



Review Article

# NO<sub>x</sub> FROM DIESEL ENGINE EMISSION AND CONTROL STRATEGIES—A REVIEW

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Increased usage of diesel vehicles and equipments are big challenges in terms of pollution and depleting diesel fuel recourses. Significant improvements in diesel emission levels have been achieved in the last 20 years. Development of diesel engine to meet new emission regulations and adopt renewable fuels is a challenging research objective. Automobile industries worldwide are continuing efforts to control emission associated issues both on road and off road diesel vehicles with developed technology for cleaner diesel fuels, quick responding electronic control units, advanced engine design and effective after treatment of exhaust. Some of the NO<sub>x</sub> control techniques are discussed in this paper.

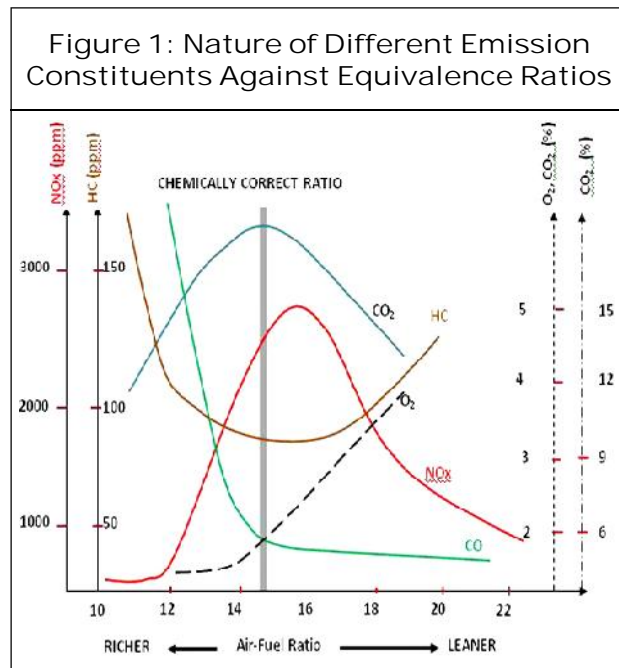
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## INTRODUCTION

Theoretical or clean diesel combustion should emit only CO<sub>2</sub> and H<sub>2</sub>O as exhaust with carbon oxidized to CO<sub>2</sub> and hydrogen to H<sub>2</sub>O. However, actual combustion under no circumstances will be theoretical due to various design and operating conditions. Fuel is not pure hydrocarbon, contain traces of nitrogen, sulphur and other chemicals and air is comprising hundreds of constituents other than O<sub>2</sub> and N<sub>2</sub>, may be amounting to 1% of total constituents. When combustion occurs between fuel and air, in real, a number of compounds appear in the engine exhaust, both as gaseous and solid. High

combustion pressure and temperatures during combustion with heterogeneous nature of air fuel mixture further complicates combustion process. Regions of rich and poor mixture ratios along with local stoichiometric composition pockets exist within the combustion chamber. Fuel rich composition produce CO, soot, dark smoke and unburnt HC, on the other hand fuel Oxygen rich regions generate NO<sub>x</sub> at high temperature conditions. Of all emission constituents, major concerns are NO<sub>x</sub>, CO, HC, Smoke (Particles and soot) and in the recent past CO<sub>2</sub> as its accumulation add to green house gases in the atmosphere (Figure 1).

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The non uniform fuel air composition during diesel combustion makes it difficult to abate all polluting constituents with one technique. Control of one emission constituent aggravates the other with whatever improvements in swirl. Other gaseous compounds known to be of toxicologic relevance are the aldehydes (e.g., formaldehyde, acetaldehyde, acrolein), benzene, 1,3-butadiene, and polycyclic aromatic hydrocarbons (PAHs) and nitro-PAHs (Heywood 1988). To prevent the environmental and health effects caused by vehicular emissions, regulations are imposed in countries worldwide. The two regulatory commissions that currently impose the most stringent emission regulations are the Environmental Protection Agency (EPA) and the European Parliament (EURO) (EPA document 2002).

