

A Urea Decomposition Modeling Framework for SCR Systems

Thomas L. McKinley
Cummins Emission Solutions

Andrew G. Alleyne
University of Illinois at Urbana-Champaign

Copyright © 2009 SAE International

ABSTRACT

Selective catalytic reduction (SCR) is allowing diesel engines to reach NO_x emission levels which are unachievable in-cylinder. This technology is still evolving, and new catalyst formulations which provide higher performance and greater durability continue to be developed. Usually, their performance is measured on a flow reactor using ammonia as the reductant. However, in mobile applications a urea-water solution is used instead, and urea decomposition by thermolysis and hydrolysis provides the required ammonia to the catalyst. It is well known that urea decomposition is incomplete by the inlet face of the converter, and this is at least one reason why on-engine performance is generally lower than would be expected from reactor tests.

Previous modeling of urea-water droplets has focused on developing detailed sub-models that can be implemented into computational fluid dynamics (CFD) codes. However, the required computational effort is not compatible with real-time, controls-oriented models. This paper addresses that gap by presenting a framework for urea-decomposition modeling which employs the so-called probability distribution function (PDF) operator method. As a starting point, this paper focuses on operators of atomization, particle-borne decomposition, and catalyst impact. Furthermore, the significance of these effects are reported over a span of temperatures and space velocities. The framework is quite general and provides a means for integrating

detailed droplet models, three-dimensional CFD codes, and experimental measurements. Moreover, the approach can be applied to other two-phase flow arrangements such as fuel injection upstream of fuel reformers and diesel oxidation catalysts (DOCs).

INTRODUCTION

There is growing interest in urea-based SCR to meet future heavy-duty NO_x emission standards. Johnson [1] indicates that this technology will be a key to achieving EPA-2010, Euro VI, and final Tier-4 emissions legislation. SCR technology continues to evolve, and modeling plays an important role in this dynamic environment. For example, models are used for mixing studies, system and component sizing, system simulation, and are even embedded within control modules. Although complete three-dimensional, reacting, two-phase, transient simulation is possible, for some applications a one-dimensional model is preferred. This is especially true of controls development and mission simulation, where the model may need to run in real-time.

In many cases, one-dimensional models are based on flow reactor experiments using NH₃ as a reductant. However, on engine test beds the NH₃ slip limited NO_x conversion efficiency is generally lower using aqueous urea solution (AUS) than gaseous ammonia [2]. This performance offset can have multiple causes such as: (1) lack of mixture uniformity; (2) slower kinetics with urea compared to NH₃; and (3) bound NH₃ in the form

of AUS or urea particles which may not interact with the catalyst surface. It has been demonstrated that, with careful design, sufficient mixing uniformity can be achieved [3, 4]. However, without entirely new dosing systems [5, 6] or additional investment in urea decomposition or hydrolysis catalysts, the obstacle of converting AUS to NH₃ remains.

The significance of this obstacle was clearly demonstrated by Chi and DaCosta [7], who measured the conversion efficiency from AUS to gaseous NH₃ through a 3.0 m long exhaust pipe for a range of gas temperatures and mass flow rates (see Figure 1). The mesh corresponds to a curve fit, while the circles represent experimental measurements. Keeping in mind that the distance from the injection point to the inlet face of the catalyst is frequently 1.0 m or less, for virtually the entire operating range only a minority of the reagent is in the form of NH₃ by the time it reaches the catalyst.

Detailed models of AUS and urea particle dynamics have been developed [8] for implementation in three-dimensional CFD codes. The end goal of that work was to improve exhaust system designs using CFD. Starting from this foundation, our objective is instead to present a framework for implementing these effects into one-dimensional models. The near term goal is to improve models for a given design, especially for controls development and mission simulation, where real-time performance is expected if not required. Over the longer term, this enables performance gains by improving tools used by controls and system integration engineers.

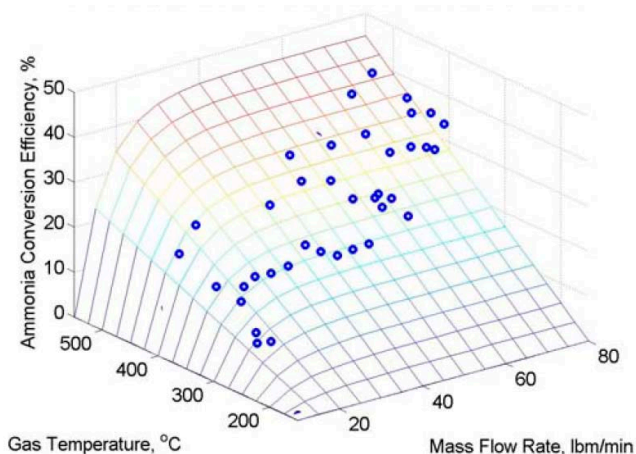


Figure 1: Conversion Efficiency of Urea to NH₃ at 3 m from Injection Location (Ref: Chi and DaCosta [7])

UREA DECOMPOSITION MODELING FRAMEWORK

As mentioned above, our objective is to provide a framework for a first order, physically-based, urea decomposition model. A starting point is to identify the

phenomena which occur, dividing the overall model into individual sub-models. This facet-wise decomposition of large problems into manageable chunks aligns with Goldberg's concept of a patchwork quilt of sub-models [9].

