

Numerical Study of Deposits Formation in SCR Systems Using Urea-Water Solution Injection

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Citation: Habchi C, Nicolle A, Gillet N (2018) Numerical Study of Deposits Formation in SCR Systems Using Urea-Water Solution Injection. J Mater Sci Nanotechnol 6(1): 104

Received Date: February 05, 2018 Accepted Date: June 07, 2018 Published Date: June 12, 2018

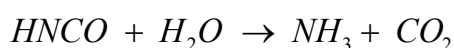
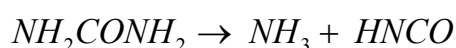
Abstract

Selective catalytic reduction (SCR) using Urea-Water Solution (UWS) is a NO_x reduction method that many diesel engine manufacturers adopt to meet new regulations for mobile applications (cars, trucks, etc.) throughout the world. However, this technology still faces many scientific and technological challenges. Indeed, the UWS injected spray can impact on the walls/mixer or lead to a marked inhomogeneity of ammonia at the SCR catalyst inlet; worse, this spray can affect the monolith by forming solid deposits. This work aims to investigate numerically such conditions using recently developed multidimensional models for the UWS evaporation and urea thermal hydrolysis upstream of a typical SCR catalyst. This study highlights the different timings of water evaporation and urea thermal hydrolysis processes in cold and hot conditions. It is found that the UWS Leidenfrost temperature is the main parameter of UWS modelling and numerical simulation. In cold conditions, for which the temperature of the walls is smaller than the UWS Leidenfrost temperature, the deposits are formed mainly on the mixer and near the pipe bends. In this case, water vaporizes first and more than 99% of the injected urea sticks on the wall forming a liquid film of pure urea. In such conditions, the main role of the mixer is to significantly increase the wall surface area covered by the liquid film, which may enhance the thermal hydrolysis processes for cold conditions. In hot conditions, for which the temperature of the walls is greater than the UWS Leidenfrost temperature, the deposits are formed on the inlet section of the monolith also because of the short distance between the UWS injector and the SCR monolith. Besides, the mixer does not play its role in such hot conditions, since the UWS spray droplets mainly splash on the mixer hot walls, and quickly leave towards the monolith. Finally, this study showed that deposits formation is an inherent phenomenon of UWS injection (whether in hot or cold conditions). They may be very problematic for the exhaust system durability since it can lead to the formation on the wall of solid by-products (Cyanuric acid or ammelide ...) due to urea polymerization. In addition, deposits generally lead to a maldistribution of ammonia upstream of the SCR catalyst brick, as shown in the particular configuration of the exhaust pipe studied here.

Keywords: Urea; SCR; Thermolysis; Deposit; Mixer; Exhaust; Model; 3D; Liquid Film; Boiling; Leidenfrost Regime

Introduction

Nitrogen oxides (NO_x) are pollutants that remain a concern for the environment and human health. Selective catalytic reduction (SCR) is an effective technology to control NO_x emissions in the exhaust automotive engines. For this, a reducing agent (typically ammonia, NH₃) is introduced upstream of the SCR catalyst. In automotive applications, a urea water solution (UWS) is often used to produce ammonia. Urea has considerable advantages over solid ammonia in terms of toxicity and handling [1,2]. However, the non-selectivity of its decomposition into ammonia and carbon dioxide (CO₂) complicates the NO_x reduction process. Currently, the most developed technology solutions are using a UWS containing 32.5% urea by weight ((European AdBlue™ or US DEF - Diesel Exhaust Fluid) [3]. This two-component mixture is injected into the engine exhaust pipe downstream of the SCR monolith (Figure 1). The reducing agent is produced by two successive reactions of decomposition: thermolysis of urea (NH₂CONH₂) and hydrolysis of iso-cyanic acid (HNCO) according to the following overall balances [4]:



Regarding the evaporation of the UWS, previous work has shown that evaporation of water is affected by the presence of dissolved urea and influences the NH₃ upstream spatial distribution, which is a crucial factor for the conversion of NO_x inside the monolith [5-8]. Song *et al.* have recently shown that the improvement in the uniformity of ammonia at the inlet of the monolith improves the effectiveness of NO_x reduction by SCR to 7% and reduces the "NH₃-slip" by 10 to 20 ppm, in addition to a reduction of the urea injection up to 15% [9]. Thus, to model the behavior of an SCR after-treatment system, several physical processes must be taken into account, mainly the evaporation of water from the UWS and thermal decomposition of urea. In addition, the proximity of the injector and the walls, due to the injection in a confined environment (small diameters of exhaust pipes, the presence of bends and mixers) promotes UWS spray interactions with the walls. These interactions can lead to the formation of liquid and/or solid deposits, as shown schematically in Figure 1, which can reduce the effectiveness of the SCR system. These deposits have been demonstrated in previous studies [10-12]. Experimental studies of Strots *et al.* revealed that the deposits can be formed over the after-treatment system from the injector to the inlet section of the SCR monolith. However, little theoretical and modeling studies have addressed this issue of forming hardened deposits [13,14]. To the authors knowledge, the only kinetic study of formation of deposits, published since the IFPEN work has focused on an anhydrous medium, neglecting the reaction processes (e.g., hydrolysis) that take place in aqueous phase [15,16]. The kinetic scheme obtained by these authors considers a polymerization of the urea via HNCO in liquid or gaseous state. Our kinetic scheme involves reactions between condensed phase species that have been well documented by Schaber *et al.* [17]. The resulting kinetic scheme provides a suitable basis for direct coupling with a CFD code. An overview of deposits formation pathways postulated in this work is presented in Figure 2. In this study, the different models described in detail by Ebrahimian *et al.* are used to investigate an industrial SCR system [15].

