Chemical Engineering Journal 264 (2015) 936–944

Contents lists available at ScienceDirect

Chemical Engineering Journal

journal homepage: www.elsevier.com/locate/cej

Simulation study of SCR catalysts with individually adjusted ammonia dosing strategies

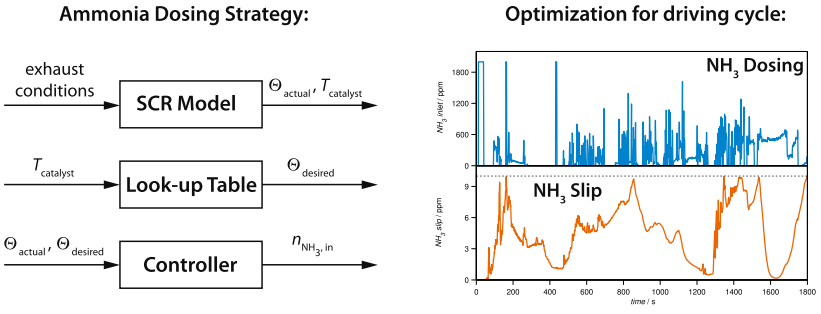
B. Opitz [a,b], M. Bendrich [b,c], A. Drochner [a], H. Vogel [a], R.E. Hayes [c], J.F. Forbes [c], M. Votsmeier [b,*]

[a] Technische Universität Darmstadt, Alarich-Weiss-Str. 8, 64287 Darmstadt, Germany
[b] Umicore AG & Co. KG, Rodenbacher Chaussee 4, 63457 Hanau, Germany
[c] University of Alberta, 116 Street, Edmonton, Alberta T6G 2V4, Canada

HIGHLIGHTS

- A practical, model-based, ammonia dosing strategy is presented.
- The dosing strategy is optimized and applied to SCR demonstration examples.
- Performance comparison of different catalyst materials (iron and copper).
- Investigating the sensitivity of the performance towards change in storage capacity.
- The examples demonstrate the importance of an individually adjusted dosing strategy.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 5 September 2014
Received in revised form 10 November 2014
Accepted 23 November 2014
Available online 2 December 2014

Keywords:
Selective catalytic reduction (SCR)
Ammonia dosing strategy
Simulation
Driving cycle
Optimization
Control

ABSTRACT

In today's vehicle applications, SCR ammonia dosing is completed using complex control algorithms that need to be parameterized for the individual catalyst technology. Owing to the complexity of the parameterization procedures, ammonia dosing is frequently completed using either oversimplified dosing strategies (e.g., constant NH_3/NO_x ratio) or strategies previously optimized for different catalysts during the early design phase's simulation and laboratory studies.

This paper presents a practical, model-based approach that allows for simulation and laboratory studies to be performed with an individually adapted dosing profile. The procedure is based on conventional feedback control strategies, where the average ammonia storage level is controlled using a temperature dependent setpoint (the desired storage level), which is interpolated from a look-up table. The approach of this paper is to optimize numerically the entries of this look-up table for a given test cycle such that the NO_x conversion is maximized while the ammonia slip is kept below a defined level. Although the optimization is performed for a particular cycle, it is demonstrated that the dosing strategies obtained in this way also work reasonably well for other test cycles.

The proposed procedure was applied in a catalyst screening simulation study that compared the performance of an Fe– and a Cu–zeolite catalyst for a given test cycle. It was demonstrated that each of the two catalysts appeared to have a better performance than the other catalyst when the specified catalyst's optimized dosing profile was applied to both catalysts. The best catalyst was only identified when comparing both catalysts using their own respective optimized dosing profile. A second simulation study used the proposed dosing procedure to compare catalysts with different ammonia storage capacities. Again, the relative ranking of the catalysts' performance was shown to depend on the dosing profile. Overall it is demonstrated that the true potential of a catalyst can only be determined when an individually adapted dosing strategy is used.

* Corresponding author. Tel.: +49 6181 59 2311.
E-mail address: martin.votsmeier@eu.umicore.com (M. Votsmeier).

http://dx.doi.org/10.1016/j.cej.2014.11.114
1385-8947/

1. Introduction

Stringent, government imposed, emission standards have resulted in continuous advances by the automobile industry to develop techniques to meet the more demanding driving test cycles. In the case of reducing NO_x emissions in diesel vehicles, selective catalytic reduction (SCR) is a successful method currently used [1,2]. In the SCR process, ammonia, the reducing agent, is generated onboard through the hydrolysis of urea, provided that the inlet gas temperature to the SCR reactor is hot enough to allow for the hydrolysis reaction to occur [3]. The SCR catalysts are able to adsorb or desorb the ammonia, which is beneficial when too much has been dosed or more is needed [4]; however, the storage capacity of ammonia in the catalyst decreases strongly with an increase in temperature [1]. Therefore, a sharp increase in load can result in a significant amount of ammonia slip. This behaviour presents the need for an optimized urea dosing strategy, where NO_x conversion is maximized, while maintaining the ammonia slip below a currently non-regulated acceptable level.

