VI CONGRESSO NACIONAL DE ENGENHARIA MECÂNICA
VI NATIONAL CONGRESS OF MECHANICAL ENGINEERING
18 a 21 de agosto de 2010 – Campina Grande – Paraíba - Brasil
August 18 – 21, 2010 – Campina Grande – Paraíba – Brazil

MODELLING OF NOX EMISSION BY NEURAL NETWORKS FROM VEHICLES EQUIPPED WITH ADVANCED AFTERTREATMENT SYSTEMS

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Resumo: *NOx emission standards are becoming stringiest over the world especially for heavy-duty vehi-cles. To comply with current and future regulations the majority of vehicle manufacturers are adopting exhaust aftertreatment systems known as Selective Catalytic Reduction (SCR) by using a catalyst based on Vanadium (Va) and a reductant agent based on ammonia. However Va is listed on the California Proposition 65 List as potentially causing cancer and alternatives are being studied. This paper presents a model based on neural networks that integrated with a road vehicle simulator allows to estimate NOx emission factors for different powertrain configura-tions, along different driving conditions, and covering commercial, zeolite and mordenite alter-natives as the base monolith for SCR. The research included the experimental study of Copper based and iron based zeolites (ZSM5 and Cuban natural mordenite). The response of NOx con-version efficiency was monitored in a laboratory for varying space velocity, oxygen, sulphur, water, NOx and* SO_2 *emulating the conditions of Diesel engine exhaust along a trip. The experi-mental data was used in training neural networks and obtaining a mathematical correlation be-tween the outputs (conversion efficiency,* N_2O *formation and ammonia slip) and inputs (space velocity, oxygen, sulphur, water, NOx and* SO_2*). The developed correlation was integrated with ADVISOR road vehicle simulator to obtain NOx emission factor and to test each SCR system installed on light-duty and heavy-duty vehicles for standardized driving cycles and real meas-ured driving cycles.*

Palavras-chave: NOx emission, aftertreatment systems, vehicles , SCR

1. INTRODUÇÃO

Current standards concerning NOx emissions of road vehicles imply a reduction of up to 90% of these emissions at the vehicle tailpipe. Japanese heavy-duty emission standard is currently 0.7 g/kWh instead of previous NOx limit of 2 g/kWh. United States current NOx limit is 0.3 g/kWh instead of previous limit of 2.7 g/kWh and, finally, Europe current NOx limit is 2 g/kWh instead of previous 3.5 g/kWh (dieselnet, 2009). To attain such emission limits, engine techniques covering exhaust gas recirculation are not sufficient and is necessary the addition of a catalyst at the tailpipe. The commercialized SCR (Selective Catalytic Reduction) catalysts used in heavy-duty vehicles are based on V_2O -WO_3-TiO_2. However Vanadium is listed on the California Proposition 65 List as potentially causing cancer and alternatives are being studied especially zeolite-based technologies. Zeolite-based technologies are generally more thermally stable - which is important to consider, if the car/truck is equipped with a Diesel Particulate Filter (DPF) which is thermally regenerated. During thermal DPF regeneration high exhaust gas temperatures occur, which would make Va "disappear" - and thus damage the SCR catalyst (apart from potential health risks associated with Va-derived / Va-containing species). Also SCR requires the use of low sulfur content fuel. Low sulphur fuel is gradually increasing within the European Union (EU) limited to a maximum of 10 ppm in 2009.

The share of Diesel vehicles in the light-duty transport sector is also increasing, especially in Europe because of low CO_2 emission per power and because in some markets it is cheaper than gasoline. So it is expected that the future light-duty vehicles also be equipped with SCR technologies (Figure (1)) due to stringiest emission limits.

