

Research Article

The Experimental and Simulation Study of Selective Catalytic Reduction System in a Single Cylinder Diesel Engine Using NH₃ as a Reducing Agent

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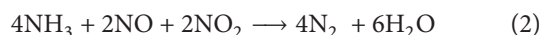
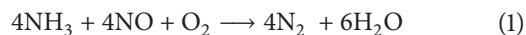
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Selective catalytic reduction (SCR) technology has been widely used in automotive applications in order to meet the stringent limits on emission standards. The maximum NO_x conversion efficiency of an SCR depends on temperature and mass flow rate of an exhaust gas. In order to assess the suitability of Cordierite/Pt catalyst for low temperature application, an experimental work is carried out using single cylinder diesel engine for different load conditions by varying ammonia induction rate from 0.2 kg/hr to 0.8 kg/hr. The simulation is carried out using AVL FIRE for the validation of experimental results. From the study, it has been found that for 0.6 kg/hr ammonia induction rate the maximum conversion is achieved, whereas, for 0.8 kg/hr, conversion is reduced due to desorption of ammonia. Also it has been found that, at 75% of load, for all mass flow rates of ammonia the conversion was drastically reduced due to higher exhaust gas temperature and higher emission of unburnt hydrocarbons. More than 55% of NO_x conversion was achieved using Cordierite/Pt catalyst at a temperature of 320°C.

1. Introduction

In NH₃ SCR system, NH₃ is inducted to the exhaust gas coming out from the engine exhaust, which will undergo various reactions to convert NO_x into free nitrogen (N₂).

The most important reactions are



Two most commonly used SCR reductants are anhydrous (dry) ammonia and aqueous ammonia or urea. Pure anhydrous ammonia is extremely toxic and difficult to store safely but does not require further conversion to operate within an SCR. It is typically favoured by large industrial SCR operators. Aqueous ammonia must be hydrolyzed in order to use, but it is substantially safer to store and transport than anhydrous ammonia. Urea is the safest to store but requires

conversion to ammonia through thermal decomposition in order to be used as an effective reductant. The temperature-programmed activity of a series of oxide-supported (TiO₂, Al₂O₃, and SiO₂) Cu catalysts formed from two different Cu precursors (Cu(NO₃)₂ and CuSO₄) for the selective catalytic reduction of NO_x using solutions of urea as a reductant [1]. These activities are compared to those found using NH₃ as a reducing agent over the same catalysts in the presence of H₂O and it is found that catalysts that are active for the selective reduction of NO_x with NH₃ are inactive for its reduction using solutions of urea. However, catalyst plays a major role in the conversion of NO_x. V₂O₅/Al₂O₃ catalysts can be used for operating temperature higher than 250°C but restricted to sulphur free application due to deactivation of the catalyst from alumina reaction with SO₃ forming Al₂(SO₄)₃. The nonsulphating TiO₂ carrier was recommended for the V₂O₅. Studies that have been carried out with V₂O₅ catalyst



FIGURE 1: Experimental test rig.

supported on TiO_2 and WO_3 used for Heavy Duty diesel Engines in Europe with numerous highway studies [2].

The studies highlighted that problems with vanadium catalyst are quickly deactivated at high temperatures above 500°C and concluded that recommended temperature window for vanadium is from 300 to 450°C [3]. Zeolite catalysts are developed to cover a wider range of temperature windows over platinum- and vanadium-based catalysts. Zeolite catalysts are developed to extend the operating temperature above 350°C over platinum and vanadium based catalysts. However, two types of zeolite catalysts were developed to cover high and low temperature windows. The high temperature zeolite covers temperature windows from 350 to 600°C . Automotive catalytic converters constantly heat up and cool down as the engine starts and stops. This requires some special material with the greatest possible resistance to change temperature and a low heat expansion coefficient. There are studies conducted on the effect of HC emission on the retarded activity of SCR catalysts [4]. They considered CuZSM5 , FeZSM5 , and $\text{V}_2\text{O}_5/\text{TiO}_2$ catalysts and compared the effect of HC emission on the DeNO_x efficiency of SCR catalyst. They concluded that the primary cause for the inhibition is the competitive adsorption of NH_3 and C_3H_6 onto the catalyst surface and the useless consumption of NH_3 by the side reaction, including the NH_3 oxidation and ammoxidation reactions during the course of NH_3/SCR reaction with C_3H_6 . Another study was conducted on the effect of unburnt hydrocarbons in the activity of automobile catalysts [5]. The study reveals that the NO_x conversion efficiency of SCR catalyst will reduce, when there is an HC in exhaust gas. When HCs are added to the flow, some C-species deposit forms on the surface. This result is consistent with a pore blocking effect which will slightly affect the NO_x removal at the highest reaction temperature. They also found that the most important effect of HC presence on the SCR activity is due to the competitive adsorption between hydrocarbons and ammonia which limits the NO_x conversion efficiency of SCR.

