

Experimental study of SCR in a light-duty diesel exhaust to provide data for validation of a CFD model using the porous medium approach

Tamaldin, N. , Roberts, C.A. and Benjamin, S.F.

Published version deposited in CURVE August 2012

Original citation & hyperlink:

Tamaldin, N. , Roberts, C.A. and Benjamin, S.F. (2010). *Experimental study of SCR in a light-duty diesel exhaust to provide data for validation of a CFD model using the porous medium approach*. SAE Technical Paper 2010-01-1177

<http://dx.doi.org/10.4271/2010-01-1177>

Publisher statement: Copyright © 2010 SAE International. This paper is posted on this site with permission from SAE International. Further use or distribution of this paper is not permitted without permission from SAE.

Copyright © and Moral Rights are retained by the author(s) and/ or other copyright owners. A copy can be downloaded for personal non-commercial research or study, without prior permission or charge. This item cannot be reproduced or quoted extensively from without first obtaining permission in writing from the copyright holder(s). The content must not be changed in any way or sold commercially in any format or medium without the formal permission of the copyright holders.

CURVE is the Institutional Repository for Coventry University

<http://curve.coventry.ac.uk/open>

Experimental Study of SCR in a Light-Duty Diesel Exhaust to Provide Data for Validation of a CFD Model Using the Porous Medium Approach

2010-01-1177

Published
04/12/2010

N. Tamaldin
Univ. Teknikal Malaysia Melaka

C. A. Roberts and S. F. Benjamin
Coventry Univ.

Copyright © 2010 SAE International

ABSTRACT

Removal of NO_x from a light-duty diesel automotive exhaust system can be achieved by SCR reactions using aqueous urea spray as the reductant. Measurements of emissions from such a system are necessary to provide data for CFD model validation. A test exhaust system was designed that featured an expansion can, nozzle and diffuser arrangement to give a controlled flow profile to define an inlet boundary for a CFD model and to approximate to one dimensional flow. Experiments were carried out on the test exhaust using injection of either ammonia gas in nitrogen or aqueous urea spray. Measurements were made of NO, NO₂ and NH₃ at inlet to and exit from the SCR using a CLD analyser. The NO and NO₂ profiles within the bricks were found by measuring at the exit from different length bricks. The spray and gas measurements were compared, and insights into the behaviour of the droplets upstream and within the bricks were obtained. Approximately half to three quarters of the droplets from the spray remained as droplets at entry to the first brick. Approximately 200 ppm of ammonia was released from the droplets to react in the first SCR brick. Between 10 and 100 ppm of potential ammonia passed through the first brick as droplets under conditions ranging from NO_x-matched spray input to excess spray. The CFD model for the gas cases gave reasonable predictions for long bricks. For short bricks, the space velocity was high and there was breakthrough of all species. Nevertheless, the reaction kinetics used, based on a scheme published in the open literature, were shown to have some ability to describe the species profiles within the bricks.

INTRODUCTION

The urea SCR (selective catalytic reduction) system shows effective performance in reducing NO_x with low impact on fuel consumption [1]. Most SCR systems require the injection of aqueous urea, which can be rapidly thermally hydrolysed to produce ammonia in the exhaust stream [2]. The ammonia produced reacts selectively with NO_x to reduce it on the SCR catalyst, typically a base metal zeolite. Such catalysts operate most efficiently above 473 K, and have the ability to store ammonia [3]. At low temperatures the NO_x reduction is substantially accelerated in presence of NO₂, which is called fast SCR [4]. For optimal performance, the level of ammonia injected into the exhaust must match the NO_x emissions over a specific time interval. Under-injection of NH₃ leads to lower NO_x conversion rate and the inability to meet the regulated emission standards. Over-injection gives NH₃ slippage, which is undesirable. In order to develop a validated CFD model to describe the processes in a light-duty diesel exhaust SCR system, data on emissions are required. This paper describes experimental measurements made in a specially designed exhaust system on an engine test bed to obtain such data and attempts to predict the measured data using a CFD model. The CFD model is based on the porous medium approach [5], which has been demonstrated for other catalyst systems. The methodology of the experiments is simple and requires species measurements upstream and downstream of the SCR catalyst as a function of ammonia input level, where the ammonia is introduced either as ammonia gas in nitrogen or as aqueous urea spray.

SCR KINETICS

The reaction kinetics used in the CFD model were based on the scheme described by Olsson et al., [6]. The scheme includes ammonia adsorption and desorption, ammonia oxidation and NO oxidation. Standard, fast and slow SCR reactions were all included and additionally N₂O formation. Formation of nitrate, however, was not included. The rate constants provided by [6] were used without further tuning.

The scheme [6] assumes that species in the gas phase at the active surface react with ammonia adsorbed on to a single type of site. The number of available sites in the catalyst must be specified and Olsson et al. quote 200 mol-sites/m³ monolith for their catalyst.