The power of this decomposition is two fold. First, after determining the information flow between sub-models, development can proceed in parallel among multiple researchers at universities, consulting firms, and in industry. Second, the sub-models allow us to integrate data from a wide range of data sources, including reactor tests, test cell measurements, and three-dimensional CFD modeling.

Figure 2 shows the framework, overlaid on a sketch of a two brick SCR converter. The phenomena are listed in the boxes, with arrows indicating the time sequence of effects that a given AUS or urea particle may experience. Each of these phenomena (atomization, wall wetting, wall deposit formation and removal, particle-borne decomposition, and catalyst impact) would be represented through a sub-model.

For a two-brick arrangement, this can be clustered into four groups:

- **Upstream of the Converter** – After injection, AUS is atomized and a portion of it strikes the wall (wall wetting). This can lead to the formation and removal of urea deposits. Those particles not striking the wall continue toward the inlet of the first brick, experiencing particle-borne decomposition along the way.
- **Within the Front Brick** – Upon entering the first brick, a portion of the particles may strike the catalyst (catalyst impact). The remainder continues through the converter channels, experiencing particle-borne decomposition in transit.
- **Within the Rear Brick** – Upon entering the rear brick, the processes present in the front brick reoccur. Specifically, this entails another round of catalyst impact and particle-borne decomposition.
- **Downstream of the Converter** – After exiting the rear brick, any remaining particles continue to experience particle-borne decomposition.

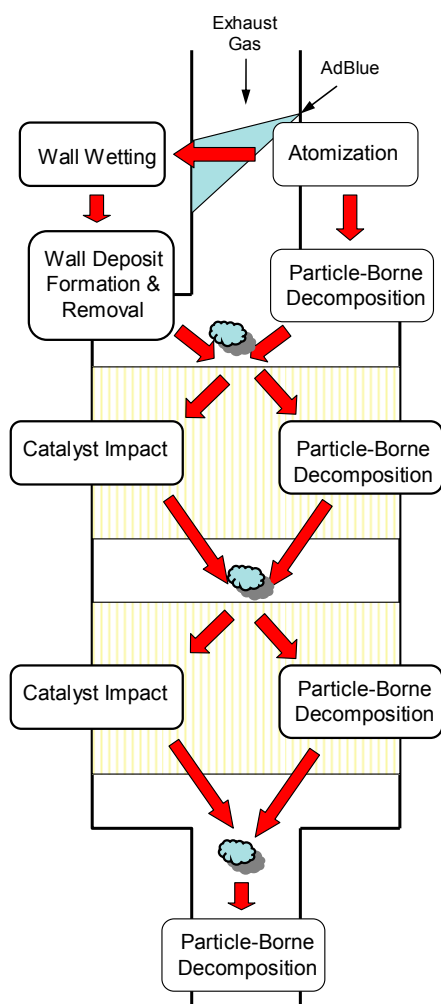


Figure 2: Urea Decomposition Modeling Framework

Before integrating the sub-models, the information flow between them must be defined. The so called *probability distribution function (PDF) operator method*, described in the following section, is used for this purpose.

THE PDF OPERATOR METHOD

The intent of the urea decomposition framework is to track the evolution of AUS and urea particles throughout the catalyst. In CFD codes, this is commonly done by subdividing the particle population into different parcels containing particles having identical properties. This so called *discrete droplet method*, proposed by Dukowicz [10], tracks parcel positions in time, and particle states (mass, internal energy, composition, and thermodynamic phase) evolve over time due to interactions with the local environment. This Lagrangian approach is quite effective when our goal is a multi-dimensional description of particle distribution.

Because it is a type of Monte Carlo simulation, it is not unusual to use hundreds to thousands of parcels, resulting in a dynamic model of thousands to tens of thousands of states. On the other hand, controls-oriented modeling and mission simulation are generally expected to run in real-time. For that reason, the number of states must remain comparatively small (e.g. less than 10 to 100), and the sheer number of states in a discrete droplet model is a major obstacle.

