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Effective Mixer Design an Important Factor In SSCR Systems for Reduction of NOx from Exhaust of Diesel Engines

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ABSTRACT

In recent decades, the environment has been seriously polluted by the hazardous exhaust components of diesel engines. The international community, which is dedicated to preserving the harmony between nature and humanity, has taken this seriously and imposed strict regulations on Diesel engine manufacturers regarding the quantity of exhaust components from Diesel engines that may apply to the standards of EURO-VI. The SCR technology attempted to reduce the problem somewhat, but the associated problems of solid particle formation on the pipe walls, ammonia slip, and incomplete NOx reduction led to the development of new technology - solid selective catalytic reduction. The use of solid ammonium salt for ammonia generation has shown better results in NOx reduction and reduction of solid particle formation compared to SCR. However, it was not possible to fully resolve the ammonia slip issue. A uniform flow rate of ammonia through the SCR catalyst can reduce NOx efficiently. In this paper, the role of mixer design in achieving a uniform flow rate of ammonia is investigated in detail. The results show that an optimized mixer design leads to efficient reduction of NOx and thus reduces ammonia slip to a great extent. When the mixer is placed near the ammonia injection point, the most homogeneous ammonia distribution is achieved for flow through the SCR catalyst.
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INTRODUCTION

The rapidly increasing demand for vehicles has badly affected the environment. Exhaust emissions of diesel engines contain extremely poisonous unburned hydrocarbons, carbon dioxide, carbon monoxide, particulate matter, sulfur dioxide, and oxides of nitrogen. Society is struggling with the adverse effects of exhaust emission components of diesel engines (Erickson et al. 2020). The removal of nitrogen oxides from the exhaust is the most difficult task.

Fig. 1 shows the exact distribution of the exhaust gas components of diesel engines which contain nitrogen, oxygen, carbon dioxide, carbon monoxide, particulate matter, unburnt hydrocarbons, and oxides of nitrogen. Due to the harmful effects of nitrogen oxides on human health, it is proposed in EURO-VI to limit NOx emissions to 0.4 $kg.KWh^{-1}$ in a steady state and 0.46 $kg.kWh^{-1}$ in the transient state (Williams & Minjares 2016).

Exhaust gas recirculation (EGR), Lean NOx trap (LNT), and selective catalytic reduction (SCR) are some of the technologies previously adopted by engine manufacturers (Praveena & Martin 2018) but could not increase the reduction efficiency by more than 75%. Selective catalytic reaction (SCR) can reduce upto 90% of NOx with a suitable filter design and catalyst. But ammonia slip, freezing of diesel exhaust fluid (DEF) at low temperature, and nozzle clogging due to cyanuric acid formation reduce the NOx reduction performance of SCR. Latha et al. (2019) reviewed various after-treatment technologies for NOx reduction and found SCR to be the most promising technology for meeting EURO-VI emission standards. Major problems observed with SCR were ammonia slip and solid deposit formation. The authors investigated that catalyst deactivation can reduce ammonia slip by placing an oxidation catalyst after SCR (Latha et al. 2019).

CONSTRUCTION OF SCR

The use of appropriate filters with suitable catalysts can improve the NOx reduction performance of catalytic converters without affecting fuel consumption and engine power (Zhang et al. 2018). The use of filters reduces the particle size of soot and at the same time reduces more than 90% of fine dust particles (Mamakos et al. 2013).

Correct positioning of various filters is an important factor in achieving the optimum amount of ammonia for NOx