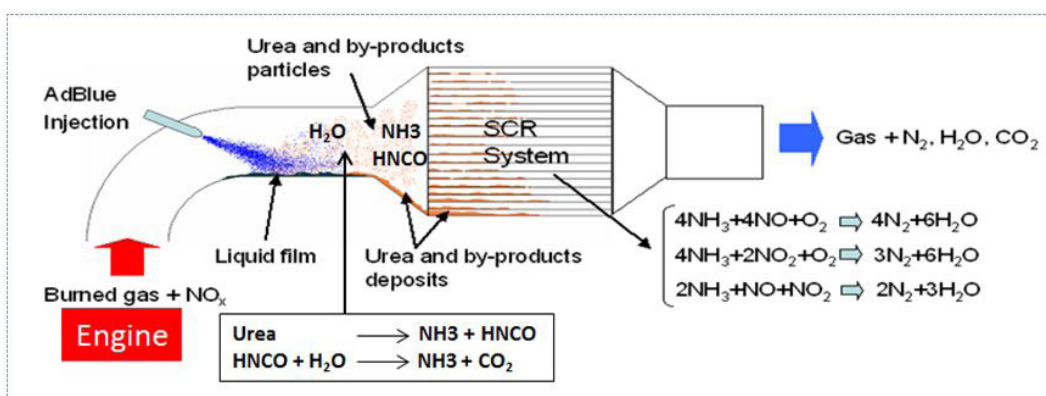


Figure 1: Schematic diagram showing the physical and chemical processes of an SCR NO_x reduction system

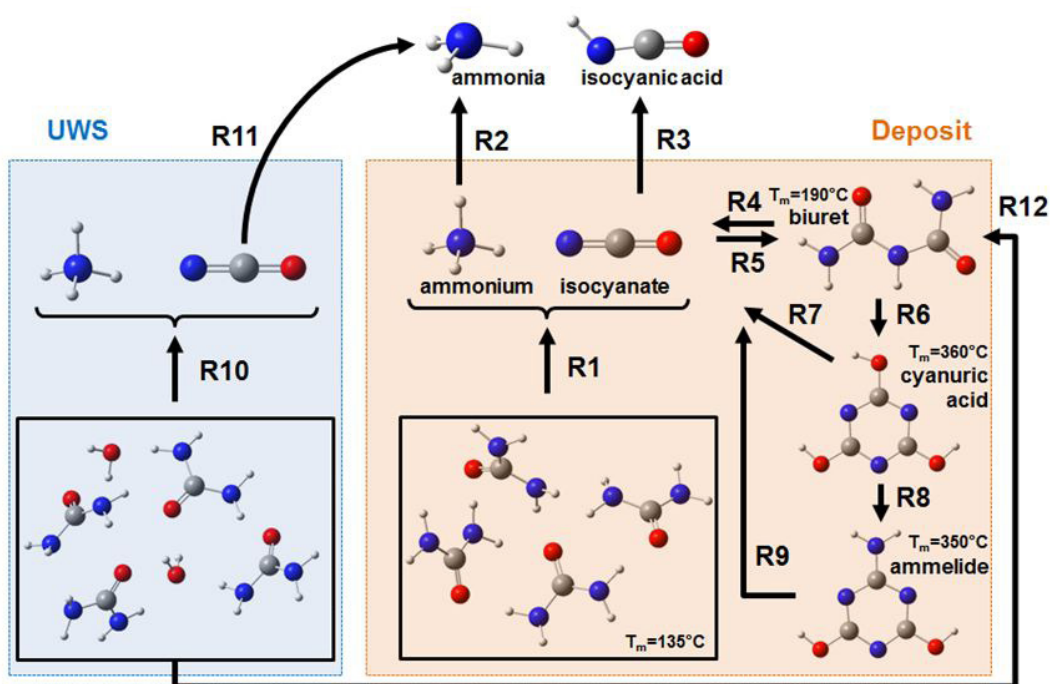


Figure 2: Main reaction pathways (R1) - (R12) to apply for the decomposition of urea in the semi-detailed kinetic model UWS-12R. The melting temperatures (T_m) of by-products (biuret, cyanuric acid and ammelide) are also shown

This article is organized as follows. The industrial configuration of the exhaust pipe and the operating conditions for the intended simulations are presented in Exhaust SCR Modelling Setup section. Then, the different physical models used in this work are briefly described in Modelling Approach Section. In particular, the main characteristics and assumptions of our multidimensional SCR modeling approach will be recalled. The simulation results are then discussed in Results and Discussion section, to draw up a report of achievements. Finally, the conclusions Summary and Conclusion section summarizes our modeling approach and highlights the different regimes of deposits formation obtained by the present numerical simulation methodology.