In the current work, Cordierite/Pt ceramic honeycomb structured catalyst has been used to assess the suitability of this material for the medium and light duty diesel vehicles as it gives resistance to temperature changes and a low heat expansion coefficient.

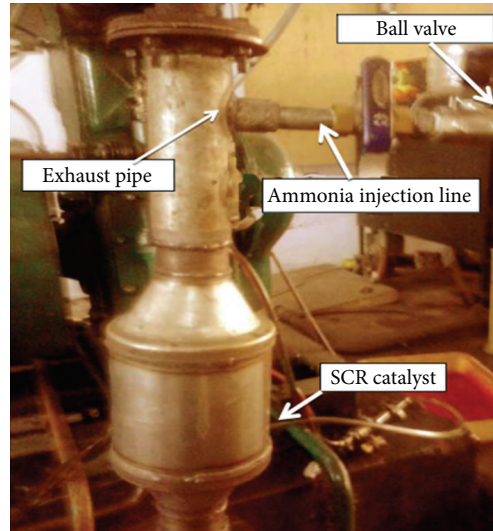


FIGURE 2: SCR setup.

TABLE 1: SCR catalyst properties.

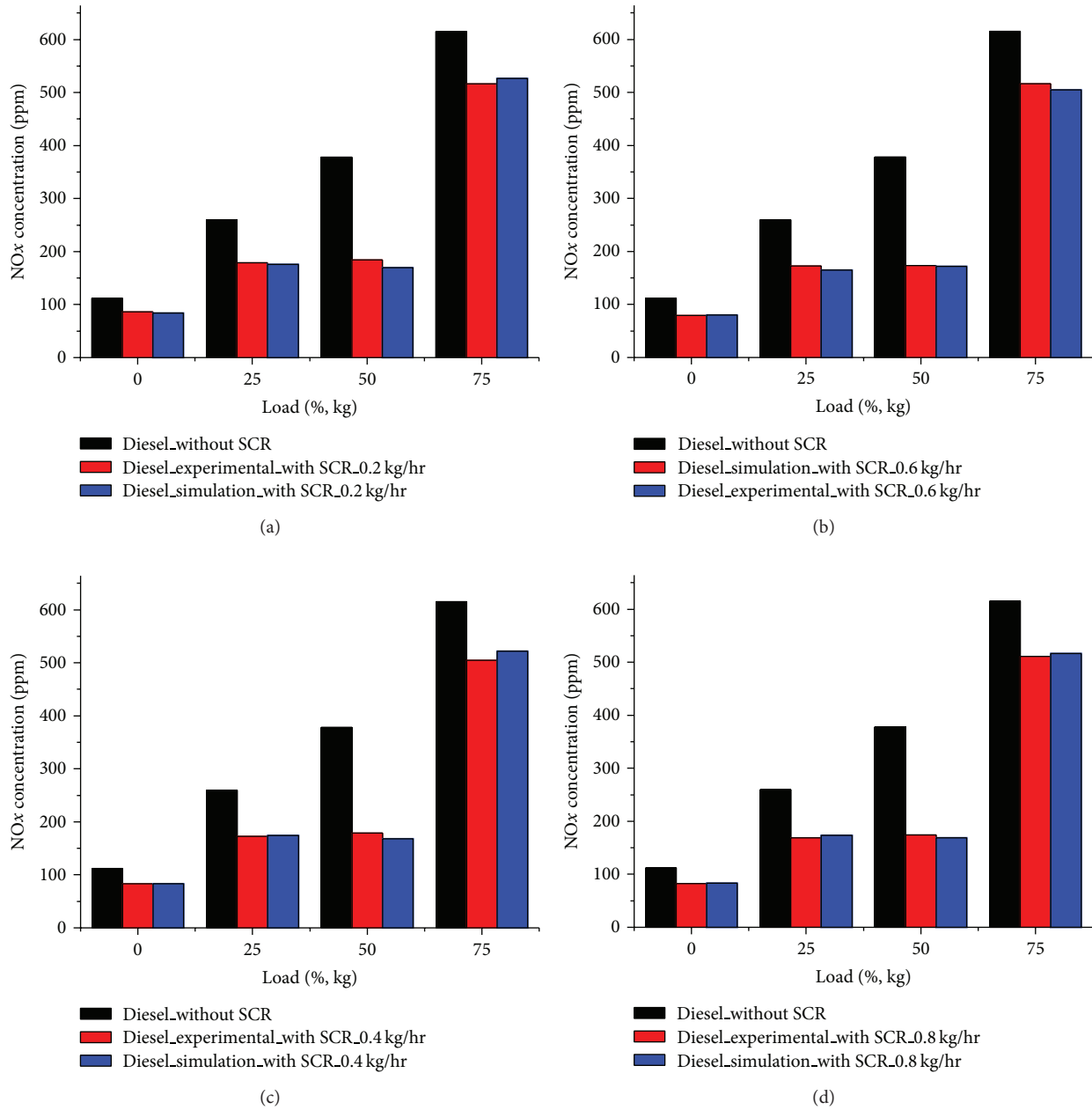
Catalyst material	Cordierite/Pt
Catalyst type	Circular honeycomb structure
Size of the catalyst	$118.4 \text{ mm} \times 127 \text{ mm}$ ($D \times L$)
Specific heat	1.46 kJ/kgK
Cell density	400 l/in^2
Volume of catalyst	1.2 L
Wall thickness	0.114 mm
Density	0.0215 kg/m^3
Thermal conductivity	3 W/mK

