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A COMPARATIVE KINETIC STUDY OF SNCR PROCESS USING AMMONIA

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Abstract - The paper presents comparative kinetic modelling of nitrogen oxides (NO_x) removal from flue gases by selective non-catalytic reduction process using ammonia as reducing agent. The computer code SENKIN is used in this study with the three published chemical kinetic mechanisms; Zanoelo, Kilpinen and Skreiberg. Kinetic modeling was performed for an isothermal plug flow reactor at atmospheric pressure so as to compare it with the experimental results. A 500 ppm NO_x background in the flue gas is considered and kept constant throughout the investigation. The ammonia performance was modeled in the range of 750 to 1250 ^oC using the molar ratios NH_3/NO_x from 0.25 to 3.0 and residence times up to 1.5 seconds. The modeling using all the mechanisms exhibits and confirms a temperature window of NO_x reduction with ammonia. It was observed that 80% of NO_x reduction efficiency could be achieved if the flue gas is given 300 msec to react with ammonia, while it is passing through a section within a temperature range of 910 to 1060 ^oC (Kilpinen mechanism) or within a temperature range of 925 to 1030 ^oC (Zanoelo mechanism) or within a temperature range of 890 to 1090 ^oC (Skreiberg mechanism).

Keywords: SNCR; Kinetic modelling; Ammonia; NO_x; Thermal $DeNO_x$

INTRODUCTION

The Selective Non-Catalytic Reduction (SNCR) is conceptually a simple process of NO_x control. At temperatures between 850 to 1175 ^oC, a particular reagent is injected and thoroughly mixed in a flue gas stream containing NO_x. A rapid gas phase homogeneous reaction occurs such that, despite the presence of excess oxygen generally required in the furnace, the reagent selectively reduces NO_x leaving oxygen untouched. The technology has many attractive features. It is simple to apply and requires no catalyst, hence no problems associated with catalyst such as catalyst poisoning, or replacement etc. It can easily be installed on existing plants, applicable in all types of stationary fired equipments, bears lower capital and running cost, generally not affected by fly ash and can be used with other NO_x emission control technologies. The ammonia and urea are the most commonly used reagents for the SNCR process of NO_x control employed usually in the combustion applications (Javed et al., 2007).

Computational kinetic modeling can be used to investigate the process of NO_x removal in post-combustion gases. In this paper an attempt is made to predict the performance of ammonia to remove nitric oxide gas from post-combustion gases assuming a constant pressure, isothermal plug flow reactor. The computer code SENKIN (Lutz et al., 1994) is used in this study using the two published chemical kinetic mechanisms, one that is used by Zanoelo (1999[a,b]) and the other that is used by Kilpinen (Kilpinen, 1997). An additional effort to assess the SKG mechanism (Skreiberg et al., 2004) for thermal $DeNO_x$ under the presented conditions was also conducted. These kinetic modeling studies were performed to provide a direct comparison of modeling predictions with the experimental measurements undertaken at the experimental setup for SNCR process at Leeds

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University UK (Irfan, 1995; Irfan and Gibbs, 1996).

The chemical kinetic code SENKIN was written at the Sandia National Laboratories, Livermore, California. It predicts the time dependent chemical kinetic behavior of a homogeneous gas mixture and runs in conjunction with the general purpose, problem independent FORTRAN chemical kinetic code CHEMKIN-II, also developed in Sandia Laboratories (Kee et al., 1994). The codes allow for the use of detailed chemical mechanisms composed of a set of finite rate elementary reactions in the gas phase. The mechanism is input by the user and so are their corresponding rate constants. The codes use a set of subroutines and data libraries to calculate thermodynamic properties. The computational solution is accomplished by a code called DASAC, which handles the solution of the governing differential equations.

CHEMICAL KINETIC MECHANISMS

The codes used allow for the choice of the chemical mechanism by the users. As mentioned earlier, the chemical kinetic mechanisms used by Zanoelo (EFZ), Kilpinen (K97) and Skreiberg (SKG) are employed in this study. The detailed mechanisms listing in Chemkin format is available on request.

