

Mixture Non-Uniformity in SCR Systems: Modeling and Uniformity Index Requirements for Steady-State and Transient Operation

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04/12/2010Thomas L. McKinley
Cummins, Inc.Andrew G. Alleyne and Chia-Fon Lee
University of Illinois at Urbana-Champaign

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ABSTRACT

Selective catalytic reduction (SCR) of NO_x is coming into worldwide use for automotive diesel emissions control. To meet the most stringent standards, NO_x conversion efficiency must exceed 80% while NH₃ emissions or slip must be kept below 10-30 ppm. At such high levels of performance, non-uniformities in ammonia-to-NO_x ratio (ANR) at the converter inlet can limit the achievable NO_x reduction. Despite its significance, this effect is frequently ignored in 1D catalyst models. The corresponding model error is important to system integration engineers because it affects system sizing, and to control engineers because it affects both steady-state and dynamic SCR converter performance.

A probability distribution function (PDF) based method is introduced to include mixture non-uniformity in a 1D, real-time catalyst model. The model is subsequently applied over a broad range of steady engine operating conditions to identify regimes where non-uniformity has greatest impact on NH₃ slip limited conversion efficiency. Non-uniformity effects during transient emission cycles (US EPA HD-FTP and SET) are also studied for a 15 liter heavy-duty diesel which targets EPA-2010 standards and uses a copper-zeolite formulation. It is shown that the required ANR uniformity index is a function of not only the engine operating conditions and emissions test cycle, but also catalytic coating characteristics.

INTRODUCTION

Diesel engines are the power-plant of choice for trucks, buses, agricultural tractors, and earth moving equipment.

Unfortunately, diesels also release significant amounts of NO and NO₂ into the atmosphere. These NO_x emissions have been linked to health effects such as respiratory tract infections and asthma. In response, aggressive NO_x emissions legislation has been enacted in Asia, Europe, and North America. A comparative assessment of NO_x control technologies by application and emissions standard was recently completed by Johnson [1], who concluded that SCR will play a leading role in diesel emissions control both on-road and off-road.

SCR is a “game-changing” technology in that it decouples engine out NO_x from tailpipe NO_x, providing an opportunity to optimize the engine-aftertreatment system combination. The preferred strategy depends on several factors such as emissions requirements, customer needs, packaging limitations, product cost, and consumable costs (e.g. fuel cost, urea solution cost). For example, increased NO_x conversion efficiency within the SCR converter can be utilized to reduce engine heat rejection, reduce engine cost, and increase fuel economy [2]. Yet another strategy is to increase NO_x conversion efficiency at part load and moderate temperatures to affect the engine out NO_x to PM ratio. This approach promotes passive filter regeneration, and can be used to reduce or eliminate the need for forced regeneration events if an upstream diesel particulate filter is used [3]. Certainly, the ability to execute any chosen strategy is limited by the achievable engine out NO_x and the achievable NO_x conversion efficiency.

It is widely recognized that achievable NO_x conversion efficiency can be sensitive to the spatial distribution of

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ammonia-to-NOx ratio (ANR) at the converter inlet, and so several mixer designs have been proposed to minimize mixture non-uniformity. Their performance can be compared by plotting the mean NOx conversion efficiency and NH3 slip over an ANR range, by measuring the NH3 slip distribution at the converter outlet and computing the corresponding ANR distribution at the inlet, or by modeling the ANR distribution up to the catalyst inlet face and then using a one-dimensional model to predict overall performance [4,5,6]. However, these methods are applicable to steady-state operation and a specific mixer design.

Moreover, it has to be kept in mind that transient test cycles are commonly used to assess emissions compliance. Since the ANR distribution affects converter dynamics [6], some method of modeling mixture non-uniformity in real-time is needed. This paper presents such a technique, using a probability distribution function to represent the inlet ANR distribution. Mixing effects are then explored over a broader range of operating conditions and catalyst characteristics than has been previously reported. Furthermore, the real-time model is used to quantify mixture non-uniformity impacts during EPA heavy-duty emissions test cycles. This includes the impact of mixer performance on both SCR system capability and sensor accuracy. In so doing, new techniques and new understandings relevant to SCR system optimization are introduced.

UNIFORMITY MEASURES AND EXPECTATIONS

For catalytic converters, the most commonly used measure of mixture non-uniformity is the uniformity index (UI). Originally introduced by Weltens et al [7] for velocity distributions, it can be applied to any property z at the converter inlet face. Specifically:

$$UI = 1 - \frac{1}{2A_{CONV}} \int_{A_{CONV}} \left| \frac{z}{\bar{z}} - 1 \right| dA \quad (1)$$

A perfectly uniform distribution has UI equal to one. Considering a bi-modal distribution to be worst case, it can be shown that the lowest possible value for UI is zero.

