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Mathematical and Artificial Neural Network Simulation of NOx Selective Catalytic Reduction in a Monolithic Reactor

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ABSTRACT: Worldwide development of industries and increase of energy consumption, have resulted fast increase in the emission of nitrogen oxides pollutants. Therefore, removal of nitrogen oxides is a very important issue. In this study, modeling and simulation of selective catalytic reduction of nitrogen oxides in a monolithic catalytic reactor at both steady-state and dynamic-state was performed. Steady-state results showed that because of intense effect of temperature on nitrogen oxides conversion and competition of the main reaction with ammonia oxidation reaction, conversion of nitrogen oxides requires a catalytic filter in the range 300-350°C. Results showed that nitrogen oxide conversion increases with decreasing gas hourly space velocity and increasing inlet nitrogen oxide concentration. At dynamic-state, the effect of changes in some parameters including gas hourly space velocity, inlet nitrogen oxide concentration, and ammonia /nitrogen oxide ratio were investigated. Also, steady-state simulation of the process was performed using an artificial neural network and conversions of nitrogen oxides and ammonia were estimated as a function of gas hourly space velocity, reactor temperature, and nitrogen oxide concentration. 96 networks with different neurons and two different activation functions in hidden layer were trained. The resulted optimum network showed maximum mean square error of about 0.01 compared to mathematical modeling results indicating high performance of neural network for prediction of process performance.

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1. Introduction

Removal of Nitrogen Oxides (NOx) pollutants from high-temperature gases is necessary for thermal power plants, waste incinerators, and internal combustion engines. They are usually removed by catalytic NOx processes in electrostatic precipitators or in the filters. In some systems, NOx is reduced selectively using ammonia which is usually performed at low pressure [1]. In gas treatment, catalytic filters are manufactured for exhaust-emission pollutants, either for fossil-fuel power plants [2, 3], or for biomasses and combustion wastes [4]. When ceramic filter materials combine with intermediates, they exhibit high yields for the separation of particulate pollutants. They also have high catalytic activity to remove NOx, CO, and CHx. The filter must remove dust and soot from the exhaust and at the same time must have an active catalyst for the process of converting gaseous pollutants passing through the internal filter structure [5]. The reduction of NOx occurs according to the general reaction (Eq. (1)), and simultaneously reaction (Eq. (2)) occurs as a result of oxidation of Ammonia (NH_3) with oxygen [1].

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

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

NO_2 formation is largely prevented because of the presence of water in the exhaust gas [6].

Tronconi [7], investigated experimentally interaction between chemical reaction kinetics and transport phenomena within monolithic reactors. The reaction of NOx was selected as benchmark and Rideal-type rate equations were assessed and their parameters were obtained. Also different geometries of honey-comb catalysts were studied. Lei et al. [8] performed modeling and simulation of Selective Catalytic Reduction (SCR) of NO in a honey-comb monolithic reactor using Computational Fluid Dynamics (CFD) method. They also used Rideal-type reaction rate equations for developing mathematical models. Their results showed that low inlet gas rate, high inlet gas temperature and high NH_3/NO mole ratio increase NO conversion. Sharifian et al. [9] presented a One Dimensional (1D) dynamic model for simulation of NOx SCR on iron/zeolite catalyst within a channel equivalent to a diesel motor.

Faghihi and Shamekhi [10], developed a feed-forward Artificial Neural Network (ANN) model for simulation of

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NOx SCR process in order to optimize the ammonia inlet flow rate. They considered three input variables of temperature, and inlet Nitrogen Oxide (NO) and ammonia concentrations. Network outputs included NO conversion and NH_3 outlet. They used 18 steady-state data series for training ANN, but did not find the optimum number of neurons in hidden layer and also didn't investigate the effect of Gas Hourly Space Velocity (GHSV) on NO conversion. Izadkhah and Farzi [11], studied mathematical and ANN simulation of ethanol pyrolysis in a tubular reactor at steady-state. An optimal network with 10 neurons in hidden layer was obtained, which had very small error with respect to the data of the mathematical simulation. In this study, mathematical modeling and simulation of the selective catalytic reduction of NOx using ammonia is carried out in a tubular and monolithic plug-flow reactor in both steady-state and dynamic-state. For this purpose, component mass balance equations were written for both conditions. To validate the model, the empirical results of the work of Schaub et al. [1] were used. Dynamic modeling of process is a novelty of this research which has seldom been found in previous works. The ANN trained and used in the present work has an additional input of GHSV investigated by Faghihi and Shamekhi [10].