The mechanism developed by Zanoelo is based on three reliable detailed kinetic models, the first by Miller and Bowman (1989) that includes 73 reactions among 19 species, the second by Glarborg et al. (1994[a]; 1994[b]) which includes 104 reactions with 22 species, and the last one by Miller and Glarborg (1996), which comprises 134 reactions and 24 species. In spite of the large differences in terms of the species and reactions, Miller and Glarborg (1996) showed that the main differences among these models can be explained in terms of the values chosen for two key parameters: the chain branching ratio, α, and the lifetime of the NNH radical, τ_{NNH}. Zanoelo found that to obtain a reasonable agreement with his experimental results he had to eliminate the reaction $NNH+O_2 = N_2 + HO_2$ and had to use the previous lifetime of NNH proposed by Miller and Bowman (1989). He argued that, although this reconciliation has a great significance for understanding of the real gas-phase kinetics of the process, it is not so important for engineering purposes where effective kinetic models can be used to design and optimize the industrial process. Zanoelo validated his mechanism for an experimental setup composed of 40 mm spheres made of silica to minimize wall catalytic reactions, which could be operated at temperatures up to 1175 ^oC and at atmospheric pressure. Fast mixing was achieved by introducing the reactants through four nozzles 1 mm in diameter (Zanoelo, 1999; Rota et al., 2001).

The second kinetic scheme used in this study is that of Kilpinen, which involves 137 elementary gas phase reactions between 26 chemical species including, NH_3, NH_2, NH, N reactions as well as the reactions involving NO and consumption of NO_2. In addition the mechanism also includes the reactions of HNO, HONO, H_2NO and NO_3 species, whereas subsets of elementary reactions of N_2-amine, N_2O, H_2-O_2 and CO are also included. Hence, the Kilpinen mechanism is based on the mechanism of Glarborg et al. (1994[a,b]; 1998) along with Miller and Glarborg (1996). Kilpinen et al. used an enlarged version including urea reactions for the reduction of nitrogen oxide emissions from biomass integrated gasification combined cycle (Zabetta et al., 2000). The aim was to study the effect of selected parameters on the reduction of nitrogen oxides in gas turbine combustors burning gasified biomass with staged air. The kinetic scheme was combined with simple flow assumptions described by ideal reactor concepts. In addition to other reactor concepts the above mentioned mechanism was also used by Kilpinen to simulate plug flow reactors using the SENKIN code. Hence, it was likely to be applicable for simple thermal $DeNO_x$ in a plug flow reactor where ammonia is used as a reducing agent to model an experimental set-up such as described below. The only difference may be that, from this detailed Kilpinen mechanism, urea related elementary reactions are excluded. Skreiberg et al. (2004) presented a detailed reaction mechanism comprising 35 species and 200 elementary chemical reactions, including the species NH_2OH, NH_2NO, H_2NN and HNN, which are not included in the other two mechanisms. The mechanism is suitable for modeling the NO reduction by primary measures in the combustion of biomass and below the temperature of 1400 K, but the performance in comparison to the other mechanisms and experimental observations has revealed some interesting results.