Exhaust SCR Modelling Setup

First, a typical passenger car exhaust line has been scanned and a CAD was performed. Figure 3 shows the main elements of the line including a stock Continental three holes injector, a mixer, followed by a shaped "gooseneck" pipe whose diameter is adapted at the entrance of the monolith. A 3D mesh of more than one million cells was generated. This grid has an inlet for the burnt gases, on the left of Figure 3 and a right exit of the monolith. The inlet section of the monolith is also shown in Figure 3. This interface operates as a wall impermeable to Adblue and urea droplets or solid by-product particles. However, the gas flows freely through this interface, as the monolith is not modelled in this study. The numerical results of 3D simulations are also recorded in this section for the purpose of subsequent analysis of the urea thermolysis (Results and Discussion section).

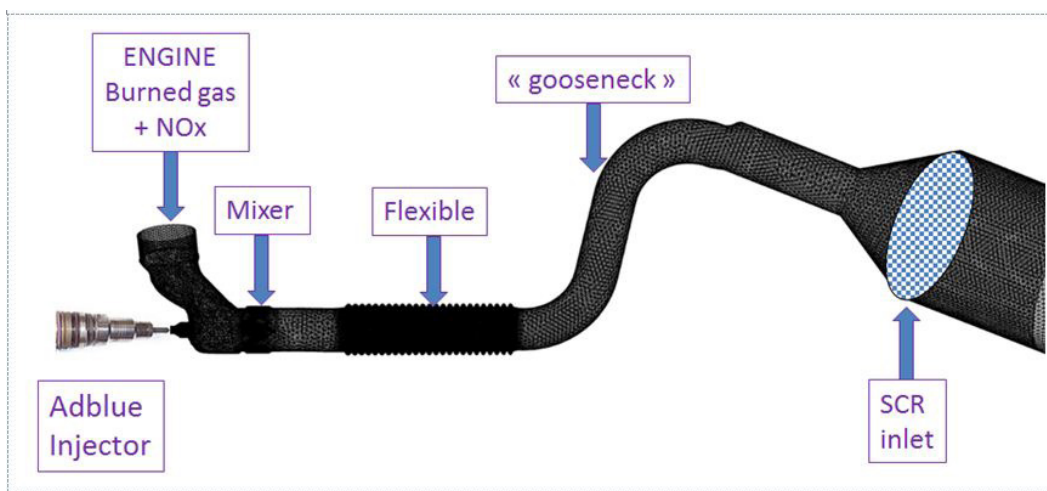


Figure 3: Exhaust line Configuration

Table 1 shows the operating conditions of the simulated three operating points. The first point 1500F corresponds to an engine speed of 1500 rev./min (RPM) and a gas flow rate of 21.3 g/s flowing at a rather cold temperature (437 K). The second point 1500C corresponds to a change in temperature of exhaust gases (582 K) with respect to the previous 1500F point. Finally, the third point corresponds to a speed variation (2500 RPM) and a gas flow rate of 46.3 g/s at a temperature (577 K) similar to that of the second point 1500C.

Engine Point	1500F	1500C	2500C
Temperature [K]	437	582	577
Flow rate of exhaust gas [g/s]	21.3	22.6	46.3

Table 1: Engine exhaust conditions

A Continental injector having three holes is used in this study. Despite their small cone angle, the three sprays quickly meet before impacting the mixer (Figure 4 which shows a view of the spray during injection). Table 2 summarizes the injection conditions. Particularly, a Rosin-Rammler distribution is specified with a relatively high factor $q=6$, in order to keep the drop size around of the measured SMD = 130 μ m.

Injector/ nozzle	Injection pressure [bar]	Gas flow rate [mg/s]	Injection frequency [Hz]	Injection duration [ms]	PDF / SMD [μ m] and q	Cone angle [°]
Continental/ 3 holes	5	8	1	5	Rosin-Rammler Distribution / 130 and 6	21 \pm 8

Table 2: Injection conditions

In addition, since we want to measure in this study the amount of deposit formed upstream of the SCR monolith by a single

injection, the duration of a simulation has been limited to one second by the specified injection frequency of the UWS. Under these conditions, the computation time was about a week using 256 cores (2.6 GHz Intel SandyBridge 3.5 GB of RAM per core) of the IFPEN supercomputer ENER110 (110 Tflops).