Moreover, the particle time scales are quite fast, in many cases shorter than the residence time of the exhaust gas in the converter. In contrast, the limiting catalyst dynamics for energy and NH_3 storage are several orders of magnitude slower. This tremendous span in time scales can lead to very inefficient simulation, because the integrator time step is limited by the fastest time scale.

Instead, we introduce the so called PDF operator method. It is also based on the concept of parcels, but only a minimum number (e.g. 100) are used to represent the entire population. Our objective is not a multi-dimensional representation, but merely a one-dimensional one. Therefore, the parcels have no position as in a Lagrangian approach, instead possessing only residence times in different Eulerian control volumes. This reduced dimensionality and the neglect of particle-to-particle interactions justifies the use of significantly fewer parcels than in the discrete droplet method.

Each parcel is represented by a *property vector* having the following components:

- ☐ Initial droplet diameter
- ☐ PDF or fraction of injected AUS represented by the parcel
- ☐ Current lifetime phase
- ☐ Progress variable for the current lifetime phase

This defines the information flow needed to integrate the different sub-models. The property vector is initialized by the atomization operator and then updated by other *operators* corresponding to each of the phenomena listed in Figure 2. In this paper, we describe only three of them: atomization, particle-borne decomposition, and catalyst impact.

As described below, these operators are algebraic rather than dynamic, so the urea decomposition model does not increase the number of states. By eliminating the states, we also eliminate the problem of vastly different time scales mentioned previously. The end result is a urea decomposition model that is very efficient, grounded in physics, and suitable for implementation in controls-oriented modeling and mission simulation.

LIFETIME PHASES AND PROGRESS VARIABLES

As described above, each parcel is represented by a four dimensional property vector. Two components of this vector are the current *lifetime phase* and a *progress variable* corresponding to that phase. Specifically, from the time of atomization to extinction, the particle can experience up to five different lifetime phases. The phases are numbered sequentially and named for the dominant thermodynamic process occurring in each phase. The progress variable is a characteristic that changes during that phase and is used to mark the transition to the succeeding phase.

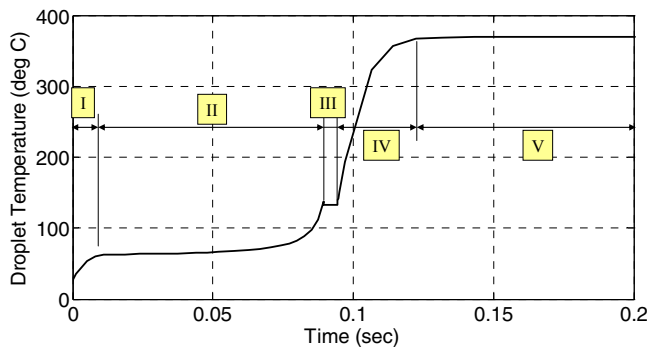


Figure 3: Five Phases of Particle Lifetimes and Their Relationship to Temperature (Illustration Only)

Figure 3 shows a cartoon of a particle temperature history, with the five phases identified by roman numerals. These five phases are:

- **Solution Heating (Phase I)** – The temperature of the droplet changes from the injection temperature to the wet bulb temperature. The progress variable is the particle temperature.
- **Drying (Phase II)** – The droplet concentration changes from 67.5% water to 0% water, resulting in a solid urea particle. The progress variable is the particle diameter.
- **Melting (Phase III)** – The solid urea particle melts to form a liquid urea droplet. The progress variable is the molten fraction.
- **Urea Heating (Phase IV)** – The liquid urea is heated from the melting point to the so-called thermolysis temperature. The progress variable is the particle temperature.
- **Thermolysis (Phase V)** – The thermolysis process continues until all of the urea has been decomposed. The progress variable is the particle diameter.

ATOMIZATION OPERATOR

The function of the atomization operator is to initialize the property vector for each parcel. The current lifetime phase is set to Solution Heating (Phase I). Since the progress variable for that phase is particle temperature, this value is set to the injection temperature. The PDF is set to be the inverse of the number of parcels, meaning that the total injected AUS volume is divided equally among the parcels. The “mid-point” cumulative distribution function by volume for the *i*th parcel is then computed from:

$$CDF_i = \frac{2i-1}{2N_p} \quad (1)$$

The initial droplet diameter for the *i*th parcel is then determined by assuming drop size follows a Rosin-Rammler distribution [11]:

$$CDF_i = 1 - \exp \left[- \left(\frac{D_{0i}}{D_{REF}} \right)^\beta \right] \quad (2)$$

PARTICLE-BOURNE DECOMPOSITION OPERATOR

The function of the particle-bourne decomposition operator is to update the property vector as it passes through a control volume. Control volume size is usually selected based on gradients in local conditions. For example, the inlet section (from the injection point to the catalyst inlet face) might be represented with a single control volume. However, the channels within the converter might be divided into as many as 20-25 control volumes to account for changes in composition and temperature.

