_2 -peaks above a radiative burner are being extensively discussed by means of kinetic reaction simulation, using the Chemkin code Premix and its internal kinetic and equilibrium data of combustion reactions, together with consideration of measuring errors of the sampling system including the 'quenching effect'. Through this discussion it can be evaluated whether the measured NO_2 -concentration in this laminar one dimensional premixed flame results only from measuring errors or -up to some extent- is really being formed in the combustion flame itself.

EXPERIMENTAL SETUP

A radiative burner with a porous burner plate has been investigated experimentally. As shown in Fig.1, fuel gas and air streams are mixed before they pass a porous plate ($\phi 90\text{mm}$) in which they are heated through heat exchange with the porous material. Near to the plate the mixed gas temperature reaches its ignition temperature and ignites. The surface of the porous plate has a high temperature, it radiates and transfers heat to the surroundings, thus lowering the peak temperature of the flame considerably. The fuel gas used for the investigations was methane. The porous burner plate (sinter metal) operates at a heat loading of $271\text{-}705\text{ kW/m}^2$. This burner system has good symmetry and can therefore be considered as a steady state laminar one dimensional premixed flame system.

Temperature and concentration fields in this combustion system were measured by thermocouples and a sample sucking system, respectively.

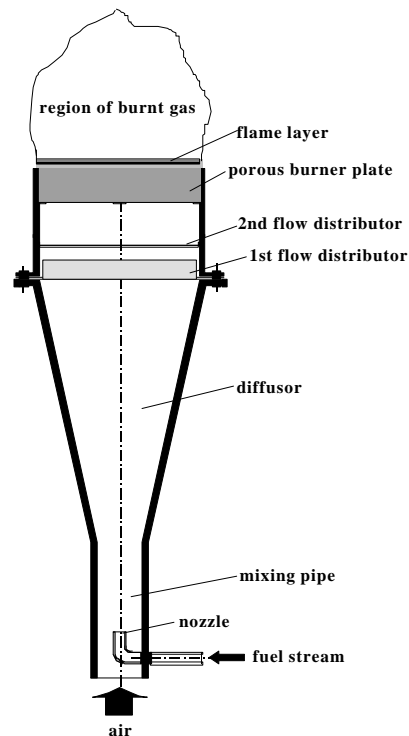


Figure 1. A radiative burner with a porous plate

RESULTS AND DISCUSSIONS

Observed Phenomenon

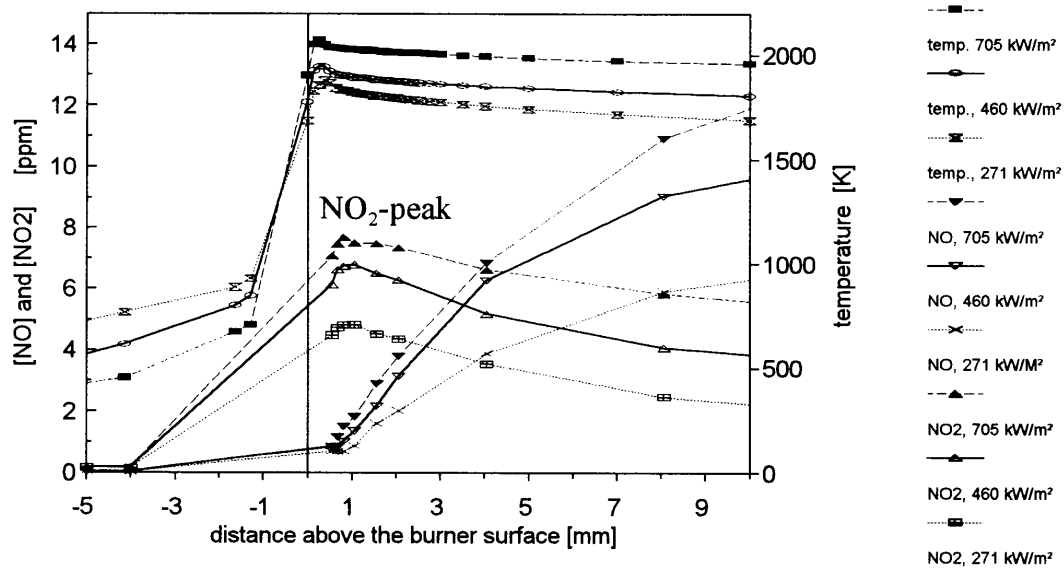


Figure 2. NO_2 -peak directly above the surface of the burner plate (air ratio=1.05)

As shown in Fig.2 typically for air ratio ≥ 1.0 , there is a relatively high NO_2 -peak immediately above the surface of the burner plate for all three heat loadings investigated. At this position, only low NO-concentrations have been formed by the prompt-NO mechanism, since much thermal NO cannot yet be present due to relatively low temperature and short residence time of the gas in the hot zone. From experiments it was found that this NO_2 -peak is independent of the material of the burner plate and also independent of the material, of the design and, therefore, of the temperature of the sampling probe head, as shown in Fig.3. Further experiments revealed that NO_2 is being formed in the blue flame zone, especially in premixed flames. This effect has been measured also with many other combustion systems.