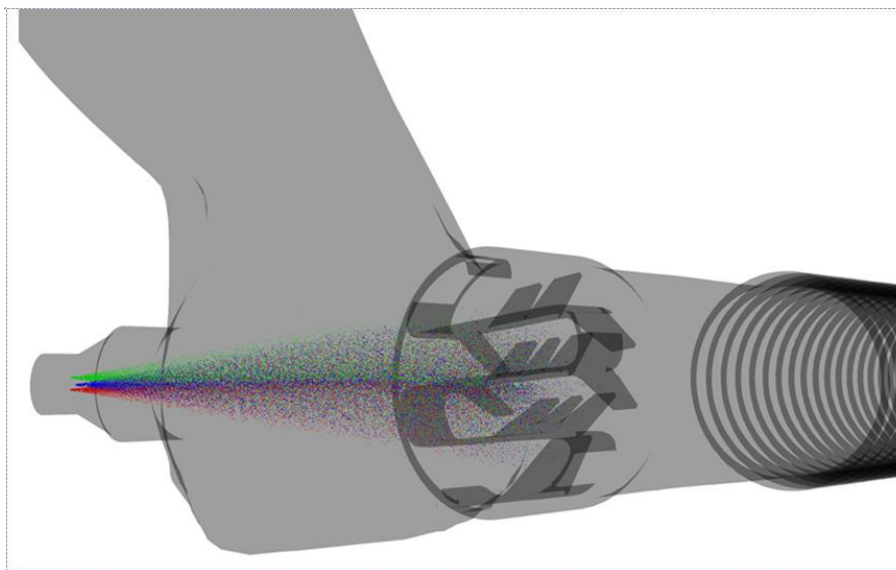


Figure 4: Zoom in on the sprays formed by the Continental three-hole injector

Modelling Approach

Ebrahimian *et al.* have proposed a set of coupled models that allows multidimensional simulation of the exhaust system upstream of the SCR monolith [15]. A first model represents the evaporation of a urea aqueous solution. In this model, called UWS-NRTL, evaporation of urea is neglected, and only water can evaporate although the evaporation of urea may actually take place as well [18]. But the evaporation of water takes into account the presence of dissolved urea by using "Non-Random Two Liquid" (NRTL) activity coefficients to model the non-ideality of the binary urea-water mixture, in particular, with high concentrations of urea. Indeed, it has been shown in [6] that the temperature of the UWS droplets increases exponentially at the end of the evaporation of water, and this justifies the hypothesis of a thermo-hydrolysis in aqueous phase.

The kinetic model that we developed is named UWS-12R because it uses a semi-detailed scheme with 12 reactions (Figure 2). It allows predicting the formation of NH_3 and HNCO upon decomposition of the urea and the formation of deposits resulting from the polymerization thereof (such as biuret, cyanuric acid and ammelide). Both models, UWS-NRTL and UWS-12R, are complementary. According to the authors' knowledge, these models differ from the previous models of literature on the following points:

1. The UWS-NRTL model considers in a simple and accurate way to account for the non-idealities of binary mixture, such as UWS; in particular, it predicts the exponential increase in the UWS temperature just before the end of the evaporation of water.
2. The UWS-12R model considers the competition between thermohydrolysis and polymerization in aqueous phase during the UWS temperature growth at the end of water evaporation in the droplets or liquid film.
3. The UWS-12R model allows the simulation of the formation of urea deposits and main polymerization byproducts.

In this study, an Euler-Lagrange approach was used: the liquid or solid phases are modeled using a Lagrangian approach, while gas is modeled by an Eulerian approach. In addition to the UWS-NRTL and UWS-12R new models, Lagrangian modeling includes a spray turbulent dispersion model and a comprehensive map for spray-wall interaction including splash and liquid film evaporation and its boiling [19-21]. The same mapping of spray-wall interaction processes (splash, rebound, liquid film, boiling ...) developed for hydrocarbons is applied to the AdBlue, with the exception of solid urea and byproducts for which the impact always leads to a rebound. This assumption can be considered acceptable for a dilute solution of urea, as the AdBlue; but it seems inappropriate for drops of pure urea or liquid film. In the latter case, the Leidenfrost regime is no longer considered as the saturation vapor pressure of pure urea is very low [18]. Besides, because of a serious lack of experimental physical and chemical data for urea and its by-products, their physical properties (density, viscosity ...) were assumed the same as those of pure water. Finally, Unsteady-Reynolds Averaged Navier-Stokes (U-RANS) numerical approach along with a standard k-epsilon turbulence model is used for the gas phase. In addition, wall-laws for gas velocity and temperature are used, taking into account a possible liquid film (or solid deposit) in the vicinity of the wall. In any case, the wall temperature is assumed to be equal to the exhaust gas temperature (Table 1). This assumption may however not be checked, in particular upon impact of the spray on the walls. The modeling of parietal conjugate heat transfer (CHT) is among the important topics of our future work.

Results and Discussion

In this study, three conditions have been simulated (Table 1). We will focus our attention on the formation conditions of NH_3 and

HNCO during the decomposition of urea and on the deposits resulting from its polymerization (such as biuret, cyanuric acid and ammeline).