## OXIDES OF NITROGEN

Nitrogen ( $N_2$ ) is a diatomic molecule and relatively inert gas that makes up about 79%

of atmospheric air. However, at high temperatures (around 1800 K and above) molecular Nitrogen ( $N_2$ ) disassociate into its atomic state (N). The single nitrogen atom (N) can be reactive and have ionization levels from plus one to plus five valence states. Thus nitrogen can form several different oxides like  $N_2O$ , NO,  $N_2O_2$ ,  $N_2O_3$ ,  $NO_2$ ,  $N_2O_5$  and  $N_2O_4$ . Actually, EPA regulates only nitrogen dioxide ( $NO_2$ ) as a surrogate for this family of compounds of Nitrogen because it is the most prevalent form of NOx in the atmosphere that is generated by anthropogenic (human) activities (Yokota *et al.* 1997). Bulk of nitrogen oxides emitted is in the form of NO (monoxide) and are oxidized in the atmosphere to  $NO_2$  (dioxide) within short time. Approximately 10 to 20% of nitrogen oxides from diesel engines are emitted as  $NO_2$  (nitrogen oxide), which is five times more toxic than NO (nitrogen monoxide). Nitric oxide (NO) and nitrogen dioxide ( $NO_2$ ) are collectively called as NOx. The amount of NO is normally dominating followed by a much smaller amount of  $NO_2$ . The other oxides of nitrogen occur in very small and insignificant quantities and readily react to NO and  $NO_2$  in IC engine combustion. Among NO (nitric oxide) and  $NO_2$  (nitrogen dioxide), NO is of primary concern because NO generally accounts for over 90 percent of the total NOx emissions from fossil fuel combustion, with the remainder being  $NO_2$ .

## SOURCES OF NOx FORMATION

The major sources of formation of NOx during combustion processes can be explained by three different mechanisms (Heywood, 1988), namely: i. Thermal NOx, ii. Fuel NOx and iii. Prompt NOx.

## Thermal NOx

Thermal NOx is produced when nitrogen reacts with excess oxygen at higher temperature (greater than 1800 K) in the combustion process. The thermal NO pathway occurs most quickly at high gas temperatures for which equilibrium chemical thermodynamics favor formation of NO, primarily through dissociation of molecular nitrogen and oxygen. The strong triple bond in the N<sub>2</sub> molecule requires high temperature to break to its radicals as N<sub>2</sub> → 2N and O<sub>2</sub> → 2O.

## Zeldovich Mechanisms of Thermal NOx

Thermal NOx require high activation energy and hence its formation rates are fast enough to be significant only at high temperatures. The thermal mechanism is most important for stoichiometric and lean premixed flames where temperatures are high. Because the NOx formation process is very nonlinear, so called hot spots, local areas with higher temperature than the average temperature have very large effect on the amount of NOx produced. Most NO<sub>2</sub> formed during combustion is quickly decomposed. Nitrogen monoxide, NO can also disintegrate to form N<sub>2</sub> and O<sub>2</sub>, but this rate very slow (Heywood, 1988). Thus, almost all of the NOx emitted is NO.

Two of the main reactions for the formation of thermal NOx are described by the Zeldovich mechanisms.

NO formed in the flame zone can be rapidly converted to NO<sub>2</sub> via

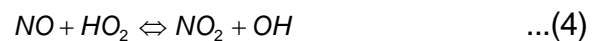


An additional reaction has been shown necessary at near stoichiometric and fuel rich mixture is



Inclusion of hydroxyl (OH) in the above Equation (3) as an extension of thermal NOx, was proposed by Lavoie *et al.* (1970).