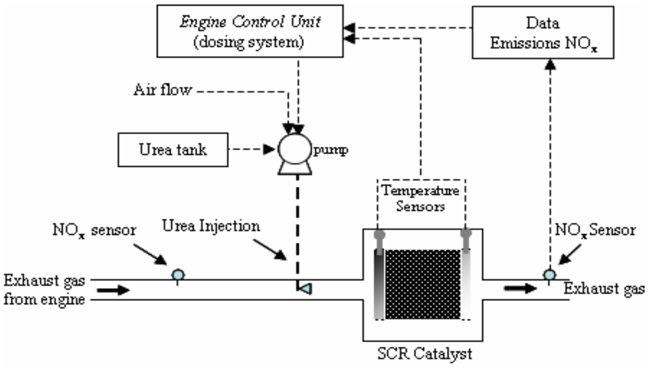

Figure.1 SCR-NO$_x$ system for diesel vehicles

The chemical reactions of SCR for NOx conversion that takes place in the presence of oxygen are as follows:

$$4NO + 4NH_3 + O_2 \rightarrow 4N_2 + 6H_2O \qquad (1)$$

$$2NO + 2NO_2 + 4NH_3 \rightarrow 4N_2 + 6H_2O \qquad (2)$$

$$6NO_2 + 8NH_3 \rightarrow 7N_2 + 12H_2O \qquad (3)$$

At lower operating temperatures, the reaction between ammonia and NOx is the predominant one. However, above a certain temperature (depending on the catalyst) the direct oxidation of ammonia by air becomes the predominant reaction leading to a decrease in NOx conversion and even increasing NOx emissions after the SCR:

$$4NH_3 + 3O_2 \rightarrow N_2 + 6H_2O \qquad (4)$$

$$4NH_3 + 5O_2 \rightarrow 4NO + 6H_2O \qquad (5)$$

The main disadvantages of SCR technology are the need of a "second storage" tank, distribution, and dosing system as well as the side production of N_2O a Green House Gas with a global warming potential of 310 (Foster et al, 2007) times the one of CO_2:

$$2NH_3 + 2O_2 \rightarrow N_2O + 3H_2O \qquad (6)$$

The focus of this research is to explore copper based and iron based zeolites (ZSM5 and Cuban natural mordenite) ability to convert NOx into harmless N_2 and water and to create a model to simulate NOx emission factor by neural networks of vehicles equipped with commercial available and future available SCR systems.

The aim of this research is to analyse new catalysts to use in SCR-NOx systems on the exhaust gas aftertreatment of diesel vehicles. The catalysts were prepared by ion-exchanged over Natural Cuban Mordenite and ZSM5 zeolites support. The catalyst stability in presence of H_2O and SO_2 of the prepared and commercial catalysts was also determined as well as the evaluation of catalyst activity at different temperatures. Finally, a neural network based approach to predict NO conversion efficiency of all catalysts were implemented to derive and input a mathematical function integrated with ADVISOR (Wipke, 1999) road vehicle simulator to obtain NOx emission factors and to test each SCR system mounted on vehicles (light and heavy-duty) for standardized driving cycles and real measured driving cycles.

2. EXPERIMENTAL SETUP

Samples of commercial vanadium based SCR (CATCO), and self made samples of copper (copper mordenite designated CuMORD; copper ZSM5 designated CuZSM5) and iron zeolites (iron mordenite designated FeMORD; iron ZSM5 designated FeZSM5) were tested.

Self made samples were prepared and characterized according to a rigorous methodology well documented in the literature (Torres-Abreu et al, 1997; R Moreno-Tost et al, 2008; Qingjum, 2003). Zeolites have an atomic ratio Si/Al = 20, and a natural mordenite from Palmarito deposit (Santiago de Cuba, Cuba), has an atomic ratio Si/Al = 5. Both were impregnated with copper and iron reaching 2-4 %wt.

After the preparation and characterizations, the samples were pelletised with a particle size of 0.25-0.40 mm. About 150 mg of pelletized solids were set into a reactor and plugged with glass wool (see apparatus diagram Figure (2)). The gas mixture allows mixing each the intended gas species concentration of oxygen, sulfuring oxide, ammonia and nitrogen oxide. Water was added to the feed, passing helium through a saturator with deionized water. Moreover, 50 ppm of the SO_2 was added to the feed to study its effect in the catalytic performance. The flows were independently controlled by channel mass flowmeters. After passing through the reactor the outlet gas was analyzed by a quadrupole mass spectrometer (Balzers GSB 300 02).

