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Study of the Effective Parameters on Selective Catalytic Reduction of NOx by Ammonia over a Vanadia-Titania Catalyst from Exhaust Gases

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Abstract

The removal of nitrogen oxides (NO_x) is crucial problem for global environment. In This study, a 3D dynamic simulation model for the application in investigation of the reaction characteristic and the phenomena of transport in the catalytic filter of the SCR reactor is introduced. In order to make an assessment on the kinetic parameters of the model by the mechanism of Eley-Ridel (ER) from experimental data, an optimization method which acts by integrating the Taguchi method, a real-coded genetic algorithm auxiliary model is proposed. With the aid of the introduced dynamic model, the impacts of the key parameters, namely operating temperature, the gas hourly space velocity, the amount of the applied ammonium and the cross section of the channel of SCR reactor on the NO_x conversion and NH_3 slip phenomena were investigated. By comparing with the experimental data available in the literature, it was validated and it is found that NO_X conversion increases with the decrease in the operation temperature, the space velocity, the concentration of H_2O, the ratio of NH_3/NO_X, and the increase in O_2 concentration. Moreover, it is evident that the reactors with the square shaped cross section have more percentage of conversion but the resident time in the corner increased.

Keywords: $DeNO_x$; SCR reactor; V_2O_5/TiO_2; Optimal design.

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Introduction

The gaseous pollutants and particles elimination in gases at high temperature which is required in the thermal power plants has always been a crucial environmental concern. Nowadays, while the NO_X emission regulation has become more and more strict, much effort has been concentrated on the development of the technology of the efficient NO_X control ($DeNO_X$) [1-3]. Among various methods, the process of selective catalyst reduction (SCR) is one of the techniques widely applied in NO_X elimination from flue gases at about 250-500 ^{0}C. The system of SCR in which urea or NH_3 are serving as a redutant, is often considered as one of the highly potential and effective technologies in accordance with NO_X emission regulation of the euro V and VI standards for the heavy duty diesel engines[4-7].

Many studies have been conducted on numerous metal oxide catalysts, such as MoO_3/TiO_2, V_2O_5/TiO_2 and CuO/Al_2o_3, for NO elimination [8-9]. However, the fact is that in order to avoid the deactivation by SO_2 and H_2O which takes place at low temperatures [10], these catalysts usually function at high temperatures (higher than 350 ^{0}C). Also, Fe zeolite SCR catalysts which are commercially available for stationary applications is shown to operate at temperature up to 600 ^{0}C. Fe zeolite SCR when exposed to high temperatures in the presence of H_2O, may be subjected to the problems of stability. When exposed to the temperatures higher than 600 ^{0}C, in a process flow with high concentration of H_2O, zeolites SCR has the tendency to be deactivated by de-alumination, while the Al^{+3} ions in the SiO_2-Al_2O_3 framework start to migrate out of the structure, the Cu/zsm-5 plays an active role in NO_X reduction within the temperature window of about 200-400 ^{0}C, but its thermal durability is insufficient [11]. It is evident that the V_2O_5 catalyst shows quite high activity for NO elimination at low temperatures, and the presented SO_2 in the flue gas does not deactivate the V_2O_5 catalyst but even can enhance its activity in the absence of H_2O [12]. Therefore, it seems that the V_2O_5 catalyst could be an excellent alternative for SCR of NO with NH_3. The vastly studied support for vanadium is TiO_2 as it is resistance to poisoning effects of sulfur and has the ability to effectively disperse the V_2O_5. But, Titania dose suffer from several limitations, such as it is expensive, sintering non-resistance, and has relatively low surface area [13, 14]. Mixed oxides containing both TiO_2 and Al_2O_3 have also been examined extensively by Centi et al. [15]. It was shown that Al_2O_3 component (with no addition of CuO to the catalyst) attracts the sulfate species which are generated and originally formed at the surface of vanadia, and as a result, this shields the active sites of vanadium. The structure of the surface of vanadyl species present in catalysts shown to have a prominent impact on the activity and the selectivity in the NOx reduction by ammonia.

SCR systems are capable of the selective reduction of NO and NO_2 even in the O_2 abundant environments, which is usually encountered in the exhaust gases of diesel motors. Anyway, such SCR systems adjustment for the practical applications needs to optimize some of the different parameters such as the position of the SCR catalyst, the space velocity, the strategy of the injection of urea. In order to make an assessment on the performance of $DeNO_x$, studies were conducted by Gieshoff et al. [16], in which the parameters such as the $NH3/NO_x$ and the NO_2/NO_x ratios over a V_2O_5 SCR catalys were varied. For the purpose of enhancement in the NO_x conversion efficiency, some studies have been carried out by the use of the common parameters in diesel engine, at different raw NO_x emission levels [17]; for example space velocity, the SCR catalyst temperature, and the oxidation catalyst volume. In addition, several dynamic investigations on the V_2O_5 SCR catalyst have been conducted with the help of a kinetic model, which has a supportive role of an Eley-Rideal (ER) mechanism for the reactions of SCR. Measurements of the kinetics of SCR on vanadium oxide supported on carbon–ceramic catalyst in the absence of both SO_2 and H_2O was performed by Valdes-Solis et al. [18], it was found that it showed the Langmuir–Hinshelwood mechanism. In contrast, Hsu and Teng [19] had made a different conclusion in which the redox mechanism (i.e. the Mars–van Krevelen model) showed a more justified correlation of experimental data than the Langmuir– Hinshelwood mechanism and Eley–Rideal mechanism, the research group [20] studied the kinetics in the absence of H_2O but after a pre-sulfated step, and concluded that rather than the Langmuir–Hinshelwood or redox mechanism, the Eley–Rideal mechanism could be the most appropriate mechanism for description of the SCR of NO with NH_3 over V_2O_5/TiO_2 catalyst. The reaction of SCR takes place in accordance with the following stoichiometry:

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

$$4\,NH_3 + 3\,O_2 \rightarrow 2\,N_2 + 6\,H_2O \quad (2)$$

Here, in a solid-catalytic reaction, the selective reduction of NO_x is made by NH_3, typically in the structures of the low pressure drop honeycomb V_2O_5/TiO_2. It can be seen that the NO is reduced according to (1), and at the same time the amount of a parallel NH_3 oxidation by O_2 (2) must be minimized. Regardless to the type of catalyst, the shape & characteristics of reactors such as the cross section of channels can have a great impact on the amount of the conversion. In addition, the state of cost- effectiveness or feasibility of construction of such a reactor with desirable characters must be noted. In this paper, just the cross section of channels is considered.

2 Experimental database

The experimental information of this model has been obtained from Due-hansen et al. [21]. In their study, they investigated the steady-state performance of vanadium-based honeycomb monolith catalysts in the SCR for elimination of NO with the help of the NH_3. In the current model, some conditions were considered for example the concentration of NO was maintained at 1000ppm and the space velocity was fixed at 8,000h^{-1}. According to the optimized experimental results

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of Due-hansen et al. [21], the inlet concentration of ammonia and the inlet oxygen concentration were set to11000ppm and 3.5% , respectively. Optimum NO_x conversion was obtained at 523 K. table 1 depicts the detailed characteristics of catalyst and its operating conditions which was employed for the study.in which, the concentrations of various components such as NO, NO_2, N_2O, NH_3, H_2O, O_2 and HNO_3 were altered in order to find their influence on the SCR functionality. Here, the properties of NO conversion with respect to temperature were measured.similar to data in Fig 3.

3 Mathematical model for an SCR reactor

In the following, an isothermal pseudo-homogeneous model is developed which results in the differential equations based on unsteady state continuity equations. The SCR reaction system is illustrated in Fig. 1 , where NH_3 is applied as a reducing agent in NO reduction. Before entering to the catalyzed bed, first, NO is diluted with N_2 and O_2 and then mixed with the NH_3 reacting agent. In the following subsections, the reaction kinetics mechanisms and the phenomena of transport in the SCR reactor are explained and formulated.

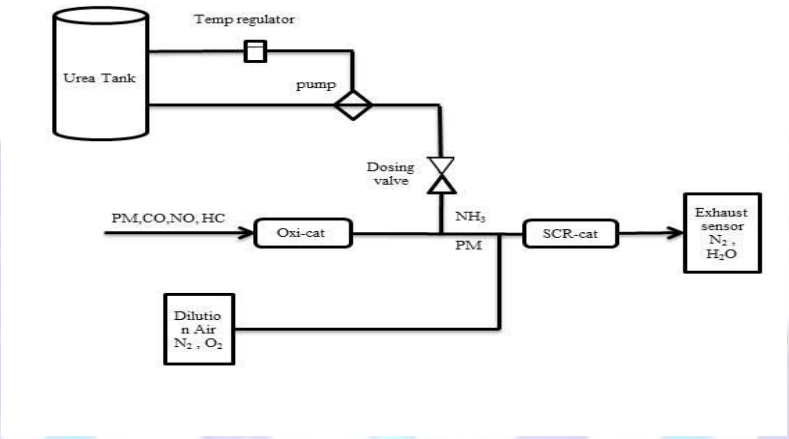

Figure 1 The SCR system for reduction of NO

3.1 kinetics model

The reactions of (1) and (2) describe the mechanism of the NO reduction reaction with the application of NH_3 over catalyst in which the rates of reaction are given by [4]:

$$r_1 = k_1 C_{NO} \frac{aC_{NH_3}}{1 + aC_{NH_3}} \quad (3)$$

$$r_2 = k_2 C_{NH_3} \quad (4)$$

In the above equations, it is assumed that the reaction constants obey the Arrhenius law as follows:

$$k_i = A_i \exp\left(-\frac{E_i}{R_g T}\right) \quad (5)$$

$$a = A_0 \exp\left(-\frac{E_0}{R_g T}\right) \quad (6)$$

In which E_i represents the activated energy (J/mol), R_g indicates the gas constant (J/ mol K) and T is the temperature (K).