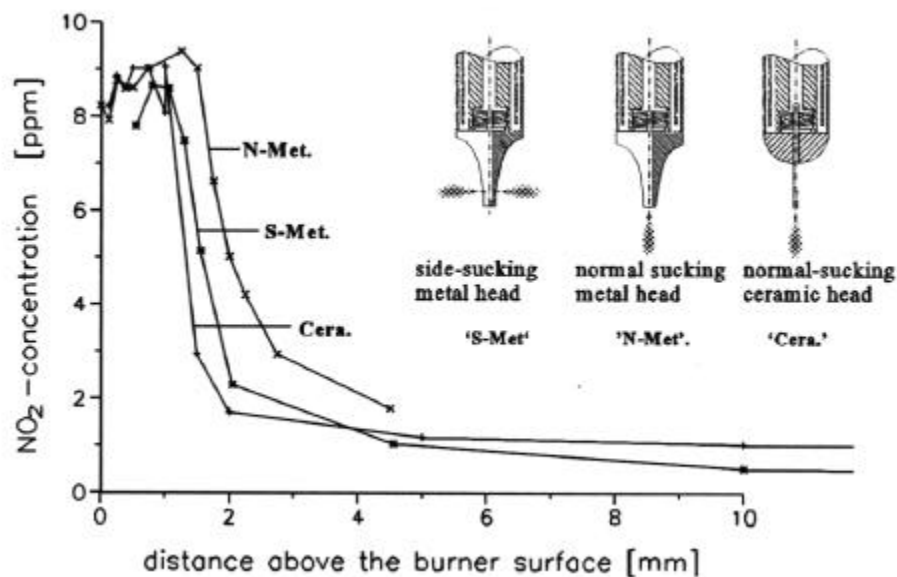


Figure 3. Independence of NO_2 -peak of the material and the design of the sampling probe head

Table 1. Analysis of the reaction rates of the four NO_x-related reactions

<i>reaction</i>		<i>NO+HO₂ = NO₂+OH</i>			<i>NO₂+M = NO+O+M</i>		
<i>X [mm]</i>	<i>T [K]</i>	<i>rf</i>	<i>rr</i>	<i>q=rf-rr</i>	<i>rf</i>	<i>rr</i>	<i>q=rf-rr</i>
-2.156	762.600	2.06e-11	1.38e-15	2.06e-11	5.30e-21	7.24e-22	4.57e-21
-1.678	798.400	1.36e-10	1.74e-14	1.36e-10	7.29e-20	1.43e-20	5.86e-20
-1.439	828.600	4.54e-10	1.02e-13	4.54e-10	4.40e-19	1.14e-19	3.25e-19
-1.200	920.600	1.49e-09	8.44e-13	1.49e-09	2.46e-17	8.29e-19	2.38e-17
-0.961	1095.000	4.43e-09	8.13e-12	4.42e-09	4.52e-15	5.88e-18	4.51e-15
-0.841	1183.000	7.97e-09	2.77e-11	7.94e-09	2.13e-14	2.74e-17	2.13e-14
-0.722	1270.000	1.27e-08	5.62e-11	1.27e-08	4.60e-14	1.08e-16	4.59e-14
-0.662	1314.000	1.45e-08	6.94e-11	1.45e-08	5.14e-14	2.27e-16	5.12e-14
-0.602	1358.000	1.47e-08	7.39e-11	1.46e-08	5.31e-14	4.44e-16	5.26e-14
-0.543	1401.000	1.24e-08	6.82e-11	1.23e-08	5.04e-14	8.51e-16	4.96e-14
-0.423	1489.000	5.41e-09	3.96e-11	5.37e-09	4.11e-14	2.66e-15	3.85e-14
-0.363	1532.000	3.33e-09	3.04e-11	3.30e-09	4.14e-14	3.84e-15	3.76e-14
-0.304	1576.000	2.27e-09	2.66e-11	2.24e-09	5.10e-14	4.65e-15	4.63e-14
-0.244	1620.000	1.73e-09	2.60e-11	1.71e-09	7.23e-14	4.98e-15	6.73e-14
-0.124	1707.000	1.28e-09	2.96e-11	1.26e-09	1.77e-13	4.60e-15	1.72e-13
-0.005	1794.000	1.01e-09	3.59e-11	9.74e-10	4.48e-13	3.84e-15	4.44e-13
0.234	1861.000	8.65e-10	4.74e-11	8.18e-10	1.05e-12	3.23e-15	1.05e-12
0.713	1791.000	1.01e-09	4.69e-11	9.65e-10	7.11e-13	3.19e-15	7.08e-13