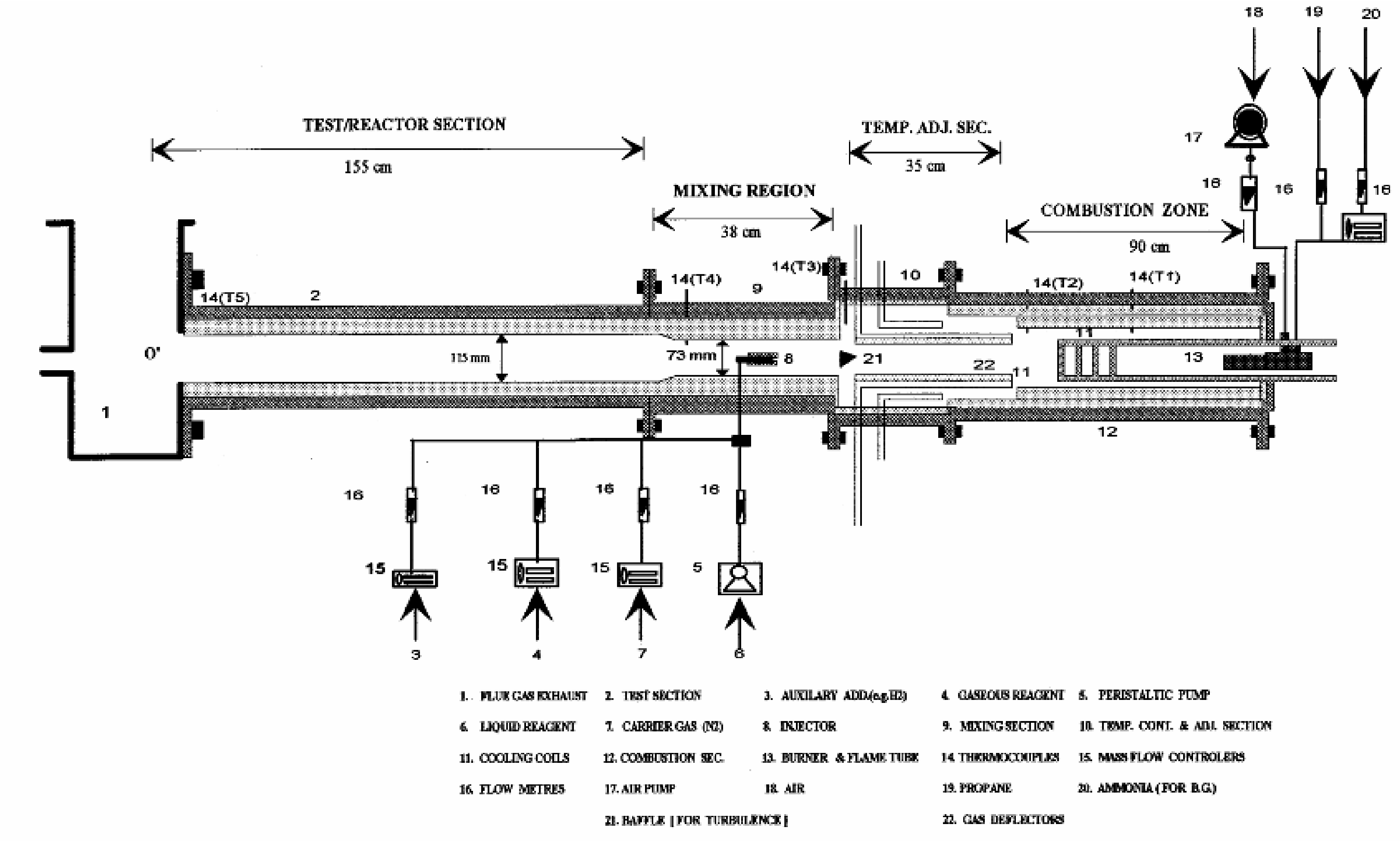

Figure 1: Schematic diagram of the experimental setup.

EXPERIMENTAL SETUP

A schematic diagram of the experimental system used in this investigation is shown in Figure 1. The flow reactor assembly, for gas phase reaction, was fabricated from stainless steel sections flanged together to form four main zones: the combustion zone, the temperature control and adjustment section, the mixing region and the reactor/test section. The combustion box was comprised of 300 mm square stainless steel section, 90 cm in length, internally lined with two layers of 1400 grade ceramic fiber boards, each 25 mm thick, giving a clear cross-section 200 x 200 mm square. It housed the combustor and silicon carbide flame tube. A nozzle mixing burner FRG-4A with a nominal rating of 116 kW was used to produce combustion products. The gas and air were rapidly mixed at the nozzle where they were burned. Due to the independent supply and control of air and gas, the excess oxygen level in the flue gas could be adjusted by means of the flow of air and gas. Flue gas mixture was produced by combustion of methane or propane that was supplied to the burner through a 25 mm ID flexible stainless steel pipe and metered by a Rotameter. The air was supplied by a centrifugal fan and flow was measured with a calibrated rotameter, having a range of 0-3800 lit/min.

To obtain the required NO levels in the flue gas, ammonia was supplied and burned in the flame in the presence of excess oxygen. The kinetics of the homogeneous, gas phase reaction of NH_3 and O_2 at high temperatures have been studied by several investigators (Wise and Frech, 1954; Miller and Bowman, 1989). The reaction is well known to produce NO at high temperatures (T>1900K) and indeed has been frequently used as a convenient method for generating NO in combustion studies (Wendt and Sternling, 1974; Chen et al., 1988). Using the established strategy, the propane was doped with ammonia, which, on burning, simulated the typical background level of NO_x from coal combustion. The ammonia supply for background NO_x was controlled and monitored by flow meter and mass flow controller. Primary ammonia injection into the burner was adjusted while sampling the exhaust gases to give 400 to 600 ppm NO_x.

