Chalmers Publication Library

(article starts on next page)

Dispersion aspects of NH$_3$-delivery strategies for NH$_3$-based SCR systems

Andreas Lundström[1], Henrik Ström[1,2]* and Magnus Skoglundh[1]

[1]Competence Centre for Catalysis, Chalmers University of Technology, Göteborg, SE-412 96, Sweden
[2]Department of Applied Mechanics, Chalmers University of Technology, Göteborg, SE-412 96, Sweden

*Corresponding author: henrik.strom@chalmers.se, Tel.: +46 (0) 31 772 13 60.

Short title for running headlines: *NH$_3$-dispersion in SCR systems*

Keywords: selective catalytic reduction (SCR), computational fluid dynamics (CFD), urea-SCR, NH$_3$-SCR

Abstract

The current work investigates the performance of an NH$_3$-based SCR-system for two different NH$_3$-delivery strategies, based on either urea or gaseous ammonia, using computational fluid dynamics (CFD). Quantification of the radial uniformity of NH$_3$ shows that the system based on gaseous ammonia is capable of providing a more even NH$_3$-profile. It is also shown that the uniformity obtainable in a urea-SCR system may be substantially enhanced by the introduction of a static mixer. Furthermore, the results indicate that the expected negative influence on the radial uniformity of a gas-based system from a decreased retention time before the catalyst is counterbalanced by an increased radial mixing. In addition, two theoretically derived "rule of thumb"-relations for estimating the performance of SCR systems are presented and their use in the interpretation of the CFD data is illustrated.

1. Introduction

For quite some time, urea-based selective catalytic reduction (SCR) has been a commercially available technique for NO$_X$-abatement in heavy-duty lean-burn engine aftertreatment [1,2]. In the urea-based system, an aqueous solution of urea (urea-water-solution, UWS) is injected into the exhaust gas system upstream of an SCR-catalyst, and the urea undergoes thermal decomposition into NH$_3$ (ammonia) and HNCO (isocyanic acid). The isocyanic acid may hydrolyze into additional NH$_3$ on the SCR-catalyst or on a hydrolysis catalyst. Ideally, each mole of injected urea releases upon decomposition two moles of NH$_3$ that may take part in the SCR-reactions [3,4]. However, due to the difficulties associated with the injection and conversion of urea (e.g. the formation of deposits and low-temperature dosing [3-6]), alternative NH$_3$-delivery strategies are presently paid extensive attention. One

1

such emerging technique is based on solid ammine salts for on-board storage of NH_3. In contrast to urea-based SCR-systems, where UWS is sprayed into the exhaust, the ammine system therefore relies on direct release of gaseous NH_3 [7]. The major benefit of this technique is the ability to dose NH_3 at low exhaust gas temperatures. A major design challenge for an SCR-system is thus to guarantee an acceptably uniform NH_3-distribution for a wide range of operating conditions, within the often very limited space available for the geometrical design of an exhaust gas system [8,9].

The aim of the current work is twofold. The first aim is to deduce theoretical "rule of thumb"-relations to assist in the design of NH_3-SCR systems. The second aim is to perform detailed CFD simulations, partly to illustrate the validity of these relations. Simulations of both a UWS spray and directly injected NH_3 are performed in order to contrast the differences in the dominating transport mechanisms for the respective system.

2. Theory

2.1 Urea decomposition as a function of the droplet retention time

The release of NH_3 in a urea-SCR system is delayed in comparison to a gas-based system (e.g. an ammine-based system) due to the required evaporation of water and decomposition of urea. In order to assess the retention time needed for a urea-SCR system to operate efficiently, one must determine the time needed for the complete decomposition of a typical droplet in the UWS-spray. This droplet life time can be calculated using the UWS-droplet model of Lundström et al. [10], as shown in Figure 1. As expected, smaller droplets and higher exhaust gas temperatures significantly enhance the decomposition efficiency. This relation between the initial droplet properties, the flow conditions and the time needed for the NH_3-release provides a quick first estimate of the retention time needed if a urea-SCR system is used instead of directly injecting NH_3.