Karlsson et al [4] showed that a higher uniformity index yielded a better trade-off between steady-state NOx conversion efficiency and NH3 slip. Johansson et al [6] extended Karlsson's work by considering uniformity indices of NH3, NOx, and ANR. It was demonstrated that the uniformity index of ANR had the strongest correlation to conversion efficiency and slip. Therefore, for the remainder of this paper, the variable z is taken to be ANR, and so UI is a measure of spatial non-uniformities in ANR.

ANR uniformity indices have been reported in the literature for several different mixer designs. Perhaps the most comprehensive work to date has been published by Oesterle et al [5], who reported values ranging from 0.68 to 0.79 without a mixer to 0.88 to 0.95 with a mixer. These values were based on NH3 mole fraction measurements at several locations just downstream of the converter outlet face, and computing the corresponding ANR at the converter inlet face. Based on CFD modeling, Karlsson et al [4] reported UI values of 0.76, 0.87, and 0.89 for three different mixer designs. From these publications, it seems that the following uniformity indices can be expected:

- **With No Mixer:** UI ~ 0.75
- **With Good Mixer Design:** 0.85 < UI < 0.90
- **With Excellent Mixer Design:** 0.90 < UI < 0.95

It is most important to exercise models within the range of interest. Therefore, in this paper UI values greater than 0.75 are examined, with particular focus on values ranging from 0.85 to 1.00. This covers a spectrum of good, excellent, and ideal (UI = 1.00) mixer designs.

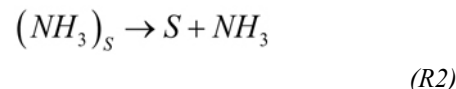
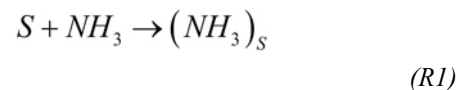
SCR CONVERTER MODEL

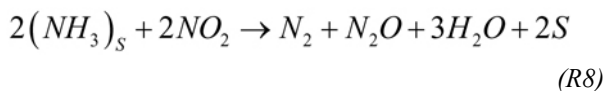
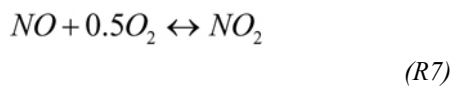
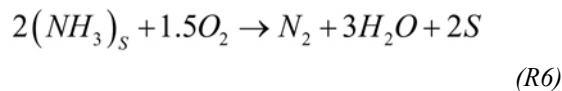
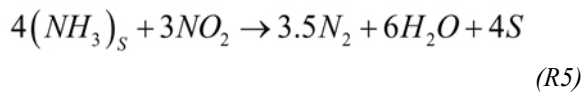
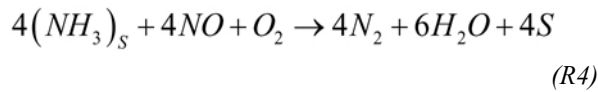
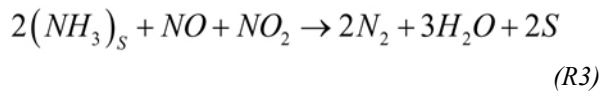
The sensitivity of SCR system performance to mixture non-uniformity is predicted using a one-dimensional model, where the converter is subdivided into a number of axial elements. A total of 24 uniform length elements are used in order to accurately resolve axial distributions in coverage [8]. A detailed description of the model is given elsewhere along with its validation against test data [9]. In brief, each element has two states: (a) catalyst temperature; and (b) NH3 coverage. Due to its short residence time, the gas phase is represented using quasi-steady equations.

Model parameters correspond to a copper-zeolite catalyst developed and quantified in great detail by Chalmers University using their flow reactor [10]. Although a non-commercial coating, it has comparable steady-state performance [11] and NH3 storage capacity [12] to published data for commercial copper-zeolite offerings.

CHEMICAL REACTIONS

The model includes the following chemical reactions:





In these equations, NH_3 refers to gaseous ammonia, $(NH_3)_S$ refers to adsorbed ammonia, and S refers to an unoccupied catalytic site.

COMPARISON TO EXISTING 1D MODELS

Previous mixture non-uniformity modeling also used a one-dimensional converter model [4,6]. However, there are significant differences in system architecture, catalyst formulation, and model spatial resolution from this prior work that are worth mentioning.

First, prior studies assumed that nearly all NOx was in the form of NO and so NO2/NOx ratio effects were neglected. This is a good assumption when the SCR system is placed immediately downstream of the turbo outlet. However, to meet current day heavy-duty EPA standards, the SCR converter is installed downstream of a diesel oxidation catalyst (DOC) and a diesel particulate filter (DPF). These components use platinum-based coatings to promote the oxidation of NO to NO2, which affects low temperature soot oxidation and SCR system performance. For this system arrangement, the model must include NO2/NOx ratio effects (reactions (R3),(R4),(R5)).