3.2 The equations of transport in the SCR reactor

Two main portions comprise the SCR reactor: the portion of free channel and the portion of catalyzed-bed where the reaction takes place. In the followings, the transport phenomena through individual part are discussed:

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The Navier-Stokes equations describe the flow in the free channel:

$$\rho\frac{\partial u}{\partial t}+\rho(u.\nabla)u=\nabla.\left[-P\,I+\eta(\nabla u)+(\nabla u)^{T}-\left(\frac{2\eta}{3}\right)(\nabla.u)I\right] \quad (7)$$

$$\frac{\partial\rho}{\partial t}+\nabla.(\rho u)=0 \quad (8)$$

It worth noting that Eq. (7) describes the slightly compressible behavior due to the gas flow, and Eq. (8) is the equation of continuity Where ρ shows the density (kg/m^3), **u** is the velocity (m/s), η indicates the viscosity (kg/(m· s)), and p denotes the pressure (Pa). In the case of the porous domain, the Brinkman equations explaines the flow [22]:

$$\left(\frac{\rho}{\varepsilon_P}\right)\frac{\partial u}{\partial t}+\left(\frac{\eta}{\kappa}+Q\right)u=\nabla.\left[-P\,I+\left(\frac{1}{\varepsilon_P}\right)\left\{\eta(\nabla u+(\nabla u)^{T})-\left(\frac{2\eta}{3}\right)(\nabla.u)I\right\}\right] \quad (9)$$

$$\frac{\partial(\varepsilon_P\rho)}{\partial t}+\nabla.(\rho u)=0 \quad (10)$$

Where ε_p represents the porosity (dimensionless) and *k* is the permeability (m^2) of the porous medium. As it can be seen in (7) and (9), the momentum-balance equations are tightly related. The term on the left-hand side of the Navier-Stokes equation corresponds to the transported momentum by convection in free flow. In the Brinkman formulism, this term is substituted by a contribution related to the drag force experienced by the fluid as it flows through a porous medium.

Table 1 The physical properties of the SCR reactor and operating condition

Parameters	Value	Units	Name
C_{NO}	1000	ppm	Inlet NO concentration
L	0.36	m	Length of reactor
r	0.1	m	Radius of reactor
r_c	0.8	m	Radius of catalyzed bed
u	0.046	m/s	Inlet velocity
NH_3/NO	1.1	-	Ratio of NH_3 to NO
a_s	60	$m^2\,g^{-1}$	Specific surface area
e_p	0.4	-	porosity
κ	1e-7	m2	permeability
R_g	8.3141	J/molk	Gas constant
P	101.325	pa	pressure
T	523	k	Temperture
GHSV	8000	h^{-1}	Gas hourly space velocity
V_2O_5	3	wt %	V_2O_5 content
F	300	Nml/min	Total flow
m	48000	#	Number of mesh

The boundary conditions include:

$$u.n = v_0 \quad \text{inlet}$$

$$u = 0 \quad \text{walls}$$

$$p = p_{ref} \quad \text{outlet}$$

The viscous stresses are neglected at the outlet, and the pressure is adjusted at 1 atmosphere. It must be reminded that the Brinkman equation is a generalization form of Darcy's law whose aim is to facilitate the boundary conditions matching at the interface between the catalyst larger pores and the permeable medium. The diffusion-convection equations at unsteady state are utilized for the description of the mass-balance equations:

$$\frac{\partial C_i}{\partial t} + \nabla.(-D\nabla C_i + uC_i) = R_i, \quad \text{i=NO, } NH_3, O_2, N_2 \text{ and } H_2O \qquad (11)$$

Where D_i is the representative of diffusion coefficient (m^2/s), c_i indicates the concentration of species, and **u** denotes the velocity vector (m/s). The term R_i is associated to the net rate of reaction for each of the species, which is a function of the reaction rates, (1) and (2), and also the reaction stoichiometry. In the free channel, the inlet conditions are equal to the inlet concentrations; and in the case of the outlet, as the diffusion coefficient and the viscosity of gas are dependent to the temperature and pressure, the convective flux condition is applied, the used corrections are mentioned in continue

$$D = 2.695\times10^{-3}\frac{\sqrt{T^3(M_i+M_o)/(2\times10^3 M_i M_o)}}{P\sigma_i\sigma_o\Omega_D} \qquad (12)$$

$$\eta = 2.699\times10^{-6}\frac{\sqrt{T(1\times10^3 M)}}{\sigma^2\Omega_V} \qquad (13)$$

In which M_i and M_o correspond to the molar mass (kg/mol), and σ_i and σ_o denote the diameters (m) of the gas species. In addition, p indicates the pressure (Pa) and Ω_D and Ω_V are representatives of collision integrals [23].

In Table 1, the nominal operating condition and the physical characteristics of the SCR reacting system are mentioned. In this account, it must be highlighted that the proposed 3-Dimensional dynamic model, which is relatively different from the previous one- and two-dimensional steady-state models of monolith reactors [24], can make it easier to investigate the phenomena of the interior transport via porous catalyst and the spatial concentration distribution in the SCR reactors.

