Effect of Injection Parameters on Evaporation and Thermolysis Characteristics of UWS (Urea-Water-Solution) in SCR (Selective Catalytic Reduction) Systems

Sadashiva Prabhu S¹, Nagaraj Shivappa Nayak² and Kapilan Natesan³

1. Canara Engineering College, Visvesvaraya Technological University, Karnataka 591702, India

2. Caledonian College of Engineering, Glasgow Caledonian University, CPO Seeb 111, Sultanate of Oman

3. Nagarjuna College of Engineering and Technology, Visvesvaraya Technological University, Karnataka 591702, India

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Abstract: UWS (optimized Urea-Water Solution) injection system is required to increase the NH_3 conversion efficiency of urea-based SCR (Selective Catalytic Reduction) system of modern automobiles. The focus of the current study is to do parametric studies by simulation in a three-dimensional model using CFD (Computational Fluid Dynamics) code AVL FIRE. Simulations were carried out to study the characteristics of evaporation and thermolysis UWS considering the effect of injection velocity, duration of injection, injection angle and for different types of injection. In the case of the injection velocities up to 20-50 m/sec, the ammonia concentration continues to increase. It is found that as the duration injection decreases, the concentration of ammonia increases. In case of continuous injection, the flow rate is less which results in lower velocity of injection, lesser atomization and slower evaporation resulting lesser conversion of UWS into NH_3 . Shorter duration of injection leads better atomization with increased velocity of injection which results in faster evaporation and thermolysis.

Key words: Evaporation, thermolysis, SCR, UWS, injection parameters.

Nomenclature

A_d	Surface area of droplet	(m^2)
A	Pre-exponential factor	$\text{kg}/(\text{m}\cdot\text{s})$
$B_{M,T}$	Spalding numbers	
C	Concentration	
C_p	Heat capacity	$\text{J}/(\text{kg}\cdot\text{K})$
D	Diameter	(m)
E_a	Activation energy	$(\text{J}\cdot\text{mol}^{-1})$
Δh	Heat of reaction	$(\text{J}\cdot\text{mol}^{-1})$
Le	Lewis number	
m	Mass	(kg)
$\dot{m}$	Mass flux	(kg/s)
$\dot{Q}$	Energy flux	(W)
Nu	Nusselt number	(hD/k)

$\dot{q}$	Specific energy flux	(W/m^2)
S	Source term	(kg/s)
Sh	Sherwood number	
t	Time	(s)
T	Temperature	(K)
u	Velocity	(m/s)
Σv	Diffusion volume	
Y	Mass fraction	

Greek Symbols

σ	Surface tension	(N/m)
h	Heat transfer coefficient	($\text{W}/\text{m}^2\cdot\text{K}$)
ρ	Density	(kg/m^3)
Γ	Diffusion coefficient	(m^2/s)
μ	Absolute viscosity	(Ns/m^2)
τ	Shear stress	(N/m^2)
ν	Kinematic viscosity	(m^2/s)
ε	Eddy viscosity	(m^2/s)

Corresponding author: Sadashiva Prabhu S, tech. associate professor, research fields: IC engines and emission control. E-mail: ss.prabhu97@gmail.com.

Subscripts

1, 2	Directions, phase
d,g	Droplet, gas
I	Interface
l,vap	Liquid, vapor
ref	Reference
g	Gas phase
m	Mass
s	Solid phase
sat	Saturation
v	Vapour
w	Wall

Superscripts

*	Characteristic
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1. Introduction

There are several methods to eliminate NO_x emitted from exhaust of diesel engines namely Humid Air Method, Exhaust Gas Recirculation, Water Injection, High Scavenging etc.. Compared to these methods of removal NO_x from the exhaust gases, the SCR (Selective Catalytic Reduction) based on UWS (Urea Water Solution) is an effective technique to reduce NO_x (Nitrogen Oxides) emitted from the diesel engines without altering the performance of the engines. UWS (containing 32.5 wt% urea, brand name: Ad Blue) is sprayed into the hot exhaust gas stream which gives reducing agent NH_3 after evaporation and thermolysis [1].

The major part the SCR system is the injection system where the injection parameters play a major role in evaporation and thermolysis characteristics. The impingement of droplets on the catalyst and on walls cannot be avoided due to slow evaporation and thermolysis, as well as the inertia of the droplets in actual exhaust configurations [2]. Birkhold et al. [3] developed a model that accounts for the injection of UWS, spray, thermolysis, spray-wall interaction etc. and implemented into CFD (Computational Fluid Dynamics) code and validated the results with experimental data of Kim et al. [4].