Cold Case: 1500F

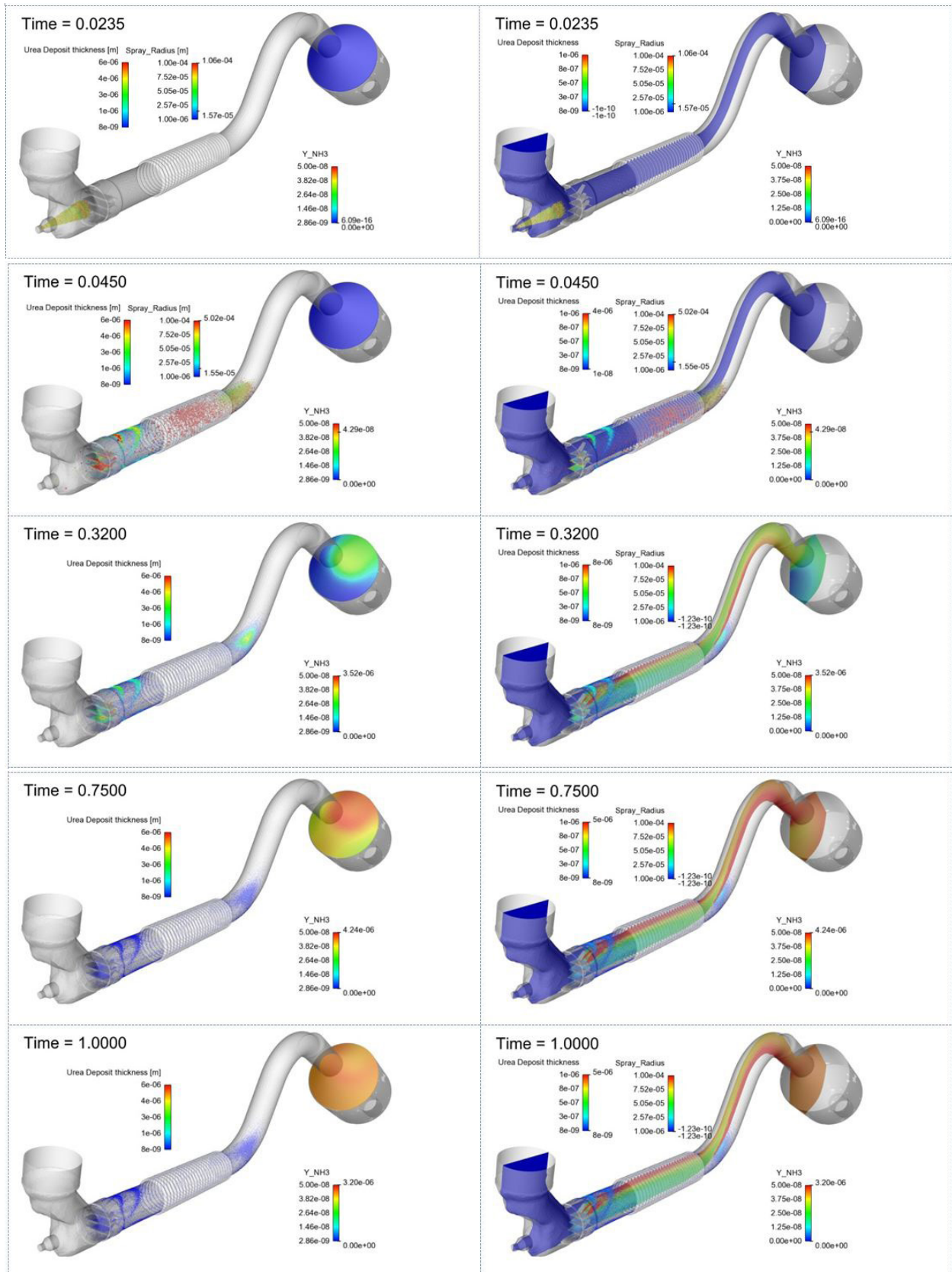


Figure 5: 1500F case - Spray injection and liquid film on the surface of the mixer and the pipe, in particular on the initial bend of the "gooseneck". Distribution of NH₃ on the monolith inlet section (left) and a longitudinal section (right). Time in seconds. All pallets indicate (on the right) the instantaneous (Min and Max) values