2. Experimental Work

Whole set of experiments were conducted at the designed injection timing of 27 deg bTDC , speed of 1500 rpm , and 17.5 compression ratio on test rig as shown in Figure 1.

Airflow measurement was done by the conventional method U-tube manometer as well as by air intake DP unit in the control panel. Engine speed measurement was sensed and inducted by inductive pickup sensor in conjunction with digital RPM indicator, which is a part of eddy current dynamometer controlling unit. The dynamometer shaft rotates close to inductive pickup sensor as an arrangement to send voltage pulse whose frequency is converted into RPM and displayed by digital indicator in the control panel. To measure the load on the engine, an eddy current dynamometer is attached to the crankshaft of the engine. An AVL-made exhaust gas analyzer was used to measure the exhaust gas emissions. With the analyzer, NO_x (ppm), CO (%vol.), UBHC (ppm), and CO_2 (%vol.) emissions were measured. A rotameter specially calibrated for ammonia was used to measure the flow rate of ammonia. The experiments were conducted at no load, 25% of full load, 50% of full load, and 75% of full load condition with diesel fuel.

The experiment is conducted on two conditions: firstly, without using SCR at the exhaust pipe and, secondly, fitting

FIGURE 3: Concentration of NO_x versus percentage of load.

SCR catalyst at the exhaust pipe. Both the readings will be tabulated and can find the DeNO_x efficiency of SCR.

3. SCR Setup

In this experimental setup, a circular shaped, honeycomb type of SCR catalyst (Cordierite/Pt, 1.2L) is fitted to the exhaust pipe with flange supports as shown in Figure 2. Sealing gaskets were placed in between two flange connections. The gasket used was a high temperature resistance type to prevent gas leakage from the exhaust system. Some minor adjustment was necessary in the final SCR exhaust assembly because of the restricted space within the cell. A half inch

barrel nipple of length of 3 inches is coupled to the exhaust pipe through which ammonia gas is passed.

A hose pipe from the ammonia cylinder is connected to ammonia rotameter, where the ammonia flow rate can be controlled by turning the regulating knob.

4. Simulation Work

The simulation has been carried out using CFD code AVL FIRE. The governing equations and reactions used in AVL FIRE are listed in [5]. The properties of the material are shown in Table 1.