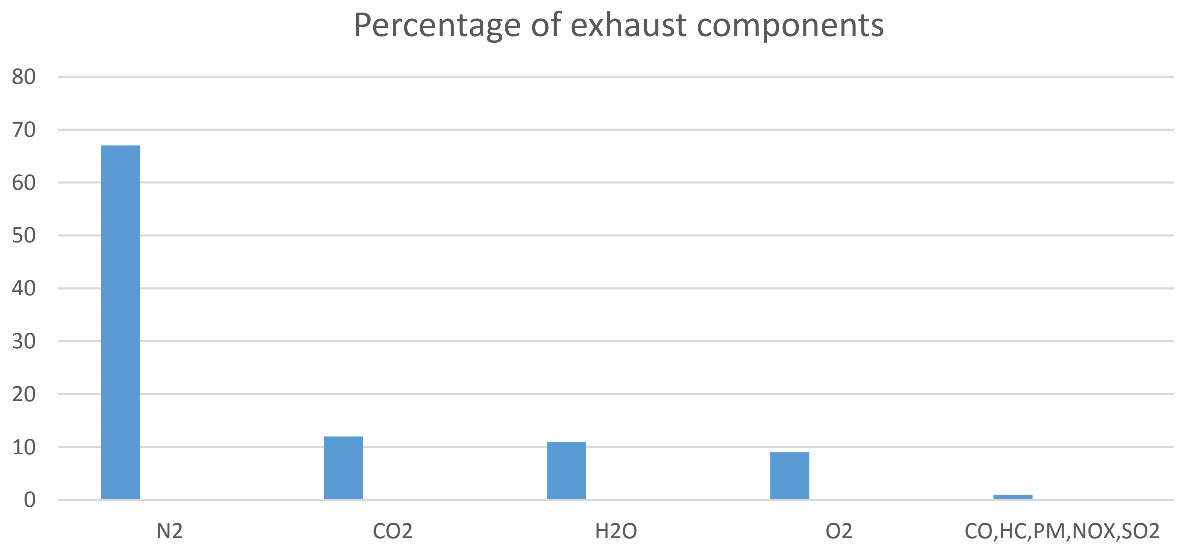

Fig. 1: Diesel engine exhaust composition (Reşitoļu et al. 2015).

reduction. DOC helps in the oxidation of hydrocarbons and carbon monoxide thus reducing their toxicity. DPF reduces the size of soot particles and periodic active regeneration burns the carbon particles from the filter thus making the exhaust carbon-free. SCR reduces the oxides of nitrogen into nitrogen in presence of a suitable catalyst.

DIESEL OXIDATION CATALYST (DOC)

17% unused oxygen from diesel engine exhaust can be used to oxidize hydrocarbons and carbon monoxide. These reactions take place in the presence of a catalyst (DOC) which increases the rate of reaction. DOC is placed before the DPF to take advantage of the temperature rise in DOC (Kurien et al. 2018). It has been studied that for every 1% oxidation of carbon monoxide, the exhaust gas temperature increases by 90°C, which supports the combustion of carbon particles in the DPF (Reşitoılu et al. 2015). DOC is a honeycomb structure of a platinum or palladium substrate surrounded by a catalyst such as V_2O_5 or TiO_2. It helps in the passive regeneration of soot particles in DPF (Wang et al. 2012). The production of NO_2 during oxidation leads to increased efficiency of SCR.

DIESEL PARTICULATE FILTER (DPF)

DPF uses a ceramic substrate in the form of a honeycomb structure. Its purpose is to trap carbon particles and burn them periodically to prevent their emission into the atmosphere. The soot remains in the filter until active or passive regeneration occurs. Passive regeneration occurs at 275°C to 360°C and active regeneration occurs at 600°C. Active regeneration takes place when the filter is clogged. Fresh fuel is allowed to inject into the exhaust pipe to increase the

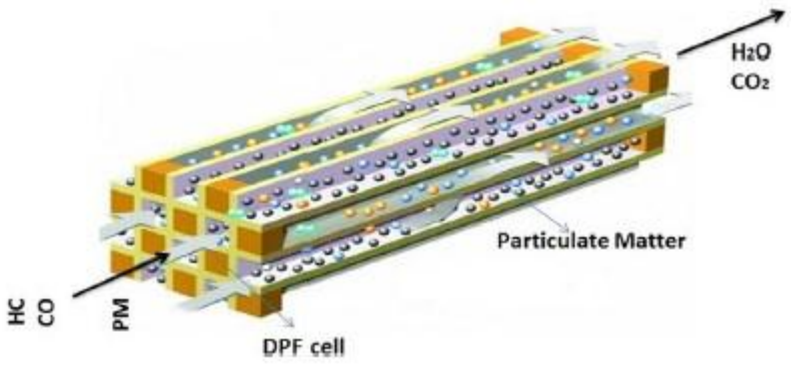

Fig. 2: Trapping of carbon particles in DPF (Mohankumar & Senthilkumar 2017a).

temperature to oxidize the accumulated soot**.** Plasma-assisted burner type DPF ensures the reduced particle size of soot (Wang et al. 2012)

Fig. 2 shows a schematic diagram of a DPF filter. It contains four openings. Exhaust gases from DOC enter through two openings, flow through the cells where carbon particles are trapped and are then discharged through opposite openings.