<i>reaction</i>		<i>NO_x+H = NO_x+OH</i>			<i>NO_x+O = NO_x+O₂</i>		
<i>X [mm]</i>	<i>T [K]</i>	<i>rf</i>	<i>rr</i>	<i>q=rf-rr</i>	<i>rf</i>	<i>rr</i>	<i>q=rf-rr</i>
-2.156	762.600	5.22e-13	3.49e-23	5.22e-13	3.123e-15	5.656e-20	3.123e-15
-1.678	798.400	1.03e-11	1.19e-21	1.03e-11	6.684e-14	5.460e-19	6.684e-14
-1.439	828.600	7.25e-11	1.67e-20	7.25e-11	5.126e-13	2.739e-18	5.126e-13
-1.200	920.600	5.30e-10	1.09e-18	5.30e-10	4.524e-12	7.643e-17	4.524e-12
-0.961	1095.000	3.19e-09	1.94e-16	3.19e-09	3.270e-11	4.291e-15	3.269e-11
-0.841	1183.000	9.15e-09	3.91e-15	9.15e-09	9.189e-11	2.235e-14	9.187e-11
-0.722	1270.000	1.48e-08	6.26e-14	1.48e-08	1.369e-10	8.293e-14	1.368e-10
-0.662	1314.000	1.63e-08	2.53e-13	1.63e-08	1.450e-10	1.441e-13	1.448e-10
-0.602	1358.000	1.55e-08	8.89e-13	1.55e-08	1.380e-10	2.346e-13	1.377e-10
-0.543	1401.000	1.29e-08	2.72e-12	1.29e-08	1.231e-10	3.609e-13	1.227e-10
-0.423	1489.000	5.77e-09	1.73e-11	5.75e-09	7.957e-11	8.235e-13	7.875e-11
-0.363	1532.000	3.82e-09	3.49e-11	3.78e-09	6.287e-11	1.215e-12	6.166e-11
-0.304	1576.000	2.88e-09	6.24e-11	2.82e-09	5.336e-11	1.789e-12	5.157e-11
-0.244	1620.000	2.44e-09	1.01e-10	2.34e-09	4.853e-11	2.586e-12	4.594e-11
-0.124	1707.000	2.09e-09	2.13e-10	1.87e-09	4.498e-11	5.118e-12	3.986e-11
-0.005	1794.000	1.91e-09	3.88e-10	1.52e-09	4.405e-11	9.487e-12	3.457e-11
0.234	1861.000	1.90e-09	6.04e-10	1.30e-09	4.698e-11	1.525e-11	3.173e-11
0.713	1791.000	1.82e-09	4.06e-10	1.41e-09	4.481e-11	1.009e-11	3.472e-11

Note: rf -- forward reaction rate 10⁶ [mol/(m³.s)]
 rr -- reverse reaction rate 10⁶ [mol/(m³.s)]
 q -- net reaction rate 10⁶ [mol/(m³.s)]

NO₂-Related Reactions and Comparison to Calculation with Chemkin

Based on the Glarborg-Hadvig[5] mechanism there are the following four NO₂-related reactions to be considered:



According to the kinetic data and the code-internal thermal equilibrium data of the reactions, net reaction rates of each reaction were calculated and compared against each other in Tab.1, where the net reaction rates, according to:

$$\text{net reaction rate}(q) = \text{forward reaction rate}(rf) - \text{reverse reaction rate}(rr)$$

in [10⁶ mol/(m³s)] are all positive. This means that, amongst the four considered reactions, reaction(1) is the only reaction which forms NO₂ in the combustion system, and the reactions (2-4) reduce nitrogen dioxide.

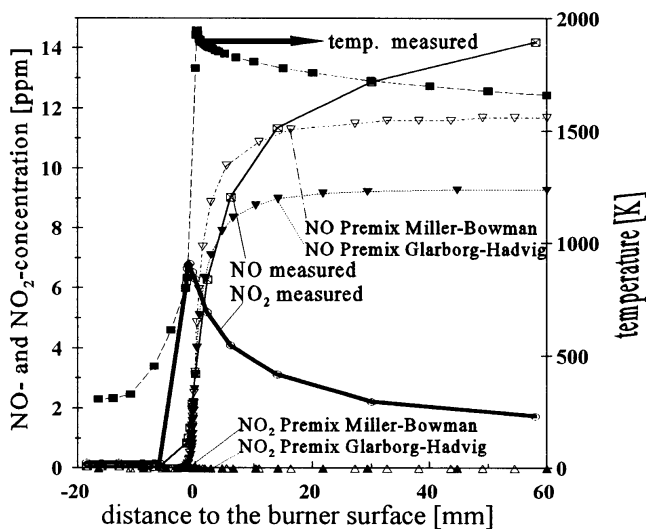


Figure 4. Comparison of the calculated NO₂-peak to the measured NO₂-peak (The measured NO- and NO₂-profile were shifted upstream by 2mm)

With the Chemkin program PREMIX, based upon a measured temperature profile for the steady state laminar one-dimensional premixed flame, a very small NO₂-peak is calculated according to the Glarborg-Hadvig or Miller-Bowmen mechanism[6] which is much lower than the measured one(Fig.4). The measured NO- and NO₂-curves in Fig.4 were shifted upstream by 2mm, in order to enable comparison with the calculated NO-curve. Furthermore, as is also shown in Fig.4, the NO₂-peak occurs always before the rise of the NO-curve (which results from prompt NO) at the position of radical-rich blue flame zone, both from calculation and from measurement. It is also remarkable that the measured NO-curve fits the calculated NO-curve quite well up to about 4-5mm (with

Glarborg-Hadvig mechanism) above the burner surface, whereas in the thermal NO formation zone the measured NO increases further, but the calculated NO-curves exhibit only a very low gradient.