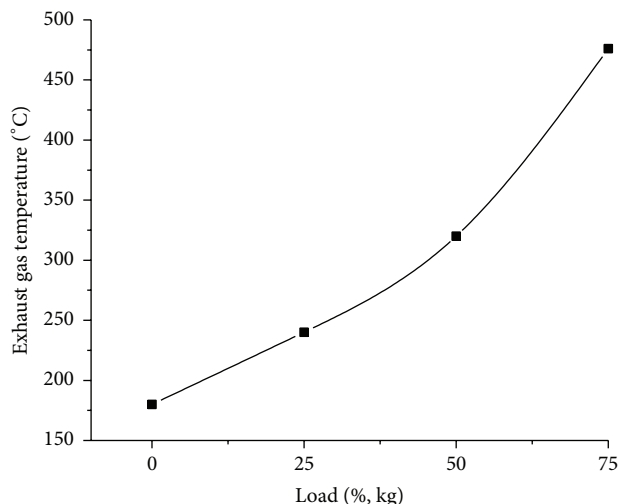


FIGURE 4: Variation of exhaust gas temperature versus percentage of load.

The mass flow rate of exhaust gas is calculated for different load conditions. It was found that as the load increases, the mass flow rate of exhaust gas decreases. This is due to change in density of gas. When load increases, exhaust gas temperature also increases, which results in lower density of a gas. The boundary conditions considered in the simulation are given in Table 2. The normalized residual values for the convergence criteria were taken as $1e-4$ for continuity, momentum, and energy equations. It took 60 iterations to reach the convergence. The maximum iteration was limited to 60, since the solution arrived at was close to the exact solution (experimental values) within these convergence criteria.

The mass fractions of different species including NO_x , CO , CO_2 , H_2 , and HC are considered from experimental values, measured by AVL Five gas analyzer. The outlet pressure was assumed to be 1 bar. The mass flow rate and temperature of exhaust gas are also given as input which is measured by experimental work. The SCR catalyst was modeled using the abovementioned dimensions and meshed in AVL Workflow manager. The simulation is carried out for different load conditions by varying ammonia flow rate from 0.2 kg/hr to 0.8 kg/hr.

5. Results and Discussion

The experiment was carried out for diesel fuel by varying load conditions. It reveals that as the load increases, the exhaust gas temperature increases. The flow rate of ammonia is also varied from 0.2 kg/hr to 0.8 kg/hr. The three major factors which have got significant effect in the conversion of NO_x are exhaust gas temperature, exhaust gas mass flow rate, and ammonia flow rate.

The SCR catalyst used in the experiment can withstand temperature range up to 500°C . Therefore, the load condition applied was limited to 75% of full load where exhaust gas temperature was found to be 486°C . At 100% of load, the temperature will go beyond 520°C , which will damage the

SCR catalyst, which in turn affects the exhaust gas analyser. The experiment has been carried out for two conditions. In the first case, SCR catalyst was not fitted at the downstream exhaust pipe. Engine was run for different load conditions and the concentration of NO_x was measured directly by AVL-made Five gas analyser. In the second case, the SCR catalyst has been fitted at the exhaust pipe. The ammonia gas was inducted to the exhaust pipe just before SCR setup. The flow rate has been varied from 0.2 kg/hr to 0.8 kg/hr. Again the engine can run in varying load conditions. The concentration of NO_x can again be measured by exhaust gas analyzer. Figure 3 explains the concentration of NO_x measured for both the conditions explained above.

From Figure 3, it is clear that as the load increases, the concentration of NO_x also increases because of higher combustion temperature. The higher combustion temperature results in higher exhaust gas temperature.

Figure 4 explains the variation of exhaust gas temperature with respect to varying load conditions. It shows that as the percentage load increases, the exhaust gas temperature also increases. At no load condition 180°C temperature was observed, whereas at 25% of full load, at 50% of full load, and at 75% of full load 240°C , 320°C , and 486°C temperatures have been observed. From the graph it can be clearly observed that, at 50% of full load for all the ammonia flow rates, maximum conversion has been observed, where NO_x concentration was reduced from 378 ppm to 169 ppm. This is because the catalyst exhibited the maximum selectivity at the temperature of 378°C . The minimum conversion is observed at 75% of full load, where NO_x concentration was reduced from 615 ppm to 526 ppm. The NO_x conversion with respect to varying load condition is explained in Figure 5.

