

## Study of Urea-Water Solution Injection Spray in De-NO<sub>x</sub> SCR System

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### ABSTRACT

Future emission limits of diesel engines require additional effort for developing adequate and advanced Exhaust Gas Aftertreatment devices. Urea-Selective Catalytic Reduction (SCR) systems are the promising approach to reduce Nitric Oxide emissions from diesel engines. Due to the toxicity and handling problems of ammonia, the non-toxic and commonly available reductant urea is used, and is injected in front of the SCR converter. This work presents a comprehensive study of Urea-Water Solution (UWS) injection spray using commercial CFD code Fire v8.3. The evaporation of water from a single droplet of UWS is investigated theoretically by Rapid Mixing and Diffusion Limit models. UWS droplets are treated with Lagrangian particle tracking and the evaporation model is extended for droplet boiling and thermal decomposition of urea. Computational Fluid Dynamics (CFD) simulations at various temperatures, injector locations, number of injector holes was carried out. Thus, this CFD study predicts the local distribution and the conversion of the reducing agent. Results of CFD study are compared with experimental values.

**Key word : SCR, CFD, Nox, Injection, Spray**

### Introduction

For many years, the legislative restriction of vehicle exhaust emissions has shown a steady trend towards more stringent limits. One of the major concerns on heavy duty diesel vehicles is the reduction of nitric oxides. This requires advanced aftertreatment techniques such as Selective Catalytic Reduction (SCR) systems. The feasibility of SCR systems is investigated and underlined by the European commission, reports that Urea-SCR systems are the most promising approach to comply with EURO IV and V emission standards. Urea-SCR has been widely used since 1980 to reduce NO<sub>x</sub> emissions from exhaust gas in stationary applications. More recently, the technology has been applied to mobile diesel engine applications such as heavy duty trucks, ships, etc. The development of Urea-SCR system is still a challenging task for mobile diesel engines [1].

Urea is preferred as reducing agent for automotive SCR applications because it possesses considerable advantages in terms of toxicity and handling. Nevertheless, the essential thermal decomposition of urea into NH<sub>3</sub> and carbon dioxide makes the DeNO<sub>x</sub> process complicated. The commercially available Urea-Water Solution (UWS) contains 32.5wt% urea (eutectic composition) and behavior of UWS droplets in a heated environment is not well understood. The theoretical investigation of the evaporation of UWS droplets is presented by some researchers [1].

Since mobile SCR systems have only a short distance between UWS injector position and catalyst entrance; which leads to possible incomplete decomposition into ammonia and HCNO and significant performance loss of SCR catalyst. The model containing both evaporation of water and thermal decomposition of urea is implemented into the CFD code Fire v8.3 [2].

### Homogeneous Urea-SCR Reactions

The formation of NH<sub>3</sub> out of UWS droplets follows several principle steps [1, 3]:

- Evaporation of water from a fine spray of UWS droplets  
 $(\text{NH}_2)_2\text{CO}(\text{l}) \rightarrow (\text{NH}_2)_2\text{CO}(\text{s}) + \text{H}_2\text{O}(\text{g})$  (1)
- Thermolysis of urea into ammonia and iso-cyanic acid,  
 $(\text{NH}_2)_2\text{CO}(\text{s}) \rightarrow \text{NH}_3(\text{g}) + \text{HCNO}(\text{g})$  (2)
- Hydrolysis of isocyanic acid,  
 $\text{HCNO}(\text{g}) + \text{H}_2\text{O} \rightarrow \text{CO}_2(\text{g}) + \text{NH}_3(\text{g})$  (3)

