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A Review on Carbon Monoxide Emission: Formation Measurement and Control Strategies

S.Karthikayan[1]*, Vimal Raj. V[2], Vishnu.N[2]

[1]Department of Mechanical Engineering, National Institute of Technology Puducherry, Thiruvettakudy, Karaikal, Puducherry-609 609, India.
[2]Department of Mechanical Engineering, Karpagam College of Engineering, Coimbatore - 641 032, India.

Abstract : Abundant increase in population across the world causes many major problems. Peoples' craze towards the automobiles is increasing nowadays. So due to this reason the pollutants from the vehicles are drastically increasing. All these pollutants are emitted through the engine exhaust gas except the HC from gasoline engines. In the conventional gasoline fuelled SI engines, in addition to exhaust emissions, unburnt fuel/HC comes from evaporation in fuel tank, fuel system and from the crank case blow by gases. Amount of sulphur in the current engine fuels such as gasoline and diesel is quite small (<500 ppm by mass) and is being lowered further below 50 ppm. Therefore, emissions of sulphur dioxides and sulphur trioxides from the engines are not considered. The amount of pollutants emitted depends on engine design, operating conditions, ambient conditions, fuel type and exhaust after treatment employed. Under idle engine conditions, CO and HC emissions are high due to fuel rich engine operations. The HC emissions under wide range of engine operating conditions are high. In diesel engines, CO emissions are quite low (0.02-0.1 percentage volume). This technical paper details the engine emissions, formation, measurement in macroscopic level and CO emissions are studied in microscopic level.

Keywords : Carbon Monoxide, lean mixture, equivalence ratio, air-fuel ratio, HCCI, Stoichiometric mixture.

Introduction

The major pollutants emitted by IC engines' are CO, HC, NO_x and PM which are commonly referred as vehicle emissions [9-22]. HC emission from diesel engine is also lower by a factor of 5-10 compared to typical SI engine. However The NO concentrations are comparable to those from SI engines. In HCCI engines efficient combustion leads to lower the CO, NO_x and soot emissions. Moreover this engine generates the same power as that of the conventional engines with less fuel consumption. In plenty of the cases the HCCI combustion was most successful and it may be an acceptable futuristic engine [14].

Sources of pollutants in spark-ignition engines

The mechanism of formation of exhaust emissions is governed by the process and chemistry of combustion. The sources of different emissions in a conventional SI engine are schematically shown in fig 1. After spark ignition, a flame propagates across the combustion chamber and burns the charge in the engine cylinder. NO and CO are formed in the bulk of burned gases during combustion and post combustion reactions

[8]. Carbon monoxide (CO) results practically due to deficiency of oxygen in the fuel air mixture leading to incomplete oxidation of the fuel. With decrease in air-fuel ratio below stoichiometric value, the formation of CO increases sharply. During expansion stoke, the downward piston motion rapidly cools the burned gases by expansion, freezing the reactions that involved during NO and CO formation. The concentrations of CO and NO are frozen at much higher values than expected from the chemical equilibrium considerations. The fuel-air mixture is rich during cold starting, engine warm-up and transients like acceleration. Engine operation in these modes contributes significantly to the CO emissions[1].

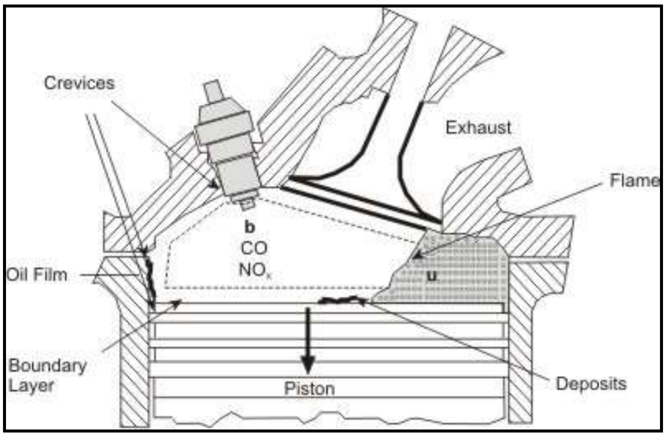


Figure 1. Sources of pollutants in spark-ignition engines [1]

As the fuel approaches cold combustion chamber walls, it is quenched, leaving a very thin quenched layer of unburned fuel-air mixture. The flame is unable to propagate in narrow passages or 'crevices' in the combustion chamber. Therefore, the mixture in the crevices between piston top land and cylinder wall above top ring, around spark plug threads, and cylinder head gasket is left unburned. Presence of liquid fuel in the combustion chamber during cold start too, contributes to unburned hydrocarbon emissions. As the pressure inside the engine cylinder decreases due to expansion, the unburned mixture from the crevices expand back into the cylinder. Simultaneously, the hydrocarbons absorbed during intake and compression strokes in the oil film and combustion chamber deposits are desorbed. The unburned mixture and fuel from these sources are partly entrained into bulky hot burned gases and are partially or completely oxidized before the gas leaves the engine. Finally when the exhaust valve opens CO, NO_x and part of the entrained unburned hydrocarbons from quench layers and cervices leaves the engine and are exhausted.