NO produced in the flame zone gets converted to NO<sub>2</sub> through



Subsequently, conversion of this NO<sub>2</sub> to NO occurs via the reaction



This reaction continue as long as all the NO<sub>2</sub> produced is quenched by mixing with cooler fluid:

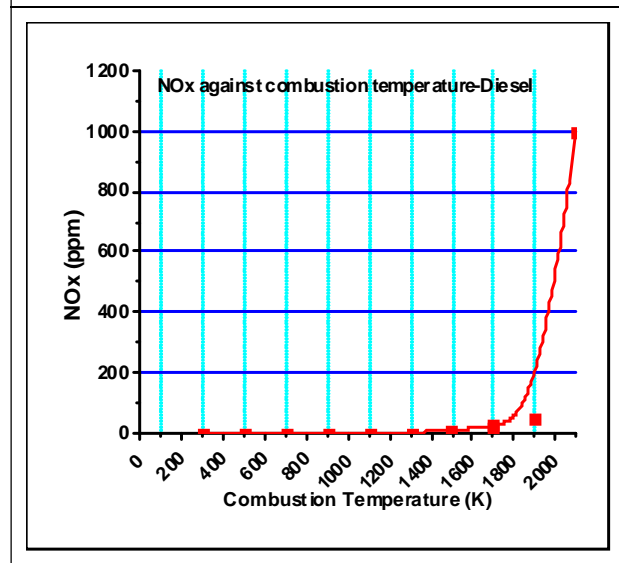
Formation rate of thermal NO is slow and is considered unimportant below 1800 K. Thermal NO formation attributed to equations (1) through (2) are considered formed in the post combustion exhaust gases. Heywood (1988), reported the relation of NO formation rate with equilibrium concentrations of O<sub>2</sub>, N<sub>2</sub> and temperature as shown below.

$$\frac{d[NO]}{dt} = \frac{6 \times 10^6}{\sqrt{T}} \times e^{\frac{-69090}{T}} \times [N_2]_e \times \sqrt{[O_2]_e} \quad \dots(6)$$

where [ ]<sub>e</sub> denotes equilibrium concentrations.

Above equation vindicate that NO formation increases exponentially with temperature. A plot of NO formation rate with temperature is shown in Figure 2. Other important factors apart from high temperature, in thermal NOx formation, are the residence time, amount of Longer the duration of high temperature burnt gases, higher will be the NOx formation. A good

Figure 2: NO<sub>x</sub> Formation Against Combustion Temperature



degree of turbulence facilitates quick mixing of air with fuel and accelerates reaction rates.

## FACTORS RESPONSIBLE FOR NO<sub>x</sub> FORMATION

NO<sub>x</sub> formation mainly depends on the temperature of the burnt gas, the residence time of the burnt gas at a high temperature and the amount of excess oxygen and turbulence (Heywood, 1988). Residence time describes how long time the combustion gas is having the high temperature. Longer the duration of high temperature burnt gases, higher will be the NO<sub>x</sub> formation. Increased pressure due to continuing combustion causes the burnt gases to experience compression. The net result is the initial burnt gases spend a longer time under high pressure and temperature as compared with the burnt gas from the later stages of combustion. This allows more time for NO to form. More the time for oxidation, more is the NO<sub>x</sub> produced. As the combustion process prolongs for a longer time period, there is more time available for NO to form in

slow speed engines as compared to high speed engines. A good degree of turbulence facilitates quick mixing of air with fuel and accelerates reaction rates.

## NO<sub>x</sub> EMISSION REDUCTION TECHNIQUES

It is an established factor that NO<sub>x</sub> formation is highly dependent on temperature and reducing peak temperatures during combustion obviously reduce NO<sub>x</sub>. Again any techniques that aim to reduce flame temperatures influence reaction rates, ignition delay, short or delayed combustion, so that emission of other constituents like CO, CO<sub>2</sub>, HC and importantly particulate emissions are also affected. Some of the cited in-cylinder techniques also cause fuel penalty. Unfortunately, there is no single method available that can improve both emissions as well as performance parameters.