Vehicles with catalytic DPF and urea SCR reduced particulates with 99.9% and 90% efficiency, respectively. The Plasma assisted burner type DPF effectively reduces particulate matter over the entire size range. Lee et al. (2015) experimentally calculated the particle number size distribution for diesel engine exhaust gases with the FMPS rapid particle size measuring device TSI 3091. They concluded that an effective particle number size can be achieved with the help of plasma-assisted burner type DPF. Mohankumar & Senthilkumar (2017a) reviewed the various technologies for removing particulate matter. He concluded that the use of a suitable regeneration method, an electrostatic precipitator, a particulate filter and a multi-hole nozzle with high injection pressure can greatly reduce particulate matter (Mohankumar & Senthilkumar 2017b). Martinovic et al. (2020) mixed the soot oxidation catalyst with the SSCR catalyst and investigated that the soot temperature can be reduced by 150°C with a significant increase in NOx reduction efficiency of the SCR catalyst. It could happen due to the optimized mass ratio of the catalyst resulting in a rapid SCR reaction (Martinovic et al. 2020).

DIESEL EXHAUST FLUID (DEF)

Because of its unpleasant odor, aqueous urea is used instead of ammonia, which, after decomposing to ammonia, reacts with NOx in the presence of the catalyst SCR and reduces a considerable amount of NOx. This aqueous urea solution, containing 32.5% urea and 67.5% deionized water, is called DEF. Spraying of DEF in the combustion chamber reduces the risk of deposits due to increased surface area, better mixing, and evaporation (Hasan Shahariar & Lim 2018). The turbulence and bounce created by the high wall temperature increase the mixing length, which helps to reduce the deposition of urea from the exhaust pipe (Hasan Shahariar et al. 2019). The aqueous urea is injected into the SCR tank using a nozzle. But the problem is that the tank has a low ammonia capacity (Qu et al. 2014a). Freezing of DEF at temperatures below -11°C is another big issue, which requires an integrated electric heater in the tank of DEF (Kurien & Srivastava 2018). DEF produces cyanuric acid and some other acids that lead to blockage of the urea nozzle, reduce the activity of the catalyst and thus reduce NOx efficiency (Zhang et al. 20116). Chan et al. (2015) investigated a predictive approach by a nonlinear model to efficiently control injection timing and DEF dosing rate. A model with an NMPC approach for SCR control was simulated and the results showed a reduction in ammonia slip and an increase in NOx reduction efficiency (Chen & Wang 2015).

SELECTIVE CATALYTIC REACTION (SCR)

It is a honeycomb structure of porous ceramic materials surrounded by catalytic components such as V_2O_5 or TiO_2. Here NOx combines with ammonia and is reduced to nitrogen. Zeolites of copper and iron are widely used catalysts in SCR out of which copper zeolite gives better results in NOx reduction due to its higher ammonia storage capacity, thermal stability, and reduced ammonia slip (Guan et al. 2014). Manganese-based catalysts are used in some applications, but they have lower NOx reduction efficiency at low temperatures (Li et al. 2017). NOx reduction efficiency acts as a performance indicator of SCR.