Evaporation from the wall film leads to further

cooling and an increasing risk of formation of melamine complexes [1]. Birkhold et al. [2] theoretically investigated the evaporation of water from a single droplet of UWS by a rapid mixing model and a diffusion limit model. The results show that the decrease in vapor pressure due to increasing concentration of urea in the droplet results in a continuous increase of the droplet temperature and a slower evaporation compared to pure water. Helden et al. [5] used water instead of UWS in a CFD study and estimated the concentration of the reducing agent from the water vapor concentration.

To the best of our knowledge, there are no much of parametric studies published on the effects varying the UWS injection parameters like injection velocity, injection duration on spray evaporation and thermolysis with consideration of wall interaction. To predict the generation and distribution of the reducing agent a detailed three-dimensional numerical model is developed and implemented in the commercial CFD code AVL FIRE [6]. The developed model for SCR design optimization is capable of accurately predicting the effect of some of injection parameters on: (1) the atomization process of the injected liquid jet; (2) the vaporization and thermolysis processes of UWS droplets in the heated and turbulent exhaust gas environment; and (3) the spray/wall interaction.

2. CFD Methodology

2.1 Evaporation Model Including Urea Thermolysis

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. The separate models for evaporation at gas and liquid phases are discussed below:

- Liquid phase:

RM model (Rapid Mixing model) [7, 8]:

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:

$$\frac{dY_u}{dt} = -\frac{\dot{m}}{m_d} Y_u \quad (1)$$

where the mass flow from liquid to gaseous phase is defined to be negative.

• Gas phase:

For the gas phase, the quasi-steady model [7, 8] is used. Evaporation rate prediction is sensitive to choice of property values. Analytical expressions for the diffusive transport fluxes are obtained by integration of transport equations for mass and enthalpy outside the droplet. The differential equations for droplet mass and temperature can be derived from energy balance as follows:

$$\frac{dm_d}{dt} = -\pi D_d \rho_{g,ref} \Gamma_{g,ref} Sh^* \ln(1 + B_M) \quad (2)$$

$$\frac{dT_d}{dt} = \frac{m_{vap}}{m_d c_{p,d}} \left(\frac{c_{p,vap,ref}(T_g - T_d)}{B_T} - h_{vap} \right) \quad (3)$$

where, $B_M = \frac{Y_{vap,s} - Y_{vap,g}}{1 - Y_{vap,s}}$ and:

$$B_T = (1 + B_M)^x - 1 \text{ and:}$$

$$x = \frac{c_{p,vap,ref} Sh^*}{c_{p,g,ref} Nu^* Le}$$

The Eq. (2) is modified as:

$$\frac{dm_d}{dt} = -\pi D_d \frac{\lambda_{g,ref}}{c_{p,vap,ref}} Nu^* \ln(1 + B_T) \quad (4)$$

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

As the temperature and the urea concentration change during evaporation, variable fluid properties are used. The droplets are assumed to be spherical

throughout the evaporation and decomposition processes [3].

In the modeling of thermolysis, there are different formulations mentioned in the literature. The reaction rate is described by an Arrhenius-type equation used by Kim et al. [4] as follows:

$$\frac{dm_{urea}}{dt} = -m_{urea} A \exp\left(-\frac{E_a}{RT}\right) \quad (5)$$

with the kinetic parameters $A = 382 \text{ s}^{-1}$, $E_a = 2.94 \times 10^7 \text{ Jmol}^{-1}$, R -universal gas constant, Birkhold et al.[2] used the experimental data from Kim et al. [4] for a parameter fit and the values for $A = 0.42 \text{ kg/ms}$, $E_a = 69,000 \text{ J/mol}$.

For thermolysis, following reaction is considered:

and the rate equation is:

$$\frac{d\alpha}{dt} = -A_r (1 - \alpha)^n \exp\left(-\frac{E_a}{RT}\right) \quad (6)$$

where the urea conversion fraction α is given by:

$$\alpha = \frac{m_{urea}(t=0) - m_{urea}(t)}{m_{urea}(t=0)} \quad \text{and} \quad \text{Buchholz} \quad [9]$$

suggests $A_r = 1 \times 10^6 \text{ s}^{-1}$, $E_a = 7.3 \times 10^4 \text{ Jmol}^{-1}$ and $n = 0.3$.