The temperature control section had a provision for 15 mm OD adjustable copper cooling tubes for coarse temperature adjustment; 1100 grade, 5 mm thick, ceramic fibre boards were attached to the cooling tube at the plane formed by the longer tubes on the inner side of the section. These boards served as gas deflectors. The test/reaction section, which was coupled onto the mixing section, had a circular cross section of 11.5 cm ID. The experiments were performed with a test section length of 135 cm to provide adequate time for reaction. To obtain reaction profiles, gas samples were taken at various locations along the length of the test section corresponding to different residence times.

The general experimental operating conditions included propane and air flowrates adjusted to give a gas velocity of 3 m/s at NTP at 3% excess oxygen whereas background NO_x concentration was kept typically 500 ppm. The temperature range studied was 800-1300^{o}C. The set-up was found to be approximately isothermal with a drop in the test section of only 10-30 ^{o}C/m. The ammonia to initial NO molar ratio could be varied between 0.25 and 3.0, whereas a maximum residence time of 1.5 seconds could be provided in the reactor section of the experimental setup for completion of reaction. The flue gas analysis was performed using an automated chemiluminescence NO_x analyzer, Servomex oxygen analyzer, and infrared N_2O, CO and CO_2 analyzers.

RESULTS AND DISCUSSION

Nitric oxide removal using ammonia SNCR was modeled for an isothermal plug flow reactor at atmospheric pressure so as to compare it with the results obtained by the experimental set up mentioned earlier. A background NO_x of 500 ppm in the flue gas is considered and kept constant throughout the investigation. The performance was modeled using the previously mentioned Zanoelo and Kilpinen mechanisms in the range of 750 ^{o}C to 1250 ^{o}C using the molar ratios NH_3/NO_i from 0.25 to 3.0 and residence times up to 1.5 seconds.

Figures 2 and 3 show 3D surface plots of NO concentration, temperatures and residence time for the two models considered at a molar ratio of 1.25. Surface plots are employed as they can accommodate larger data in a single plot, which otherwise may require several 2D plots. In addition, they could demonstrate the interdependence of NO concentration on temperature and residence time concurrently. A dip showing NO reduction with temperature is visible in the NO concentration versus temperature plane. The third plane, in addition, shows the effect of residence time and indicates that NO reduction diminishes beyond about 300 ms. This may be shown more clearly by taking a projection of the surface plot on the residence time and temperature plane. The three projected lines indicate the residual NO concentration of 50, 100 and 150 ppm, corresponding to 90%, 80% and 70% reduction with the innermost corresponding to higher reduction efficiency. The bold line parallel to the temperature axis is indicative of the fact that 80% of NO reduction efficiency may be achieved if the flue gas is given 300 ms to react with ammonia, while it is passing through a section within a temperature range of 910 to 1060 ^{o}C (Kilpinen mechanism) or within a temperature range of 925 to 1030 ^{o}C (Zanoelo mechanism). Hence 300 ms may be considered as an optimum residence time corresponding to 80% reduction.

Below 850 °C the NO concentration remains unaffected at all residence times. A similar trend is obvious at higher temperatures where a diminished ammonia performance is observed and, if this trend is extrapolated, ammonia seems to produce NO instead to decreasing it. Thus, the modeling using both the mechanisms exhibits and confirms a temperature window of NO reduction with ammonia.

The insensitivity of NO concentration below 750 °C may attributed to lesser contribution of the reactions such as $H + O_2 \rightleftharpoons OH + O$ and $O + H_2O \rightleftharpoons OH + OH$, which are strongly temperature dependent, have a low rate below 750 °C. Thus, OH is not replenished fast enough to convert NH_3 to NH_2 and chain termination reactions such as $NH_2 + HNO \rightleftharpoons NO + NH_3$ and $OH + HNO \rightleftharpoons H_2O + NO$ compete with the branching sequence, thus limiting the NO reduction. These are detrimental to NO reduction because they consume NH_2 and OH radicals, without producing any further radical; whereas for any substantial reduction to occur, enough OH and O radicals are required to convert NH_3 to NH_2. Thus, below 750 °C, the rates of reactions such as $NH_3 + OH \rightleftharpoons NH_2 + H_2O$ and $NH_3 + O \rightleftharpoons NH_2 + OH$ limit the overall process.