2. Methodology

For modeling of NOx SCR, a monolithic reactor containing honey-comb vanadia/titania filter catalyst was considered. Specifications of the catalytic filter are presented in Ref. [1]. A pseudo-homogeneous isothermal model was obtained at steady-state using component mass balance equations. Modeling assumptions include steady-state, isothermal plug-flow reactor, one-dimensional concentration changes, and ideal gas law. The resulting ordinary differential equations were solved numerically using Runge-Kutta method. The effect of different parameters including GHSV and NO inlet concentration at fixed NH_3/NO mole ratio was assessed and their optimal values were determined.

At the next step, dynamic modeling of the process was performed and Partial Differential Equations (PDEs) were obtained for concentration changes of NO and NH_3 assuming 1D isothermal plug-flow reactor. Steady-state results were used as initial conditions. The method of lines [12] was used for the numerical solution of PDEs. The effects of disturbances on different parameters such as GHSV, NO inlet concentration, and NH_3/NO mole ratio on NO conversion were investigated. Finally, simulation of the process was done by training and then using a feed-forward ANN [11], based on the results of steady-state simulation with the mathematical model. In this work, NO and NH_3 conversion were estimated by the network as a function of GHSV, inlet temperature, and NO inlet concentration. Modeling data were divided into three sets of training (60%), validation (15%), and test (15%) and the ANN was trained using Levenberg-Marquardt back-propagation algorithm. Different topologies of ANN were examined and the optimum activation functions in hidden and output layers and also optimum number of neurons in hidden layer were obtained.

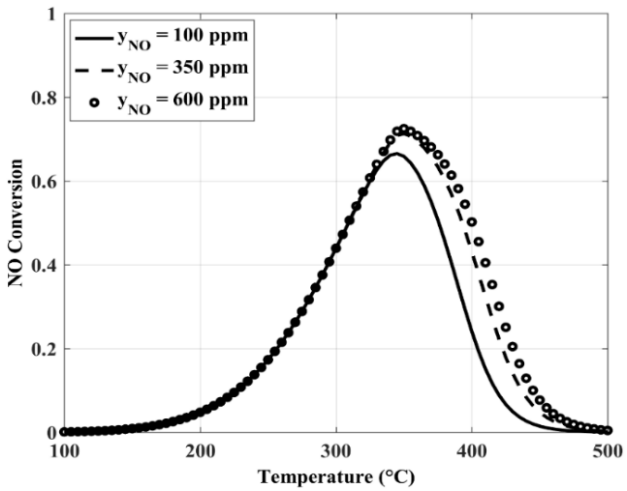

Fig. 1. Profiles of NO conversion at different temperatures and NO inlet concentrations, GHSV=12000 h^{-1}, $y_{NO} = y_{NH_3}$

3. Results and Discussion

For validation of the mathematical model, simulations were performed at steady-state for GHSV of 11000 h^{-1}, NO inlet concentration of 350 ppm and NH_3/NO mole ratio of 1 [1]. Maximum error was about 8% which is acceptable considering assumptions made for process modeling.

The effect of NO inlet concentration and temperature at fixed GHSV of 12000 h^{-1} and NH_3/NO mole ratio of 1 on the conversion of NO is shown in Fig. 1. As can be seen, it increases with increase of NO inlet concentration. It is concluded that the optimum temperature for maximum NO conversion is 330°C for different NO concentrations.

For the assessment of the effect of GHSV on the transient response of the catalytic reactor, it was changed from 16000 to 8000 h^{-1} at a constant temperature of 330°C. Inlet gas contained 7.6% O_2, and 350 ppm NO and NH_3. As it is evident from Fig. 2, NO and NH_3 conversions are increased and reached new steady-state values after 0.2s which is due to

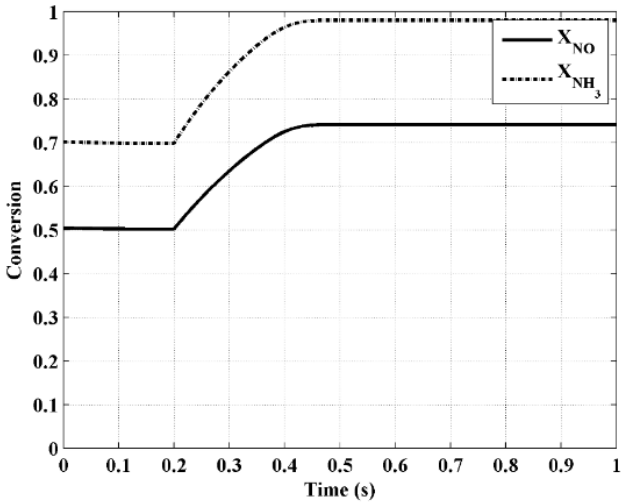

Fig. 2. Response of NO and ammonia conversion to a step-change on GHSV from 16000 to 8000 h^{-1}, $y_{O_2} = 7.6\%$, $y_{NO} = y_{NH_3} = 350\,\text{ppm}$ and T=330 °C