Second, previous studies corresponded to a vanadium-based catalyst formulation. In contrast, this work assumes a copper-zeolite formulation. Moreover, the coating is quite active, having good low temperature conversion efficiency but also significant activity for oxidation of adsorbed NH3 to N2 (reaction (R6)).

Lastly, there are differences in model spatial resolution. The current model uses 24 axial elements, while the prior study used only three. However, with appropriate tuning of model parameters, it is still possible to get good model results with three elements [8].

Both one-dimensional models were originally derived for a uniform ANR distribution, but subsequently extended to represent the inlet ANR distribution. This technique is described below.

ANR DISTRIBUTION MODEL

As mentioned above, for a given mixer design and steady-state operating condition, either experimental measurements or CFD predictions can be used to estimate the spatial distribution of ANR over the catalyst inlet face. In this paper, we seek a general way of representing the distribution without requiring detailed modeling or measurement. Moreover, the chosen method should be compatible with real-time simulation, since the intent is to examine mixture non-uniformity effects during transient emissions test cycles.

To achieve the objectives stated above, ANR variation is represented by a probability distribution function (PDF) rather than a spatial distribution. In selecting an appropriate PDF, it must be kept in mind that local ANR values are bounded. First of all, ANR distribution is only of interest when the inlet NOx is non-zero. This in turn means that local ANR values must be non-negative. Moreover, assuming there is at least some mixing of exhaust gas and reductant, the local ANR will be below some finite value. Thus, a PDF with finite support, ranging from zero to some maximum finite value, is desired. This eliminates many commonly used PDFs such as the normal distribution.

The beta distribution is frequently used for bounded random variables. Although the distribution is only defined over the interval from zero to one, it can be related to ANR using the equation:

$$ANR = X \cdot ANR_{MAX} \quad (2)$$

The distribution has two independent parameters a and b . The PDF is defined as:

$$f_X(x) = \frac{1}{B(a,b)} x^{a-1} (1-x)^{b-1} \quad (3)$$

where:

$$B(a, b) = \int_0^1 x^{a-1} (1-x)^{b-1} dx = \frac{\Gamma(a)\Gamma(b)}{\Gamma(a+b)} \quad (4)$$

The beta distribution can represent either symmetric or asymmetric distributions. However, setting $a = b$ gives a symmetric distribution with a mean value of 0.5. Since little is known of the distribution function shape for actual mixers, this assumption is adopted in this paper to reduce the degrees of freedom. With this restriction, setting:

$$ANR_{MAX} = 2 \cdot ANR_{AVE} \quad (5)$$

ensures the PDF is centered on the intended mean ANR.

Figure 1 shows the PDF for different values of parameter a under the restriction that $a = b$. Figure 2 shows the corresponding relationship between the parameter a and the uniformity index. Comparing the two figures, it can be seen that when $a = 1$, the PDF is a uniform probability distribution and $UI = 0.75$. This roughly corresponds to the case where no mixer is used.

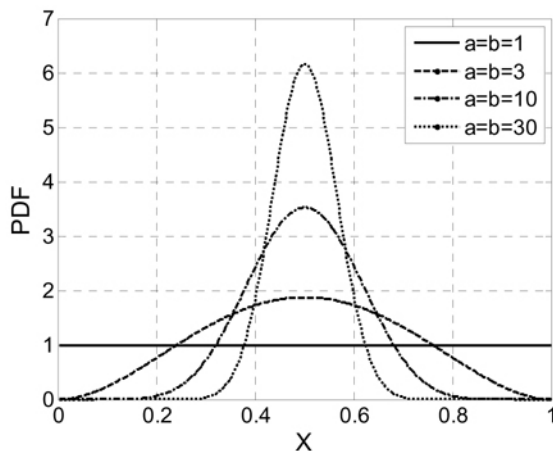


Figure 1. Beta Distribution PDF

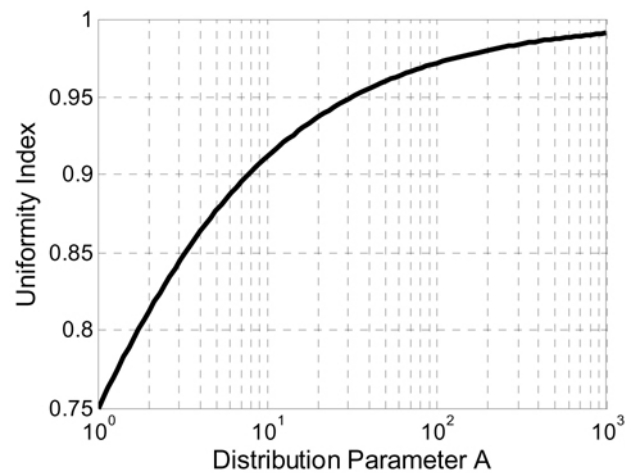


Figure 2. Relationship Between Parameter A and UI

MODEL EXTENSION FOR NON-UNIFORMITY

The end goal is to compute the mean values of NOx conversion efficiency (η_{NOX}) and NH3 slip (y_{NH3}) using the PDF and the 1-D converter model. Since η_{NOX} and y_{NH3} are nonlinear functions of X , it is not possible to accomplish this directly. Instead, the model must be extended to include these nonlinearities.