Because the wall temperature (437 K) is, in this case, far below the Leidenfrost temperature of the UWS (taken in this study equal to that of water (553 K), since the pure urea saturation vapour pressure is very low, even at high temperatures), the impact of the AdBlue spray led to the formation of a liquid film on the surface of the mixer and the pipe, in particular on the initial bend of the "gooseneck" (Figure 5). These results highlight the interest of a mixer for cool operation. Indeed, this component can significantly reduce the number of spray droplets arriving at the inlet of the SCR catalyst. This conclusion is in agreement with experimental observations by Lecompte *et al.* on a very similar configuration [22]. In Figure 5, we can see the urea deposits left on the wall after the complete evaporation of the water. One can also distinguish the spatio-temporal maldistribution of NH_3 on the monolith inlet section (left side of Figure 5) and a longitudinal section (right side of Figure 5). Ammonia is formed mainly from urea deposited on the mixer and at the "gooseneck" entrance. The blades of the mixer and the "gooseneck" appear to be oriented one with respect to the other so as to promote the spatial homogenization of ammonia at the monolith inlet. The maximum concentration of NH_3 obtained is very low (<10 ppm), as shown in Figure 6 and the palettes showing on their right side, the instantaneous (Min and Max) values for each image). Indeed, over 99% of the injected urea is found deposited on the wall, as shown in Figure 6. After one second, more than 40% of the deposits are in the form of the "Dry-urea" obtained after water evaporation, and the rest of deposits are in form of "ammonium isocyanate" in liquid state. Therefore, thermolysis of the urea deposits actually lead to a very low molar fraction of NH_3 and H₂NCO in the SCR monolith inlet section, as depicted in Figure 7. This low thermohydrolysis can lead to the formation on the wall of solid by-products due to the polymerization of urea and that are very problematic for the longevity of the exhaust system.

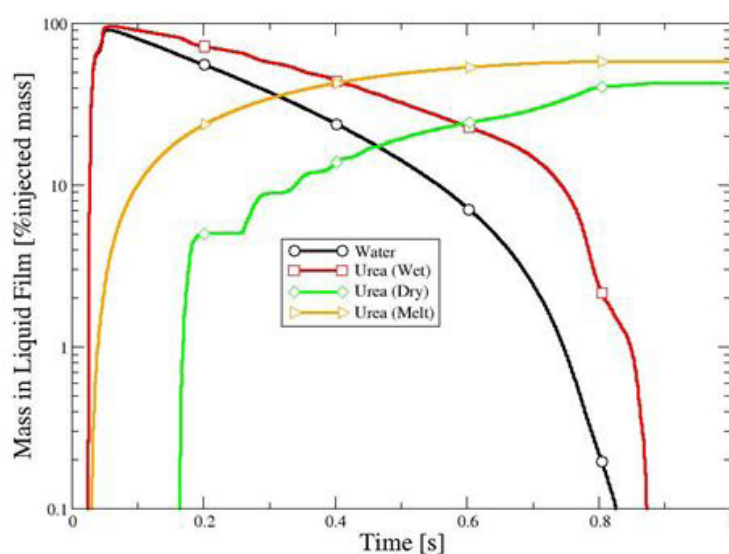


Figure 6: 1500F Case – Mass of water and urea de-posit-ed on the internal walls of the exhaust pipe (as a percentage of the injected urea compound (29% urea of 8 mg total injected mass). Urea (Wet) stands for urea in water. Urea (Dry) stands for "dry urea deposit" obtained after the water evaporated completely. Urea (Melt) stands for "ammonium isocyanate" in liquid state

Hot Cases: 1500C and 2500C

For these cases, a relatively high temperature (around 580 K, Table 1) is used for the inlet gas but also for all the walls. In these conditions, the impingement of the spray on the mixer does not lead to the formation of a liquid film as the AdBlue Leidenfrost temperature (taken equal to 553 K) is lower than the wall temperature. No deposit was formed because the AdBlue droplets rebound/splash on the hot walls of the mixer and quickly leave towards the SCR monolith, as shown in Figure 8. This behaviour is in accordance with the experimental observations of Smith *et al.*, for instance. Figure 8 clearly shows that the spray residence time is smaller in the 2500C case for which the gas flow rate is twice the case 1500C [11]. In both cases, the magnitude of the obtained residence time is very close to the experimental observations performed by Lecompte *et al.* on a similar exhaust configuration with an optical access at the location of the monolith [22]. In addition, Figure 8 shows the accumulated deposits on the inlet section of the monolith due to the small distance between the latter and the nozzle of the injector. Using a similar exhaust configuration to ours, Zheng *et al.* reported that high temperature deposits cannot be completely eliminated up to about 700 °C. Moreover, their tests show that deposits in an exhaust system can become powdery and able to detach from the walls, and released into the environment through the SCR monolith [10]. Even for this relatively hot conditions, a significant amount of deposits are still accumulated on the inlet face of the monolith. The spatial distribution of these deposits occupies a small central area which can reduce the NO_x conversion efficiency of the SCR catalyst (Figure 8). Moreover, the numerical results shown in Figures 8 and 9 also show a decrease in the deposits mass when the engine speed (respectively, the gas flow rate) increases from 1500 RPM to 2500 RPM (respectively, 21.3 g/s to 46.3 g/s). However, it may be noted in Figure 9 a slight increase of the cyanuric acid (Cya) deposit in solid state (as the melting temperature of Cya, 633 K is greater than the operating temperature, 577 K of the 2500C case), which

may become problematic for the longevity of the monolith.