## LITERATURE REVIEW ON SOME OF THE IN-CYLINDER TECHNIQUES

Several techniques have been researched and developed to abate hazardous emission constituents from diesel engines at the source level. Some of such extensively investigated techniques are:

- Variations of Injection Pressure and Nozzle Geometry
- Pre-Mixed Combustion
- Water Injection or combinations of two or more of above.
- Retarded and split fuel injection
- Exhaust Gas Recirculation (EGR)

### Early In-Cylinder Injection

Early In-Cylinder injection is achieved by a portion or entire fuel is injected using a separate injector, or the same injector (Aoyama *et al.*, 1990; and Reitz *et al.*, 1998), during intake or early during compression stroke. Air fuel homogeneity is expected when ignition occurs but impingement is a problem as found from the literature. This method can reduce NO<sub>x</sub> up to 90-98% as compared to conventional diesel combustion but a huge amount of HC and CO is produced and fuel consumption deteriorates. Soot emissions are reported to be low.

### Modulated Kinetics

A fairly new concept to this alternative combustion system is Modulated Kinetics (MK) combustion (Yokota *et al.*, 1997). Here the fuel is injected directly into the combustion chamber near or after TDC. Large amounts of cooled EGR are used for extended ignition delay. Despite significant mixture heterogeneity a very low NO<sub>x</sub> emissions are achieved. Problems of wall impingement are avoided in this technique with some control of the combustion phasing.

### Retarding SOI

Retarding SOI is reported as another technique in reducing NO<sub>x</sub> (Reitz *et al.*, 1998). Literature indicate that retarded fuel injection produce very low levels in nitrogen oxides (NO<sub>x</sub>) and soot emissions, but higher carbon monoxide (CO) and unburned hydrocarbons (HC) emissions and a significant penalty in fuel efficiency. When retarded injection was carried out with EGR, NO<sub>x</sub> and soot were found to be very low but fuel consumption and CO and HC emissions increased. It was reported that

complete premixed combustion was not possible to achieve and only 55% of fuel mass was burnt during this phase and with EGR this was extended up to 70%. Reduced overall combustion duration, reduced diffusion controlled phase, reduced NO<sub>x</sub> and dry soot emissions, a marginal increase in bsfc due to EGR (8% for 40% EGR), huge increase in CO and HC emissions were major findings from the experiments. In general, it can be concluded that the strategy of retarding SOI combined with high EGR rates is convenient at light engine load for reducing NO<sub>x</sub> and dry soot emissions, at the cost of a significant increase in fuel consumption and in emissions of CO and HC.

### Multiple or Split Injection Method and Injection Rate Shaping

Split fuel injection involves reducing splitting the injection as two or more events which can lead to a reduction in the ignition delay in the initial fuel pulse. This leads greater fraction of combustion to occur later in the expansion stroke. As majority of NO<sub>x</sub> occurs during pre-mixed stage, the net amount of NO<sub>x</sub> formed during the split fuel injection is lowered (Gao *et al.*, 2001). Multiple injections method is found to be very effective at reducing particulate emissions at high load, and combined technique of multiple injections with EGR is effective at intermediate and light loads. However, increased particulate emissions due to EGR causes increased engine wear due to degradation of lubricant. Increased Brake Specific Fuel Consumption (BSFC) is another concern. Split injection up to 5 splits, are experimented (Wang *et al.*, 2007) in combination with EGR. The adoption of the multiple fuel injection strategy and EGR results in lower power output and higher

residual fuel amount as penalties. This requires a balance between fuel injection timing (start of injection, injection duration), the fuel injection amount at each injection duration and EGR rate. Some of the common findings are:

- NO<sub>x</sub> emissions are reduced effectively but increased the soot at a given SOI.
- For the same NO<sub>x</sub> amount emitted, split injection combustion results in to more soot than EGR.
- EGR cooling reduced NO<sub>x</sub> significantly with a small penalty in soot emission.