All of the above approaches are implemented in CFD code AVL FIRE under spray evaporation and gas phase reaction modules. The approach developed by Birkhold et al. [2] has been established in numerical code and default values of input parameters, frequency factor A_r and activation energy E_a are given as mentioned above.

2.2 Mass, Heat Transfer and Evaporation

In the present work, a uniform gas model derived by Dukowicz [9] is applied. The model assumes spherical symmetry, a quasi steady gas film around the droplet, uniform droplet properties, phase and thermal equilibrium at the droplet surface. The mass balance of

the droplet is given by:

$$\frac{dm_d}{dt} = A_d \dot{m}_{vap} \quad (7)$$

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 given by:

$$m_d c_{p,d} \frac{dT_d}{dt} = \Delta h_{vap} A_d \dot{m}_{vap} + \dot{Q} \quad (8)$$

By introducing a specific surface energy flux $\dot{q} = \frac{\dot{Q}}{A_d}$ the balance equations of the mass and energy can be re-arranged as:

$$\frac{dm_d}{dt} = \dot{Q} \frac{m_{vap}}{q} \quad (9)$$

$$m_d c_{p,d} \frac{dT_d}{dt} = \dot{Q} \left(1 + \Delta h_{vap} \cdot \frac{m_{vap}}{q} \right) \quad (10)$$

Eqs. (9) and (10) define the transient behavior of the droplet provided that the expressions $\dot{Q}$ and $\frac{m}{q}$ are known. The energy transfer flux $\dot{Q}$ is calculated as:

$$\dot{Q} = A_d h_d (T_g - T_d) \quad (11)$$

h_d is a general heat transfer coefficient. The mass and energy fluxes transferred between the individual droplets and the gas phase are applied as additional source terms in the balance equations of the fluid flow in their Eulerian formulation, it is possible to formulate any urea decomposition reaction as homogeneous gas phase reaction.

2.3 Wall Film Modeling

Deposition of droplets leads to a wall film which is modeled with a 2D finite volume method in numerical code. Gas and wall film flow are treated as separate single phases, coupled by semi-empirical boundary conditions. The film is transported due to shear forces, gravity and pressure gradients.

Continuity equation:

$$\frac{\partial \delta}{\partial t} = \frac{\partial(\delta u_1)}{\partial x} + \frac{\partial(\delta u_2)}{\partial y} + \frac{1}{\rho A_{cell}} S_m \quad (12)$$

The usual step would be to employ the momentum equations for solving the velocity components u_1 and u_2 . Let us assume that the velocity components are known as well as the source term.

Now, the convective terms $\frac{\partial(\delta u_1)}{\partial x}$, $\frac{\partial(\delta u_2)}{\partial y}$ is evaluated and Eq. (12) can be solved explicitly.

The viscosity increases with increasing urea concentration and leads to an increase of the film thickness and shear stress and a tendency to a laminar film profile [6].

2.4 Film Velocity Profiles

One of the key features of the FIRE wall film model is the use of analytical film velocity profiles instead of a momentum equation. As inertial effects are ignored, the profiles introduced here are steady state. As long as the film is thin and no transient forces are taken into account, steady state conditions are reached within fractions of a second. The distribution of shear forces across the film directly relates to the velocity profile. Using Boussinesq hypothesis for turbulent eddy viscosity ϵ , the velocity profile of film due to applied shear force τ can be written as:

$$\frac{\tau}{\rho} = (\nu + \epsilon) \frac{\partial u}{\partial y} \quad (13)$$

Finally, the mean film velocity, which is necessary for equation is obtained by integrating over film thickness:

$$u_f = \frac{\delta}{6\mu} 2\delta \left(\rho g - \frac{dp}{dx} \right) + 3\tau_I \quad (14)$$

where τ_I is interfacial shear force.

2.5 Evaporation and Thermolysis from Wall Film

The multi-component model developed for evaporation and thermolysis of UWS droplets which

are impinging at the walls. The UWS droplets are heated up and due to the low vapor pressure of urea compared to the vapor pressure of water, the water evaporates first from the droplet which leads to a spatial urea concentration gradient with a maximum at the droplet surface. Thus solved urea at the droplet surface causes a decrease of the vapor pressure of the water. For this purpose, a two stage process is assumed, where evaporation of water takes place initially from the multi species film and if the mass fraction reduces by 5%, then thermolysis of urea takes place based on an Arrhenius approach.