Koebel et al. studied the evaporation of urea solution in the exhaust gas stream; a solid or molten urea will be produced after evaporation of water from the droplet. Scaber et al. found that urea melts at 1330C and thermal decomposition of urea into NH<sub>3</sub> and HCNO started. Helden et al. numerically estimated the mixing efficiency of injected urea solution in the hot gas stream and uniformity of urea concentration in the catalyst with water as reducing agent of NO<sub>x</sub>. Munnannur and Liu used single kinetic rate model in conjunction with homogeneous finite rate chemistry calculations to model thermolysis and hydrolysis of urea with reasonable agreement of experimental data reported by Kim et al. Birkhold et al. developed a comprehensive model that accounts for the injection of UWS and spray in hot gas, spray wall interactions and implemented into CFD. Van Helden et al. used water instead of UWS in a CFD study and estimated the concentration of the reducing agent from the water vapor concentration. Wurzenberger and Wanker modeled the thermal decomposition as a homogeneous gas phase reaction, which followed the evaporation of UWS to water vapor and gaseous urea. Chen and Williams and Deur et al. assumed the thermal decomposition to occur instantaneously relative to the evaporation rate.

Cremer et al assumed a fast decomposition of urea after the evaporation of pure water. So it is not yet completely known what is the behavior of UWS droplets in the heated environment.

**CFD Methodology**

**Fluid Flow and Spray:** Since spray simulation comprises multi-phase flow phenomena, it requires the simultaneous solution of the conservation equations of both the gas and the liquid phases. Spray model of the liquid phase is based on a statistical method, the Discrete Droplet Method (DDM), as discussed by Dukowicz. Droplet parcels are introduced in the flow domain at a given spatial position with specified size, velocity, temperature, and number of droplets in the parcels. Using these initial conditions, the model equations of the momentum, trajectory, heat and mass transfer of each single droplet can be solved and the balance equations can be found in AVL FIRE user's manual [14].

**Basic Equations for Mass and Energy:** Assuming drag and gravity are the only relevant forces, the momentum balances equation of droplet parcels is given by,

$$\frac{du_{i,d}}{dt} = \frac{3}{4} \cdot \frac{\rho_g}{\rho_d} \cdot \frac{C_D}{D_d} \cdot |u_{i,g} - u_{i,d}| \cdot (u_{i,g} - u_{i,d}) + \left(1 - \frac{\rho_g}{\rho_d}\right) \cdot g_i \tag{4}$$

where  $u_{i,d}$  and  $u_{i,g}$  are the velocity components in the direction  $i$  of the droplets and the gas flow, respectively.  $\rho_d, \rho_g$  represent the densities of the droplets and the gas.  $D_d$  is the diameter of the droplet and  $g_i$  the gravitational acceleration. The general drag coefficient  $C_D$  is evaluated (Schiller and Naumann [15]) as a function of the droplet Reynolds number  $Re_d$

$$C_D = \frac{24}{Re_d} (1 + 0.15) Re_d^{0.687} \tag{5}$$

$$= 0.44 \quad \begin{matrix} Re_d < 1000 \\ Re_d \geq 1000 \end{matrix}$$

The integration of Equation (4) leads to the droplet velocity. Consequently the instantaneous position of the droplet parcel can be evaluated using

$$\frac{dx_{i,d}}{dt} = u_{i,g} \tag{5}$$

where  $dx_{i,d}$  is the component  $i$  of the droplet position vector in space domain.

**Evaporation of UWS Droplet:** The influence of urea on the evaporation of water from a UWS droplet is investigated theoretically by different evaporation models considering droplet motion and variable properties of UWS and the ambient gas phase. To evaluate the influence of dissolved urea on the evaporation of water in liquid phase, three different evaporation models are used:

**Liquid Phase**

**Rapid Mixing Model (RM Model)**

Within the RM model infinite high transport coefficients are assumed for the liquid phase, resulting in spatial uniform temperature, concentration and fluid properties in the droplet, but the quantities will change in time. The variation of urea concentration of the droplet can be evaluated by

$$\frac{dY_u}{dt} = - \frac{\dot{m}_{vap}}{m_d} Y_u \tag{6}$$

**Diffusion Limit Model (DL Model):**

Neglecting internal convection, only diffusive transport of energy and mass are assumed. The diffusion equation for species and energy in the droplet is solved considering

variable fluid properties.