2.2 Radial mixing as a function of the driving speed

In addition to the obtainable NH_3-levels at the catalyst entrance, the radial uniformity is also of great importance. The mechanisms for radial mixing of NH_3 are very different in urea-SCR and gas-based SCR, since the droplets possess inertia and have a different radial diffusivity to that of the gaseous NH_3. In Figure 2, the turbulent diffusivity of the UWS droplets is shown as a function of the droplet size and the Reynolds number of the continuous phase. The calculation is based on a relation between the turbulent particle diffusivity and the Stokes number-corrected turbulent viscosity [11,12]. Also plotted for comparison is the molecular turbulent diffusivity of a gaseous species.

In general, it is seen that the turbulent diffusivity (and hence the radial mixing) is better the smaller the UWS droplets and the higher the exhaust gas Reynolds number. However, for UWS droplets larger than approximately 100 μm, the effect of the droplet inertia dominates and the degree of gas phase turbulence has no significant influence. It should be stressed here as well that increasing the Reynolds number means decreasing the retention time in the exhaust pipe (i.e. less time for radial mixing) due to a higher gas phase velocity.

3. CFD modelling

Simulations of a generic exhaust gas system, presented in Figure 3, are performed using the commercial CFD code ANSYS Fluent 13.0. The same injection point is chosen for both the UWS-spray and the gaseous NH_3. The UWS dosing is 2 mg/s, and for the gaseous NH_3 the same molar rate of NH_3 as from the ideal decomposition of urea is used (22 μmol/s).

Two different driving conditions (33 and 55 km/h) at an exhaust gas temperature of 456 K are investigated for both the UWS case and the case with gaseous NH_3. In addition, all simulations are performed with and without the inclusion of a static mixer. A summary of all cases is provided in Table 1.

The continuous phase is modelled using the RNG k-ε model with non-equilibrium wall functions [13]. Mixing of the gas phase components is accounted for via an effective turbulent diffusivity, calculated from the modelled turbulent viscosity [14].

3

The modelling of the UWS droplets employs Lagrangian particle tracking with a set of heat and mass balances to monitor the droplets' temperatures, sizes and urea contents as they move in the system [10,11]. The UWS droplet sizes are chosen according to a Rosin-Rammler distribution that has been fitted to experimental data from a commercial spray. The initial conditions for the UWS are a droplet temperature of 293 K and a urea content of 32.5 wt.-% in water. No models for wall wetting or film formation are included due to the doubtful results such models would produce for the complex UWS chemistry [3,13]. Turbulent dispersion is modelled using the discrete random walk (DRW) model [15], since the turbulence transport of the UWS droplets has been shown to have a significant effect on both the NH_3-uniformity and on the urea decomposition efficiency [13].

4. Results and Discussion

4.1 UWS decomposition

Decomposition results for the UWS-spray cases (cases 1 to 4) are presented in Figure 4. As can be seen, the inclusion of a static mixer has a large effect on the decomposition efficiency of the UWS-spray. It is further clear that a longer retention time (i.e. lower driving speed) is beneficial for the UWS decomposition. Since the ambient temperature is the same in all cases, the decomposition rate should only be a function of retention time and dispersion of the UWS. These results obtained in the CFD simulations are thus consistent with the predictions from Figures 1 and 2.

An interesting observation is that the release of water is more or less unaffected by the static mixer and a change of driving speed. This is due to the much faster evaporation rate of water compared to that of urea. Thus, the droplet retention times are sufficiently long to include the entire water evaporation phase in all four cases.

Decomposition efficiencies for the UWS cases are presented in Table 2. The best performance, a decomposition efficiency of urea into NH_3 of 31%, is observed at 33 km/h with a static mixer. All other cases display lower decomposition efficiencies. According to

Figure 1, the lifetime of a typical UWS particle of around 40 μm is approximately 0.8 s. Here, the retention time in the pipe is on the order of 0.1 s. This shows that qualitative results may to some extent be obtained a priori to predict the attained low decomposition efficiencies of the system.