Evaporation wall film is based on Fick's law of diffusion which is given by:

$$\dot{m} = \left[\frac{\rho_v (D_{12} + D_t)}{1 - c_t} \right] \frac{\partial c}{\partial y} \quad (15)$$

This is the evaporation mass flux ($\text{kg}/(\text{s}\cdot\text{m}^2)$), or evaporation rate, which depends primarily on the following parameters:

(1) Concentration gradient has probably the greatest influence on evaporation rate. If the gas above the film is already saturated with vapor, the gradient goes to zero and evaporation declines;

(2) The influence of temperature, reflected by the temperature dependent properties c_t and D_{12} and time dependant properties D_t .

Wall film energy equation and the evaporation routines are coupled in this study through a time step adaptation to avoid numerical instabilities and to solve the steep gradients of heating and evaporation of the wall film. The derived models implemented in the numerical code help to predict the real processes during the layout of exhaust tube configurations and injector mounting positions with respect to the spatial distribution of the reducing agent at the upstream of the catalyst. Birkhold et al. [2] suggested the model for urea decomposition rate which is in the form:

$$\frac{dm_{urea}}{dt} = -Area D_{filmpatch} A \exp \left(-\frac{E_a}{RT} \right) \quad (16)$$

Fig. 1 The dimensions of CFD model

They utilized the experimental data from Kim et al. [4] for a parameter fit and get $A = 0.42 \text{ kg/ms}$, $E_a = 6.9E + 3 \text{ J/mol}$, $n = 1$. $D_{filmthick}$ is the local film thickness.

3. CFD Model

The model developed by Kim et al. [4] is used for our studies. Fig. 1 show the dimensions CFD model.

4. Results and Discussion

4.1 Effect of Injection Velocity

Droplet evaporation: the evaporation of single droplet is studied earlier by Wang et al. [10] is for larger droplet compared those which are really seen in SCR applications. Grout et al. [11] used the dependency of d^2 -law and obtained the expression for evaporation time. During evaporation and subsequent thermolysis process the parameters of UWS injection play major role. Birkhold et al. [2] did numerical studies on UWS evaporation and thermolysis behavior using CFD code AVL FIRE and validated with experimental results of Kim et al. [4] at different exhaust gas flow rates and temperatures.

In our work, for the experimental set up developed by Kim et al. [4], the simulations are done to optimize the NH_3 formation in the SCR converter. The initial droplet size is $50 \mu\text{m}$ and UWS is injected at start time 0.1 s and end time 0.4 s. There are no correlations to predict the droplet sizes of UWS with injection velocity. But Kim et al. [4] have given some experimental values at the downstream of the injection point but they are not exact values to be used in our simulation. Obviously at the nozzle tip, the velocity will be higher. So a range of velocity 20-50 m/s is chosen to study the behavior of UWS at different temperatures.

Figs. 2a-2c indicate the faster evaporation UWS at higher injection velocity and higher temperature. The higher velocities, i.e., > 50 m/s droplets do not find sufficient time for evaporation and thermolysis and simulation diverges in such cases. The droplet evaporation and their size diminishing along the length of SCR converter are shown in the Figs. 2a-2c.

Evaporation from wall: Nagaraj et al. [12] numerically predicted that more film deposition occurs

Fig. 2 Comparison of droplet evaporation characteristics with various injection velocities at (a) 573 K (b) 623 K and (c) 673 K .

Fig. 3 Evaporated film mass at various injection velocities.

at lower temperatures. At temperature 300°C , the evaporation behavior is studied for different injection velocities. As the injection velocity increases, the droplet size reduces which enhances the evaporation (Fig. 3).

Thermolysis behavior: NH_3 distribution for various injection velocities at 573 K, 623 K and 673 K are compared in Figs. 4a-4c. The NH_3 conversion efficiency is found to be high at elevated temperature. At higher injection velocities, the spray penetration increases and droplet size decreases which leads to incomplete evaporation and thermolysis due to decreased residence time and reduced inertial effect. However, as the droplet size decreases with increase in injection velocity, resulting in better atomization of the droplets which may lead to faster evaporation and thermolysis. At 673 K, maximum NH_3 concentration is for injection velocity in the range of 40-50 m/s. Figs. 4a-4c show NH_3 concentration at transient injection conditions for temperatures 573 K, 623 K and 673 K, respectively.