Using θ as fraction of total sites, the surface coverage relationship for ammonia stated by Olsson is

$$\frac{\partial \theta}{\partial t} = \sum [S_j R_j] \quad (1)$$

S_j is the stoichiometric coefficient of the surface species in reaction j and R_j is the reaction rate (mol/mol-sites/s).

This is interpreted within Star-CD, which is the software used to develop the model, as

$$\epsilon \rho_{gas} \frac{\partial \theta}{\partial t} = \frac{\epsilon \rho_{gas}}{\Omega} (R_{adsorption} - R_{desorption} - R_{consumption}) \quad (2)$$

where all rates, R , are in mol NH₃/m³ monolith /s and Ω is mol-sites/m³ monolith. Density of the exhaust stream is ρ_{gas} and ϵ is the void fraction of the catalyst monolith.

EXPERIMENTAL TEST RIG

The light-duty diesel engine on the test bed was a 1998 cc 4-cylinder engine equipped with common rail injection system. A single engine condition was investigated, 1500 rpm and 6 bar brake mean effective pressure (BMEP), and the tests were run to steady state. An ultra low sulphur diesel fuel, Swedish Class Diesel Carcal 55 was used throughout.

A diagram of the test exhaust is shown in Fig. 1. When the DPF (Diesel Particulate Filter) was placed in the more usual location downstream of the DOC (Diesel Oxidation Catalyst), some of the NO₂ produced was reduced to NO by soot on the filter. Hence, the DOC was placed downstream of the DPF as in Fig. 1. This established an NO/NO₂ ratio in the exhaust stream of about 40/60. An excess of NO₂ can be advantageous [7,8,9,10]. Urea spray was injected into the expansion box and nozzle arrangement to promote homogeneous mixing of the urea droplets for ammonia conversion. Alternatively ammonia gas, either 4% or 5% in

N₂, was introduced after the DOC, and was also well mixed by its passage through the expansion box and nozzle. The nozzle provided a uniform flow profile to the long diffuser sited upstream of the SCR bricks. This in turn provided approximately uniform flow to the SCR bricks. The inlet boundary of the CFD model coincided with the nozzle exit and the long 10° diffuser avoided the need to model flow maldistribution that occurs with short diffusers [11].

The SCR bricks were 91 mm in length and four were available, so that the optional configurations 1SCR, 2SCRs, 3SCRs and 4SCRs could be studied. Fig. 1 illustrates a case with 3 SCR bricks installed, two in the first can and one in the second. Gas analysers sampled from the instrumentation modules sited both upstream and downstream of the SCR.

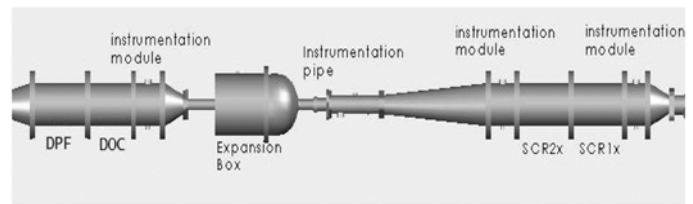


Fig.1. Test exhaust system: urea spray was introduced into the expansion box; NH₃ gas in nitrogen was introduced into the first instrumentation module.

EMISSION MEASUREMENTS

A Horiba EXSA analyser was used to monitor NO_x levels from the engine. Measurements were made of NO, NO_x and NH₃ upstream and downstream of the SCR catalyst using a Horiba MEXA 1170NX CLD-based analyser. This analyser had two operation modes: NO/NO₂ mode and NO_x/NH₃ mode. In NO/NO₂ mode, both NO and NO₂ could be measured while in NO_x/NH₃ mode, total NO_x and NH₃ readings were obtained. Although designed for such measurements, this analyser was an early model without ammonia scrubbers fitted and careful interpretation of the results was necessary. The operating principle of this analyser is illustrated in Figure 2.

When NO₂ is present the MEXA can measure NO_x and NO₂ correctly only in the absence of ammonia. This is because NO₂ can react with ammonia on the NO_x converter within the analyser [12]. This unintended reaction causes an apparent loss of NO_x as the reacted NO₂ is not accounted for. In NH₃ mode, the SUM (NO + NO₂ + NH₃) read from the analyser is correct, but NO_x is too low and hence NH₃ by internal subtraction within the analyser is too high. In NO mode, the SUM (NO + NO₂) is too low, but NO is correct, so that NO₂ by internal subtraction is too low.

In order to interpret the analyser readings the following procedure was used. Upstream of the SCR, NO and NO₂ measurements were made in the absence of ammonia and it was assumed that gas phase reactions prior to the SCR were negligible, so that these readings were valid also in the presence of ammonia. The SUM value read from the analyser in the presence of ammonia was valid, so the ammonia level could be found by manual subtraction.