The particle-bourne decomposition operator is simply reapplied to each control volume in the exhaust system model, starting with the control volume farthest upstream and continuing sequentially to the control volume farthest downstream.

As mentioned above, the parcels do not have physical positions, they only have residence times in Euclidean control volumes. By assuming that the particles move with the gas and are uniformly distributed within each control volume, the residence time can be determined from:

$$\Delta t_{cv} = \frac{\rho_{cv} V_{cv}}{\dot{m}} \quad (3)$$

The technique for updating the property vector varies depending on the current lifetime phase. Updates continue until the total time in all phases equals the residence time in the current control volume. The resulting property vector is then passed to the neighboring downstream control volume.

Algebraic relationships derived from a particle dynamics simulation are used to update the property vector. Details of that simulation and its validation are given in the Appendix. Specifically, the simulation was programmed into MATLAB/SIMULINK and applied over a broad range of operating conditions to develop a database of droplet lifetime histories. To avoid adding states to the urea decomposition model, we simply interpolate in these lifetime histories rather than repeating the dynamic calculations. This is closely related to the “equation-free” approach for multi-scale modeling proposed by Kevrekidis et al [12], where a simulation is used as a computational experiment, thereby allowing the modeler to concentrate on macro-scale system behavior rather than micro-scale effects.

The following subsections describe the update procedure for each lifetime phase.

SOLUTION HEATING (PHASE I) – During Phase I, the particle is approaching the wet bulb temperature, defined as the temperature where the heat transfer rate equals the latent heat demand associated with evaporation. It is a function of the temperature, pressure, and water mole fraction of the gas. In the urea decomposition model, it is estimated using a three-dimensional table in these parameters.

The Phase I duration is defined as the time needed for the particle to come within one deg C of the wet bulb temperature from some beginning temperature. It is then correlated to the initial particle diameter through:

$$\tau_I = C_{\tau_I} \cdot D_0^2 \quad (4)$$

In this equation, the subscript i has been suppressed from the initial particle diameter. The empirical constant C_{τ_I} is determined from a four dimensional table in gas temperature, (beginning) particle temperature, water mole fraction, and gas pressure.

Figure 4 shows a surface plot of C_{τ_I} at gas conditions of 110 kPa and 0% H₂O. The valley in this plot corresponds to injection temperatures within one deg C of the wet bulb temperature. Figure 5 compares the Phase I duration from the complete particle dynamics simulation (labeled as *actual*) and the estimate using Eq (4) and look-up tables (labeled as *interpolated*). This comparison is for 100 random operating points covering

the following range: gas temperature – 200 to 700 deg C; injection temperature – 0 to 80 deg C; air-fuel ratio - 20 to 100; ambient humidity – 0 to 0.070 kg H₂O/kg dry air; gas pressure – 80 to 110 kPa; and initial drop diameter – 1 to 120 microns. Clearly, Eq (4) gives accurate results over a broad range of conditions.

If the residence time is shorter than the Phase I duration, then the progress variable of particle temperature is set to be:

$$T_P^{j+1} = T_P^j + (T_{WB}^j - T_P^j) \frac{\Delta t_{CV}}{\tau_I} \quad (5)$$

Note that this assumes a linear transition from initial to wet bulb temperature, which is conservative.

On the other hand, if the residence time exceeds the Phase I duration, the current lifetime phase is set to drying (Phase II) and the progress variable is set to the initial particle diameter. The residence time in the control volume (Eq (3)) is decreased by the Phase I duration (Eq (4)), and the calculations described in the following subsection are performed.

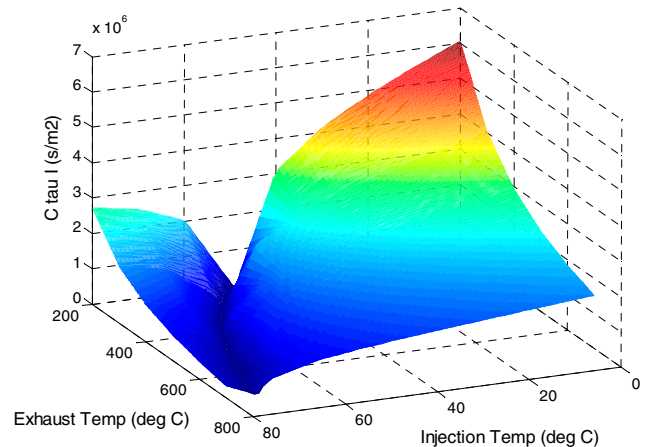


Figure 4: Map of C_{τ_I} versus Injection and Exhaust Temperature (110 kPa Pressure, 0% H₂O)