4.2 Turbulent dispersion

To quantify the uniformity of the released NH_3 independent of the NH_3-delivery method (UWS injection or direct injection of NH_3), the so-called uniformity index is used [16]:

$$\gamma_i = 1 - \frac{1}{2A} \int_S \frac{|C_i - \bar{C}|}{\bar{C}} dS \tag{1}$$

The uniformity indices obtained in all simulation cases are listed in Table 3. In all cases the static mixer improves the radial mixing. In the cases with directly injected gaseous NH_3 (cases 5 to 8), the uniformity results are almost independent of the driving speed. This implies that a shorter retention time due to an increased exhaust gas velocity is compensated for by an increased radial mixing.

In all cases the UWS uniformity results are lower than those for the directly injected NH_3. This result is due to the different radial transport mechanism for the UWS droplets to the gaseous NH_3. When the droplets are large (upon injection), the evolution of their trajectories is governed by their inertia rather than by the gas phase turbulence. As the UWS particles become smaller due to the evaporation of water and the decomposition of urea, the influence of turbulence on their motion increases. Eventually, the particles attain a turbulent diffusivity similar to that of a gas phase species (cf. Figure 2). Even though the diffusivity of the UWS droplets approaches the turbulent diffusivity of the gas phase in the limit of small particle diameters, the time delay before this happens is crucial. Because of the relatively short retention time in the exhaust pipe compared to the UWS particle lifetime (cf. Figure 1), the particles do not shrink in size quickly enough for the more efficient turbulent gas-phase mixing to have time to disperse the NH_3 in the radial direction. The uniformity of the NH_3 that reaches the catalyst entrance in the case of a UWS-spray is therefore very sensitive to the

penetration of the spray into the pipe (i.e. spray momentum and inertia of the injected UWS droplets). The design of the UWS spray system must thus be done with uttermost care, in terms of injection angles, mixing devices etc.

5. Conclusions

Simulations of a point injection of both a urea-water solution (UWS) spray and direct injection of gaseous NH_3 (e.g. from an ammine-based SCR system) are performed and evaluated on the basis of the obtained radial uniformity of NH_3. The following conclusions can be drawn from the present work:

There is a significant difference between the urea-SCR and the NH_3-SCR systems in terms of the obtainable uniformity index, γ, of released NH_3 over the catalyst inlet cross-section. For the investigated configuration, injection of gaseous NH_3 is more beneficial ($\gamma = 0.74$ on average, compared to $\gamma = 0.55$ for the UWS-spray).

Turbulent radial dispersion of gaseous NH_3 scales with the Reynolds number of the exhaust gas in such a way that the uniformity index remains similar for both driving speeds investigated (33 and 55 km/h).

A static mixer improves the radial mixing in both the urea-SCR and in the direct-injected gaseous NH_3-system.

The UWS radial dispersion is dominated by inertial effects on the droplet motion and is thus more sensitive to the design of the system (e.g. location and direction of injector and mixer).

Furthermore, the results from the numerical simulations confirm the validity of two theoretically derived figures illustrating the time needed for decomposition of a UWS droplet and the efficiency of the radial mixing in an SCR system. These "rule of thumb"-illustrations can be used early in the system design phase to provide qualitative input on the expected performance of a suggested urea-SCR system and/or an SCR-system based on direct injection of gaseous ammonia.