It was found that, up to 50% load, NO_x conversion keeps on increasing, whereas, at 75% load, DeNO_x efficiency is drastically reduced. Similar pattern of graph is observed for all the ammonia mass flow rate conditions. It was found that the experimental values are in close agreement with simulation results. The increase in NO_x conversion is due to increase in exhaust gas temperature. At 50% load, maximum conversion is obtained since the exhaust gas temperature is 320°C where the catalyst exhibits maximum selectivity, due to which fast SCR reaction takes place, which in turn converts NO_x into free nitrogen (N_2). It was found that, for 0.2 kg/hr of ammonia induction, at no load condition, up to 25% of NO_x conversion is achieved. Similarly for 0.4 kg/hr, 0.6 kg/hr, and 0.8 kg/hr of ammonia induction rate, 26%, 29%, and 27% of NO_x conversion are achieved as shown in Figure 5. Similarly at 25% load, for 0.2 kg/hr, 0.4 kg/hr, 0.6 kg/hr, and 0.8 kg/hr ammonia induction rate, 31%, 33%, 36%, and 34% of NO_x conversion are achieved. A slight increment in NO_x conversion is achieved at 25% load, because of increased exhaust gas temperature and lower mass flow rate of exhaust gas. At 50% load, maximum conversion is obtained for all the mass flow rates. It was found that, at 0.2 kg/hr ammonia induction, 55% of NO_x conversion is obtained. Similarly for 0.4 kg/hr, 0.6 kg/hr, and 0.8 kg/hr ammonia induction, 55%, 55%, and 54% of NO_x conversion

TABLE 2: Boundary conditions used in the simulation work.

Solver control discretization				
Calculation of boundary conditions: extrapolate	Calculation of derivatives: least square fit	Iteration method: simple	Decoupled domains: no	Realizability constraints: no
Equation control				
Momentum and continuity: yes	Turbulence: K-epsilon	Energy: yes	Viscous heating: yes	Pressure work: yes
Differencing scheme				
Momentum: central differencing	Continuity: central differencing	Turbulence: upwind	Energy: upwind	Scalar: upwind
Convergence criteria per time step				
Maximum number of iterations: 60		Minimum number of iterations: 10		