The interval of temperature studied ranged between 30ºC and 550oC. Species inlet concentrations (volume based) ranged from: 2-17% O_2, 0-16% H_2O, 0-50 ppm SO_2, 150-1600 ppm NO and 150-1600 ppm NH_3. These were found to be the typical values for Diesel engine exhaust (Wipke, 1999). Ca. 600 measurements were gathered for each of the 5 catalysts.

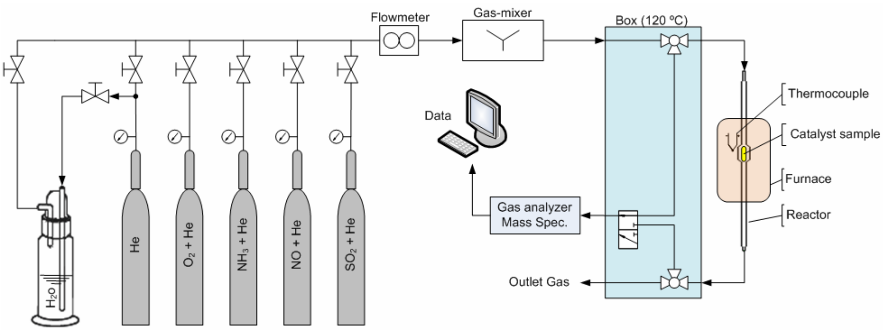

Figure 2. Apparatus diagram of the reaction system SCR.

To extrapolate the data measured in the reactor to a real catalyst, the space velocity parameter was used. Space velocity of a real catalyst in the Diesel vehicle exhaust was referred to the wash coat volume of the SCR catalyst that is 20% of the catalyst volume (Koltsakis, 1999). The typical engine-out data was converted to typical space velocity referred to the active part of the catalyst, and these values of space velocity were the ones used in the experimental part of the research:

$$GHSV = \frac{EMF(g.s^{-1})}{M_E(g.mol^{-1})} * \frac{R(\mathrm{J.mol^{-1}\ K^{-1}})T(K)}{P(J.l^{-1})} * \frac{1}{V(l)*0.2} * 3600(s.h^{-1}) \qquad (7)$$

EMF stands for exhaust mass flow, ME stands for molar mass of exhaust flow, R is the universal constant for gases, T is the temperature of exhaust gases, P is the exhaust gas pressure. GHSV values ranged from 40 to 250 h^{-1}.

Exhaust gas temperature of Diesel engines is typically below 400ºC. CATCO exhibits the highest conversion efficiencies and achieves rapidly the maximum conversion of 95%-99%. Both CATCO and iron based catalysts roughly maintain their maximum conversion efficiency after 400ºC. Mordenite catalysts exhibit a significant decrease, especially for copper based, revealing that the mechanism of NH_3 oxidation is becoming dominant (see equations (4), (5) and (6)).

3. NEURAL NETWORKS APPROACH

No application of Artificial Neural Networks (ANN) was found in the literature for the SCR system and subsequent integration with a road vehicle simulator. This innovative approach was explored by the authors in the current research. The measured results obtained from the experimental apparatus described above for Cu,Fe-zeolite catalyst and commercial catalyst were used to train an ANN. The main goal is to obtain the bias and weights for each neuron and the activation functions for hidden and output layers with the lowest Mean Squared Error (MSE). 15% of the data was not used to train the ANN, but instead to test it. The best ANN (Figure (3)) will be used to construct the correlation function between the inputs and outputs, and was integrated in a road vehicle fuel consumption and emission model.