Possible Measuring Errors

Several parameters may affect the measured NO₂-concentration. In order to investigate the formation of NO₂ in a combustion system, one must first consider measuring errors caused by the used sample-sucking-system. Those measuring errors may be divided into two categories: One is due to the total piping of the sampling system, including the analysing apparatus; the other may be

related to the 'quenching effect' occurring immediately near to the entrance of a cooled sampling probe.

The first category of measuring errors was investigated systematically in our earlier work[7] which reveals that, with the usual moisture content of 18% in the CH₄ flue gas and with our measuring conditions(13.5 l/h), only about 30% NO₂- and 70% NO-concentration of the sampled gas reaches the analyser(Fig.5), and that the transformation degree of NO to NO₂ is about 2% in the used sample-sucking system.

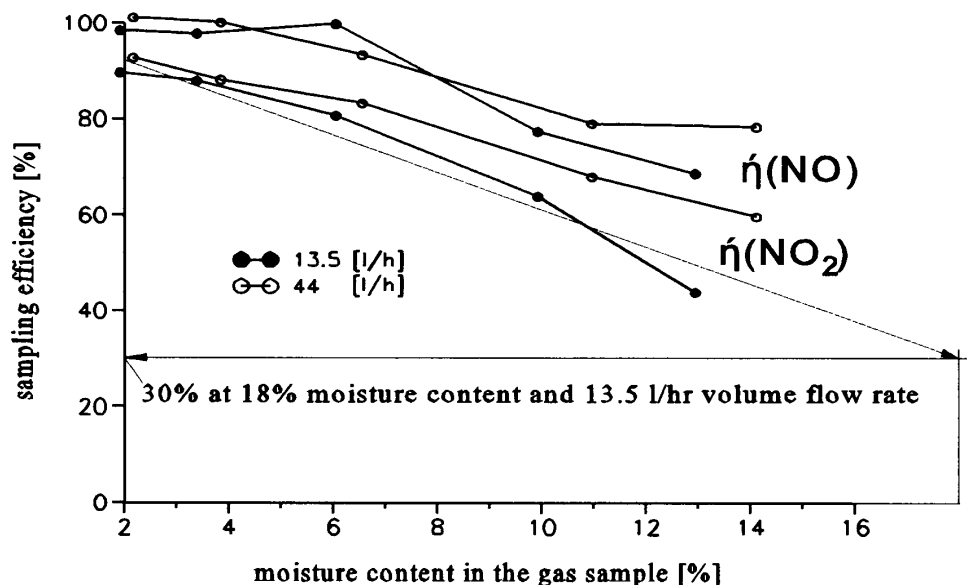


Figure 5. Loss of NO₂ in the sampling system because of ad- and absorption

The so-called 'quenching effect' means that during sampling a hot gas stream, which consists of many components and radicals, into a cooled probe, combustion products may undergo fast quenching reactions inside the cooled probe head. In particular for nitrogen dioxides, this effect may be thought to be significant because of the following radical reaction:



which has a negative activation energy. As the temperature decreases, the reaction rate of this reaction increases. The HO₂-radicals needed for the quenching effect result from a combining reaction $\text{H} + \text{O}_2 \Rightarrow \text{HO}_2$, where the H-radicals originate from hydrocarbon fuel.

This 'quenching effect', which is also called as 'HO₂-mechanism', will be discussed now in detail.

Analysis of the 'Quenching Effect'

In our sample-sucking probe, the gas sample stream is cooled down rapidly by means of a cooling fluid with temperature T_p, so that the reactions will be stopped. This cooling fluid must be maintained above a certain temperature(e.g 80°C) to prevent water steam condensation from the

sampled burnt gas. The temperature of the gas sample can, therefore, be calculated from the equation:

$$t(x) = T_f + (t_{\text{inlet}} - T_f) \exp(-cx) \quad \text{eqn.(1)}$$

where $c = 154 [1/m]$ (when the diameter of the probe is 2mm and the sucking velocity of the sample gas stream amounts to 13.5 l_N/hr), by assuming that the cooling fluid temperature T_f remains constant.