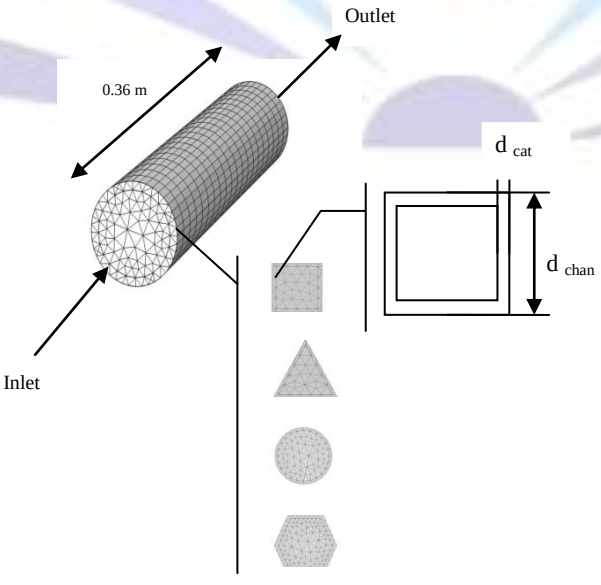

Figure 2 The schematic graf of the SCR reactor with various cross section

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In the resented article, in order to simulate the 3D dynamic behavior of the SCR, finite element software is employed. In the following simulation studies, the fine mesh and Direct (mumps) solver with time step of 0.1 s are utilized and the error control of relative tolerance of 1×10^{-5} and absolute tolerance of 1×10^{-7} are considered. The mesh grid of the formulated 3D model is illustrated in Fig. 2. For the purpose of the PDE model parameters estimation such as activated energies, constants of the reaction rate and frequency constants, an optimization algorithm is introduced. Due to the possibility of the direct operation over real numbers and no need to the encoding and decoding procedures, the use of the real-coded genetic algorithm (RCGA) seems to be easier. Moreover, the RCGA has showed to benefit from the high efficiency and accuracy for optimization of the process. For application of the real-coded genetic algorithm (RCGA) [25] for the SCR model, the objective function was set as $\phi = \sum_{m=1}^{n} (X_{NO_{calc_m}} - X_{NO_{exp_m}})^2$ in which X_{NOcalc} represents the output of the model, and X_{NOexp}, which corresponds to the experimental data, both are adopted from Due-hansen[21], before going through the optimization procedure. With setting the RCGA parameters as N = 200 (population number), P_r = 0.2 (possibility of reproduction) and G = 100 (number of generations).

Fig. 3 and Table 2 depict the results of the fitting procedure. Calculated values of NO conversion show good accordance with the experimental data. The values of E1 and E2 were set in the ranges of published values (57 -65 and 81 -87 kJ /mol, respectively, both in [26-27]). Also the constants of the reaction rate are confined in the range of $0.7\times10^{6} \leq k_1 \leq 1.3\times10^{6}$ ، $6.7\times10^{7} \leq k_2 \leq 9\times10^{7}$.

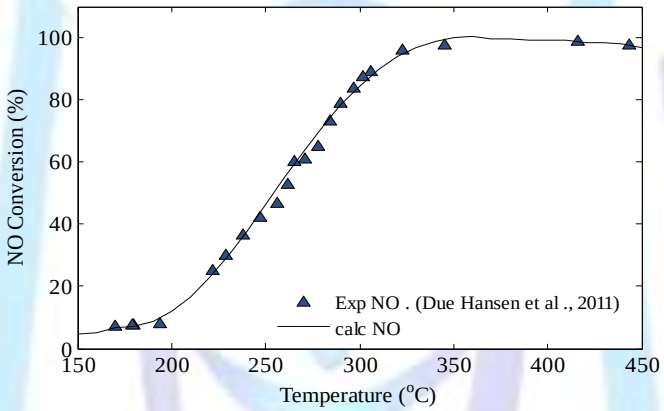

Figure 3 Result of fitting in model validation, experimental data from $3V_2O_5/TiO_2$, NO conversion(%) vs . operating temperature (k), y_{NO}=1000ppm,y_{NH3}=1100ppm,y_{O2}=3.5 vol%,y_{H2O}=2.7 vol%,GHSV=8000 h^{-1}, u=0.046 $m.s^{-1}$

Table 2 Estimated parameter values in rate equations

Kinetic parameter	Estimated value	Unit
k_1	9.97×10^{6}	s^{-1}
k_2	7.68×10^{7}	s^{-1}
E_1	5.76×10^{4}	J/mol
E_2	8.42×10^{4}	J/mol
A_0	2.67×10^{-17}	m^3/mol
E_0	-2.35×10^{5}	J/mol
Φ	2.64×10^{-2}	-

The analysis of the kinetic for the individual SCR rate approaches was carried out for a various exhaust conditions. In continue, a comprehensive analysis is done with the aim of the determination of the effects the various parameters such as the temperature, the space velocities, and the concentration of O_2, H_2O, NO_2, and NH_3, on the efficiency of NO_x conversion over an SCR $DeNO_x$ catalyst of the heavy duty diesel exhaust gases.

3.3 Effect of temperature

Only at specific temperatures, the surface reaction of NO_x reduction is effective. Hence, the simulations were carried out for five exits, in order to find out the specific range at which the reactions show effectiveness. The corresponding

temperature profiles of the temperatures 225°C, 250°C, 275°C, 300°C,& 325°C with the variation in the rate of molar flow and the temperatures are brought. In the case of a composite exhaust gas including 3.5% O_2, 2.7% H_2O, 1000 ppm NO, and 1100 ppm NH_3, Fig 4 illustrates the comparison made between the flow rates of ammonia and nitrogen oxide (NO) along the length reactor. It can be clearly seen from the picture that, with the increase in the temperature of exhaust the reduction of NO shows enhancement. For instance, at a volume of 15 units, the NO levels are10.85e-8 and 3.2e-8 (moles /sec), respectively, for the temperatures of 225 °C and 250 °C. Further increase in temperature does not result in the enhancement of NO reduction. by the analysis of the figure it can be concluded that, at a location of 15 volume units, the NO level is about 2.9 moles/sec. therefore, it can be concluded that in order to have the best reduction of NO with ammonia for the given NO emission level, the optimal temperature of the exhaust gas would be 250°C.