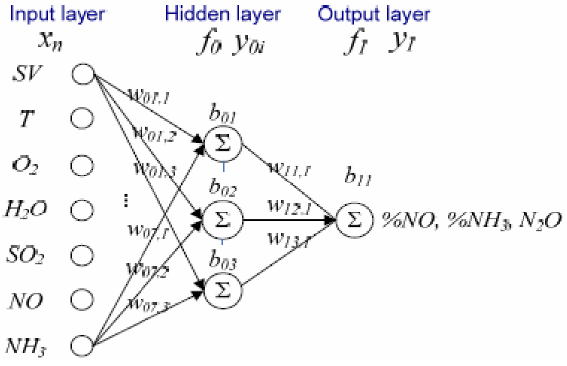

Figure 3 ANN architecture of SCR model network

ANN design is constituted by an input layer with 7 neurons, a single hidden layer with 3 neurons, and one output layer with one neuron, for each output considered. Stochastic back propagation with the Levenberg-Marquardt algorithm (MatLab®) was used to train the network. The activation functions used are:

$$f_0 = \log sig(n) = \frac{1}{(1+\exp(-n))} \tag{8}$$

For the hidden layer and for the output layer:

$$f_1 = \tan sig(n) = \frac{2}{(1+\exp(-2n))-1} \tag{9}$$

The derived correlation for the ANN is:

$$y_{0i} = \log sig(w_{0n,i} * x_n + b_{0i}) \tag{10}$$

$$y_1 = \tan sig(w_{0i,1} * y_{0i} + b_{11}) \tag{11}$$

$$\%NO, \%NH_3, N_2O = \tan sig\left(\sum\left(w_{0i,1} * \log sig\left(\sum\left(w_{0n,i} * x_n\right) + b_{0i}\right)\right) + b_{11}\right) \tag{12}$$

Different weights and biases are generated for each different output (y1). For example the mathematical correlation that describes NO conversion efficiency for FeMORD by using ANN approach is: _tan sig[(log sig((-0.4245SV-0.1349T+0.0309O2+4.9501H2O-0.406SO2+2.8184NO-0.3412NH3-4.521))*(-1.5597)+ log sig(1.2677SV-3.1144T+2.5672O2+0.3678H2O-0.6827SO2-0.1104NO+0.1376NH3+1.322))*(12.4344)+ log sig((-0.3581SV-2.8388T+3.0426O2-5.567H2O+4.3161SO2-2.9476NO+5.5501NH3-2.223))*(6.8171)-10.84)]._ This result is between -1 and 1 but is converter to NO conversion efficiency values using the correspondence -1 equivalent to 0% and 1 equivalent to 100%.

All mathematical correlations derived from ANN application were used in a road vehicle simulator exhaust aftertreatment module. The resultant application was capable of predict the NOx conversion efficiency (%), the NOx out emissions, the NH_3 emisions and N_2O emissions (in ppm, g/s, g/km or g/kWh) for several driving conditions, including the standard ones. However, in this paper will only present the results of NOx emissions and N_2O formation.

3.1. Validation

A total of about 600 set of measurements were performed, of which 85% were used to train and validate several networks. The networks with low sum squared error were used to obtain the correlation function between the inputs and outputs. Stochastic back propagation was used to train the network, the number and size of the hidden layers being determined empirically according to error value and stability observed. A single hidden layer containing 3 neurons proved the best solution for the application. The 15% were used to test the SCR ANN model.

The errors where compared to the training data for the network used. The statistical analysis of the results indicates that the R^2 value for the test, training and validation data was higher, more than 0.67, and this indicates that ANN-based model developed in this work can predict the (%) NO conversion efficiency to SCR system of a Diesel vehicle.

3.2. Integration with ADVISOR

The correlation functions obtained in ANN were integrated with the road vehicle fuel consumption and emissions model ADvanced VehIcle SimulatOR (ADVISOR) which was developed at the National Renewable Energy Laboratory (NREL). Besides the addition of new SCR functions, a real measured driving cycle in a bus line of Porto city, Portugal (Silva et al, 2006) for heavy-duty vehicles and ETC (Europe Transient Cycle, FIGE version according to dieselnet, 2009) were included in ADVISOR database.

The standard cycles (NEDC for light-duty, UDDS (Urban Dynamometer Driving Schedule), HWFET (HighWay Fuel Economy Test), ESC and ETC for heavy-duty engine), a constant speed of 120 km/h HDC (Highway Driving Cycle) and a real measured driving cycle (Porto City). Main characteristics of each driving cycles, such as total duration, average speed, distance, and so forth, are shown in Table (1).