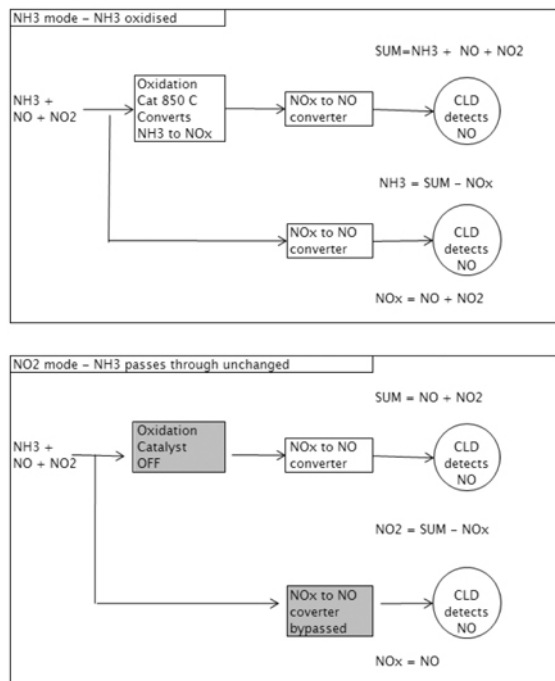


Fig.2. Schematic illustration of operating principle of gas analyser.

Downstream of the SCR brick in the presence of ammonia slip, only NO can be measured reliably. The SUM upstream minus the SUM downstream, however, gives a measure of (NH₃ + NO_x) consumed by the SCR bricks. Furthermore, an assumption can be made that NO_x and ammonia are mainly consumed on a mol/mol basis during SCR reactions. This assumption neglects ammonia oxidation and the slow SCR reaction, but is valid as a first approximation for the temperature in the tests reported here. Hence half of (NH₃ + NO_x) consumed is either ammonia or NO_x consumed. NO consumed is available directly from the difference between upstream and downstream measurements. NO₂ consumed is finally found from the difference between NO_x consumed and NO consumed.

The two items of directly measured data downstream that can be compared with CFD predictions are NO and the slip of (NH₃ + NO₂), which is found from measurements of SUM (NH₃ + NO₂ + NO) minus NO.

When injecting 4% or 5% ammonia gas in N₂, the input level can be determined from a calibrated flowmeter and the known exhaust mass flow rate. This value can be used to check upstream measurements. When injecting urea spray, the potential ammonia injected can be determined from the spray mass flow rate. Comparison of this with the measured ammonia level upstream of the SCR will indicate the mass of spray that has released its ammonia between the spray point and the emissions measurement location.

The calculated magnitude of the Potential SUM upstream (potential NH₃ + NO + NO₂) minus measured SUM downstream should indicate total consumption of (NH₃ + NO + NO₂). This implicitly assumes that no droplets pass through the SCR bricks. By comparison of the 1SCR case with urea injection with NH₃ gas injection it is possible to deduce what happens to the droplets within the SCR brick.

Tests were carried out using either 1, 2, 3 or 4 SCR bricks with ammonia gas in nitrogen; but only 1 SCR and 4 SCR configurations were investigated when using urea spray. The measurements were made as a function of input ammonia level.

RESULTS: 1 SCR brick, 4 SCR Bricks, ammonia GAS AND UREA SPRAY

Fig. 3 shows a summary of the experimental emissions measurements. The 5% ammonia gas in nitrogen experiments were carried out at low ammonia input levels that avoided excessive ammonia slip. The spray experiments were carried out at high potential ammonia input levels because the spray unit used was designed for heavy duty use but was used at the lower end of its range for light duty. The 4% ammonia gas in nitrogen experiments on one SCR brick spanned the entire range of input ammonia levels, from levels insufficient for completion of the reactions to excess.

In Fig. 3 the 5% ammonia gas results (o symbol) meet the spray results (x symbol) between 500 and 600 ppm NH₃ input levels for four SCR bricks. The 4% ammonia gas results (o symbol) agree with both the spray and the 5% ammonia gas results for NO for one SCR brick. The NO₂ and NH₃ levels after one SCR brick with spray are too high and do not agree with the 4% ammonia gas results. This is because urea droplets are able to survive passage through one brick and this is not accounted for in the methodology described above. This difference can be exploited to deduce how much NH₃ exits from one brick in droplet form, and this is discussed further below.

Fig. 4 shows the potential amount of ammonia injected into the system by the spray and the amounts released from the droplets to be detected as ammonia gas at the measuring point

upstream of the SCR catalysts. The amounts retained as droplets at this point are also indicated.

$$\text{NH}_3[\text{potential, spray input}] - \text{NH}_3[\text{measured upstream of SCR}] = \text{NH}_3[\text{entering SCR as droplets}]$$

The two experiments with 1 and 4 SCR bricks gave different results, attributable to experimental variation. A larger amount of potential ammonia is retained as droplets as the urea spray rate increases.