NOx conversion = [1 – (NOx out/ NOx in)] × 100% (Peng et al. 2015)

Ammonia slip occurs when either too much ammonia is produced that cannot react with the catalyst due to low temperature or when the catalyst has degraded (Latha et al. 2019). Torp et al. (2021) developed a mathematical model for the design of a reaction that combines two layers of ammonia slip catalyst. Chemical kinetics and N_2O formation were observed in a high-flow reactor. The authors concluded that mixing SCR catalyst with oxidation catalyst results in less ammonia slip and high NOx reduction efficiency (Torp et al. 2021). Baleta et al. (2017) investigated that by designing the DeNOx system for uniform ammonia production, optimized ammonia slip can be achieved. A mathematical model was created to analyze the chemical reactions that occur during the movement of exhaust gases from the catalytic converter. Ammonia slip, NOx reduction, and droplet formation were also taken into account. For the simulation, the CFD code AVL FIRE was used. The authors concluded that the optimized droplet size and injection direction can reduce ammonia slip and increase the NOx reduction efficiency of SCR catalyst (Baleta et al. 2017). Hebbar et al. (2014) reviewed the various technologies used to reduce NOx from diesel engine exhaust gases. They discussed the advantages and disadvantages of the technologies used and concluded that EGR, LNT, and SCR do not meet the EURO-VI emission standards and hence new technologies are required to seek better emission control (Hebbar 2014). Qi et al. (2016) examined the parameters that influence the reduction of NOx by SCR. They discussed the effects of urea's ammonia storage capacity, the reaction mechanism involved, the urea dosing system, and the types of catalysts used for reducing NOx in SCR. They concluded that SCR can be effective in reducing NOx through the use of an efficient control system (Qi et al. 2016). Chatterjee et al. (2017) discussed the optimal design of DOC, DPF, and SCR to reduce the maximum amount of NOx from the exhaust pipe. The authors briefly discussed the control of ammonia slip with an ammonia slip catalyst. FTIR was used to analyze the composition of exhaust gases (Chatterjee et al. 2017). Zhang et al. (2016) suggested that the SCR catalyst should be used in two homogeneous cells, assuming identical reactions in each cell. Optimal control of urea injection has been suggested for minimal ammonia slip. A dynamic algorithm was developed to calculate the SCR load. The authors concluded that an optimal SCR system size ratio leads to minimized ammonia slip (Zhang et al. 2016). Blending diesel with hydrogen can reduce the NOx by 70% when the optimum crank angle remains 43.2^{o} (Lamas & Rodriguez 2017).

SOLID SELECTIVE CATALYTIC REACTION (SSCR)

SCR is one of the best technologies used by diesel engine manufacturers to meet the emission standards of EURO-VI. However, it has some limitations that reduce the NOx reduction efficiency of SCR. The problems associated with it are listed in Table 1

Table 1 shows the problems faced by engine manufacturers when using SCR. These problems affect the performance of the catalyst and thus limit the efficiency of NOx reduction.

The solutions to most of the above-mentioned problems related to SCR lie in new technology, Solid selective catalytic reaction (SSCR). In SSCR, we use solid ammonium salt to produce ammonia. Kim et al. (2014) investigated that low-temperature urea freezing and deposition of solid particles on pipe walls were severe problems for SCR systems. They used solid urea and ammonium carbonate to generate ammonia as a substitute for aqueous urea. When comparing ammonium carbonate and urea in solid form, they found that the problem of solid particle formation occurs when using solid urea and is eliminated when using ammonium carbonate (Kim et al. 2014). Ammonia is directly injected with help of a nozzle into the exhaust pipe just before SCR. It eliminates the freezing problem of DEF, the early deactivation of the catalyst, and thus increases the NOx reduction efficiency of SCR. Due to the shorter mixing time and low decomposition temperature of the ammonium salt, the problem of nozzle blockage is also eliminated (Zhang et al. 2018). Another

Table 1: Problems associated with SCR.

S. No.	Problems associated with the use of SCR.
1	Deposition of solid particles on the pipe walls.
2	Freezing of DEF below -11^{o}C.
3	Limited available volume for catalyst.
4	Undesirable ammonia slip.
5	The formation of a uniform mixture in SCR is quite difficult.
6	DEF needs to be filled frequently before the fuel tank gets empty.