Table 1. Driving cycles used (dieselnet, 2009)

Drive cycle	UDDS (urban)	HWEFT (road)	NEDC	Porto bus line	HDC	ETC
Maximum speed (km/h)	91	96	120	38	120	91
Average speed (km/h)	32	78	34	11	120	60
Distance (km)	12	17	11	8	10	29
Time (s/min.)	1369/23	765/13	1180/20	2466/41	300/5	1799/30
Maximum acceleration (m/s^2)	1.48	1.43	1.06	2.23	0	3.83
Max downgrade (%)	0	0	0	-15	0	0
Max upgrade (%)	0	0	0	10	0	0
Average grade(%)	0	0	0	3	0	0
Idle (s)	259	6	298	696	0	75

In order to test the integrated model, a heavy-duty vehicle of weight 18000 kg equipped with a pre-Euro engine with 10 liters and a maximum power of 173 kW, and a SCR of 15 liters, and a light-duty Euro I vehicle of weight 1050 kg equipped with a 1.7 liters, 60 kW engine and SCR of 3 liters were used. The volume of SCR was chose according to the empirical ratio of SCR volume being 1.5-2 times engine displacement (Conway et al, 2005). The next section shows the obtained results.

4. RESULTS AND DISCUSSION

Both heavy and light-duty vehicles with and without a SCR system were simulated. NOx emissions values were compared with emissions limits to analyze the corresponding emission standard classification of the vehicles after SCR fitting. As previously mentioned other driving cycles, including real measured ones, were simulated and NOx emission values again compared with emission limit, for example: Euro IV regulation (limit 3.5 g/kWh) and EPAMY2004 (limit 2.7 g/kWh).

Emissions for light-duty vehicles are expressed in grams of pollutant per unit of travelled distance, g/km, and emissions for heavy-duty vehicle are expressed in grams per mechanical energy delivered by the engine, g/kWh. The Figure (4), show the performance of the SCR system for, respectively, (ESC + ETC) and (NEDC), European standard cycles.

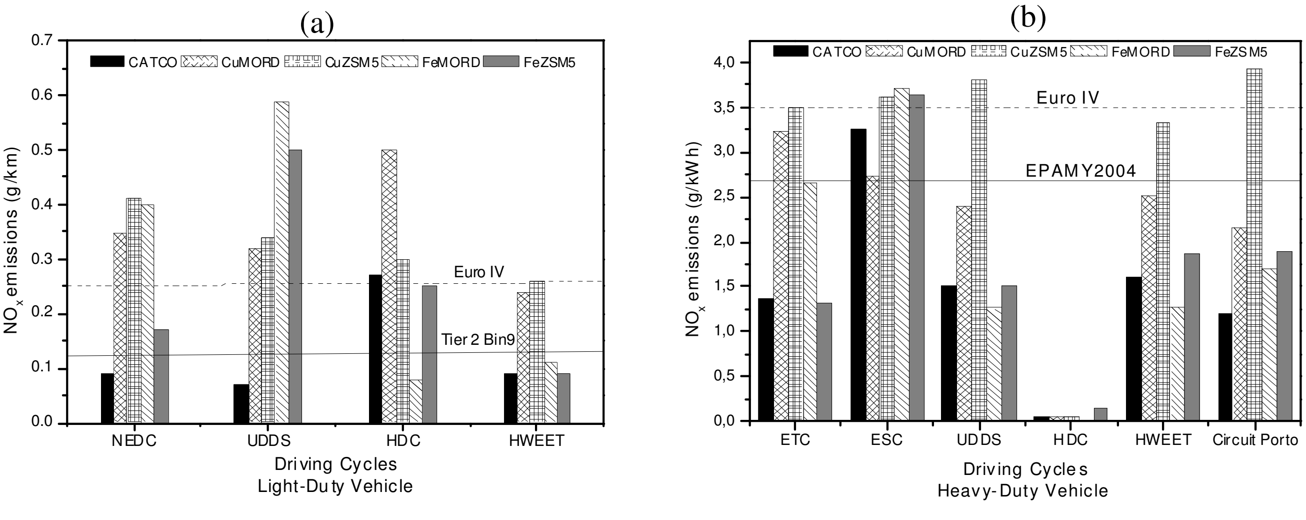

Figure 4. Light-duty (a) and Heavy-duty (b) vehicle average NOx emissions.