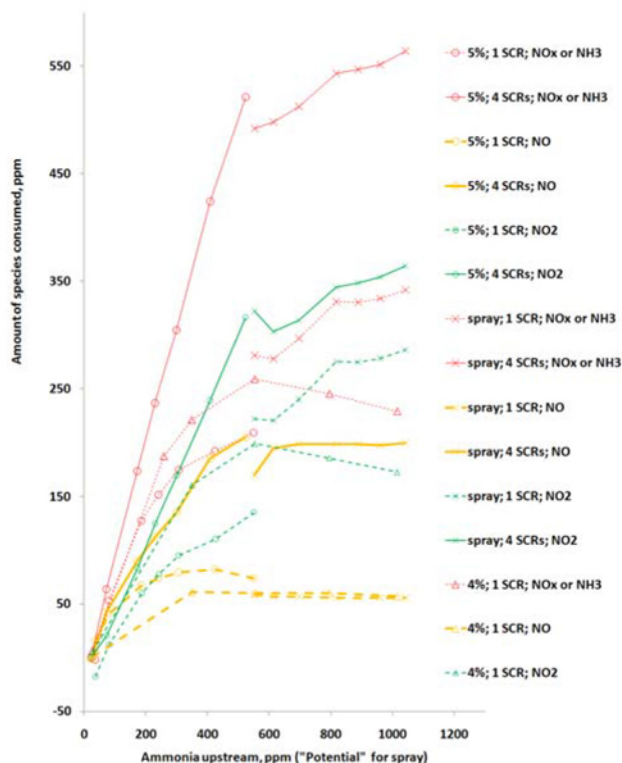


Fig.3. Summary of measurements with 1 and 4 SCR bricks. Dashed lines 1 SCR; continuous lines 4 SCR. Urea spray (x); 5% ammonia gas in nitrogen (o); 4% ammonia gas in nitrogen (Δ). Feint red NO_x or NH₃; medium green NO₂; bold yellow NO.

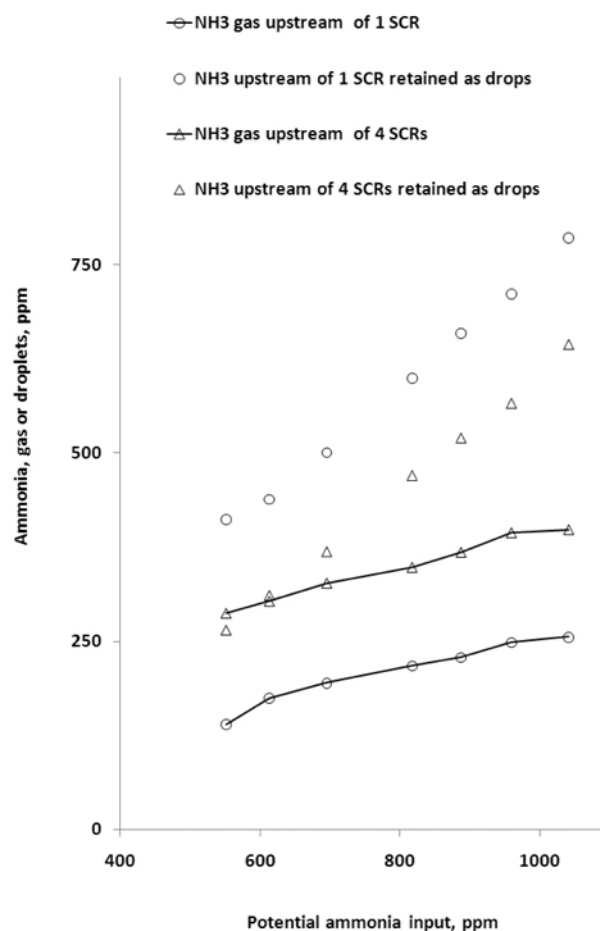


Fig.4. Ammonia released from spray upstream of SCR bricks.

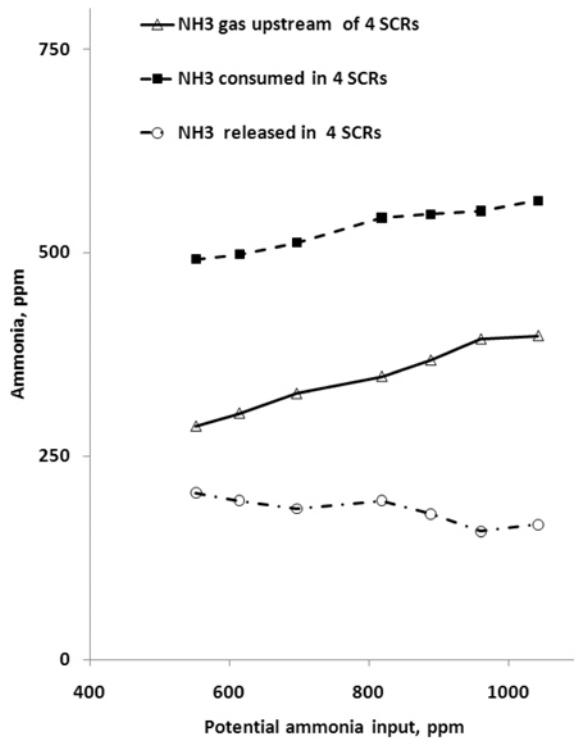


Fig. 5. Ammonia released from spray within 4 SCR bricks, deduced from NH_3 measured upstream and NH_3 consumed.