References

1. M. Koebel, M. Elsener, M. Kleemann (2000) Catal. Today 59:335-345.
2. M. Koebel, M. Elsener, G. Madia (2001) SAE Tech. Paper 01-3625.
3. P.M.Schaber, J. Colson, S. Higgins, D. Thielen, B. Anspach, J. Brauer (2004) Thermochim. Acta 424:131-142.
4. L. Stradella, M. Argentero (1993) Thermochim. Acta 219:315-323.
5. A. Lundström, B. Andersson, L. Olsson (2009) Chem. Eng. J. 150:544-550.
6. A. Lundström, T. Snelling, P. Morsing, P. Gabrielsson, E. Senar, L. Olsson (2011) Appl. Catal. B 106: 273-279.
7. G. Fulks, G. B. Fisher, K. Rahmoeller, M.-C. Wu, E. D'Herde, J. Tan (2009) SAE Tech. Paper 01-0907.
8. S.-J. Jeong, S.-J. Lee, W.-S. Kim (2008) Environ. Eng. Sci. 25:1017-1036.
9. A. Munnannur, Z.G. Liu (2010) SAE Tech. Paper 01-0889.
10. A. Lundström, B. Waldheim, H. Ström, B. Westerberg (2011) Proc. Inst. Mech. Eng. Part D J. Automob. Eng. 225:1392-1398.
11. J. S. Shirolkar, C.F.M. Coimbra, M. Queiroz McQuay (1996) Prog. Energy Combust. Sci. 22:363-399.
12. D. C. Wilcox (1998) Turbulence Modeling for CFD. DCW Industries: La Cañada, CA.
13. H. Ström, A. Lundström, B. Andersson (2009) Chem. Eng. J. 150:69-82.
14. V. Yakhot, S.A. Orszag, A. Yakhot (1987) Int. J. Heat Mass Transfer 30:15.
15. D. I. Graham, P. W. James, Int. J. Multiphase Flow (1996) 22:157-175.
16. H. Weltens, H. Bressler, F. Terres, H. Neumaier, D. Rammoser (1993) SAE Tech. Paper 930780.
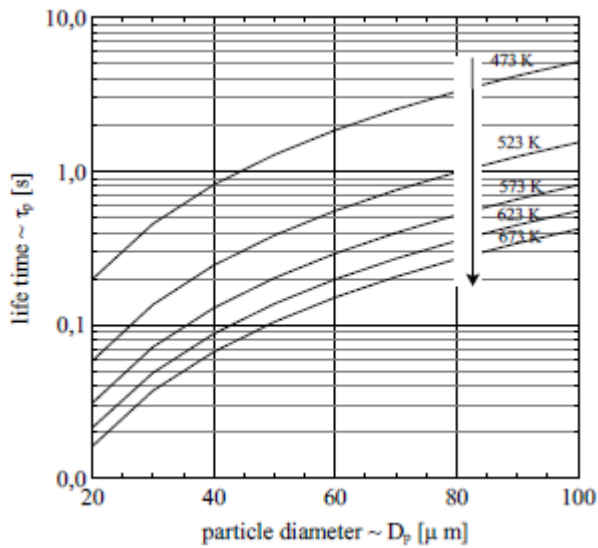
Figures

Fig. 1 UWS droplet life time as a function of initial particle diameter and ambient temperature. The life time of the droplet is defined as the time needed to obtain 99% conversion based on the total mass. The arrow indicates the direction of increasing temperature.
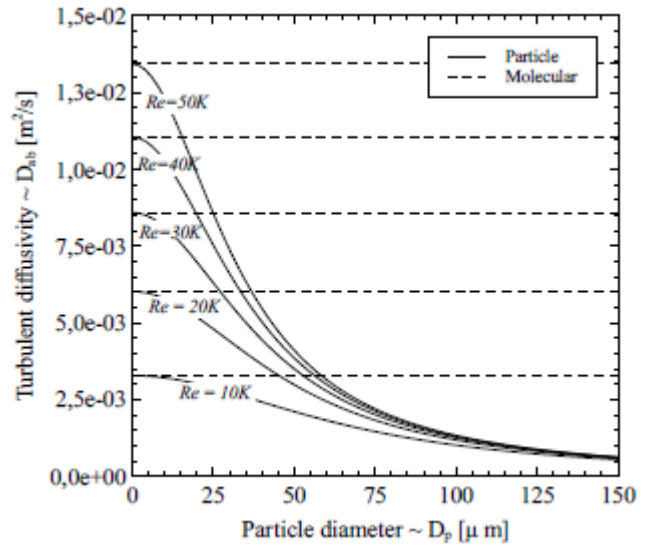