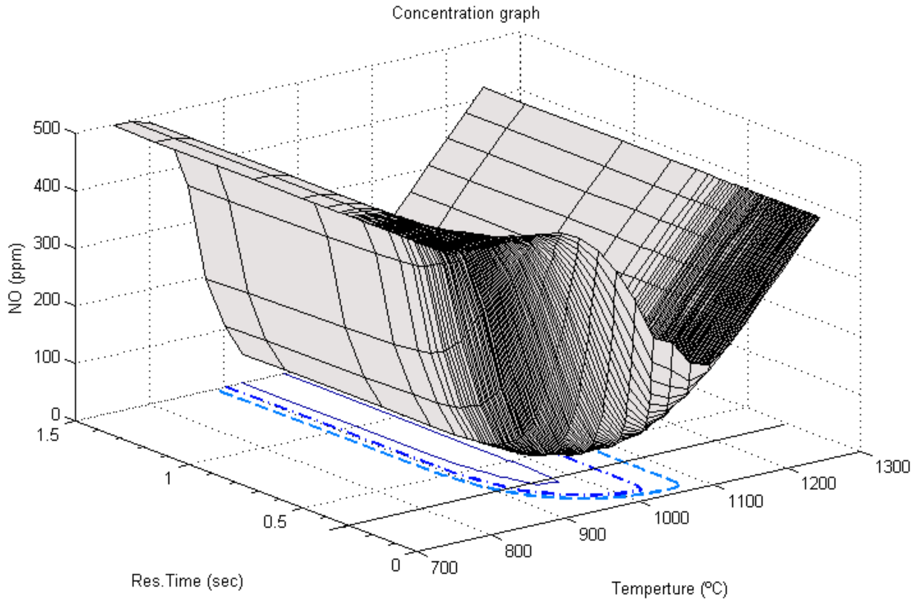

Figure 1: 3D surface plots of NO concentration, temperatures and residence time for the Zanoelo mechanism at a molar ratio of 1.25.

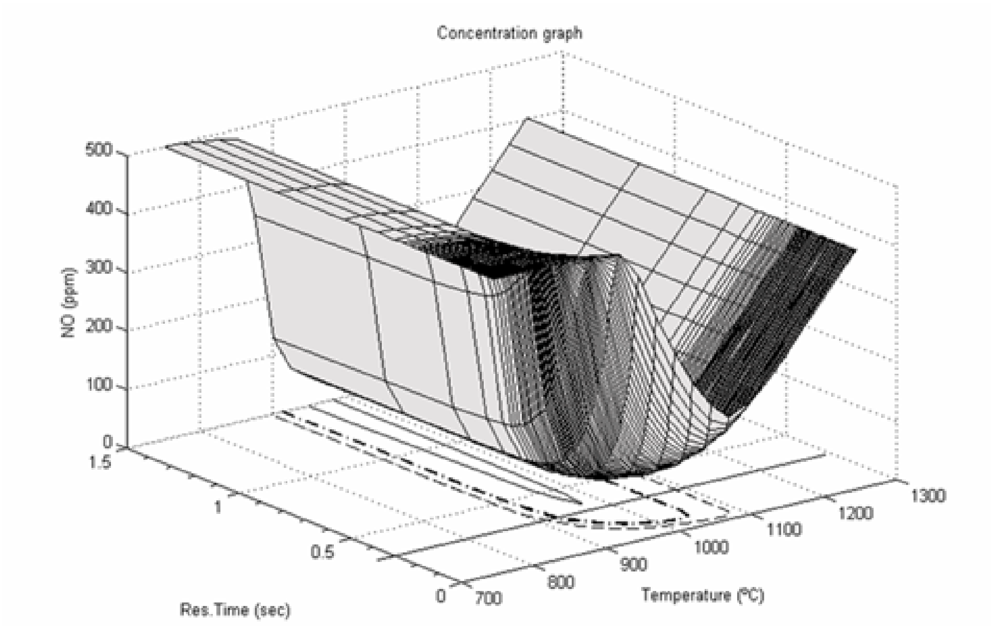

Figure 2: 3D surface plots of NO concentration, temperatures and residence time for the Kilpinen mechanism at a molar ratio of 1.25.