#### Injection Rate Shaping

Reduction of NO<sub>x</sub> using split injection and injection rate shaping was also reported as an effective method with marginal loss of soot and fuel penalty. A simulation carried (Ghaffarpour *et al.*, 1996), suggests combined effects of intercooler and rate shaping results in a reduction of NO<sub>x</sub> by 50% for some operating conditions. A 20% reduction of NO<sub>x</sub> for injection rate shape, 15% for intercooler and 30% for injection rate shape and intercooler were reported with 5% soot increase and without penalty on engine performance.

#### Premixed Combustion

Premixed combustion is one of the most researched and promising technique in compression ignition engines. Premixing of mixture provides homogeneous charge during combustion and it is achieved in different ways. Such in-cylinder techniques emerged as very effective in reducing NO<sub>x</sub> emissions at significant levels along with soot emissions. Some of such techniques reported in the literature which are identified in different names are:

- Active Thermo-Atmosphere Combustion (ATAC),
- Activated Radical (AR) combustion,
- Compression-Ignited Homogeneous Charge (CIHC),
- Homogeneous-ChargeCompression-Ignition (HCCI),
- Premixed Lean Diesel Combustion (PREDIC),
- Premixed-Charge Compression Ignition (PCCI),
- Uniform Bulky combustion System (UNIBUS),
- Controlled Auto-ignition (CAI),
- Premixed Compression-Ignited (PCI) combustion,
- Modulated Kinetics (MK) combustion.

All these methods involve a low temperature, distributed reaction of fuel-lean and/or highly diluted mixtures by auto-ignition with some variations in achieving the objective in two and four stroke engines, with liquid and gaseous fuels

#### Port Injection

Port injection technique involve mixing the fuel with upstream air in the intake port and this homogenous charge is induced in to combustion chamber during compression stroke to get auto-ignition close to Top Dead Centre (TDC). This method has the advantage of diesel like efficiency with low NO<sub>x</sub> and PM. However ignition control over the wide range of load and speed is difficult to achieve. High hydrocarbon (HC) and carbon monoxide (CO) emissions are other associated disadvantages.

### Homogeneous Charge Compression Ignition (HCCI)

Many researchers have described Homogenous Charge Compression Ignition (HCCI) as a combination of SI and CI combustion (Najt *et al.*, 1983; Christensen *et al.*, 1998; and Thring *et al.*, 1998). HCCI combustion is a combined effect of spark ignition and compression ignition processes to give higher thermodynamic efficiencies and lower oxides of nitrogen emissions in internal combustion engines. HCCI combustion SOC is a complex phenomenon achieved by controlling the temperature, pressure, and composition of the air/fuel mixture. This is a challenging aspect and two methods to control SOC are proposed. First strategy is to the temperature of the air/fuel mixture, and the second involve changing the auto-ignition properties of the air/fuel mixture. EGR is also employed for charge dilution in some of the literature. HCCI combustion can result in a highly efficient engine with very low oxides of nitrogen emissions while using a low octane fuel.

### Water Injection

Injection of water into the combustion zone results in to reduced temperature. Such methods are proved effective in gas turbine engines and some literatures are available where it is employed to IC engines also. Four major approaches for introducing water have been reported in the literature (Yoshihara *et al.*, 1996).