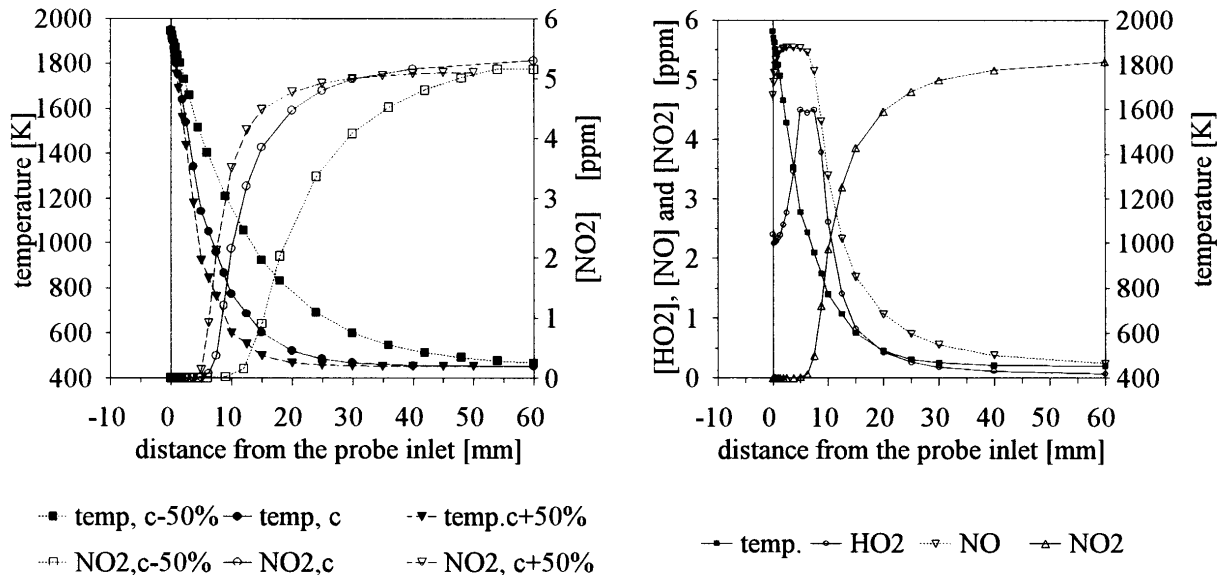


Figure 6. Simulation of the ‘quenching effect’

Fig.6 shows the simulation results concerning the ‘quenching effect’ from the Chemkin Premix program and Glarborg-Hadvig mechanism by applying a cooling fluid temperature of $T_f = 180^\circ\text{C}$. The needed input gas concentrations for the cooling simulation in the probe head were derived from a Premix calculation without cooling at the position of 0.234mm where NO_2 -peak occurs. At this position the calculated NO concentration amounts to 4.027ppm at a temperature of 1945K. Fig.6(left) shows the influence of the cooling speed on the formed NO_2 concentration through the ‘quenching-effect’. The ‘temp.c-50%’ and ‘temp.c+50%’ means that the constant c in eqn.(1) is by 50% lower or by 50% higher than $c = 154 [1/m]$, respectively. By faster cooling, NO_2 is formed earlier in the cooled probe. But at the position of 60mm from the probe inlet, almost the same NO_2 concentration of about 5-6ppm has been formed. Fig.6(right) reveals that NO- and HO_2 -concentrations undergo a peak at the beginning of the cooling, and then decrease rapidly. According to reaction(1), the NO_2 -concentration rises to about 5-6ppm. This NO_2 -concentration resulting from the ‘quenching effect’ would be included in the measured NO_2 -concentration. This cooling simulation reveals further, that at a long distance from the probe inlet, NO_2 may be transformed to NO by a few ppms.

We may now summarize that the measured 7ppmNO (460kWm^2 , air ratio=1.05) actually represent a NO_2 -concentration of about 20ppm NO_2 in the gas sample, from which 5-6ppm is formed by ‘quenching effect’ in the cooled probe head. Where could the excess NO_2 -concentration of about 15ppm come from?

The reason for this weak ‘quenching effect’ in NO_2 is the lack of NO, since this NO_2 is formed only from NO according to reaction(1) in the calculation. For the combustion conditions

present, there are only about 4ppm NO calculated at 0.23mm above the burner surface according to the known mechanisms, so maximal only 6ppm NO₂ can be calculated due to the ‘quenching effect’, including the further formation of NO in the cooled probe.

There are two questions to be raised: (1) If there were higher NO concentration at the sampling position, which NO-concentration has to be sampled in the sampling gas in order to form the NO₂-concentration of about 20ppm by the ‘quenching effect’? (2) Because NO formation is highly sensitive to combustion temperature, could this additional NO be explained from a real combustion temperature being somewhat higher than the measured one used for the Premix calculation?

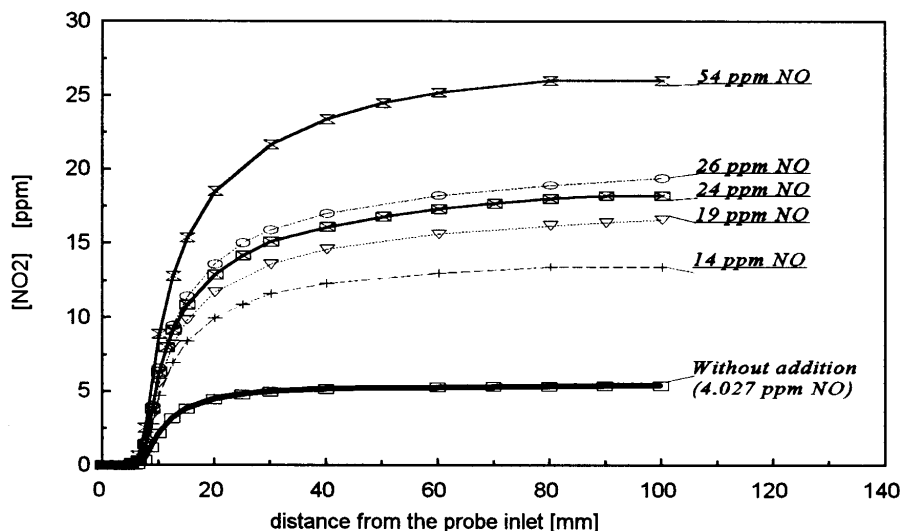


Figure 7. Formed NO₂ by the ‘quenching effect’ with higher [NO]₀ (Gas temperature according to eqn.(1), with $c=154$ [1/m])