The pre-Euro heavy-duty vehicle after being equipped with a SCR system is compliant with Euro IV regulation independently of SCR based material. For ETC driving cycles, both CATCO and FeZSM5 catalysts showed higher performance NO conversion (80%) than others catalysts. They also demonstrated more stability under condition of high average load factors and very high exhaust gas temperatures. All catalysts showed NOx conversion efficiency above 60% for the ESC test cycle. However the CuMORD exhibited the highest performance of NOx conversion (75%).

Regarding the non-standard driving cycles (representative of more realistic driving conditions) for the real measured Porto cycle, except for copper-zeolite catalysts, NOx values are lower than those obtained in the standard cycles (compliant with Euro V 2g/kWh limit). For that cycle, in the case of CuMORD, its performance could be explained by the sharply decrease of the conversion efficiency in the temperature observed experimentally, above 400 ºC, which occurs 14 % of the time. This effect is partially overcome due to the fact CUMORD has a good tolerance to water and SO_2 for temperatures below 400ºC. For CuZSM5, the temperature is not the main issue, but the space velocity, that highly affects its performance, and averages ca. 111x103 h^{-1} in Porto driving cycle, and water intolerance could explain its "poor" performance (average water concentration is 3.93 vol.% in Porto driving cycle). CATCO and iron based catalysts exhibit a high average NO conversion efficiency, respectively, 90.4% and 81%, in the Real Urban Circuit of Porto. Generally, among the zeolite-catalysts, the FeMORD revealed the best performance (low level of NOx out g/kWh) in all the non-standard simulated driving cycles. This can be explained by its performance being similar with CATCO performance for several space velocities.

Figure (5) shows N_2O emissions for different SCR. Mordenite based catalysts do not emit N_2O and therefore are not represented. In all driving cycles the FeZSM5 catalyst demonstrated low N_2O formation, with maximum value ca. 0.9 g/km in the HWEFT driving cycle.

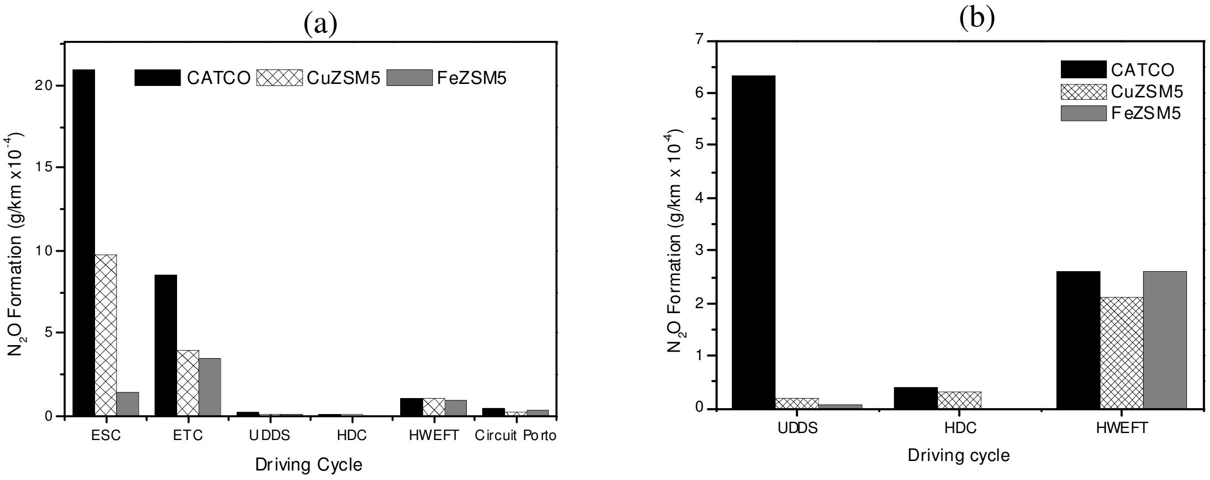
(a) (b)

Figure 5. Light-duty (a) and Heavy-duty (b) vehicle average N_2O emissions.