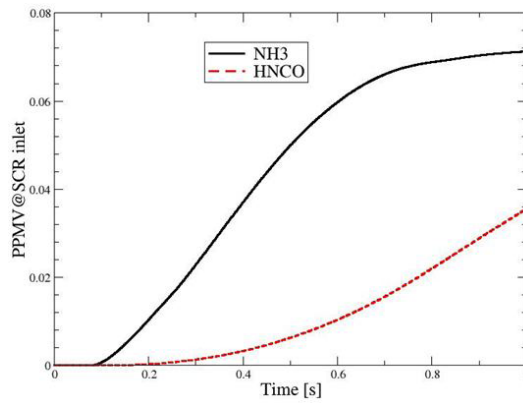


Figure 7: 1500F Case – Mole fraction of NH₃ and H₂CO in the SCR monolith inlet section

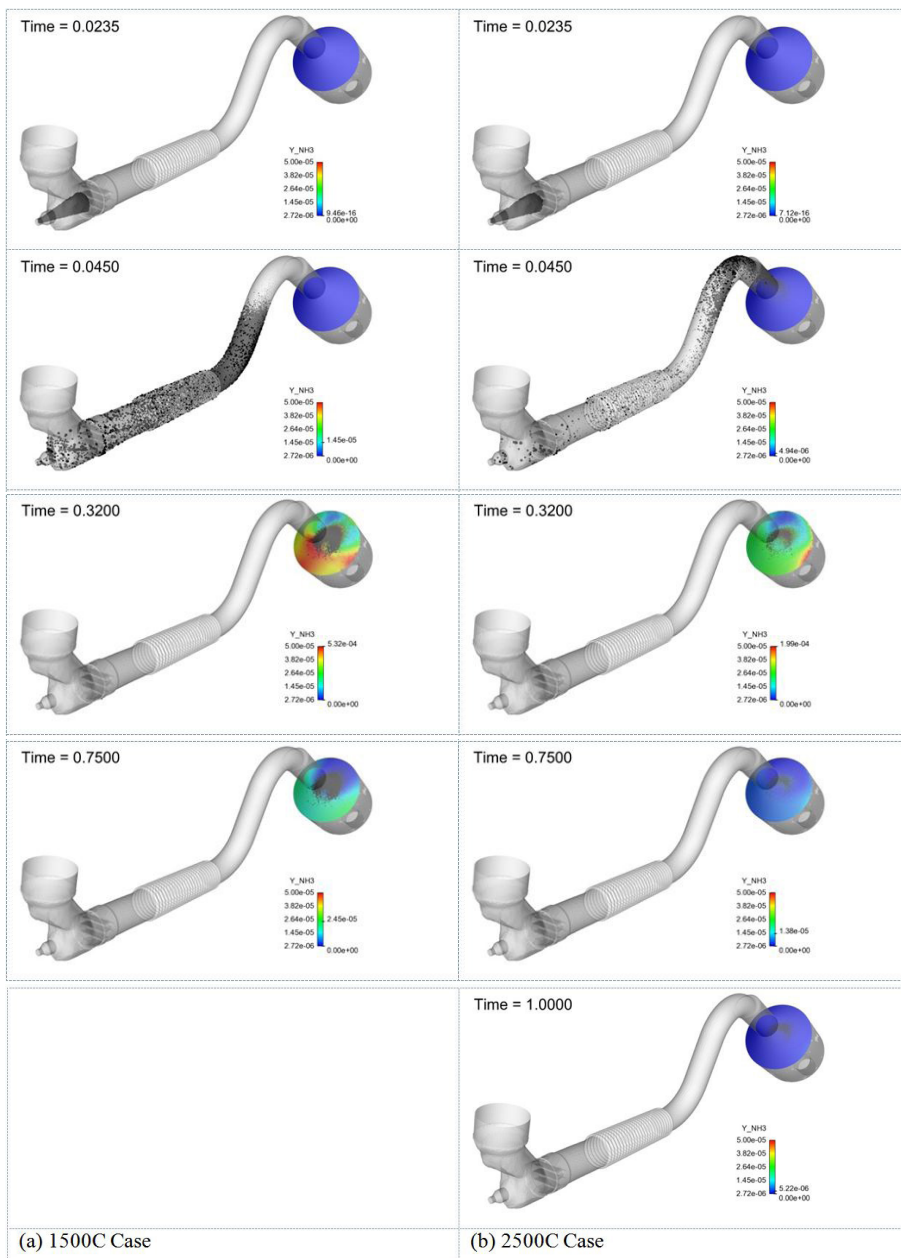


Figure 8: Spray distribution, urea deposit and NH₃ mass fraction at the monolith inlet face. Time in seconds. All pallets indicate (on the right) the instantaneous (Min and Max) values