To answer the first question, calculations were performed for higher NO-concentrations in the probe input stream. Results in Fig.7 show that at least 24ppm NO are needed in order to form about 20ppm NO₂ through the ‘quenching effect’. To answer the second question, further calculations were performed under adiabatic conditions(Fig.8). Under adiabatic condition with the highest combustion temperature([Ar]=0%), there are only 11.2 ppm NO-concentration calculated at the position 0.234mm above the burner surface with the Chemkin-Premix program and the Glarborg-Hadvig mechanism. Adding 2%, 5%, 10% and 15%Ar to the adiabatic combustion system, the inert gas Ar does not take part in the reactions, but acts as a heat sink in the combustion system. When the Ar-concentration increases, the temperature of the combustion system decreases. When the Ar-concentration reaches 15%, the adiabatic temperature is 110-130°C lower than the one without Ar-addition at the burnt gas region, but still higher than the measured temperature (corrected according to Fischer’s methode[8]). For this adiabatic case with 15%Ar, the calculated NO concentration amounts to 4.7ppm and 24ppm at 0.23mm and 15mm above the surface, respectively. From this result one can clearly see that the 24ppm NO, which is at least needed for the ‘quenching effect’ in order to form the measured NO₂-concentration, can not be transferred against the flow direction from 15mm away to the sampling position at 0.23mm.

It can be concluded now that, against the usual opinion, the ‘quenching effect’ cannot be the main reason for the measured high NO₂-concentration in the investigated system.

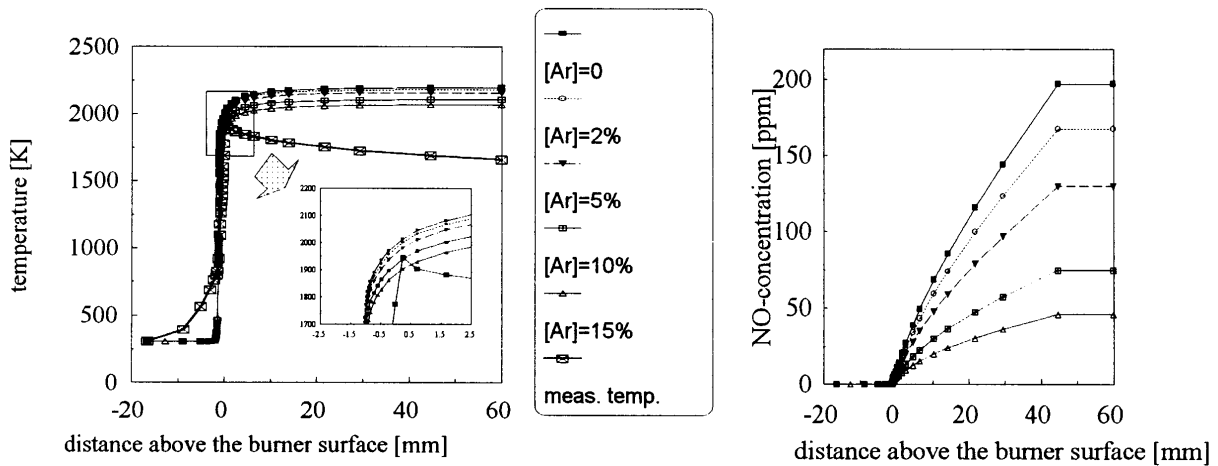


Figure 8. Formation of adiabatic NO corresponding to different temperature profiles

Further Experimental Results with Additives

As mentioned in [3], additives exert an influence on the NO_2/NO ratio. Further experiments were made in order to observe which influences could be related to the NO_2 -formation.

With CO-combustion, if carbon monoxide is premixed with dry air, there are no HO_2 -radicals in the system. Therefore, reaction(1) cannot occur. Because of shortage of H-atoms in the system, the four CO-combustion reactions from the Glarborg-Hadvig mechanism:



get reduced to two of them: reactions (5) and (6). As a consequence, the temperature measured was much lower and the flame was unstable. This can be seen from the lower temperature in Fig.9. In this case there was little NO_2 measured and no NO_2 -peak exists. But with addition of NO to CO-combustion, the NO_2 -peak (the first peak) exists. The second peak in the diagram can be attributed to further combustion of CO in this region due to instability of the flame(Fig.10).

This observation confirms again, that there must be other ways forming NO_2 in the combustion flames besides the HO_2 -mechanism, but consisting with the transformation of NO to NO_2 .