$$\frac{\partial Y_u}{\partial t} = \frac{\Gamma_u}{r_d^2} \left[ \frac{\partial^2 Y_u}{\partial w^2} + \left( \frac{2}{w} + \frac{1}{\rho_d} \frac{\partial \rho_d}{\partial w} + \frac{1}{\Gamma_u} \frac{\partial \Gamma_u}{\partial w} + \frac{r_d}{\Gamma_u} \left( w \frac{dr_d}{dt} - u_r \right) \right) \frac{\partial Y_u}{\partial w} \right] \tag{7}$$

$$\frac{\partial T_d}{\partial t} = \frac{a_d}{r_d^2} \left[ \frac{\partial^2 T_d}{\partial w^2} + \left( \frac{2}{w} + \frac{1}{\lambda_d} \frac{\partial \lambda_d}{\partial w} + \frac{r_d}{a_d} \left( w \frac{dr_d}{dt} - u_r \right) \right) \frac{\partial T_d}{\partial w} \right] \tag{8}$$

with

$$a_d = \frac{\lambda_d}{\rho_d c_{p,d}} \tag{9}$$

$u_r$  denotes a radial convective velocity which accounts for the variable density field within the droplet.

**Effective Diffusion Model(ED Model):**

The ED model accounts for internal circulation due to forced convection. It considers internal circulation by an empirical correction of the transport co-efficients  $\Gamma_u$  and  $\lambda_d$

**Gas Phase:**

For the gas phase the quasi-steady model [17] is used. The differential equations for droplet mass and temperature can be derived from energy balance as follows.

$$\frac{dm_d}{dt} = -\pi D_d \rho_{g,ref} \Gamma_{g,ref} Sh^* \ln(1 + B_M) \tag{0}$$

$$\frac{dT_d}{dt} = \frac{\dot{m}_{vap}}{m_d c_{p,d}} \left( \frac{c_{p,vap,ref}(T_g - T_d)}{B_T} - h_{vap} \right) \tag{1}$$

$$B_M = \frac{Y_{vap,s} - Y_{vap,\infty}}{1 - Y_{vap,s}}$$

and

$$B_T = (1 + B_M)^x - 1, \text{ where } x = \frac{c_{p,vap,ref} Sh^*}{c_{p,g,ref} Nu^* Le} \tag{2}$$

$$\frac{dm_d}{dt} = -\pi D_d \frac{\lambda_{g,ref}}{C_{p,vap,ref}} Nu^* \ln(1 + B_T)$$

with

$$B_T = \frac{c_{p,vap,ref}(T_g - T_s)}{h_{vap}}$$

The RM and DL models describe the physical limits of infinite high and only diffusive transport. As the temperature and the urea concentration changes strongly during evaporation, variable fluid properties are used. For the calculations, it is additionally assumed that no crystallization of urea occurs. The droplets are assumed to be spherical throughout the evaporation and decomposition processes.

**Mass, Heat Transfer and Evaporation**

In the present work, a transfer temperature, uniform gas model derived by Dukowicz is applied. The model assumes spherical symmetry, a quasi steady gas film around the droplet, uniform droplet properties, and phase and thermal equilibrium at the droplet surface. The mass balance of the droplet is given by

$$\frac{dm_d}{dt} = A_d \cdot \dot{m}_{vap} \tag{13}$$

where, the time derivative of the droplet mass  $m_d$  depends on the specific vapor mass flux  $\dot{m}_{vap}$  vaporizing from or condensing on the droplet surface  $A_d$ . The energy balance of the droplet is

$$m_d \cdot c_{p,d} \frac{dT_d}{dt} = \Delta h_{vap} \cdot A_d \cdot \dot{m}_{vap} + Q' \tag{14}$$