In terms of validation of the integrated model, for heavy-duty vehicles, results for commercial vanadium based catalyst can be found in the literature, e.g. (Conway et al, 2005; Leaners and Proppel, 2005; Goo et al, 2007; Sluder et al, 2004). Eminox guarantees that Euro I heavy-duty vehicles after SCR retrofitting can achieve Euro IV or Euro V limits, which is consistent with our results. Conway et al, found an average experimental conversion efficiency of about 80% for average trip exhaust gas temperatures of 250ºC (average temperature of Porto cycle is 220ºC, and average NOx conversion efficiency was 81%), values also consistent with G. Leaners et al work.

For N_2O emissions (see Figure (5), the works Goo et al, 2007; Sluder et al, 2004 report experimental dynamometer values below 100 ppm which is consistent with our results (average 3 ppm, maximum 30 ppm for Porto cyle). Figure (5) shows N_2O emissions for different SCR. Mordenite based catalysts do not emit N2O and therefore are not represented. Summarizing, the best alternative for replacement of CATCO seems to be iron based zeolites that combine lower NOx and N_2O tailpipe emissions.

No experimental data was found in the literature for light-duty vehicles equipped with SCR systems. The Euro I light-duty vehicle after being fitted with a SCR comply with Euro V in the cases of CATCO and FeZSM5 and with Euro III in the remain cases. In terms of standard accomplishment, FeZSM5 seems to be the best candidate for CATCO replacement. Regarding average values on other driving cycles (see Figures (4a), Euro IV limit of 0.25g/km and Tier2 Bin 9 limit 0.12 g/km are represented for reference), it is unclear which is the best alternative for replacing CATCO. In UDDS the best SCR is CuMORD, for the constant 120 km/h cruise control speed the best is FeMORD and for HWEFT is FeZSM5.

5. CONCLUSIONS

The pre-Euro heavy-duty vehicle after being equipped with a SCR system is compliant with Euro IV regulation (limit 3.5 g/kWh) independently of SCR based material. Regarding other driving cycles (representative of more realistic driving conditions) it is observed that in the real measured Porto cycle except for CuMORD, NOx values are lower than those obtained in the standard driving cycles (compliant with Euro V 2g/kWh limit).

The Euro I light-duty vehicle after being fitted with a SCR comply with Euro V in the cases of CATCO and FeZSM5 and with Euro III in the remain cases. In terms of homologation FeZSM5 seems to be the best candidate for CATCO replacement. Regarding average values on other driving cycles it is unclear which is the best alternative for replacing CATCO. In UDDS the best SCR is CuMORD, For the constant 120 km/h cruise control speed the best is FeMORD and for HWEFT is FeZSM5.Regarding N_2O emissions mordenite based SCR do not emit this pollutant.

The collection of on-board data in real vehicles and real road conditions will be of extremely importance for further validate the developed integrated model and to better understand the real NOx emission versus standard compliant one.

The results of the research show that the correlations obtained with artificial neural networks integrated with a road vehicle simulator produce average NOx results for commercial vanadium based catalyst consistent with experimental data and can be a powerful tool for SCR simulation of Diesel vehicles equipped with advanced aftertreatment systems. This way NOx emission factor simulation and N_2O formation for several SCR technology and different driver behavior/traffic conditions is possible.

6. ACKNOWLEDGEMENTS

The authors would like to acknowledge the National Council for Scientific and Technological Development (CNPq) and FUNCAP (*Fundação Cearense de Apoio ao Desenvolvimento Científico e Tecnológico*). Ministerio de Educación y Ciencia of Spain (project NAN2004-09267-C03-01). The authors would like to acknowledge FCT (National Science and Technology Foundation) through the national project POCI/ENR/57450/2004. Special acknowledgments to the laboratory research team of University of Málaga, Spain laboratory leaded by professor Enrique Castellón that were very useful to the fulfillment of the experimental phase of this research.

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