In today's vehicle applications, ammonia dosing is completed using complex control algorithms that need to be parameterized for the individual catalyst technology [5]. Feed-forward control strategies based on the SCR catalyst surface reactions with some compensation for ammonia storage are commonly used for open-loop control [6]. For instance, a Nominal Stoichiometric Ratio (NSR) map with a limiter for ammonia slip peaks has been investigated as an open-loop or feed-forward control strategy [7]. A feed-forward controller accounting for the steady state ammonia usage and storage level compensation using observers, was also examined [5]. Closed-loop feedback strategies with either an ammonia sensor [8] or NO_x emissions feedback have been developed [7]. A different approach uses a neural network model for the SCR catalyst and a multi objective genetic algorithm to optimize an ammonia dosing strategy [9]. In this context, numerical simulation has become an important tool for the development of control strategies [10].

Owing to the complexity of the control calibration, an SCR system nowadays is generally developed using the following two steps: First, the individual catalytic converter components of the exhaust emissions system and the arrangement of the components in the system are optimized. In this stage, laboratory tests and simulation studies are performed using either oversimplified SCR dosing strategies (e.g., constant NH_3/NO_x ratio) or a dosing strategy from a previous exhaust system. In a second step, the SCR dosing strategy is optimized for the system layout established in the first step. The obvious disadvantage of this two-step approach is that the influence of the control strategy on the system design is neglected. If, for example, the relative ranking of two SCR catalysts depends on the dosing strategy, the outcome of a catalyst screening will depend on the chosen dosing strategy.

The purpose of this paper is to report an investigation on the importance of applying an optimized SCR dosing strategy in the early catalytic converter development phase. In this respect, we present a simulation-based optimization method that allows for an automated adaptation of the ammonia dosing strategy for a particular catalyst. The idea is that, for a given test cycle, the entries of a look-up table in the dosing strategy are optimized to maximize NO_x conversion under the constraint of keeping the ammonia slip below a defined limit.

Although in our procedure the entries of the look-up table are optimized for a single driving cycle, it is shown that the obtained dosing strategy also works surprisingly well for other test cycles not considered during the optimization.

The proposed optimization procedure is applied in two simulation studies that investigate the extent of the influence of the ammonia dosing strategy on the performance ranking of different catalysts. In the first example, an iron– and a copper–zeolite SCR catalyst are compared. In the second example, a comparison of catalysts with different ammonia storage capacities is presented. In both cases, it is shown that the relative ranking of the catalysts depends on the dosing strategy, and that the optimal catalyst design can only be found if each catalyst is evaluated using its own individually adjusted dosing profile.

2. Optimization of ammonia dosing control strategy

The dosing strategy of this paper is based on control strategies traditionally used in vehicle SCR applications. At each time instance, the average storage level is controlled to a setpoint obtained from a look-up table that lists the desired storage level as a function temperature. The approach of this paper is to optimize the entries of this table numerically, so that for a particular driving cycle the NO_x conversion is maximized under the constraint that the average and maximum ammonia slip stay below a given limit.

2.1. Ammonia dosing control strategy

The block-diagram of the controlled system is illustrated in Fig. 1. In this set-up, the amount of ammonia added to the SCR catalyst at every time instance is dictated by an optimized look-up table, which relates average catalyst temperatures to optimal target average ammonia surface coverages. The usage of this table and functionality of the proposed dosing strategy is described in more detail via the following algorithm:

(1) Starting at a given time instant (t), ammonia ($n_{NH_3,in}(t)$) is injected into the exhaust gas stream in front of the catalyst. At this time instant, the SCR model is used to calculate the output variables including the average catalyst temperature ($T_{catalyst}(t)$) and actual average ammonia surface coverage ($\Theta_{act.}(t)$).
(2) The look-up table is then used to determine the setpoint, or desired average ammonia surface coverage ($\Theta_{des.}(t)$), for the current catalyst temperature via linear interpolation between table entries. Knowing the actual average ammonia surface coverage ($\Theta_{act.}(t)$), the deviation from the setpoint ($e(t)$) can be determined.
(3) The amount of ammonia to be added to the system at the next time instant can then be calculated as:

$$n_{NH_3,in}(t+\Delta t) = \begin{cases} [\Theta_{opt.}(t) - \Theta_{act.}(t)] \cdot \sigma \cdot V, & \text{if } \Theta_{opt.}(t) > \Theta_{act.}(t) \\ 0, & \text{otherwise} \end{cases} \quad (1)$$

where $\Theta(t)$ represents the average ammonia surface coverage, σ the number of active sites per reactor volume and V the catalyst

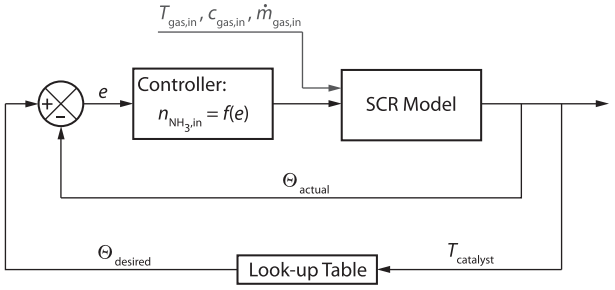


Fig. 1. Block-diagram of the model-based feedback loop using the optimized look-up table.