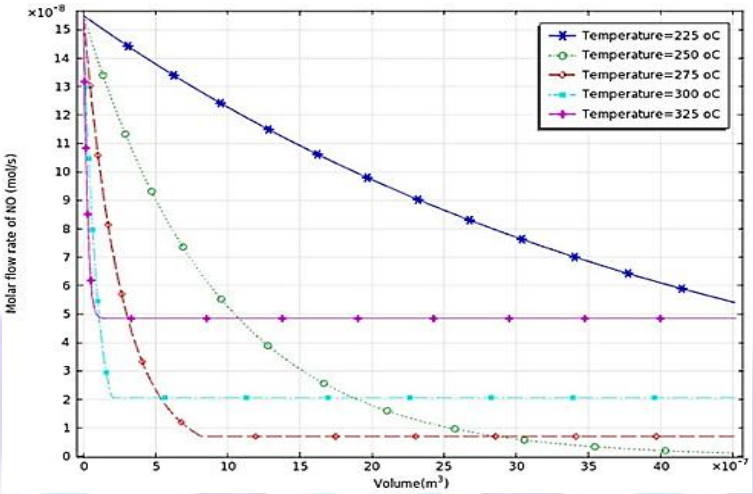

Figure 4 The effect of temperature on molar flow rate of NO (mol.s^{-1}) under different volumes of SCR reactor.

3.4 Effect of space velocity

Before the performance of the optimum design, the influence of GHSV on the efficiency of the SCR reactor in NO elimination was investigated. In the case of heterogeneous fixed-bed catalysis which has a typical use in automobile after treatment, the residence time of the exhaust gases is defined in terms of the volumetric flow rate and the volume of the catalyst. The space velocity is equal to the volumetric gas flow rate at STP divided by the total volume of the catalyst, which is shown in continue:

$$GHSV = V_{exhaust} / V_{catalyst} \quad (14)$$

Here, GHSV is the abbreviation of the gas hourly space velocity in [1/h], $V_{exhaust}$ corresponds to the volumetric flow rate of the exhaust gas in [m^3/h], and $V_{catalyst}$ indicates the catalyst's volume in [m^3]. Since by reduction of the space velocity, the residence time of the exhaust gas increases, this leads to enhancement of the performance of the SCR $DeNO_x$, but it may also result in various problems in the process of the vehicle installation. Also, since this method increased the volume of the catalyst, it may increase costs of production. The space velocity impact on the conversion of NO_x for the composite exhaust gas which contains 3.5% O_2, 2.7% H_2O, 1000 ppm NO, and 1100 ppm NH_3 is depicted in Fig.5. As it can be clearly seen in Fig, 5, the SCR $DeNO_x$ performance shows a strong dependence on the space velocity. Especially, the light off temperature (LOT), in which the SCR $DeNO_x$ performance reaches to 55% conversion rate, shows an enhancement from 225 to 300 °C with the increase in the space velocity from 5,000 to 25,000 [1/h]. Moreover, it must be noted that at temperatures higher than 430 °C, the oxidizing property of NH_3 is more effective, and as a result, the SCR $DeNO_x$ performance slightly decreases. It is also observable that by reduction of the space velocities, NO_x conversions increase. This is due to the fact that by increase in the space velocity, the residence time of the exhaust gas on the surface of the catalyst decreases [28].

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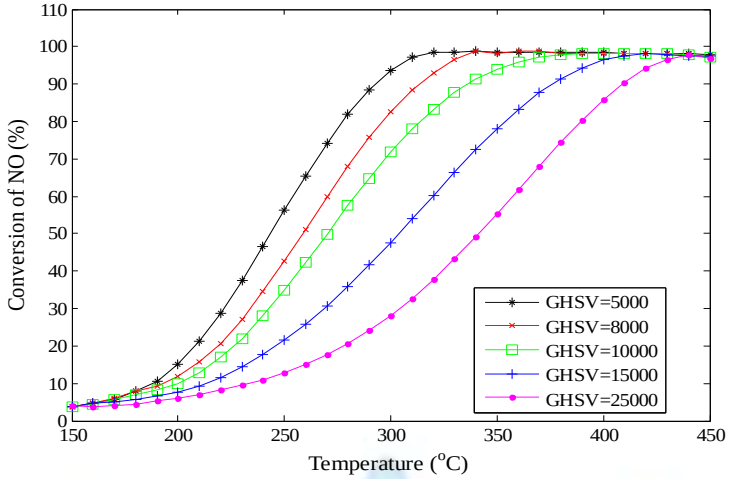

Figure 5 The effect of space velocity on the NO conversion(%) under different temperature.