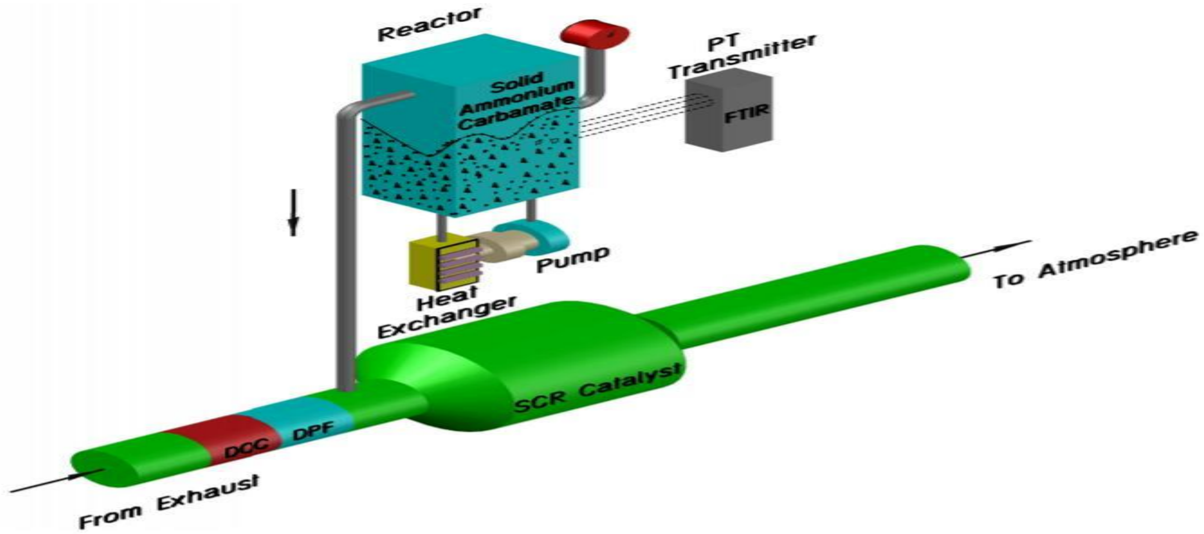

Fig. 3: Schematic diagram of SSCR system (Yadav & Srivastava 2021).

advantage of using solid ammonium salt for ammonia production is the better mixing of ammonia with exhaust gases (Krüger et al. 2003). Using the use of solid SCR increases the efficiency of NOx reduction, but at the same time power required for heating, and the salt also increases (Kim et al. 2014). Integrated hybrid systems can reduce ammonia slip and increase NOx reduction efficiency to a great extent (Guan et al. 2014).

Fig. 3 shows the arrangement of filters and accessories used in an SSCR system. It includes a DOC to oxidize hydrocarbons and carbon monoxide, a DPF to trap carbon particles, a tank containing solid ammonium carbamate, a heat exchanger, a pump, and an SCR catalyst. To reduce ammonia slip, an ammonia slip catalyst (ASC) is used after SCR.

Woo et al. (2021) investigated that insufficient heat transfer is a major challenge in the design of an ammonium carbamate pyrolysis reactor. They proposed a numerical model describing the sublimation phase of ammonium carbamate. The modeling also considered the effect of the collapse of ammonium carbamate and its sinking downward due to gravity. The program CFD at ANSYS FLUENT 19.2 was used for the analysis. The authors concluded that the prediction of the spatial temperature distribution is a herculean task since it depends on the internal structure of ammonium carbamate and the phenomena of collapse in the reactor (Woo et al. 2021).

Several parameters affecting the NOx reduction performance of SSCR have been studied. Some of these parameters are listed in Table 2

Table 2 contains most of the parameters of the SSCR system that affect performance. When these parameters are optimized, the best results can be achieved in meeting the EURO-VI emission standards.

Qu et al. (2014b) studied different parameters of the influence of SSCR ammonia spray using AVL FIRE 3D software when mixing ammonia and NOx in SCR catalyst. They concluded that better mixing, higher exhaust flow, and high temperature, resulting in less ammonia slip, can be optimized by the correct positioning of the nozzle (Qu et al. 2014c). The flow rate of exhaust through the nozzle depends upon the opening time of the nozzle, frequency, and back pressure. Using the nozzle opening control on the MOTOTRON platform using MATLAB achieves an effective flow rate (Qu et al. 2014a, 2014b, 2014c).