By introducing a specific surface energy flux  $\dot{q} = \frac{Q'}{A_d}$

the balance equations of the mass and energy can be rearranged to

$$\frac{dm_d}{dt} = Q' \cdot \frac{\dot{m}_{vap}}{\dot{q}}$$

$$m_d \cdot c_{p,d} \cdot \frac{dT_d}{dt} = Q' \cdot \left(1 + \Delta h_{vap} \cdot \frac{\dot{m}_{vap}}{\dot{q}}\right)$$

Equations 14-15 define the transient behavior of the droplet provided that the expressions  $Q'$  and  $\frac{\dot{m}_{vap}}{\dot{q}}$  are known.

The energy transfer flux  $Q'$  is calculated

$$Q' = A_d \cdot \alpha \cdot (T_g - T_d)$$

$\alpha$  is a general heat transfer coefficient, which is typically derived from Nusselt correlations. The fraction of specific

heat to mass flux at the droplet surface  $\frac{\dot{m}_{vap}}{\dot{q}}$  is evaluated

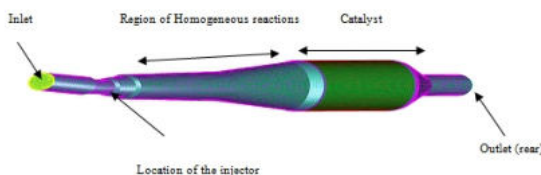
based on analogy considerations. It is assumed that the differential equations and boundary conditions for the heat and mass transfer problems are similar. Based on a rigorous theoretical study, Dukowicz derived the following expression,

$$\frac{\dot{m}_{vap}}{\dot{q}} = \frac{Le \cdot (w_{vap,d} - w_{vap,\infty}) \cdot (1 - w_{vap,d})^{-1}}{h_\infty - h_d - (h_{vap,d} - h_{g,d}) \cdot (w_{vap,d} - w_{g,d})}$$

**Cfd Model**

The amount of urea injected into gas stream is set to produce a stoichiometric amount of ammonia inside the converter. The present study allows to judge different SCR exhaust system configurations with respect to conversion and local distribution of reducing agent. For our study emphasis is given to on Hydrolysis and thermolysis of UWS which are contributing for the formation and distribution of ammonia. The simulation is done considering the gas phase reaction module of Fire v8.3 to characterize the ammonia distribution along entry to the SCR. The study has been carried out for proper positioning of the urea injector, entry shape of the SCR and geometry of the injector (Fig. 1 for SCR domain).

Figure 1: Typical SCR system for CFD study



For the calculation of the injection of UWS, the models for evaporation and thermal decomposition are implemented into the CFD code Fire v8.3. UWS droplets are treated with Lagrangian particle tracking, which solves the equation of motion for parcels of droplets with identical properties using the Discrete Droplet Method of Dukowicz.

Table 1: Properties of Urea-Water Solution

Density[ kg/m <sup>3</sup> ]	1089
Boiling point temperature[ °C ]	104
Thermal conductivity[ W/m K]	0.57
Specific heat[ kJ/kg K]	3.4
Surface tension[N/m]	0.0717
Latent heat of vaporization[ kJ/kg ]	2258

Table 2: Exhaust Gas Properties and Spray Parameters

Species	Mass fraction of the species at the Inlet	Mass fraction of the species at the Outlet
NO	6.7553E-04	0
NO <sub>2</sub>	5.1709E-04	0
NH <sub>3</sub>	1E-10	0
H <sub>2</sub> O	7.7E-02	0
O <sub>2</sub>	1.3E-01	0
CO <sub>2</sub>	9.0E-02	0
HCNO	1E-10	0
N <sub>2</sub>	0.701807	1

Table 3: Exhaust Gas Properties and Spray Parameters

Exhaust	
Exhaust temperature	573723 K
Wall temperature	300 K
Flow rate	0.23575 kg/sec
Spray	
Injection velocity	27 m/s
Injection temperature	293 K
Injection mass in the SCR domain	0.4gms
Injector locations	0.9-1.35 m (from exit)