3.5 Effect of the NH_3/NO_x ratio

The emitted NH_3 in the ambient air has the ability to react with sulfuric acid and nitric acid and form the salts of ammonium sulfate and ammonium nitrate, these compounds are important components of undesirable particulate matter [29]; also, they have an unpleasant odor. so, the amount of emitted NH_3 normally is required to be less than 5-10 ppm. Fig. 6 shows the influence of NH_3/NO ratio on the NO reduction performance of the SCR reactor. In this study, the NH_3/NOx ratio is varied from 0.5 to 1.3. In real vehicles, it is supposed that the test exhaust gas includes 2.7% H2O and 3.5% O2 and has a space velocity of 8,000 [1/h]. In Fig. 6 the impact of the NH_3/NO_x ratio on the NOx conversion when there is no NO_2 (i.e., only the standard SCR reaction is taken into consideration) is illustrated. When the ratio of NH_3/NO_x is equal to 0.5, since the reductant NH_3 is not enough for conversion of the exhaust NO into N_2 through the standard reaction, the maximum NO_x conversion would be 45%, (Eq. (2)). By the increase of NH_3/NO_x ratio from 0.8 to 1.3, the conversion rate of NO_x into N_2 reaches to 98%. However, when the operation temperature goes below 240 K the larger ratio of NH_3/ NO leads to the more significant NH_3 slip phenomenon this is due to the presence of excess NH_3.

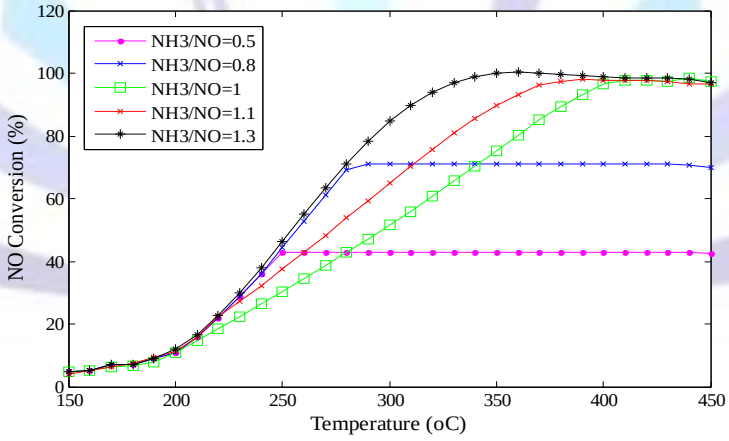

Figure 6 The effect of NH3/NO ratio on NO conversion(%) under different temperature

3.6 Effect of H_2O concentration

It is evident that the presence of H_2O leads to inhabitation of some of the reactions in the process of SCR [30]. Despite the fact that the effect of H_2O is not directly associated to the SCR $DeNO_x$ performance, the released heat of the exothermic process of adsorption, and as a result the absorbed heat in the desorption phase, may have a significant impact on the SCR catalyst. The NO_x conversion is studied at different temperatures and concentrations of H_2O in order to determine the influence of H_2O on the SCR performance. The results are shown in Fig. 4. For such simulation involves the assumption that the exhaust gas which includes 3.5% O_2, 1000 ppm NO, and 1100 ppm NH_3 was fed into the SCR converter with the

space velocity of 8,000 [1/h]. It is observable that the NO_x conversion decreases with the increase in the concentration of H_2O. In the case with the exhaust gas temperature of 250 ^{0}C, the NO_x conversion yields are 48, 26, and 17% for the H_2O concentrations of 1, 5, and 10%, respectively, at the same time the values of LOT are respectively, 240, 270, and 285 ^{0}C for 1, 5, and 10% concentrations of H2O.

This hindering behavior of H_2O could be due to the competition tendency of H_2O for taking part in the reaction with NH_3 on the reaction sites of the catalyst [31].

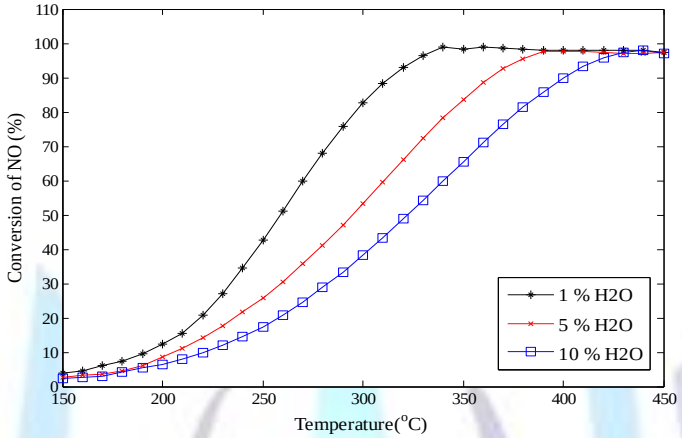

Figure 7 The effect of H_2O on NO conversion (%) under different temperature.

3.7 Effect of O_2 concentration

Exhaust gases emitted from diesel engines, generally contain O_2, whose volume is ranging from 2 to 17%, this amount of oxygen does not react with the fuel in the chamber of combustion. The results of NO_x conversion efficiency at different O_2 concentrations ranging from 0.08 to 10% with the other exhaust gas conditions of 2.7% H_2O, 1000 ppm NO, 1100 ppm NH_3, and a space velocity of 8,000 [h^{-1}] are illustrated in Fig. 8. It can be easily observed that, the NO_x conversion yield increases by temperature and approximately reaches to 98% in the temperature range of 340 and 430 ^{0}C, above this temperature, due to the oxidizing characteristics of NH_3, the efficiency shows a slight reduction. Anyway, remember that, in the temperature range mentioned here, the concentrations of O_2 greater than 5% have no profound influence on the NO_x conversion. Hence, in actual diesel exhaust gases, we can conclude that the O_2 concentration impact is not regarded as an crucial factor in the standard SCR reaction expressed in Eq. (1). But, in cases with the O_2 concentrations lower than 5%, the NO_x conversion has shown a dramatic decrease since the standard SCR reaction can't succeed in the process of NO conversion [21].