The dip observed in the surface plot shown in Figures 2 and 3 may be explained keeping in view the fact that from 850 to 1050 °C the mix of branching and terminating is just right to cause the growth in chain carrier concentration. With the increase in temperature, the reaction path leading to chain branching reaction HNO + M $\rightleftharpoons$ H + NO + M becomes more important than the one leading to termination reactions such as OH+HNO $\rightleftharpoons$ H_2O+NO. This can be explained on the basis of the activation energy of the former, i.e., 48680.0 kJ/kmole, which is greater than that of the latter which is 0 kJ/kmole (Miller et al., 1981; Miller and Bowman, 1989). The H radicals from the former reaction may react with either molecules of oxygen or water, causing more chain branching H + H_2O $\rightleftharpoons$ OH + H_2 or H + O_2 $\rightleftharpoons$ OH + O coupled with O + H_2O $\rightleftharpoons$ OH + OH. Due to this branching sequence, enough radicals are produced to drive reactions NH_3 + OH $\rightleftharpoons$ NH_2 + H_2O and NH_3 +O $\rightleftharpoons$ NH_2 + OH and significant NO reduction is thus achieved within the optimum temperature range.

The high temperature end of the window may be explained on the basis that, when the temperature is increased beyond 1250 °C, the OH radical concentrations start building up more vigorously, due to the branching sequence. This excessive increase in OH concentration may now initiate NH formation and the reaction NH_2 + OH $\rightleftharpoons$ NH + H_2O may start competing with reaction NH_3 + OH $\rightleftharpoons$ NH_2 + H_2O. Therefore, the NH radical concentration starts building up and the NO formation sequence starts competing with the NO reduction sequence. Once NH is formed, the high temperature oxidation reaction sequence leads directly to nitric oxide formation. At sufficiently high temperature, the oxidation sequences become significantly dominant and may result in a net increase in NO concentration.

At the optimum residence time and same molar ratio, Figure 4 compares the temperature windows corresponding to the three mechanisms investigated and experimentally measured data. As can be seen, the temperature window for 80% reduction for the Zanoelo mechanism ranges from 920 °C to 1035 °C with the peak efficiency of reduction of about 90% at 960 °C. For the same temperature window, the Kilpinen mechanism shows an overall 5% lesser reduction with a peak at 80% located near 975 °C. The experimental data collected show a slight deviation from both of them, with a smaller window of reduction from 930 to 1040 °C for an efficiency of NO reduction of only 60%, though the peak remains at about the same position with maximum efficiency of about 75%. Though the Kilpinen mechanism is closer to the experimental data, the deviation of results of both the models from experiment may be explained on the basis of mixing of ammonia and its overall effect. The SENKIN that incorporates the above mechanisms assumes an isothermal plug flow reactor setup and thus assumes uniform temperature and complete mixing. In a practical setup, despite the measures taken for mixing the reagent and flue gas, there still remains a degree of un-mixedness and this may be a reason that the experimental profile deviates from the predicted profile. The different temperature windows of the two mechanisms may be due to the fact that the Kilpinen mechanism incorporates all the possible reactions and thus the contribution of even a single reaction towards the final results should not remain unaccounted for. The SKG mechanism exhibits similar trends at lower temperature but it deviates from the general trend at the higher temperatures. It is observed that 80% reduction occurs between temperatures of 890 to 1090 °C, thus exhibiting a broader temperature window. In general, all three reaction schemes enabled us to represent the SNCR process characteristics and agree with the experimental observation qualitatively.

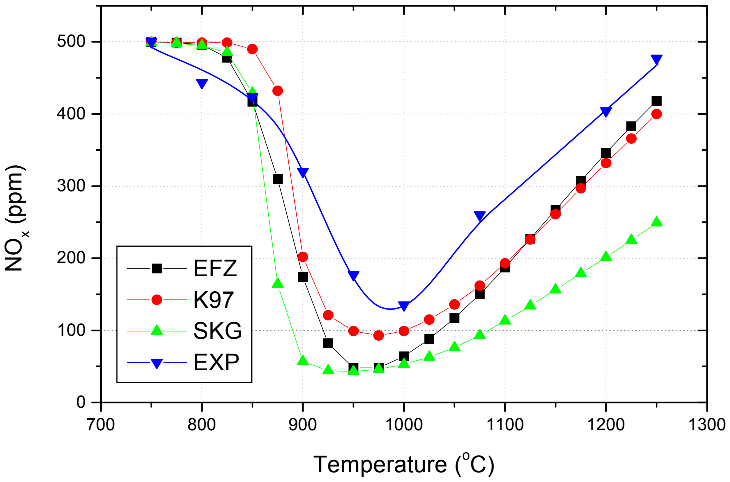

Figure 3: Comparison of temperature windows for the three mechanisms investigated together with the experimentally measured data at optimum residence time and molar ratio.