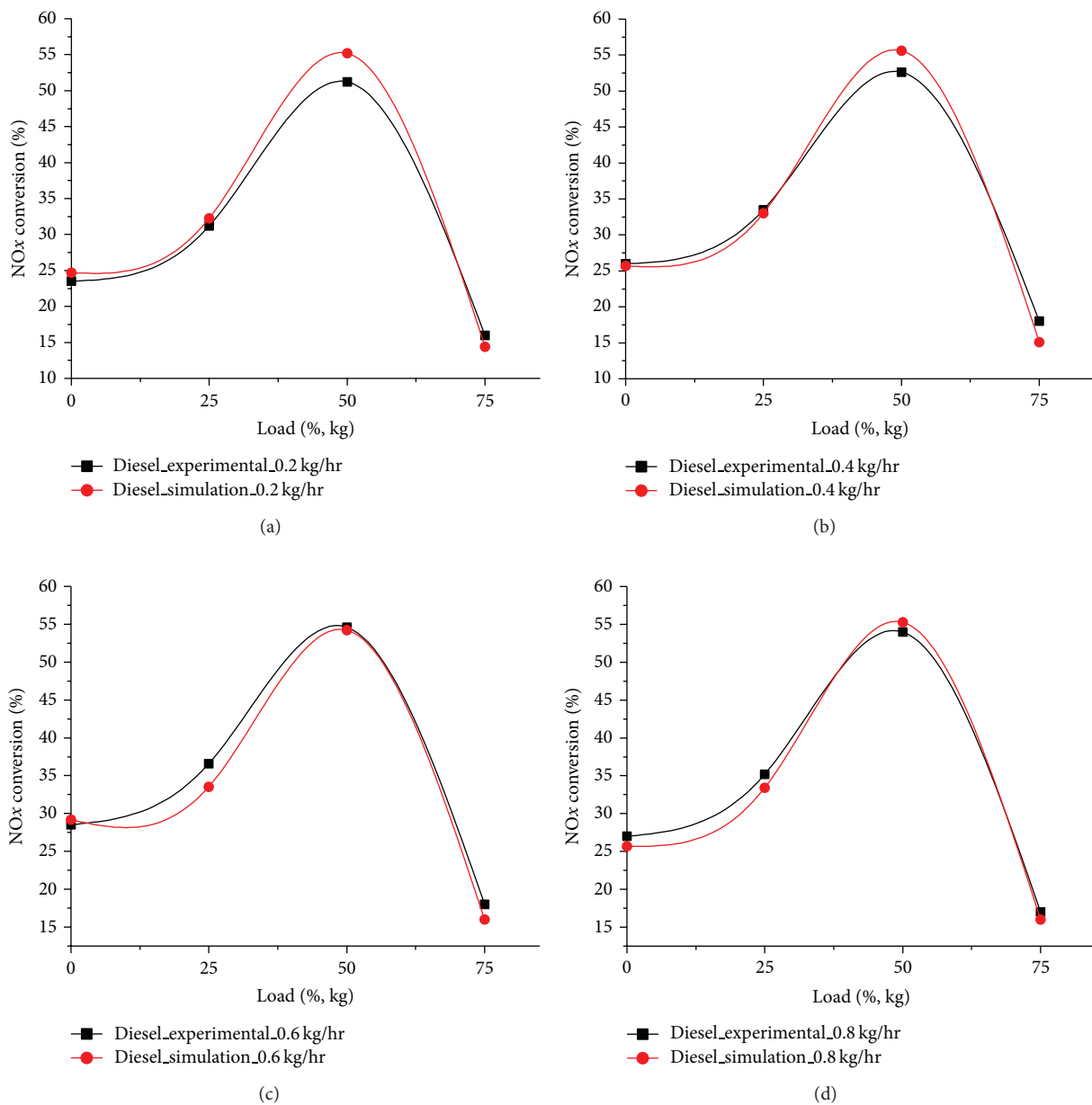


FIGURE 5: NOx conversion versus percentage of load.

are obtained. The conversion is more here due to higher exhaust gas temperature and higher reaction rate.

But at 75% load, the NO_x conversion is reduced drastically due to increased exhaust gas temperature. At very high temperature ($>450^\circ\text{C}$) the catalyst selectivity goes on decreasing due to sintering effect of the catalyst [6] and desorption of ammonia [7] through the walls of monolith. Another reason for the lower NO_x conversion at 75% load is the increased emission of hydrocarbons as explained in the Introduction part. Figure 6 explains the concentration of unburnt hydrocarbons for different load conditions.

At 75% load 34 ppm of HC can be liberated, when compared with no load where just 13 ppm of HC emission is found. When there is more emission of HC at higher temperature, the carbon particles deposit at the pores of the catalyst, which results in lower NO_x conversion.

The NH_3 conversion is another parameter, which will greatly influence NO_x conversion. The concentration of ammonia can be calculated in different ammonia flow rate. Four samples of 1N HCL acid had been taken in beakers. Ammonia gas was passed through these samples for different ammonia flow rates varying from 0.2 kg/hr to 0.8 kg/hr. The titration was carried out against 1N NaOH. Also a blank titration was carried out for 1N HCL acid without ammonia gas passing through it, against 1N NaOH. The readings were tabulated and the concentration is calculated by subtracting both the readings. It has been found that, as the ammonia flow rate increases, the concentration of ammonia also increases. From Figure 7, it can be noted that, for 0.2 kg/hr ammonia flow rate, 400 ppm ammonia concentration is obtained. Similarly for 0.4 kg/hr, 0.6 kg/hr, and 0.8 kg/hr ammonia flow rate, 450 ppm, 550 ppm, and 600 ppm ammonia concentrations are obtained.

Ammonia slip is the major disadvantage in SCR system, which will harm the environment and cause health problems. Hence, NH_3 conversion is the major focus of study in the current work. Figure 8 explains the NH_3 conversion with respect to varying load conditions.

From Figure 5, it was found that there is no much difference in NO_x conversion by varying ammonia flow rate from 0.2 kg/hr to 0.8 kg/hr. Hence, the study is focused on to find out the optimum ammonia inlet concentration which will result in the maximum NH_3 conversion. From Figure 8, it has been found that, using 0.2 kg/hr ammonia flow rate, the maximum conversion of 24% of NH_3 conversion was achieved. Since the ammonia flow meter used in the experiment can measure a minimum of 0.2 kg/hr ammonia flow rate where ammonia concentration was found to be 400 ppm, the simulation work has been extended by again minimizing the ammonia inlet concentration, to find out the maximum NH_3 conversion efficiency.