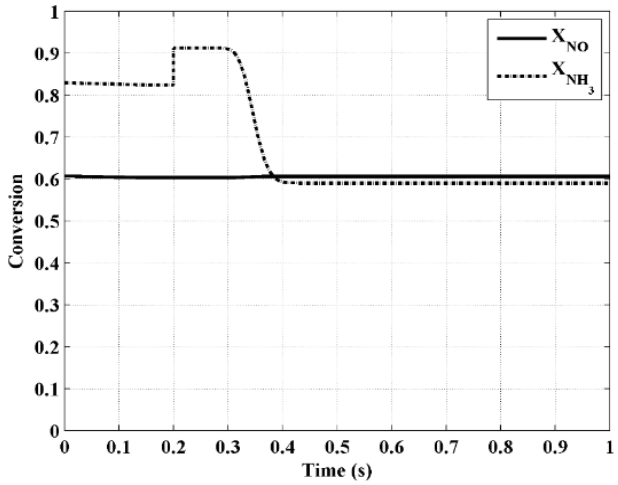

Fig. 3. The response of NO and ammonia conversion to a step-change on NH3/NO mole ratio from 1 to 2, GHSV=12000 h^{-1}, $y_{NO} = y_{NH_3} = 350$ ppm , $y_{O_2} = 7.6\%$, T=330 °C

the increasing the residence time of the reactants.

Also, NH_3/NO mole ratio was changed from 1 to 2 with GHSV=12000 h^{-1}, 350 ppm NO and 7.6% O_2, and reactor temperature of 330°C whose response is shown in Fig. 3. As shown, NO conversion has increased slightly, but the conversion of NH_3 firstly increased and then decreased significantly. It is due to the lower concentration of NH_3 compared to NO when its concentration increased which is then decreased because of increasing reaction rates. Thus, the optimum value for this parameter is 1 which is also confirmed by Lei et al. [8].

For modeling with ANN, 2087 data series generated by steady-state mathematical simulation of the process were utilized for training, validation, and test of ANN as described before. It was found that the network with tangent hyperbolic

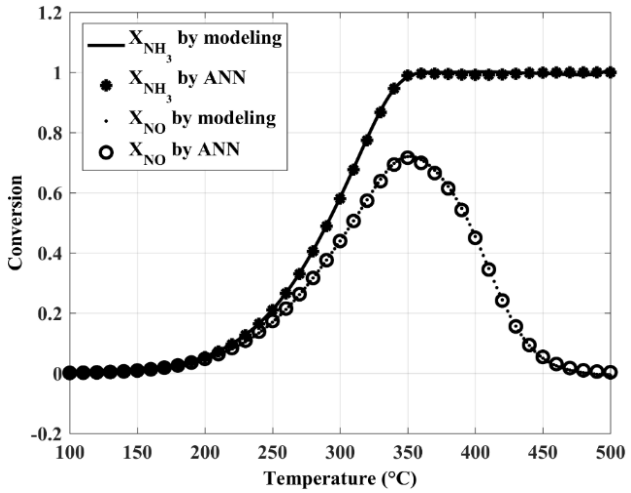

Fig. 4. Comparison of NO and ammonia conversion values resulted from the mathematical model with outputs of the optimal ANN using the same inputs, GHSV=11000 h^{-1}, $y_{NO} = y_{NH_3} = 350$ ppm , $y_{O_2} = 7.6\%$

and linear activation functions for hidden and output layers, respectively, generated results with minimum deviation from mathematical modeling (Fig. 4). Also, the optimum number of neurons in the hidden layer was obtained as 16. The advantage of using trained ANN instead of mathematical model is that it does not need numerical solutions of differential equations and so generates outputs more rapidly.

4. Conclusion

Based on the mathematical modeling and simulation at steady-state, it can be concluded that selective catalytic reduction of NO with NH3 in a catalytic filter must be performed in the temperature range of 300-350°C. Also, the simultaneous increase of NO and NH3 inlet concentrations or reduction of GHSV results in increasing of NO conversion. At dynamic-state, response of NO conversion to a step change on GHSV showed the secend-order trajectory and thus a proportional-integral controller could be used for process control. Response to a step change on simultaneous change of NO and NH3 inlet concentrations was auto-tuning which is due to the constant value of NH3/NO mole ratio. Finally, modeling of the system with a feed-forward ANN showed that the optimum trained network predicted outputs of mathematical simulation with maximum error of less than 0.02 which shows high potential of ANNs for prediction of theoretical or empirical results and can be used instead of mathematical model.

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