Guardiola et al. (2020) investigated that the problem of cross-sensitivity of NOx sensors can be reduced by estimating the ammonia slip and NOx reduction efficiency of SCR catalyst, and then passing this information to the control unit. SCR load plays an important role in reducing ammonia slip. This load estimation can be done by preparing a model related to NOx reduction and mass conservation between the SCR inlet and outlet. Using the Kalman filter, the authors developed an algorithm to estimate ammonia slip along with SCR constraints. The authors concluded that SCR load is an important factor in the estimation of ammonia slip (Guardiola et al. 2020). The efficient nozzle design results in a longer mixing time and an even flow of ammonia through the SCR catalyst, reducing ammonia slip (Qian et al. 2019).

ROLE OF EFFECTIVE MIXER DESIGN IN ACHIEVING THE BEST SSCR PERFORMANCE

It is investigated that a mixer must be included in the SSCR system for achieving the uniform flow of ammonia while moving through the SCR catalyst. A compact system can be designed for SSCR to achieve the best NOx reduction efficiency by using mixers and reducing the length of the chamber (Tan et al. 2018). Use of a mixer, near the urea injection point results in a uniform flow rate of ammonia through the SCR catalyst which increases the NOx conversion efficiency of SCR (Tian et al. 2015, Wardana et al. 2019). The commonly used mixers in the SSCR system are-

(i) Line mixer
(ii) Swirl mixer
(iii) Combination of line and swirl mixer

Mehdi et al. (2019) investigated that the combination of linear and swirl mixer gives the lowest ammonia slip and the highest NOx reduction efficiency due to the uniformity of ammonia and temperature distribution. For this purpose, the authors created a mathematical and optimized the data by

Table 2: Parameters affecting SSCR performance.

S.No.	Parameters influencing SSCR performance
1.	Position of ammonia injector
2.	Number of holes in ammonia injector
3.	Mixing length
4.	Uniformity of ammonia flow while moving through SCR catalyst
5.	Design of mixer for uniformity of ammonia flow
6.	Mass flow rate
7.	Temperature at SCR
8.	Pressure at SCR
9.	Flow rate
10.	Mixing time of ammonia with SCR
11.	Storage capacity of SCR catalyst
12.	Engine speed

simulation after analyzing CFD (Mehdi et al. 2019). Birkhold et al. (2007) investigated the evaporation phenomena and urea droplet decomposition at different exhaust gas temperatures (Birkhold et al. 2007).

Tayamon & Wigren (2016) proved that NOx emissions from diesel engines can be controlled with a good control model. Using a recursive prediction error method, the authors created a model and used it to design a controller with 17 parameters. They concluded that the designed control model can help to keep the ammonia flow uniform, and thus increase the NOx reduction efficiency (Tayamon & Wigren 2016).

Li et al. (2017) conducted a bench test on high-performance diesel engines using solid metal ammine salt and proved that the proper design of the SCR chamber can increase the DeNOx efficiency of the catalyst. For the same injection parameters, the SSCR system was found to be 24.6% more efficient in NOx reduction than the SCR system (Li et al. 2017). Phaily et al. (2014) experimented to determine the suitability of the Cardierite/Pt catalyst. The authors conducted tests with different loads and different induction rates of ammonia. Then, a simulation was performed on AVL FIRE to validate the results. The authors, finally concluded that the ammonia induction rate of 0.6 kg/hr gives optimum results for NOx conversion efficiency. (Phaily et al. 2014). In 2015, Xin Mei Yuan et al. reviewed the previous literature regarding SSCR performance and interpreted that proper mixer design for uniform ammonia flow, modeling, calibration, and various control strategies result in lower ammonia slip and better NOx performance of SCR catalyst (Yuan et al. 2015). Jeong et al. (2005) investigated that a small distance between the catalyst inlet and the engine exhaust leads to insufficient evaporation and thermolysis. A higher thermolysis rate and good mixing of ammonia and exhaust gases can lead to better NOx reduction efficiency and lower ammonia slip.

Fig. 4 shows the arrangement of the mixer in the SSCR system. Ammonia generated due to the decomposition of ammonium carbamate enters the exhaust pipe with help of an ammonia nozzle. Here the flow of ammonia remains turbulent. To obtain uniform ammonia flow, a mixer is placed after the nozzle and before the SCR catalyst. For oxidizing unused ammonia, an ammonia slip catalyst is used.