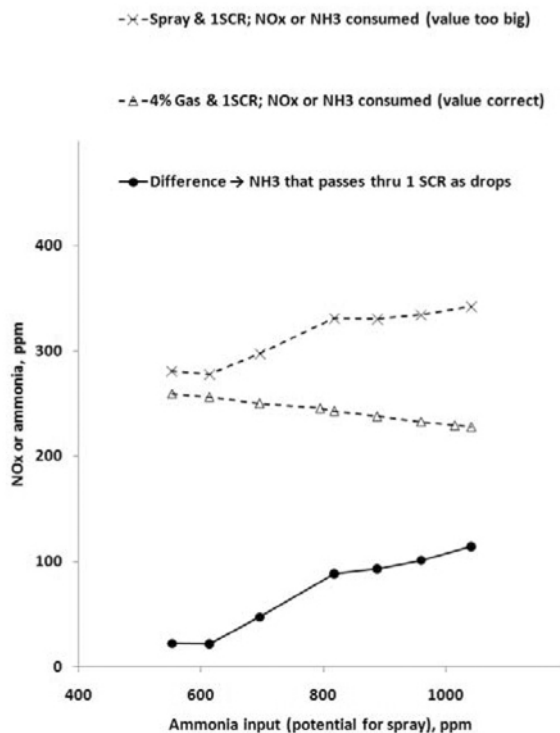


Fig. 6. Ammonia passing through 1 SCR brick in droplet form.

Fig. 5 shows that in the case of 4 SCR bricks, a further amount, 200 ppm or less, of potential ammonia is released from droplets within the bricks to be consumed by the SCR reactions. This is deduced from the difference between the measured NH_3 gas entering the bricks and the amount of NH_3 consumed within the bricks. The amount released within the bricks is lower at higher urea spray flow rates, probably because the excess urea spray lowers the brick temperature.

$$\text{NH}_3[\text{consumed in 4SCRs}] - \text{NH}_3[\text{measured upstream of 4SCRs}] = \text{NH}_3[\text{released from droplets within 4SCRs}]$$

Fig. 6 uses the discrepancy between the spray case and the 4% NH_3 gas case seen in Fig. 3 to deduce the amount of ammonia that passes through one brick in droplet form. The deduced value of ammonia consumed in the spray case is too large because the total species consumption is found from the difference between the potential total species upstream minus the measured total species value downstream, with the latter neglecting the droplets that can still be present after just one SCR brick. Hence,

$$\text{NH}_3[\text{consumed, spray}] - \text{NH}_3[\text{consumed, gas}] = \text{NH}_3[\text{emitted from 1SCR as droplets}]$$

This amount of ammonia leaving the SCR in the form of droplets increases as the spray rate increases. When 1000 ppm of potential NH_3 is injected, about 10% of this survives passage through one SCR brick to exit in a form other than NH_3 gas, most probably still in droplet form.

CFD MODELLING

The CFD model applied the Olsson et al. kinetic scheme [6] discussed above. The model was based on the porous medium approach [5, 13]. Heat transfer was prescribed between separate blocks of computational cells that represented the fluid and solid properties of the SCR catalyst. This was managed for the porous medium by enthalpy source terms coded into a user subroutine in Star-CD, version 3.26. The heat of reaction was also accounted for in the user subroutine. Mass transfer of species i between the bulk fluid and the solid phase was imposed in the simulation by scalar source terms in another user subroutine. The solid phase was at the channel wall or in the washcoat pore. These source terms are seen on the RHS of equations (3) and (4) below.

$$\varepsilon \frac{\partial(\rho_{\text{air}} C_{\text{ig}})}{\partial t} + \frac{\partial(\rho_{\text{air}} U_s C_{\text{ig}})}{\partial z} = -K_{\text{mi}} \rho_{\text{air}} A_v (C_{\text{ig}} - C_{\text{isol}}) \quad (3)$$

$$\varepsilon \frac{\partial(\rho_{\text{air}} C_{\text{isol}})}{\partial t} = \frac{\varepsilon}{V_w} [K_{\text{mi}} \rho_{\text{air}} A_v (C_{\text{ig}} - C_{\text{isol}}) + M_i R_i] \quad (4)$$

C_{ig} and C_{isol} are the species concentrations in gas and solid phase respectively, both modelled in the same computational cell. K_{mi} is the mass transfer coefficient, and A_V/ϵ is the wetted surface area per unit volume of air in the catalyst channel whereas A_V/V_W is the wetted area per unit volume of air in the washcoat pore. $M_i R_i$ is the species reaction rate per unit volume of monolith. U_S is the superficial velocity in the porous medium, that is $\epsilon U_{channel}$. Further CFD modelling details are given in [5] and [13].

The CFD simulations are compared with the data in Figs. 7 and 8 for the injection of 4% and 5% ammonia gas in nitrogen.