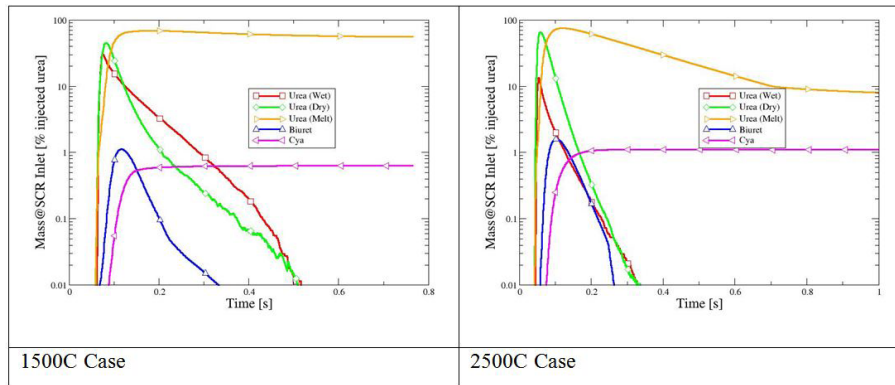


Figure 9: Urea mass deposited at the inlet face of the SCR monolith (as a percentage of the injected urea compound (29% urea of 8 mg total injected mass). Urea (Wet) stands for urea in water. Urea (Dry) stands for "dry urea deposit" obtained after the water evaporated completely. Urea (Melt) stands for "ammonium isocyanate" in liquid state. Cya stands for cyanuric acid

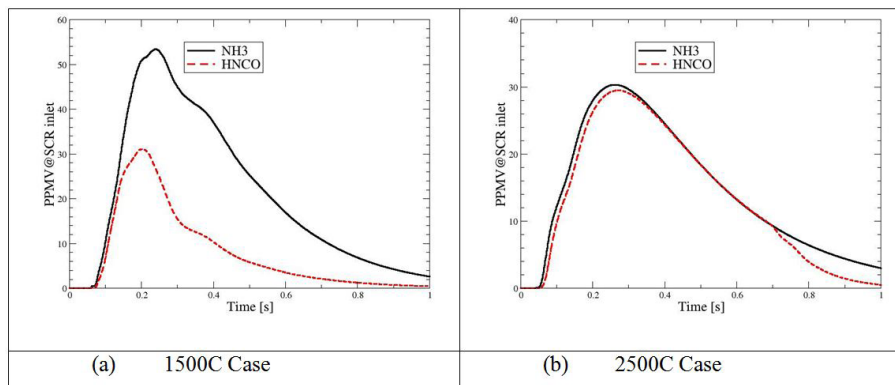


Figure 10: Mole fraction of NH_3 and HNCO in the SCR monolith inlet section

Finally, Figure 10 shows the temporal evolution of the integral over the monolith inlet section of the molar fractions of NH_3 and HNCO . This highly transient evolution is due to the rapid accumulation of urea at the inlet face of the monolith. The higher urea conversion for the 1500C case (up to 50% of NH_3 and 30% of HNCO) than the 2500C case, depicted in Figure 10(a) may be explained by the higher residence time of the urea in the exhaust pipe. Also, it is worth to note that the same amount of NH_3 and HNCO are obtained in the 2500C case, as shown in Figure 10(b), which is not the case for the 1500C case. This may highlight the need to use CFD tools to design effective SCR systems.

Summary and Conclusions

In this paper, a multi-dimensional numerical study of an exhaust line used in an SCR system was performed. The investigations have focused on the injection of the aqueous urea solution, the spray-wall interaction, the thermohydrolysis of urea to ammonia and iso-cyanic acid and also on the analysis of the conditions of formation of deposits on the internal surfaces of the exhaust pipe.

To this aim, a global modeling approach was applied, including the following recently developed models:

1. UWS-NRTL model for the evaporation of UWS spray and liquid film.
2. UWS-12R model for the thermohydrolysis of urea and deposits formation.

The study highlighted two regimes of formation for deposits in the exhaust pipe depending on temperature and gas flow (i.e., engine speed):

1. For cold conditions, urea is deposited on the inner surfaces (mixer, pipe) upstream of the SCR monolith in the form of liquid urea and ammonium isocyanate. These numerical results show the interest of a mixer for cold operation. Indeed, it has been shown experimentally by Lecompte *et al.* that this component can significantly reduce the number of droplets that can reach the entrance of the SCR catalyst [22].
2. For hot conditions and more specifically when the wall temperature is above the AdBlue Leidenfrost temperature (553 K), the spray droplets mainly splash on the mixer hot walls, and quickly leave towards the monolith. In this regime, a small amount of solid cyanuric acid (Cya) is formed in the central area of the inlet section of the monolith. This deposit can be very problematic for the longevity of the exhaust system. In addition, the amount of Cya seems to increase with engine speed.

Finally, this study showed that the formation of deposits is inherent to the injection of a liquid solution of urea to the exhaust (either in hot or cold conditions). The polymerization of these deposits leads to a maldistribution of ammonia at the inlet face of the SCR catalyst, in the particular configuration of the exhaust pipe studied here.

Acknowledgements

This work was performed in the framework of the UREENOX project (ANR-11-VPTT-002) funded by the French National Research agency ANR (Agence Nationale de la Recherche). The authors would like to thank Mr. S. Laabidi (IFPEN) for providing the CAD used in this article and Mr. A. Frobert and Mr. S. Raux (IFPEN) for reviewing this document and for valuable discussions that we had during the UREENOX project.

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