

A reduced model for the evaporation and decomposition of urea–water solution droplets



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ABSTRACT

Selective catalytic reduction with urea–water-solution is commonly used in the automotive sector to reduce nitric oxide emissions. Detailed modelling of the exhaust gas system in front of the catalyst can help in optimizing this process. Such simulations include many different physical effects and chemistry on strongly differing spatial and time scales, thus require a high amount of computational reserves. Therefore, simplified or reduced models are needed to describe some of the processes which can then be added in the overall simulation. To develop reduced models the evaporation and decomposition of a droplet of urea–water-solution and the corresponding gas phase chemistry in hot exhaust gas are analysed using detailed simulations. It is shown that the process can be separated into three main phases, namely water evaporation, urea decomposition and reactions in the gas phase, which, for the conditions considered, do not significantly couple with each other. For the first two phases a simple model is developed, which calculates the mass and energy source term for a droplet evaporation process depending only on temperature and water content of the surrounding exhaust gas and the initial diameter of the droplet. The time scales and entropy production of the gas phase chemistry are determined for typical gas mixture compositions and initial conditions and based on these results a skeletal mechanism for chemical kinetics is generated. It can describe the gas phase chemistry for temperatures up to 800 K. In addition, it is found that gas phase chemistry at temperatures up to 1000 K can be modelled without the need to resolve the boundary layer of the small droplets.

1. Introduction

Selective catalytic reduction (SCR) is a process commonly used in the automotive sector to reduce the emission of nitrogen oxides (NO_x) in combustion engines (Skalska et al., 2010). In this process ammonia, reacts with nitrogen oxides in the presence of catalysts to form molecular nitrogen. Ammonia is produced by decomposing urea from urea–water-solution (UWS, 32.5 wt% urea in water) injected into the exhaust gas system. A homogeneous distribution of ammonia in front of the catalyst is crucial for effective reduction of NO_x . Detailed simulations of the exhaust gas system are helpful to optimize this process.

Currently Reynolds-averaged Navier–Stokes (RANS) equations are commonly used for multiphase-CFD simulations of the SCR system, but the resulting ammonia homogenization strongly depends on the turbulence model used (Fischer et al., 2012). As this complex process is highly unsteady, an improvement can be expected from large eddy simulations (LES). LES are better suited to simulate the unsteady multi-scale problems found in reacting turbulent multi-phase flows (Riber

et al., 2009; Vijayaraghavan, 2006), but the computational requirements are still too high for typical applications in the industrial design process of SCR systems (Fischer et al., 2012).

Independent of the turbulence model, such simulations have to include a multitude of potentially coupled physical processes like diffusion and convection in both liquid and gas phase, evaporation and chemical reactions. In addition, these processes happen at strongly varying spatial and time scales. Covering all the scales and processes within the framework of a single simulation is very costly in terms of computer time and storage requirements. Thus, simplified or reduced models are needed to reliably describe some of the complex processes within a UWS spray with a reasonable amount of computer time (Dukowicz, 1980).

UWS evaporation and decomposition is a complicated process. It was found in experiments that a UWS droplet behaves similar to a droplet composed of two liquids that have strongly differing vapour pressures. The progression of the droplet diameter over time shows a strong resemblance of the D^2 law used to describe simple evaporation

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Nomenclature

Symbol	Description
A	pre-exponential factor for Arrhenius equation
D	droplet diameter
D_0	initial droplet diameter
E_a	activation energy
F	chemical source term
h	specific enthalpy
h_{dec}	specific enthalpy of decomposition
I	identity matrix
J	Jacobian matrix
j	diffusion flux density
$j_{q,c}$	heat flux density due to conduction
k_1	gradient of D^2 ratio for water
k_2	gradient of D^2 ratio for urea
m	mass
\dot{m}	mass flow rate
M	molar mass
\bar{M}	mean molar mass
n	number of moles of species
n_s	number of species
n_r	number of reactions
p	pressure
p_a	atmospheric pressure
\bar{R}	gas constant
R	vector of chemical reaction rates
r	spatial coordinate
r_D	droplet radius
r_{dec}	reaction rate of urea decomposition
s	specific entropy

	stoichiometric matrix
t	time
t_j	characteristic time
T	temperature
v	velocity
w	mass fraction
x	molar fraction
Z	matrix of eigenvectors of the Jacobian matrix