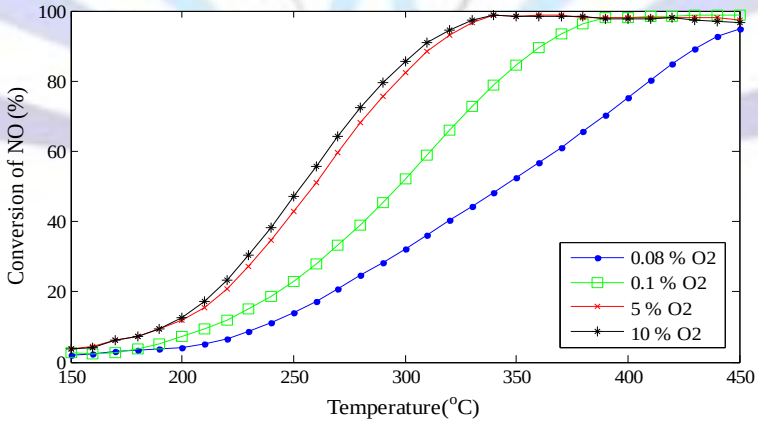

Figure 8 The effect of O2 on NO conversion (%) under different temperature.

3.8 The effect of cross section

After the design of the reaction mechanism and the assessment of data and examining their accordance with the experimental model, here, we discuss about the physic and the geometry of the model. First, the geometry of the SCR reactor system is designed in 3D space and then the geometrical shapes are investigated on the basis of the same geometrical coordinates defined for the part of reaction engineering software. In order to identify the desirable type of the reactor cross section, first of all, the four commonly used cross sections in this industry (including circle, hexagonal

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(honeycomb) triangular and squares) are applied. The data related to the reactor stimulation and the information of each individual parts of the reactor are listed in table 1. After the application of the boundary condition of the problem and the dominant transport equations, it's the time to define the mesh of the geometry of the problem, whose related information is recorded in table 3. A scheme of a meshed reactor with a circular cross section is depicted in Fig. 2. Also, in Fig 9 the concentration profiles with different cross sections are shown. The results of the 3D model with the same size of the catalyst and equal cross sections shows a good accordance for one channel. It can be seen that the reactor with a square cross section has a greater percentage of NO conversion but the time of residence increases in the corners. The results of the comparison among the different cross sections are tabulated in table 3.

Table 3 effect of different cross section on concentration of NO (outlet)

Cross Section	Ratio Of Area	Concentration Of NO
Circle	1	3.97×10^{-4}
Square	1.127	2.56×10^{-4}
Hexagonal	1.06	5.83×10^{-4}
Triangle	1.288	5.56×10^{-4}

Now that we know that the reactor with a square cross section is more desirable for more reduction of NO, therefore we start to stimulate the reactor with a square shape channels. A picture of such a porous reactor is depicted in Fig 11. After meshing of the model, we discuss about the results of the stimulated reactor. In the below figure, the distribution of the nitrogen oxide in the reactor volume is illustrated. It can be observed that, in the beginning of the reactor, the reductive conversion of NO is much faster than its end. This could be due to the decrease in concentration gradient. Finally, with the help of the SCR reactor with square shape cross section, the achievement to the conversion percentages higher than 95.3% is possible.

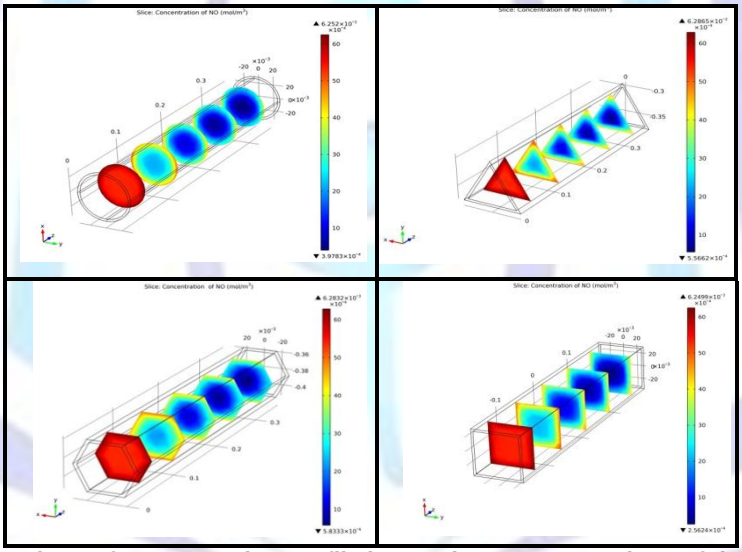

Figure 9 comparison of concentration profile in a various cross sections of the SCR reactor.