PDF DISCRETIZATION

The converter simulation was revised to include several copies of the 1D model, all running in parallel (see Figure 3). Each 1D model is given an ANR equal to the mean ANR times a scale factor. The scale factor is selected based on the assumed distribution, a procedure that can be envisioned as representing the continuous PDF with a histogram or probability mass function. Each portion of the histogram is called a *bin*, and the number of bins will be signified by nb .

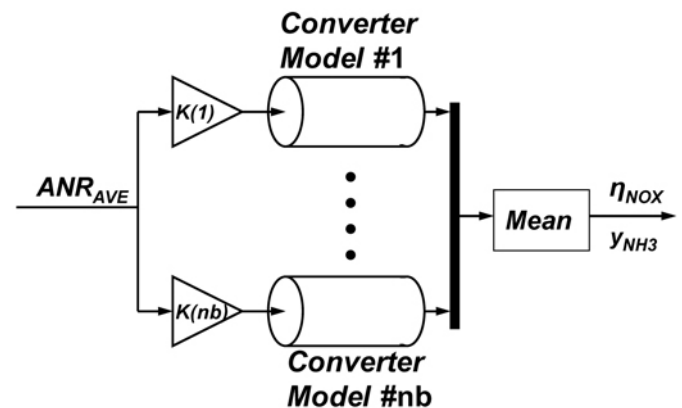


Figure 3. Parallel Model Structure

Bin bounds are chosen so there is equal probability that the random number X belongs to any given bin. Thus, the lower bin bounds are:

$$X_L(1) = 0 \quad (6)$$

$$X_L(m) = X_H(m-1) \quad \forall m \in [2, nb] \quad (7)$$

The upper bin bounds are:

$$X_H(nb) = 1 \quad (8)$$

$$X_H(m) = F_X^{-1}\left(\frac{m}{nb}\right) \quad \forall m \in [1, nb-1] \quad (9)$$

A single value of X is then used to represent each bin. Specifically:

$$\bar{X}(m) = \frac{\int_{X_L(m)}^{X_H(m)} f_X(x) dx}{\int_{X_L(m)}^{X_H(m)} f_X(x) dx} = nb \int_{X_L(m)}^{X_H(m)} f_X(x) dx \quad (10)$$

The ANR for each bin is then set to be:

$$ANR(m) = 2\bar{X}(m) \cdot ANR_{AVE} \quad (11)$$

The number of bins must be chosen carefully. Fewer bins require less computational effort, but give less accurate representation of the distribution. To that end, the following error criteria were applied:

- Error in mean of $X < 1\%$
- Error in coefficient of variation in $X < 5\%$
- Error in UI $< 0.2\%$

It was found that ten bins were sufficient to meet these requirements, with resolution determined by the error in coefficient of variation.

COMPUTATIONAL SPEED

Although ten parallel models increase simulation run time by roughly an order of magnitude, it is still possible to simulate

the SCR converter in real-time. In particular, after optimizing the model described in [9], the 10 bin $\times$ 24 axial element simulation ran two times faster than real-time. These results are for an ordinary laptop computer (Intel Core 2 Duo CPU, 1.80 GHz clock rate with 0.99G RAM), with the simulation coded into MATLAB 2007a.

Further improvements in run-time can be achieved with two more changes. First, the model is running in interpreted mode, and code speed can be increased by compiling it. Second, as stated above it is possible to achieve reasonably good model accuracy with fewer axial elements, and this change alone could increase code speed by a factor of two to ten.

NON-UNIFORMITY IMPACTS ON STEADY-STATE PERFORMANCE

The impact of mixture non-uniformity was predicted at 13 different steady-state operating points for a 5.9 liter diesel engine using a 15.6 liter SCR converter. SCR inlet conditions are reported in [Table 1](#). Model parameters, which were adjusted to represent end-of-useful life conditions, are given in the [Appendix](#). Throughout the rest of this paper, NH3 slip is reported in units of ppm and the NOx conversion efficiency is reported as a decimal. In particular, the NOx efficiency is based on the inlet and outlet mole fractions of NO + NO2.