**Results And Discussion**

**Spray Validation:** To evaluate the influence of variation of exhaust gas temperature, varying nozzles, and injector mounting positions, some known spray parameters are essential for CFD simulation as boundary conditions. In the present simulation, a UWS of 0.4gms is injected in the SCR domain for 0.3 secs from an orifice of 0.1mm outer diameter and an injection velocity of 27m/s with average particle sizes of 8E-005m. The spray distribution is shown in the Figs 2(a) and (b). It is observed that axial velocity varies with respect to droplet size. Small droplets are accelerated due to momentum transfer between exhaust gases and spray droplets.

**Evaporation and Thermolysis of the Droplets:** The influence of urea on the evaporation of UWS has been studied. The decrease in vapor pressure due to increase in concentration of urea in the droplet results in continuous increase in droplet temperature and slower evaporation as compared to water [18]. Fig. 3 shows the variation of mass fraction of water with residence time. The initial water mass fraction in the exhaust gas is found to be 0.077 and it starts increasing up to residence times of 0.4secs at all temperatures 300, 350 and 4000C. The overall water mass fraction increases in the initial stages, i.e. till 0.4 secs and it starts decreasing at the later stages due to evaporation and the consumption of water, during the conversion of HCNO into NH<sub>3</sub>.

**Droplet Density:** The properties of 32.5% solution of urea are provided in Table 1. A spray cone angle of 179° and injection velocity of 27m/sec are maintained in the simulation. The scattering of the droplets along the length of SCR converter with residence times at 300°C are depicted in Fig 3(a) and (b). Complete evaporation takes place after 0.5 secs. Spray droplet density during initial stages of spray is around 1050kg/m<sup>3</sup> at the entrance. During evaporation, spray density reduces and the droplets start disappearing in the converter. Simulations done at higher temperatures 350°C and 400°C have not shown any appreciable changes.

Figure 2: Variation of a) droplet diameter, b) density at 350°C

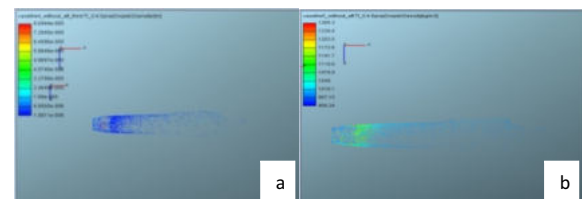
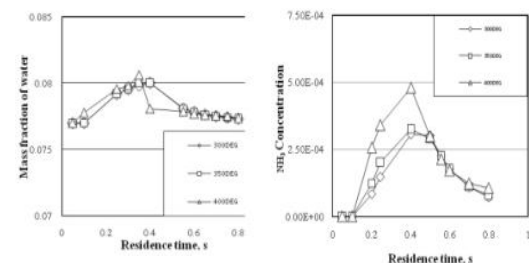