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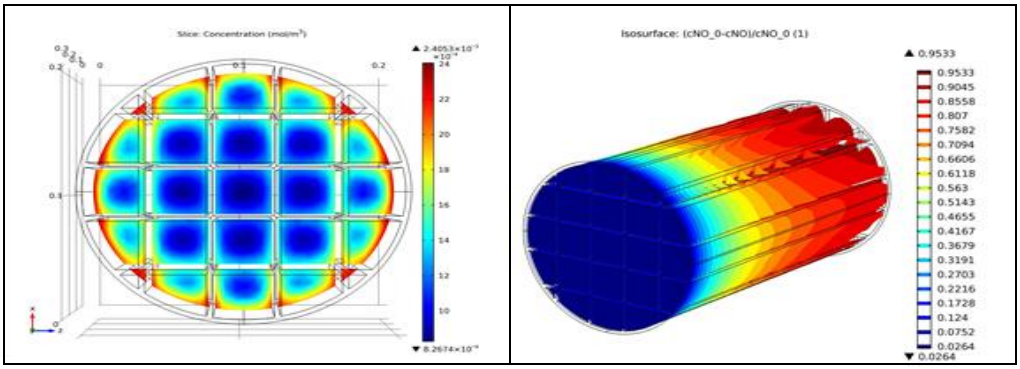

Figure 10 3D concentration plot for NO in the SCR reactor with square channel(right), at the middling slice section of the SCR reactor(left).

5 Conclusion

In order to examine the characteristics of the SCR $DeNO_x$ performance, in the presented study, a computational investigation has been made by varying the parameters such as temperature, space velocity and the O_2, H_2O, NO_2, and NH_3 concentrations and also the cross section. After the introduction of the SCR model with kinetic parameters by RCGA, with the aim of the development of SCR catalysts in Diesel exhaust environments, a numerical investigation is performed on the extensive range of parameters. The possible conclusions which can be deduced from this study are as follows:

1) In order to make the dynamic simulation of the SCR reactor for NO reduction, the 3D mathematical model capable of describing different phenomenon such as the chemical reactions, the compressible flow of gas, the temperature-dependent viscosity and the diffusion coefficient, and the porous flow inside the catalyzed bed is introduced. The mentioned 3D dynamic model is verified and it is confirmed to be in good accordance with the experimental data. Also, for optimum search and a neural network as the decision support, a real-coded genetic algorithm model has been proposed for process optimization. It was shown that this algorithm is effective in estimation of parameters and design of the SCR reactor described by PDE model in its optimal manner. As a result, this model could be useful in enhancement of the entire examination of the effects of the key factors (such as the GHSV, operating temperature, activity energy, constant of reactions and the ratio NH_3/NO) on the function of NO reduction.

2) The obtained results of the stimulation indicate that the NO conversion into N2 is profound only when the reaction takes place in the middle range of temperature i.e. about 523k. Moreover this conclusion can be made that at the temperature of 250^0C the catalytic conversion shows high effectiveness and the converter succeeded in reduction of the pollutants to a very minimum level.

3) Since the residence time of the exhaust gas on the surface of the catalyst would be decreased, the increase in the space velocity leads to the decrease in the performance of SCR $DeNO_x$. Also, at temperature higher than $440\ ^0$C, the NO_x conversion decreases as the oxidizing characteristics of the SCR reactions would be profound.

4) The concentrations of O_2 more than 5% does not have any dramatic influence on the conversion of NO_X, anyway, in the situations with O_2 concentrations lower than 5%, NO_X conversion dramatically decreases.

5) With the increase in the H_2O concentration, the SCR DeNOx performance decreases. This inhibition characteristic of H_2O has been explained to be the result of the competition properties of H_2O with NH_3 on the sites of the V_2O_5 catalyst.

6) As the ratio of NH_3/NO_x increases from 0.08 to 1.1, the performance of SCR $DeNO_x$ improves because the fast SCR reaction also takes place in the reaction of NO_x conversion. However, due to the NH_3 inhabitation behavior and oxidation effects, as the ratio of NH_3/NO_x exceeds the value of 1, the NO_x conversion decreases.

7) Among the various investigated cross sections for SCR reactor for reduction of NO, the square shape cross section possesses the higher degree of conversion, about 97.7%, however, the residence time on the corners increases.

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NOMENCLATURE

A_i	frequency constants,s^{-1}
a	reaction constant,$m^3.mol^{-1}$
c_i	concentration,mol/m^3
D	diffusion coefficient,m^2/s
d_{cat}	thickness of catalyst layer,mm
d_{chan}	width of channel,mm
E_i	activated constants,$j.mol^{-1}$
GHSV	gas hourly space velocity, h^{-1}
k_i	reaction constants, s^{-1}
M_i	molar mass,$kg.mol^{-1}$
N	number of population
p	pressure,pa
R_i	net reaction rate
R_g	gas constant,$j.mol^{-1}k^{-1}$
r_i	reaction rates,$mol.m^{-3}.s^{-1}$
T	temperature,k or ^oc
u	inlet velocity, $m{\cdot}s^{-1}$
V	volume of reactor,m^3
$X_{NO_{calc}m}$	model predicted
$X_{NO_{exp}m}$	experimental data
ε_p	porosity of catalyst
ρ	density,$kg.m^3$
η	viscosity,$m^2.s^{-1}$
κ	permeability,m^2
σ_i	diameter of gas species,m
ϕ	error function
Ω_D	coliision integral
Ω_v	coliision integral

Subscripts

SCR	selective catalytic reactor
g	gas
LOT	light off temperature

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