4.2 Effect of Injection Duration

Evaporation behavior: to study the behavior of UWS evaporation and thermolysis for various injection durations, the injection duration ranging from 0.1 s to 0.5 s is set in the simulation. The quantity 0.33 gms of UWS is injected for different injection durations resulting different velocities of injection. It is observed that from Fig. 5a that the evaporation is faster for shorter injection duration. The slope of the curve

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Fig. 4 Variation of NH_3 concentration with different injection velocities for pulsed injection (a) 573 K (b) 623 K (c) 673 K.

referring for injection duration 0.1-0.2 s shows the highest evaporation rate while the same is found decreasing as injection time increases. It is also evident from Fig. 5b that droplet diameter decreases from original size 50 μm at the onset of injection and it

Fig. 5 Variation of NH_3 concentration with different injection durations at (a) Liquid mass evaporated (b) Variation of droplet diameter (c) Mean NH_3 mass fraction at 673 K.

vanishes at 0.9 s for injection duration 0.1 s. Thermolysis characteristics show that NH_3 concentration increases with injection durations (Figs. 5a-5c).

4.3 Effect of Injection Angle

Injection angle is the key factor in spatial distribution on UWS in exhaust gas environment, mixing, heat transfer between UWS and exhaust gas. As the injection angle increases, the radial distribution increases. This results in better atomization and distribution of UWS with increased ammonia concentration (Fig. 6a). The radial distribution causes better mixing and increased residence time. Hyun et al. [13] have studied the evaporation and thermolysis behavior for angles -3° , 15° and 45° .

The average film area and film thickness were plotted and results reveal that no film formation occurs at injection angle below 80° and it increases till 140° . The two major parameters like film area and film thickness are plotted with respect to residence time (Figs. 6c-6d). The film formed exists only for few fraction of second and disappears as evaporation and thermolysis progresses from wall.

4.4 Effect of Type of Injection

Injection system in SCR system can be of two types namely continuous and pulsed injection with desired frequency. In continuous type injection, the desired quantity is injected continuously with low injection pressure and lesser spray penetration. Uniformity of mixing across the cross section cannot be attained for such kind of injection. The continuous injection is modeled and simulation is done using required boundary conditions.

The simulation results obtained for the same shows the localized urea decomposition products along the length of SCR (Figs. 8a and 8b). In pulsed type of injection, the required quantity of UWS is injected for a particular duration and for such cases, radial distribution will be uniform and axial uniformity is attained by the turbulence of the exhaust gas. The results of the same are shown in Fig. 7. Mean concentration of NH_3 with residence time in SCR domain is plotted and shown in Figs. 7a and 7b for both continuous and pulsed injection system. Mean mass

Fig. 6 Variation in (a) NH_3 concentration at 0.8 m from injection point; (b) NH_3 mass fraction for various injection angle; (c) film area; (d) average film thicknesses at different injection angles at 673 K.

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Fig. 7 NH_3 formation for continuous and pulsed injection system at transient 673 K.

Fig. 8 Effect of temperature in the formation at NH_3 at 673 K for (a) pulsed injection; (b) continuous injection.

fraction of NH_3 for continuous injection found to be lower.

In case of pulsed injection system, the highest concentration found to be 280 PPM (by mass) for residence time 0.9 s at 400 °C and it is 180 PPM for continuous injection with 30° injection angle. The increased concentration of NH_3 is found for pulsed injection is due to shorter duration of injection which ends within 0.4 s.

Shorter duration of injection leads better atomization

with increased velocity of injection which results in faster evaporation and thermolysis. In case of steady state injection, lower velocity of injection leads to lesser atomization which may cause slower evaporation of UWS and more oxidation of NH_3 .

5. Conclusions

In this work, a three-dimensional numerical simulation of UWS is done to evaluate the effects of various parameters along with exhaust gas temperature on evaporation and thermolysis behavior of UWS. The following conclusions are drawn from this work:

- (1) Higher injection velocity causes better atomization and increased evaporation and thermolysis. For UWS injection velocities up to 20-50 m/s the NH_3 concentration continue to increase
- (2) At higher exhaust gas temperature, the higher injection velocity of UWS resulting higher NH_3 conversion for lesser residence time. At lower temperatures, the UWS requires more residence time and the injection velocity should be less;
- (3) Evaporation from wall increases as injection velocity increases. As the angle of injection increases, the evaporation and thermolysis processes will increase due to better dispersion of UWS and NH_3 concentration continues to increase.
- (4) Shorter duration of injection in pulsed flow gave higher NH_3 concentration compared to that of continuous injection.

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