Kurien & Srivastava (2019) investigated various exhaust after-treatment systems. The authors proposed a fixed reducing agent for ammonia formation to avoid the problems associated with SCR systems. They suggested that effective control of ammonia flow can be used with an optimized mixer design to reduce the problem of reduced NOx reduction efficiency (Kurien & Srivastava 2019). Zhang et al. (2016) reviewed the available literature on SSCR technology and its advantages over SCR. They found that SSCR has higher NOx conversion efficiency, high ammonia density, and decomposition of salt occurs at low temperatures. Proper mixing of ammonia and exhaust gases and optimized flow rate of ammonia through SCR catalyst can lead to improved NOx reduction efficiency (Qu et al. 2014b). Ma et al. (2019) prepared a model explaining chemical kinetics and the flow field. They used this model to simulate a copper zeolite scripted system, specifying the boundary conditions of temperature, exhaust pressure, and mass flow rate through a bench test. They investigated the NOx reduction efficiency using an orthogonal experimental design, where the effect of nozzle injection angle and flow rate of ammonia was determined. It was concluded that fixed injection angle and chamber design for a uniform flow rate of ammonia through SCR catalyst can increase NOx reduction efficiency (Ma et al. 2019).

The use of layers of perforated plates of SCR catalyst results in a uniform flow of ammonia thus increasing NOx reduction efficiency but at the same time, a little pressure drop is observed during the process (Wardana et al. 2019). Koc et al. (2018) explored that an even ammonia flow can be achieved with an optimally designed mixer. A model based on the concept of the Eulerian-Lagrangian approach was created and the results were validated by CFD analysis using the HEEL software (Koc et al. 2018). Solid particle formation on exhaust pipe walls is greatly reduced by maintaining the uniform flow of ammonia through the SCR catalyst. The use

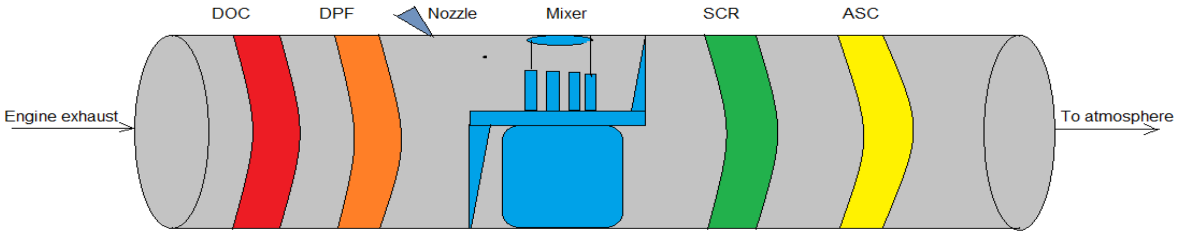

Fig. 4: Schematic diagram of SSCR with mixer.

of a mixer gives good results in direction of the achievement of uniform flow (Wang et al. 2021).

CONCLUSION

SSCR technology has very quickly reached EURO-VI emission standards for Diesel Engines. The uniformity of the ammonia stream is an important factor in achieving the better formation of droplets and evaporation as it moves through the SCR catalyst. The aim of obtaining a better ammonia flow can be achieved using a suitable mixer. Line mixer, swirl mixer, and combination of line and swirl mixer are widely used in SCR systems. But the combination of line and swirl mixer gives the best results in terms of efficient SCR performance. Several techniques have been discussed in this article to obtain a uniform ammonia flow. Finally, it can be concluded that the use of a combination of line and swirl mixer gives the best urea conversion due to the lower residence time of the droplets. The use of these mixers makes it possible to obtain an even distribution of flow and temperature. The uniform temperature distribution results in better catalyst performance and less deposit formation. The study provides sufficient information for researchers working on the reduction of NOx emission from the exhaust of diesel engines. The reduction of ammonia slip by using a suitable mixer will certainly help diesel engine manufacturers in achieving stringent emission norms.

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