A direct comparison of measured NO levels at exit from one SCR brick with CFD predictions is made in Fig. 7. Measured values of $(NH_3 + NO_2)$ are also compared. Fig. 8 shows the corresponding plots of results for four SCR bricks. For both brick sizes, agreement is seen to be fairly good at low ammonia input, but less good when excess ammonia was injected.

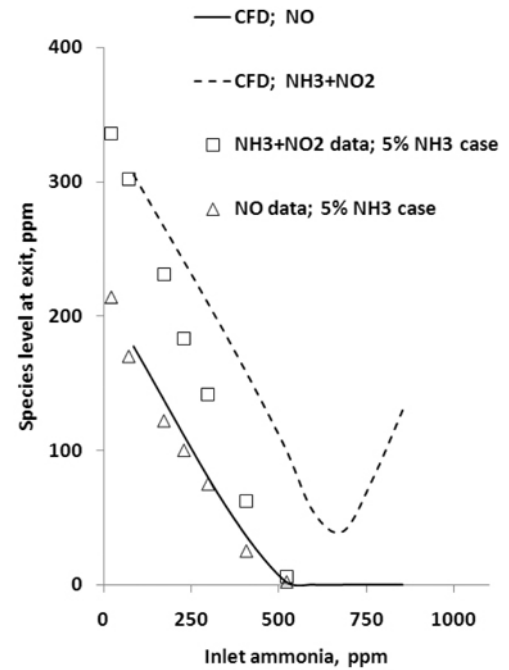


Fig.8. CFD and data comparison for species levels at exit from four SCR bricks.

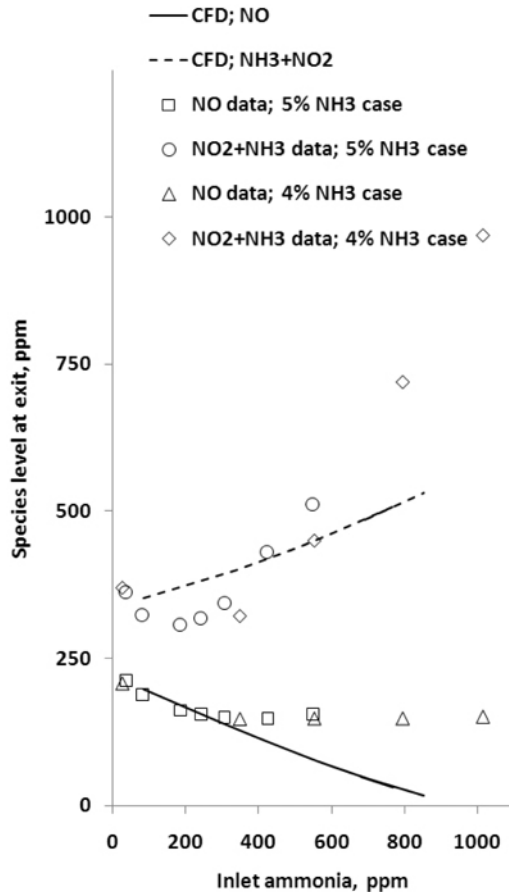


Fig.7. CFD and data comparison for species levels at exit from one SCR brick.

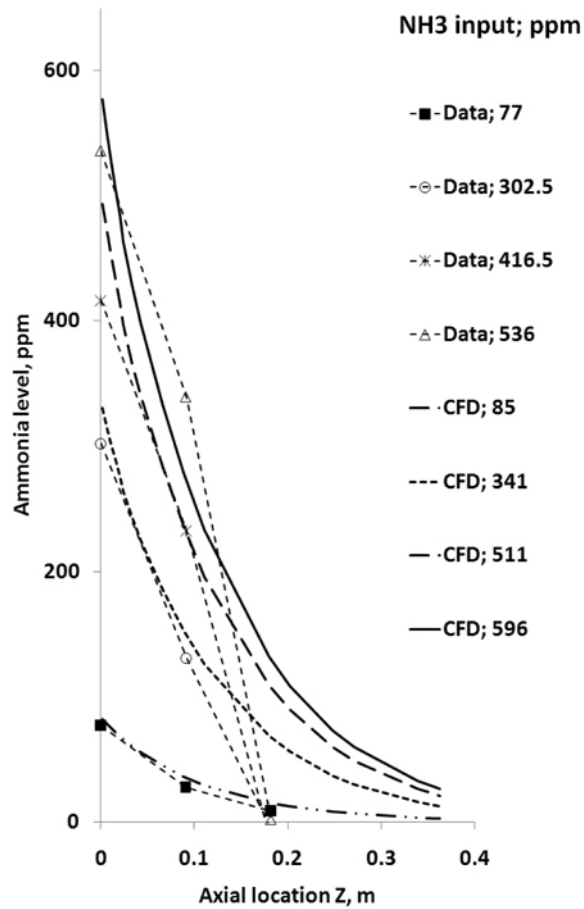


Fig.9. Simulations of NH_3 level in SCR compared with data for different ammonia gas input levels.