The ammonia inlet concentration was varied from 100 ppm to 400 ppm to find out the maximum NH_3 conversion. It has been observed that, below 100 ppm ammonia inlet concentration, the NO_x conversion as well as NH_3 conversion is drastically reduced. Hence, ammonia inlet concentration was limited to 100 ppm. Figure 9 explains the

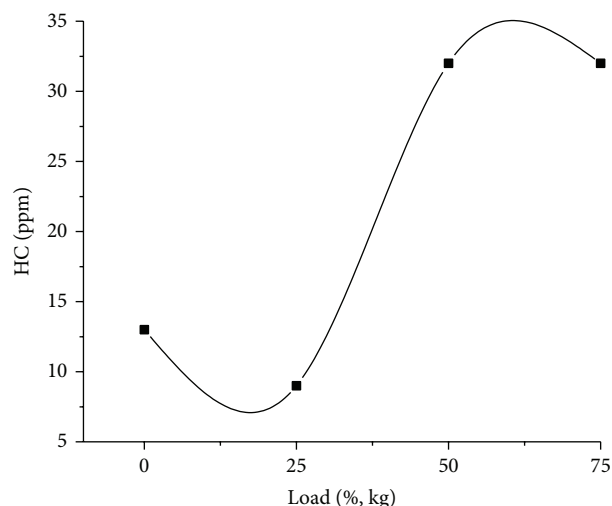


FIGURE 6: Hydrocarbon emission versus percentage of load.

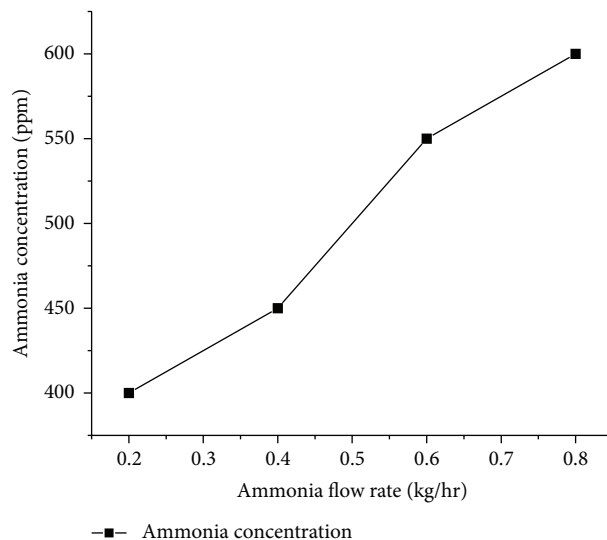
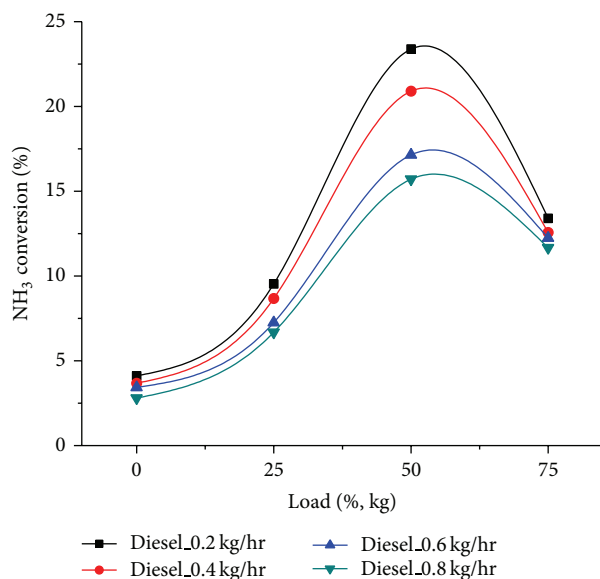
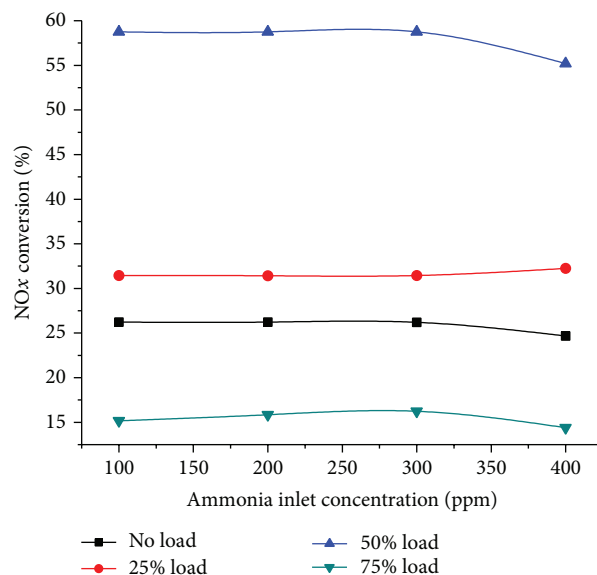
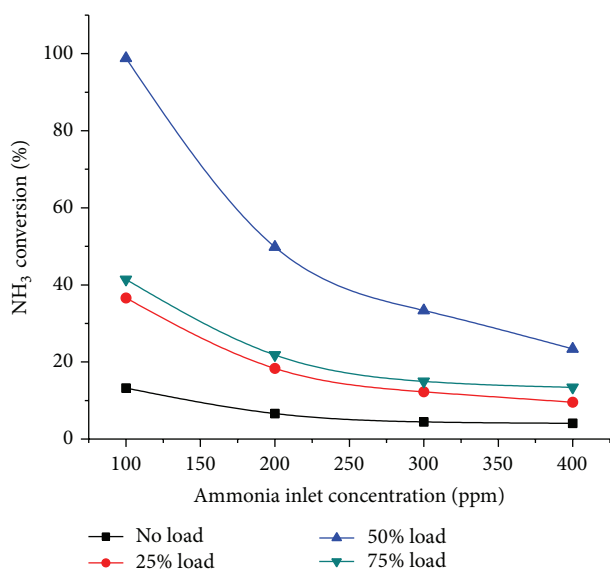