Figures 5 and 6 show the change in concentrations of N_2O, NH_3, NO and CO with respect to increasing molar ratio for the Kilpinen and Zanoelo mechanisms, respectively. The computed profiles show that, in both the cases, there is no noticeable formation of nitrous oxide and carbon monoxide. N_2O remains below 10 ppm whereas CO remains below 5 ppm for both of the mechanisms. This indicates that, unlike urea or cyanuric acid, ammonia SNCR does not result in formation of other pollutants like carbon monoxide and nitrous oxide.

The ammonia slip behavior is quite different when modeled by the two mechanisms. The concentration of ammonia in flue gases rises sharply beyond a molar ratio of 0.75 in the case of the Kilpinen mechanism and reaches a value greater than 125 ppm at a NH_3/NO ratio of 3.0. In contrast, the predicted ammonia rise is rather slower in the case of Zanoelo, where the concentration profile straightens up and approaches only about 60 ppm at a molar ratio of 3.0. Based on ammonia slip considerations, both the mechanisms suggest that the NH_3/NO ratio should be below 1.5 where 90% of ammonia reduction may still be achievable keeping ammonia in the flue gas below 50 ppm.

The same can further be optimized if NO concentration versus molar ratio of the two mechanisms is plotted together with the experimental measurements, as shown in Figure 7. The experimental measurements show that, beyond a molar ratio of 1.25, the NO concentration profile becomes almost parallel to the molar ratio axis, indicating a diminished effect of ammonia addition. The same trend is observed in the case of modeled profiles, with a difference that the predicted profiles merge together and straighten up beyond a molar ratio of 1.75. The Zanoelo mechanism always predicts lower concentration compared to the Kilpinen mechanism below NH_3/NO ratios of 1.75. Though the experimental and modeled profiles show similar trends, prediction by SENKIN for the two mechanisms investigated overestimates the performance of ammonia by 15%. This may be attributed to the fact that the SENKIN code assumes complete and rapid gas mixing, whereas flow is assumed to be linear and all the transport normal to the flow axis is neglected. In addition, the heat losses to the walls of the flow duct are assumed to be negligible and uniform isothermal conditions are assumed. In actual experimental conditions, these assumptions are not perfectly true and hence the lesser ammonia reduction performance is observed.

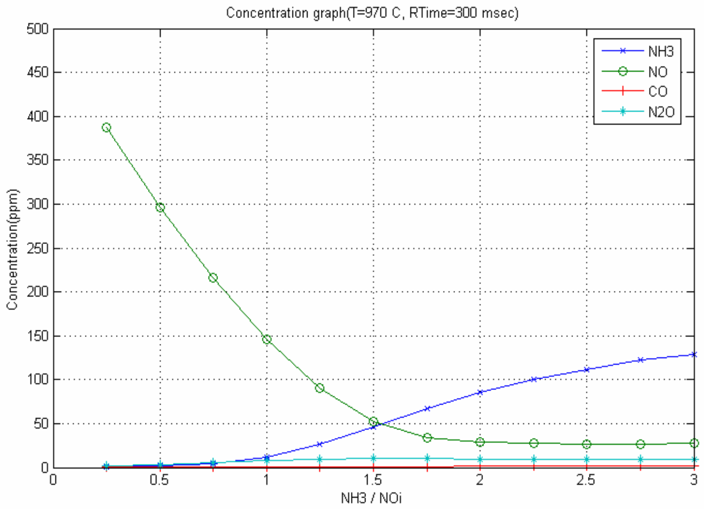

Figure 4: Concentrations of N_2O, NH_3, NO and CO with respect to increasing molar ratio for the Kilpinen mechanism.