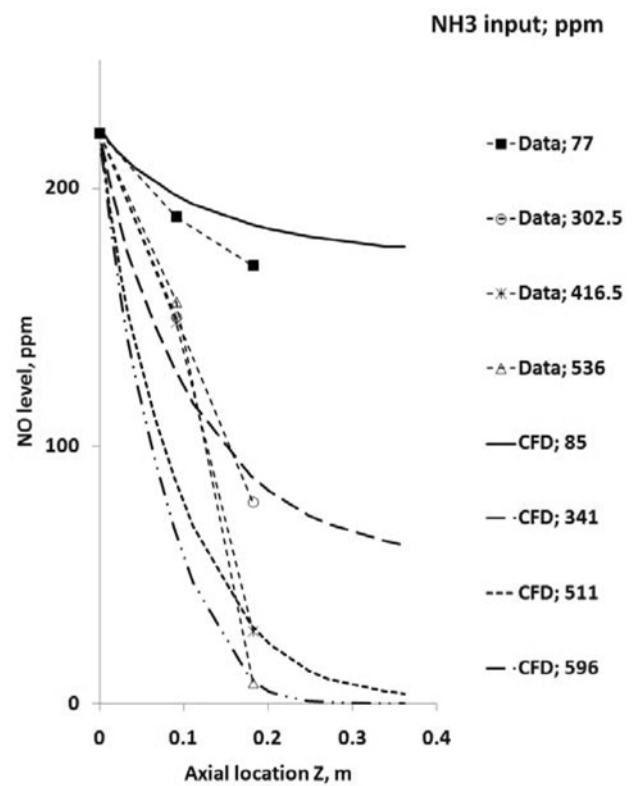


Fig.10. Simulations of NO level in SCR compared with data for different ammonia gas input levels.

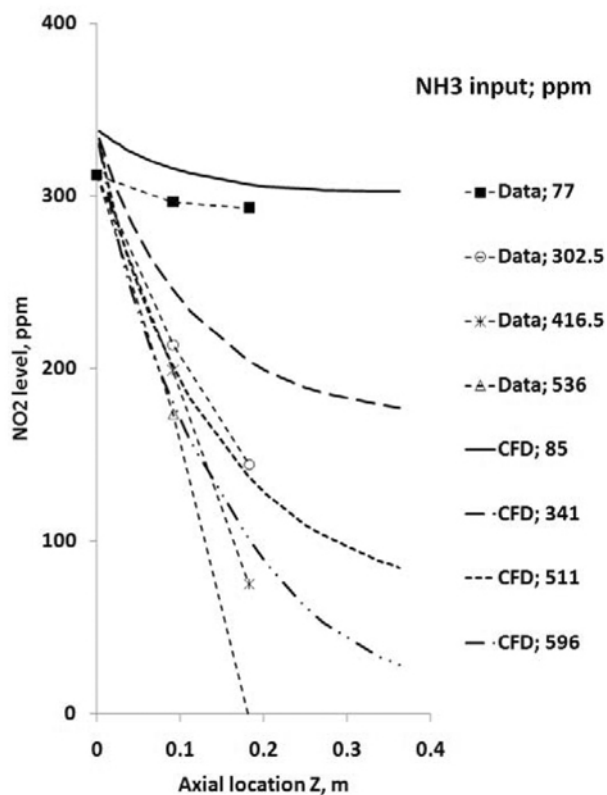


Fig.11. Simulations of NO₂ level in SCR compared with data for different ammonia gas input levels.

Measurements showed that reactions were complete after two SCR bricks for the cases investigated. A comparison of CFD simulations with measurements is shown in Figs. 9, 10 and 11 where NH₃, NO, and NO₂ levels are plotted against axial distance along the bricks for different levels of NH₃ gas injected. Agreement overall is seen to be fairly good, although NH₃ slip after two bricks is predicted in Fig. 9 that was not observed in experiments. In Fig. 10 agreement for NO is good after two SCR bricks but not good after one brick. NO₂ agreement is better after one SCR brick than two bricks in Fig. 11. The kinetic scheme was applied as presented by Olsson et al. [6] and it is not known how similar the catalysts were, although a change was made to the total ammonia storage capacity to a value appropriate for the catalysts used in the experiments here. Thus overall agreement is remarkably good. The NO profiles in Fig. 10 illustrate how fortuitous agreement at the catalyst exit does not necessarily prove full agreement with the CFD model.

SUMMARY/CONCLUSIONS

This investigation has compared SCR system performance using injected urea spray with performance when ammonia gas in nitrogen was injected. In these studies the NO₂/NO ratio was 60/40 approximately and all reactions with

ammonia were complete after 182 mm length of catalyst. The major findings in this study are:

Care is required in interpretation of readings from a CLD analyser intended to measure NO and NO₂ in the presence of ammonia. The methodology described in this paper, however, enables amounts consumed to be found. Knowing input amounts from independent measurements, values of NO, NO₂ and NH₃ both upstream and downstream of the SCR bricks can be deduced.

When ammonia was input as urea spray, between half and three quarters of the droplets remained as droplets at entry to the SCR brick.