FIGURE 7: Variation of NH_3 concentration with respect to inlet ammonia flow rate.

NH_3 conversion rate with respect to varying ammonia inlet concentration.

It was found that as the ammonia inlet concentration reduces, the NH_3 conversion rate increases. At 50% of full load and 100 ppm ammonia inlet concentration, 98% of NH_3 conversion was achieved. Similarly for 100 ppm ammonia inlet concentration, at no load condition, at 25% of full load, and at 75% of full load 13%, 36%, and 41% of NH_3 conversion were achieved. This reveals that 100 ppm ammonia inlet concentration is the optimum concentration, which will result in the maximum NH_3 conversion. The corresponding NO_x conversion is also given in Figure 10. It has been found that there is slight increment at 50% of load, where 58% of NO_x conversion was achieved. Hence, it can be concluded that the Cordierite/Pt catalyst will give a maximum of 58% of NO_x conversion at 100 ppm of ammonia inlet concentration.

FIGURE 8: NH₃ conversion versus percentage of load.FIGURE 10: NO_x conversion versus ammonia inlet concentration.FIGURE 9: NH₃ conversion versus ammonia inlet concentration.

6. Conclusions

- (1) It has been found that, up to 0.6 kg/hr ammonia induction, the NO_x conversion goes on increasing, whereas, at 0.8 kg/hr, NO_x conversion is slightly reduced due to desorption of ammonia and blockage of pores in a catalyst.
- (2) At 50% load and for all the ammonia flow rate conditions, >50% of NO_x conversion has been achieved due to higher exhaust gas temperature, where catalyst shows better selectivity, which results in conversion of NO_x into free nitrogen (N₂).
- (3) At 75% load and for all ammonia flow rate conditions, the NO_x conversion has been reduced drastically,

because of ammonia desorption and increased emission of unburnt hydrocarbons.

- (4) It was found that as the ammonia inlet concentration reduces, the NH₃ conversion goes on increasing, where a slight increment in NO_x conversion is only observed. It concludes that Cordierite/Pt catalyst will give a maximum of 58% of NO_x conversion at 100 ppm of ammonia inlet concentration.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

Acknowledgments

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