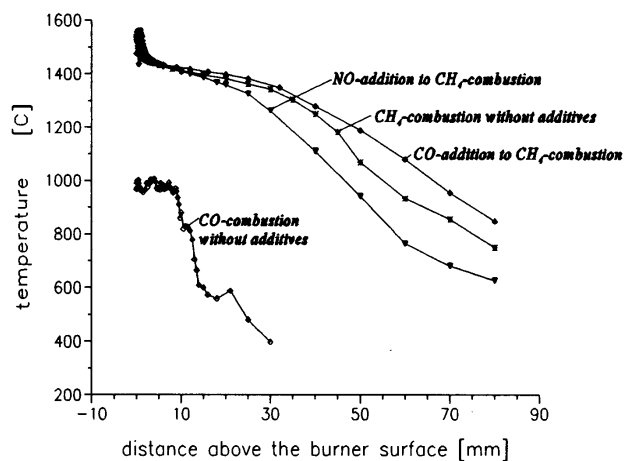


Figure 9. Experimental temperature profiles with CO-combustion and CH_4 -combustion

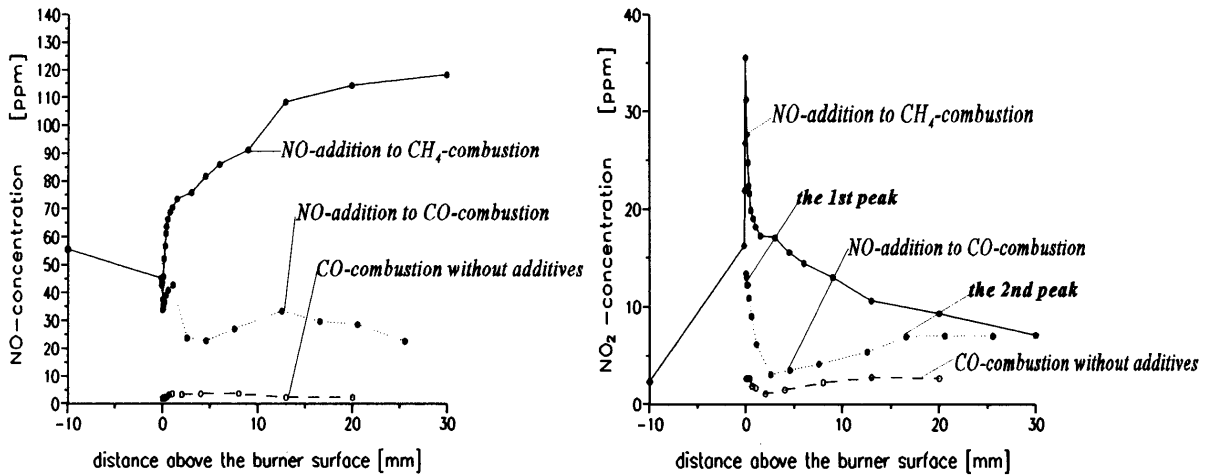


Figure 10. Effect of NO-addition to CH₄- and CO-combustion

In another series of tests, NO was supplied to the unburnt methane/air mixture as an additive with an initial concentration of $[NO]_0$. This added NO-concentration $[NO]_0$ is analysed by sampling unburned premixed gas mixture. Figs. 9-11 show that NO-addition to CH₄-combustion decreases the temperature slightly but increases the NO₂-peak greatly. At the position where the NO₂-peak occurs, there was less NO-concentration measured than added $[NO]_0$. If the curve is corrected to $[NO]_0=0$ by $[NO] = [NO]_{\text{measured}} - [NO]_0$ as is drawn, there is a negative region of NO-concentration formed where NO₂-Peak exists. This presents that NO-formation is related to the NO₂-formation. This can be partially explained as due to the 'quenching-effect'.