- Water fumigation along with intake air
- Separate injectors for direct injection into the engine
- Mixing of water and fuel before the injection (unstabilized emulsion)

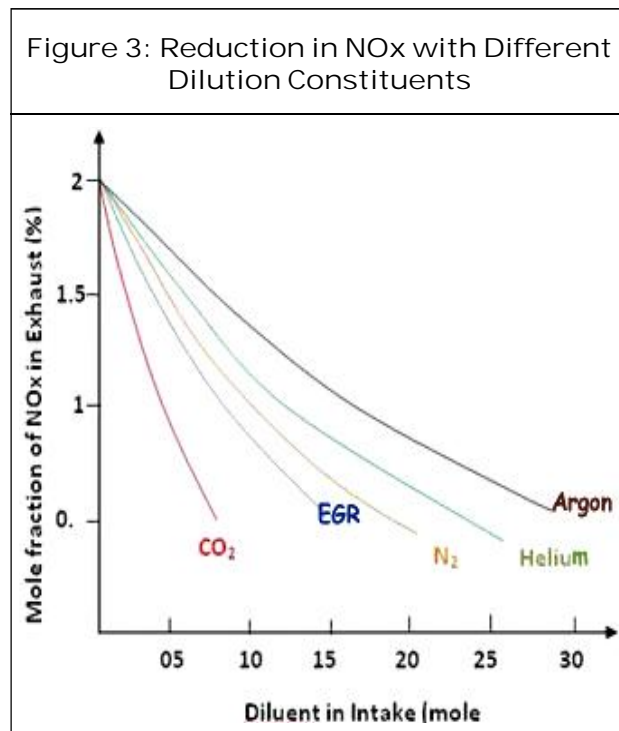
- Stabilized emulsions as a single-phase.

An emulsion is defined as a mixture of two or more generally insoluble liquids and it is called as stabilized when droplet do not separate. Under high speed and shearing the solid particles from liquid and this is referred as unstabilized emulsion (Abu-Zaid *et al.*, 2004; and Armas *et al.*, 2005). Several methods are tested by different authors introducing water into the combustion process, stabilized or unstabilized and have primarily determined that water-in-fuel emulsions are most effective in reducing NO<sub>x</sub>, BSFC, and result in lower increases of CO and UBHC emissions (Samec *et al.*, 2000; Tauzia *et al.*, 2010; and Gorkem *et al.*, 2013).

Summary of literature reviewed indicate water injection in any form like diesel emulsion, fumigation or direct injection in to cylinder, it is observed that water addition to fuel lowers the flame temperature and suppresses thermal NO<sub>x</sub> formation. HC and CO emissions and SFC increase depending upon the method of water induction method and load and speed variations. Most of the work reported increase of engine torque, effective power, efficiency, smoke density and a decrease in Specific Fuel Consumption (SFC), NO, CO<sub>2</sub> emissions. When EGR is employed along with EGR, result in decreased smoke and NO<sub>x</sub> emission without any noticeable change in the efficiency and also without increase of other pollutants. This method has the possibility of reducing NO<sub>x</sub> at heavy load regions where the application of EGR is difficult. 40 to 50% of NO<sub>x</sub> reduction is achieved at 70% accelerator opening with negligible difference of fuel consumption.

### Charge Dilution Method

Charge dilution by selected constituents is widely used strategy employed for low flame temperatures and low NO<sub>x</sub> emissions in diesel engines (Donahue, 2000). Literatures provide information on use of carbon dioxide (CO<sub>2</sub>), nitrogen (N<sub>2</sub>), argon, water/steam or Exhaust Gas Recirculation (EGR). These diluents reduce the peak flame temperature and consequently reduce the rate of NO formation at a given engine operating condition. CO<sub>2</sub> is one of the effective diluents in improving NO<sub>x</sub> reductions (Figure 3). Literature indicates with 6% CO<sub>2</sub> admission NO<sub>x</sub> emission reduce approximately 50%. Smoke emission increased around 60% and CO emission increased approximately 8.5 times from its base level. Engine torque, power, BMEP and SFC have deteriorated approximately 5.9%, 5.5%, 6%, 3.3% respectively (Ladommatos *et al.*, 1998). No significant losses in performance parameters were reported.