Greek symbols

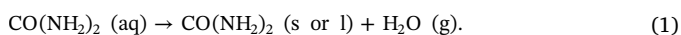
Symbol	Description
β	temperature exponent for Arrhenius equation
δ	threshold for entropy estimation
λ	eigenvalue of Jacobian matrix
Λ	diagonal matrix of eigenvalues of Jacobian matrix
ρ	density
ϕ	mass flux density
Ψ	state vector of complete system

Sub- and superscript

Symbol	Description
i	index for specific species
D	droplet
g	gas
l	liquid
v	evaporating species
vap	evaporation
∞	ambient properties

processes (Wang et al., 2009; Mikhil et al., 2016; Wei et al., 2016; Musa et al., 2006). For this reason, a typical approach to model this behaviour is to assume that urea and water behave like a mixture of ideal liquids which evaporate into the exhaust gas (Birkhold et al., 2007). There are alternatives to an evaporation model, for example using an Arrhenius expression, to describe the urea decomposition. It was found that an evaporation model provides the best fit to experimental data (Abu-Ramadan et al., 2011) and it is relatively easy to measure and fit a vapour pressure curve for urea and to integrate such a model into CFD simulations (Lundström et al., 2011). Besides the general similarity of experimental data with an evaporation model, the quantitative results vary strongly (Ryddner and Trujillo, 2015). Possible causes are different experimental conditions and hard to quantify effects like the influence of the support fibre holding a droplet (Chauveau et al., 2008).

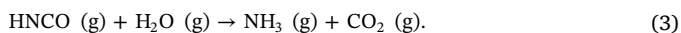
Overall, the whole process can be seen as a combination of the following three steps (Yim et al., 2004; Koebel et al., 2000): Evaporation of water from a UWS droplet:



Thermolysis of urea into ammonia (NH_3) and isocyanic acid (HNCO), modelled as evaporation:



Gas phase chemistry, mostly hydrolysis of isocyanic acid:



To reduce complexity in simulations of the exhaust the evaporation of droplets is often simplified. One possibility is to use a film model, for example based on a 1/3 rule (Sparrow and Gregg, 1958), to determine the evaporation rate instead of resolving the gas phase close to the droplet. The film model essentially represents the boundary layer around the droplet. This allows a Lagrangian approach where droplets

move through the computational domain and interact with the gas-cell they are in via this film model (Torres et al., 2003). In addition a parcels approach (Apte et al., 2003) can be used where a large amount of droplets is replaced by a number of droplets with similar properties. Within such a framework the liquid phase inside the droplet can still be resolved and it is only coupled via the film model.

The data in literature differs whether the hydrolysis of isocyanic acid is relevant in the homogeneous gas phase at SCR conditions. There are kinetic parameters available for a single step reaction based on measurements (Yim et al., 2004; Aigner et al., 1995) which predict significant reaction rates. Other authors expect isocyanic acid to be a stable compound and that a catalyst is required for the hydrolysis (Koebel and Strutz, 2003; Piazzesi et al., 2006). For reactions of ammonia with nitrogen oxides without a catalyst several mechanisms can be found in literature (Faravelli et al., 2003; Glarborg et al., 1995; Song et al., 2016; Skreiberg et al., 2004). Such mechanisms are optimized and used for the simulation of selective non-catalytic reduction (SNCR) of NO_x , which happens at higher temperatures (Caton and Xia, 2004). As UWS is also used in this context as a source for ammonia, the temperature range in this study is extended above the typical values found under SCR conditions and also covers the lower part of SNCR conditions.

This paper focuses on the analysis of the physical processes and chemical kinetics at SCR and SNCR conditions based on detailed one-dimensional simulations of droplet and ambient gas. This information is used for the development of a reduced model capable of describing reliably the process of the evaporation and decomposition of a UWS droplet in the surrounding exhaust gas. The first part of the paper deals with the physical time scales and properties of a UWS droplet during the evaporation. Based on the main observation that the evaporation of water, decomposition of urea and gas phase chemical reactions are decoupled for most of the considered temperature range, a simple

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