Figure 3: Variation of mass fraction of water with exhaust gas temperature

Figure 4: Variation of ammonia concentration exhaust gas temperature

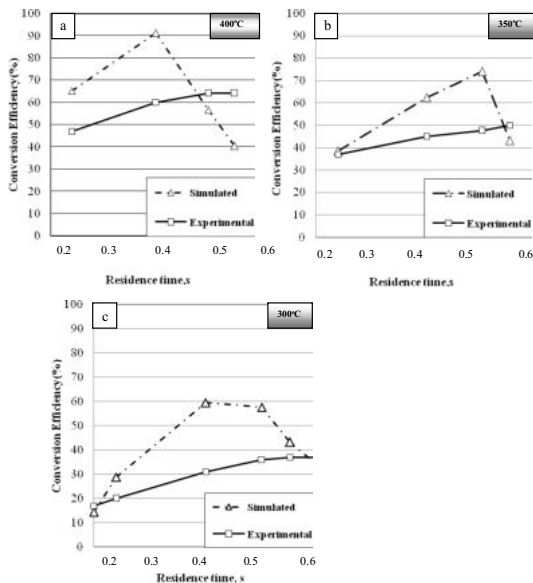


Ammonia Distribution in SCR Domain

(i) NH<sub>3</sub> Variation with Temperature

In the CFD domain, simulated the urea injection along entry to SCR converter and homogeneous urea hydrolysis reaction corresponds to a residence time of 0.4secs (i.e., before SCR catalyst). An analysis is carried out for exhaust gas temperatures of 300, 350 and 400°C. The simulation results showed that the ammonia concentration is increasing at 400°C (Fig.4) which is due to the effect of temperature on thermolysis of the urea droplets. The observed NH<sub>3</sub> conversion efficiency is found to be varying with exhaust gas temperature. The simulated NH<sub>3</sub> conversion results are compared with experimental values of Kim et al. [7] carried out at exhaust gas velocity of 10m/s, Fig.5a, b and c.

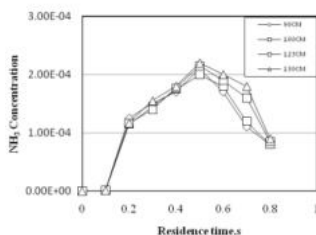
Figure 5: Comparison of simulated and experimental results at a) 400°C b) 350°C c) 300°C



An average axial velocity of 10m/s and mass flow rate of exhaust gas as 0.235kg/sec is considered in the CFD simulation. The observed values of ammonia formation are altered to "NH<sub>3</sub> Conversion Efficiency" term, which is the ratio of amount of ammonia produced as per experimental values or from simulated results with respect to gas flow rate and injected urea to corresponding value for 100% urea conversion. The simulated NH<sub>3</sub> conversion efficiency is compared with the experimental results obtained by Kim et al. [7] at various temperatures for residence times up to 0.6secs. The simulated values found higher compared to experimental values at all temperatures. These variations could be due to uniform distribution of UWS in SCR domain and increase in rate of evaporation. The average differences found to be 10.6%, 21% and 35% at 400°C, 350°C and 300°C respectively, Fig. 5(a)-5(c).

(ii) NH<sub>3</sub> Variation with Injector Positions

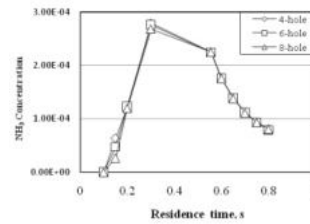
Figure 6: Variation of ammonia concentration with different injector locations (distance measured from the exit of the converter) at 350°C



Ammonia concentration values for different injector locations are plotted in Fig.6 at an exhaust gas temperature of 350°C. The initial concentration of ammonia is 1E -10 which is increasing during initial stages till 0.5 secs due to the gas phase reactions like thermolysis of urea. The observed maximum ammonia concentration is 2.2E-4 (Fig 6). The simulation results revealed that maximum NH<sub>3</sub> conversion takes place at injector locations of 115-125cm. For the shorter distances the space for mixing and residence time will reduce which may lead to incomplete evaporation, thermolysis and subsequently less ammonia formation. For the larger distances the ammonia which is formed may oxidize which reduces the DeNO<sub>x</sub> effect.