### Exhaust Gas Recirculation

EGR process involves bypassing a calculated volume (mass) of engine out exhaust back to engine to mix with fresh intake. Exhaust gases, mainly consist of CO<sub>2</sub> and H<sub>2</sub>O, which are already combusted during previous cycle, they do not burn again when they are recirculated. Application of EGR alters the normal combustion process in several ways. The physical properties of charge mixture, A/F ratio, start of Ignition and Heat release pattern change significantly affecting emission and performance factors.

Three effects of EGR are identified as responsible for NO<sub>x</sub> reductions (Ladommatos *et al.*, part II 1996 and III 1997; and Abd *et al.*, 2001), namely: the dilution, the chemical, and the thermal effects. Replacement of intake O<sub>2</sub> fraction due to replacement with CO<sub>2</sub> is referred to as the dilution effect, was reported as the most significant one influencing the combustion process and the formation of emissions. The chemical effect involving the dissociation of CO<sub>2</sub> to form free radicals was reported to have a minor impact on combustion and emissions. The thermal effect, which arises from the higher specific heat capacity of CO<sub>2</sub> in comparison to the replaced O<sub>2</sub>, was shown to have negligible effects on combustion and emissions.

The application of EGR results in modifying both the composition (addition of CO<sub>2</sub> and H<sub>2</sub>O) and physical conditions (temperature, density) of the inlet charge. Replacement of intake air by EGR decreases the A/F ratio which has a tendency to shorten ignition delay. Reduced Availability of O<sub>2</sub> due to admit of EGR also increase ignition delay due to increased mixing timing between injected fuel and the



fresh oxygen (Shahed *et al.*, 1981; Ladommatos *et al.*, 1998; and Abd-Alla *et al.*, 2002). Longer Ignition delay causes a shift in the location of start of combustion resulting in to more time for the fuel spray to penetrate and the auto-ignition locations to be shifted towards the wall of the combustion chamber. This results in an increase in the volume of the combustion flames, in turn the larger amounts of gases that absorb more energy released by combustion, leading to lower combustion temperatures. As a consequence of this shift in auto ignition locations, the whole combustion process also shifts further towards the expansion stroke. This results in the products of combustion spending shorter periods at high temperatures leading to lower thermal NO<sub>x</sub> formation as well as lower rate of soot oxidation (Jacobs *et al.*, 2003).

Even though EGR is known as one of the most representative methods for reducing NO<sub>x</sub> emissions without significant modifications in diesel engines, EGR cannot be applied alone. Increased soot emissions and fuel penalty are common indication in all reports if EGR alone is applied for NO<sub>x</sub> control. With higher the ratios of EGR higher are the degradation in combustion quality and fuel penalty (Ladommatos *et al.*, 2000). Increased piston-cylinder wearing, deterioration of efficiency, operational stability are the difficulties associated (Jinyoung *et al.*, 2001). EGR temperature also plays an important parameter while employing EGR at higher load and ratios (Abd-Alla *et al.*, 2002).

#### Ethanol Fumigation

Fumigation implies the mixing of alcohol with fresh air stream well before the mixture is being admitted into the cylinder. Fumigation of alcohol fuel is more convenient method as it

can implemented with minimum modification to the existing engine (Bilgin *et al.*, 2002; and Anil Singh *et al.*, 2007). Alcohol may be fumigated using a carburetor, vaporiser or an injector along with a separate fuel tank line and controls. Ethanol lowers the flame temperature due to its higher latent heat which favors low NO<sub>x</sub> formation.

#### CONCLUSION

A glance at the history of emission reductions suggests that the demand to decrease NO<sub>x</sub> levels still critical and is a challenge. The future Euro VI levels are set to 0.08 g/km, indicating remarkable 55% reductions from present Euro V norms for passenger cars and above 75% for heavy duty vehicles. This fact implies that NO<sub>x</sub> reduction is very critical. Research attempts should emphasize and focus on effective techniques to meet the Euro VI standards in general and Euro V standards for India in particular (EPA-2004). 🌀

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