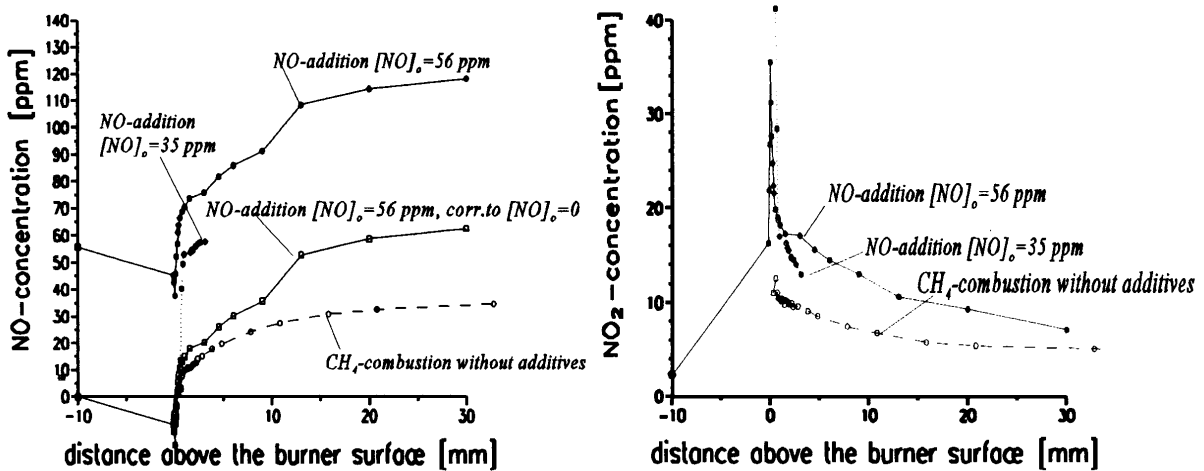


Figure 11. Contribution of NO-addition to the NO₂-formation in CH₄-combustion

In the burnt gas region, where thermal NO-formation predominantly takes place, both NO- and CO-addition will intensify the NO-formation(Fig.12). For the NO₂-formation, however, NO-addition intensifies but CO-addition reduces it(Fig.12), although CO-addition leads to a higher and NO-addition to a lower temperature than that by CH₄-combustion without any additives(Fig.9). Those effects reflect, therefore, not temperature but species influences.

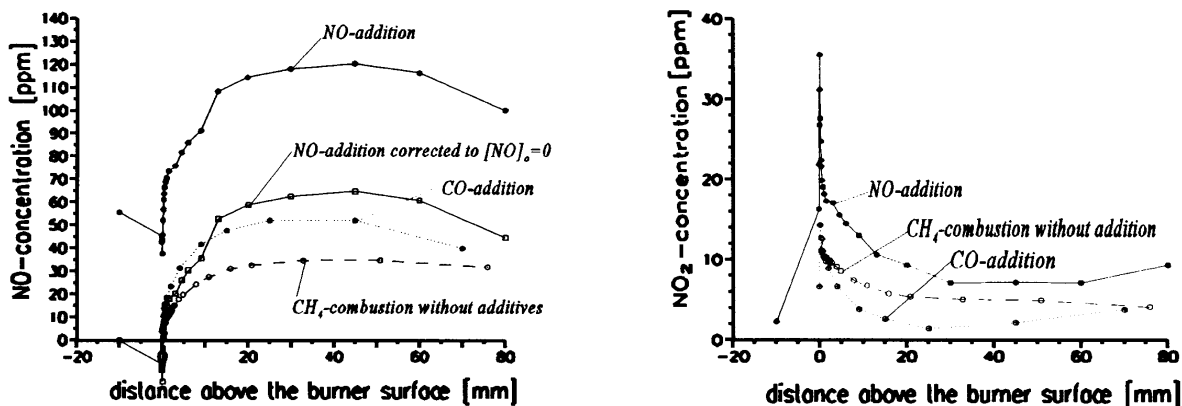


Figure 12. Difference between NO-addition and CO-addition to CH₄-combustion

CONCLUSIONS

From all the discussions above, one can conclude:

calculated $[\text{NO}_2] < [\text{NO}_2]$ resulting from quenching effect \approx detected $[\text{NO}_2] <$ real gas $[\text{NO}_2]$.

By means of the Chemkin program Premix, there is little NO₂ (<1ppm) calculated according to the Glarborg-Hadvig mechanism or to the Miller-Baumen mechanism formed in the flame. The 'quenching effect' was simulated, by which NO₂ is formed from NO. There could be 5-6ppm NO₂ resulting from the 'quenching effect' under the experimental conditions because of low NO concentration at the sampling position where the NO₂-peak occurs. This low NO-concentration has been mainly formed by the prompt-NO mechanism, since much thermal NO cannot yet be present as a result of relatively low temperature and short residence time.

The detected NO₂-concentration amounts to 7ppm, whereas the real NO₂-concentration in the sampled gas probe could be threefold higher because of ad- and absorption in the sampling system, according to the results of our earlier work. Supposing that all measured NO₂ would result from the 'quenching effect', there must be at least 24ppm NO in the gas sample. This 24ppm NO-concentration at the position where the gas is sampled can neither be formed at the highest possible adiabatic temperature, nor be transferred from downstream into this one dimensional laminar reaction layer. Regarding to the temperature influence on NO-formation, calculation results show that 24ppm NO cannot be formed till 15mm away in downstream direction, as in the case of adiabatic combustion with 15%Ar as a heat sink, where the gas temperature is still higher than the measured one. Moreover, there are also NO₂-peaks measured even if there are no HO₂-radicals in the combustion system, as in CO-combustion with NO-addition.

As a result, it must be concluded, that the four NO₂-related reactions from literature[5] and their kinetic data are obviously not adequate for quantitative simulation of the NO₂ profile in a combustion process.

Further experiments reveal that NO-addition to CH₄-combustion increases the NO-concentration and NO₂-peak, the latter can be partially attributed to the 'quenching effect' in the flame area. As for the NO₂-formation in the burnt gas region, NO-addition intensifies but CO-addition reduces it, although CO-addition leads to a higher temperature and NO-addition to a lower temperature than that by CH₄-combustion without any additives. Those effects reflect not temperature but species influences.

It can be concluded, therefore, that the HO₂-mechanism responsible for the quenching effect is not sufficient to explain the measured NO₂-peak at the radiative burner. Besides this reaction,

there must be other mechanisms for the formation of NO₂ in the combustion system which are until now not known.

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