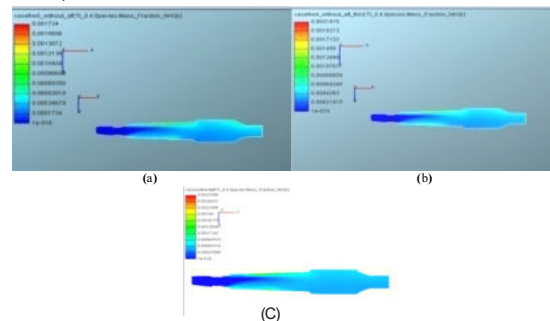
(iii) NH<sub>3</sub> Variation with Number of Injector Holes

Figure 7: Variation of ammonia concentration with 4, 6, 8-hole injectors



Similarly, the spray structure of urea solution varies with number of holes in the injector intern the uniformity of the NH<sub>3</sub> concentration distribution. The uniformity of 2-hole injectors was shown to be much lower than that of other multi hole injectors because an excessively increased droplet mass flux through each hole accompanies strong droplet inertia and allows deep penetration of injected droplet into exhaust gas stream causing flow along the surface wall of the diffuser. The injectors with greater number of holes can spray droplets more effectively than the injector with smaller number of injector holes more effectively at early stages of injection causing high efficiency of space utilization. In the present work, 4-hole, 6-hole and 8-hole injectors are used. The conversion of ammonia showed similar trend in all the three cases (Fig.7); it indicates number spray holes do not have much impact on NH<sub>3</sub> distribution except at the early stages. Fig. 8 demarcates the distribution of NH<sub>3</sub> in the SCR domain at different exhaust gas temperatures.

Figure 8 (a-c) Ammonia distributions in the SCR domain at 300°C, 350°C and 400°C



Conclusions

In the present work, ammonia distribution in the SCR domain is studied for different (a) exhaust gas temperatures, (b) geometry of SCR injector and (c) positioning of the urea injector. Droplet density during initial stages of spray i.e. at the SCR domain entrance is around 1050kg/m<sup>3</sup>. During evaporation, its density reduces and the droplets almost start disappearing. It is observed that maximum NH<sub>3</sub> conversion takes place while positioning the urea injector at 115-125 cm

from exit of the converter. The overall results showed that ammonia concentration is increasing up to residence time of 0.4secs and decreasing thereafter because of the oxidation effect in SCR catalyst. Simulation results are compared with experimental values obtained by Kim et al.[7] for some similar initial conditions. NH<sub>3</sub> conversion efficiency of 80~90% is observed during the simulation and is in-line with current trends.

#### NOMENCLATURE

a	thermal conductivity	[m <sup>2</sup> /s]
A	area	[m <sup>2</sup> ]
B <sub>M,T</sub>	Spalding numbers	
c <sub>p</sub>	Heat capacity,	[J/kg K]
C <sub>D</sub>	drag co-efficient	
D	diameter,	[m]
E <sub>a</sub>	Activation energy,	[J/mol]
g	gravity	[m/s]
Δh	heat of reaction	[J/mole]
h	specific enthalpy,	[J/kg]
Le	Lewis number	
m	mass,	[kg]
$\dot{m}$	Mass flow	[kg/s]
p	pressure	[Pa]
Q'	Energy flux	[W]
N <sub>u</sub>	Nusselt number	
Re	Reynolds number	
q'	specific energy flux	[W/m <sup>2</sup> ]
r	radius,	[m]
$\dot{r}$	rate of reaction, s	[mol/m <sup>3</sup> ]
R	universal gas constant	[J/mol K]
Sh	Sherwood number	
t	time,	[s]
T	temperature,	[K]
u	velocity,	[m/s]
w	dimensionless radius, (r/r <sub>a</sub> )	
w	vapour mass fraction	
Y	mass fraction	

#### Greek symbols

α	heat transfer coefficient	[W/m <sup>2</sup> /K]
λ	heat conductivity,	[W/m K]
ρ	density	[Kg/m <sup>3</sup> ]
Γ	diffusion coefficient,	[m <sup>2</sup> /s]
λ	heat conductivity	[W/m/K]

#### Subscripts

s	ambient
d,g	droplet, gas
l,vap	liquid, vapor
s	droplet surface
D	drag
d	droplet
g	gas phase
i	reaction index
vap	vapor

#### Superscripts

*	characteristic
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