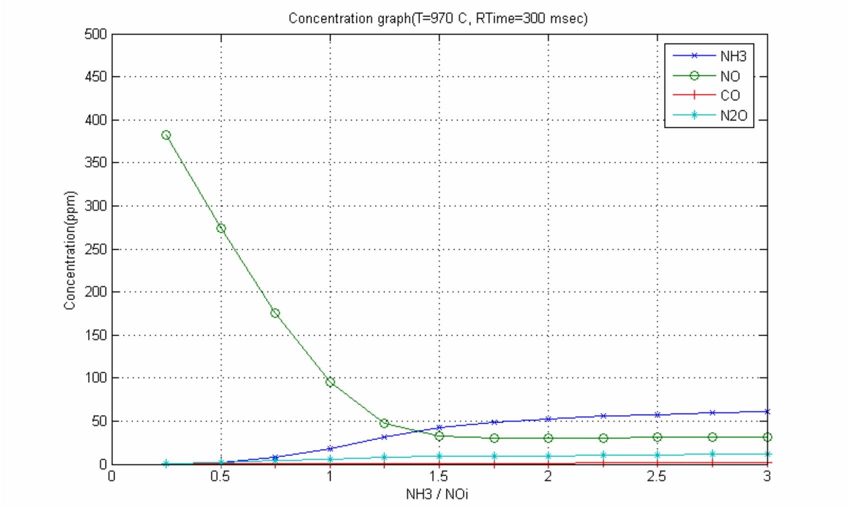

Figure 5: Concentrations of N_2O, NH_3, NO and CO with respect to increasing molar ratio for the Zanoelo mechanism.

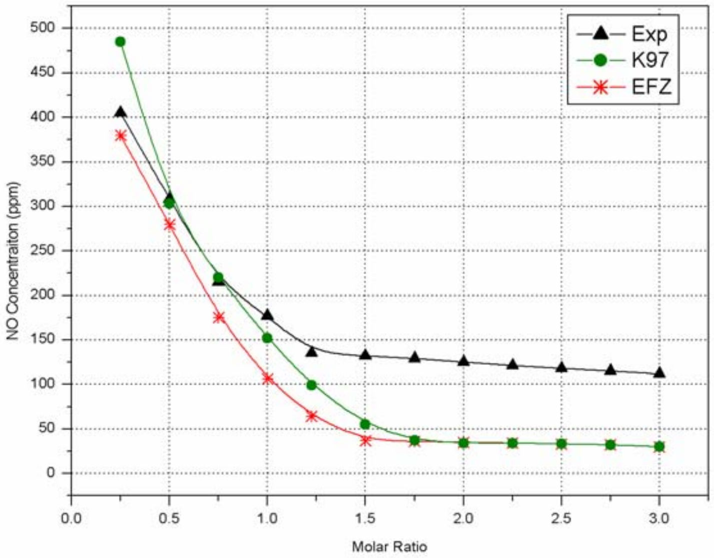

Figure 6: Comparison of NO concentration versus molar ratio of the two mechanisms and the experimental measurements.

CONCLUSIONS

A kinetic modeling study was performed to provide a direct comparison of modeling predictions with the experimental measurements undertaken at an SNCR experimental flow reactor at Leeds University, UK. The computer code SENKIN is used in this study using the Zanoelo mechanism, Kilpinen mechanism and Skreiberg mechanism. It was observed that the three mechanisms exhibit and confirm a temperature window of NOx reduction with ammonia used as a reducing agent. Qualitatively all the mechanisms are in agreement with most of the experimental observations. Some variations observed may be explained on the basis of experimental limitations on mixing of reducing agent with the flue gas. In a practical setup, despite the measures taken for mixing the reagent and flue gas, there still remains a degree of un-mixedness and this may be a reason that the experimental profiles deviate from predicted profiles.

In relation to the Zanoelo mechanism and Skreiberg mechanism, the species concentrations predicted by the Kilpinen mechanism were found to be more consistent with those observed in our plug flow isothermal SNCR experimental setup with the exception of the ammonia slip concentration. The concentration of ammonia in flue gases rises sharply beyond a molar ratio of 0.75 in the case of the Kilpinen mechanism and reaches a value greater than 125 ppm at a NH3/NO ratio of 3.0. In contrast, the predicted ammonia rise is rather slower in the case of

Zanoelo where the concentration profile straightens up and approaches only about 60 ppm at a molar ratio of 3.0.

The study also suggested that chemical kinetic mechanisms that were tried and tested for specific independent systems might effectively be employed to predict the trends in different environments and diverse experimental conditions.

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