About 200 ppm of NH₃ were released from the droplets of urea spray to react with NO_x within the SCR bricks.

It was estimated that between 10 and 100 ppm of potential ammonia survived passage through one 91 mm length SCR brick in droplet form in the tests reported here.

CFD simulations using the porous medium approach and based on a kinetic scheme published in the open literature have been shown to have some ability to describe the steady state tests performed here with injection of ammonia gas in nitrogen.

The model has been used to predict species profiles along the length of the SCR bricks and moderate agreement with data has been obtained.

REFERENCES

1. Lambert, C., Hammerle, R., McGill, R., Khair, M. et al., "Technical Advantages of Urea SCR for Light Duty and Heavy Duty Diesel Vehicle Applications," SAE Technical Paper 2004-01-1292, 2004.
2. Thompson, J., Op De Beeck, J., Joubert, E., and Wilhelm, T. "Case Studies of Urea SCR Integration on Passenger Cars; Monitoring of Urea Inside the Tank During Hot and Cold Environment Test Missions," SAE Technical Paper 2008-01-1181, 2008.
3. Tennison, P., Lambert, C., and Levin, M. "NO_x Control Development with Urea SCR on a Diesel Passenger Car," SAE Technical Paper 2004-01-1291, 2004.
4. Koebel, M., Elsener, M., Madia, G. "Reaction pathways in the selective catalytic reduction process with NO and NO₂ at low temperature," Ind. Eng. Chem. Res. 40: 52-59, 2001
5. Benjamin, S.F., Roberts, C.A. "Three-dimensional modelling of NO_x and particulate traps using CFD: A porous medium approach," Appl. Math. Modelling 31: 2446-2460, 2007

6. Olsson, L., Sjoval, H., Blint, R.J. "A kinetic model for ammonia selective catalytic reduction over Cu-ZSM-5," *Applied Catalysis B: Environmental* 81: 203-217, 2008
7. Takada, K., Kusaka, J., Daisho, Y. "Empirical and numerical study of the improvements in NO_x reduction by a urea-SCR system attainable by controlling the relative proportions of NO and NO₂," *Review of Automotive Engineering*, JSAE Technical paper 4-28-1-41, 2007
8. Narayanaswamy, K., and He, Y. "Modelling of Copper-Zeolite and Iron-Zeolite Selective Catalytic Reduction (SCR) Catalysts at Steady State and Transient Conditions," SAE Technical Paper [2008-01-0615](#), 2008.
9. Schmeig, S.J. and Lee, J.-H., "Evaluation of Supplier Catalyst Formulations for the Selective Catalytic Reduction of NO_x with Ammonia," SAE Technical Paper [2005-01-3881](#), 2005.
10. Devadas, M., Krocher, O., Elsener, M., et al. "Influence of NO₂ on the SCR of NO with ammonia over Fe-ZSM5," *Applied Catalysis B:Environmental* 67: 187-196, 2006
11. Benjamin, S.F., Clarkson, R.J., Haimad, N., and Girgis, N.S. "An Experimental and Predictive Study of Flow in Axisymmetric Automotive Exhaust Catalyst Systems," SAE Technical Paper [961208](#), *SAE Trans. J. Fuels & Lubricants* 105: 1008-1019, 1996
12. Shah, S.D., Mauti, A., Richert, J.F.O., Loos, M.J. et al., "Measuring NO_x in the Presence of Ammonia," SAE Technical Paper [2007-01-0331](#), 2007.
13. Benjamin, S. F., Roberts, C. A. "The porous medium approach applied to CFD modelling of SCR in an automotive exhaust with injection of urea droplets." IMechE Conference Internal Combustion Engines: Performance, Fuel Economy and Emissions, London, 2007

and financial support. Current funding by EPSRC, UK is also gratefully acknowledged. Financial support for N. Tamaldin was received from UTeM, Malaysia.

CONTACT INFORMATION

S. F. Benjamin
s.benjamin@coventry.ac.uk

ACKNOWLEDGMENTS

The authors would like to thank Emcon Technologies, Jaguar-LandRover and Johnson Matthey, for their technical

The Engineering Meetings Board has approved this paper for publication. It has successfully completed SAE's peer review process under the supervision of the session organizer. This process requires a minimum of three (3) reviews by industry experts.

All rights reserved. No part of this publication may be reproduced, stored in a retrieval system, or transmitted, in any form or by any means, electronic, mechanical, photocopying, recording, or otherwise, without the prior written permission of SAE.

ISSN 0148-7191

doi:[10.4271/2010-01-1177](https://doi.org/10.4271/2010-01-1177)

Positions and opinions advanced in this paper are those of the author(s) and not necessarily those of SAE. The author is solely responsible for the content of the paper.

SAE Customer Service:

Tel: 877-606-7323 (inside USA and Canada)

Tel: 724-776-4970 (outside USA)

Fax: 724-776-0790

Email: CustomerService@sae.org

SAE Web Address: <http://www.sae.org>

Printed in USA