Fig. 2 Turbulent diffusivity of a UWS droplet as a function of particle diameter and the gas phase Reynolds number (solid lines). Also plotted are the corresponding molecular diffusivities (dashed lines).
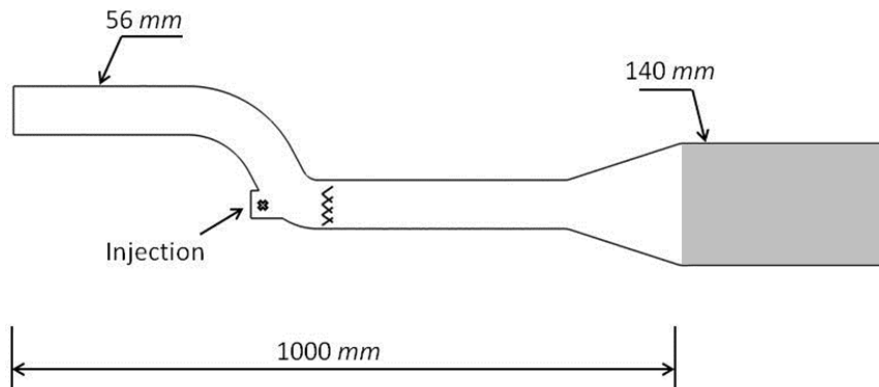

Fig. 3 Exhaust gas system geometry used in the CFD simulations. Indicated measures illustrate the diameter of the corresponding section. The injection point is located on the centreline of the pipe. The distance from the injector to the mixer is 100 mm, and the distance from the mixer to the substrate is 525 mm. The location of the substrate is indicated by the shaded grey area.
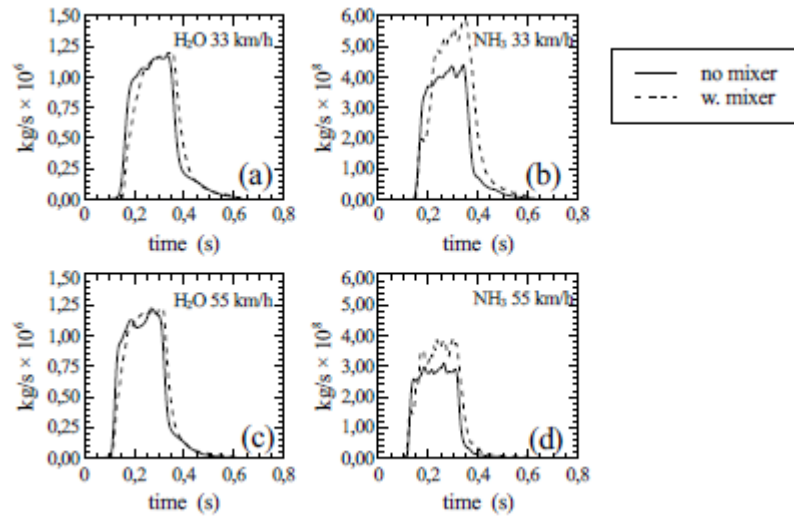
Fig. 4 Released water and ammonia for cases 1 to 4. (a) released water 33 km/h, (b) released NH_3 33 km/h, (c) released water 55 km/h, (d) released NH_3 55 km/h

Tables

Table 1. Summary of all investigated cases

Case	Speed (km/h)	$u_{in,gas}$ (m/s)	UWS/NH₃	Mixer
1	33	13	UWS	no
2	33	13	UWS	yes
3	55	17	UWS	no
4	55	17	UWS	yes
5	33	13	NH₃	no
6	33	13	NH₃	yes
7	55	17	NH₃	no
8	55	17	NH₃	yes

Table 2. Decomposition efficiencies based on total mass and injected urea

Case	Driving Speed (km/h)	Mixer	Total decomp. eff. (%)	Urea decomp. eff. (%)
1	33	no	75.2	23.7
2	33	yes	77.6	31.0
3	55	no	72.7	16.1
4	55	yes	74.1	20.4

Table 3. Obtained uniformity index, γ

Case	Speed (km/h)	$u_{in,gas}$ (m/s)	UWS/NH₃	Mixer	γ
1	33	13	UWS	no	0.54
2	33	13	UWS	yes	0.57
3	55	17	UWS	no	0.49
4	55	17	UWS	yes	0.58
5	33	13	NH₃	no	0.72
6	33	13	NH₃	yes	0.77
7	55	17	NH₃	no	0.71
8	55	17	NH₃	yes	0.77