Variation of CO concentration in the exhaust of conventional SI engine with fuel/air equivalence ratio

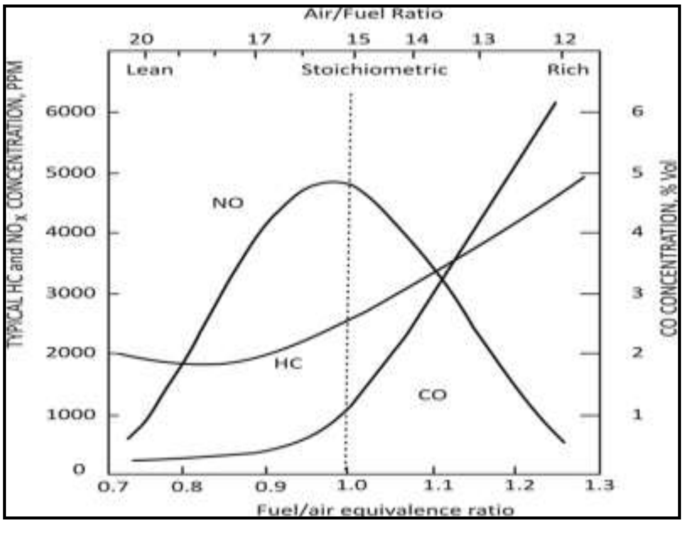


Figure 2. Variation of CO concentration in the exhaust of conventional spark- ignition engine with fuel/air equivalence ratio [2]

Air-fuel ratio is one of the most important engine variables that affect the exhaust emissions in SI engines as shown in fig 2. The SI engine operation is preferred near stoichiometric mixtures as it provides a smooth engine operation. Carbon monoxide emissions reduce with increase in the air-fuel ratio as more oxygen gets available for combustion. Higher air-fuel ratios to a certain critical value tends to reduce all the three pollutants (CO, HC, NO_x).

Formation of Carbon Monoxide

Carbon monoxide emissions rise due to deficiency of oxygen when the fuel-rich mixture burns. A two-step process may be used to approximate complete combustion of hydrocarbons to form carbon dioxide as the final product. First step is the conversion of hydrocarbon to CO, during which several oxidation reactions occur involving formation of intermediate species like smaller hydrocarbon molecules, aldehydes, ketones etc. Reaction 1 describes the oxidation of a hydrocarbon, RH where R stands for the hydrocarbon radical which may be represented by,

Reaction 1

$RH \rightarrow R \rightarrow R + O_2 \rightarrow RCHO \rightarrow CO$

Reaction 2 is the conversion of CO to CO_2 provided sufficient oxygen is available. One of the principal reactions for conversion of CO to CO_2 is,

Reaction 2

$CO + OH \longleftrightarrow CO_2 + H$

Reaction 2 is quite fast and at high temperatures it is continuously under equilibrium. During combustion and expansion stroke until the temperature decreases to about 1800K, the C-O-H system is more or less under chemical equilibrium. At the late of expansion stroke, conversion of CO to CO_2 is kinetically controlled and takes place at a slower rate. The rate of controlling reactions for oxidation of CO to CO_2 especially at temperature lower than 1800K are the following three body recombination reactions,

Reaction 3

$H + H + M \leftrightarrow H_2 + M$

Reaction 4

$H + OH + M \leftrightarrow H_2O + M$

Reaction 5

$H + O_2 + M \leftrightarrow HO_2 + M$

Studies on kinematics of CO formation established that the CO emitted is higher than the equilibrium concentrations corresponding to the temperature and pressure conditions at the end of expansion [3].The non-equilibrium kinetically controls concentrations of H and OH from the reaction 3 to reaction 5 which was used for the prediction of equilibrium of CO concentration by the reaction 2. The calculated CO values termed as partial equilibrium values are compared with the measured and the equilibrium values on fig 3. The equilibrium concentrations were calculated corresponding to peak cycle temperatures and pressures, and corresponding to exhaust conditions. The measured CO amount from several sources at different air fuel ratios and for different fuel composition and volatility were used. Measured data are correlated by a single curve. The CO increases sharply as the air fuel ratios decreases below stoichiometric value. For lean mixtures, CO concentration is very small, less than 0.1% by volume. The partial equilibrium calculations gave good correlation with the experimental values for the whole range of air fuel ratios. For the rich mixtures, the equilibrium concentrations calculated at peak pressure are quite close to the experimental values. For the stochiometric and lean mixtures however, partial equilibrium CO predictions have a better agreement with the measured values.

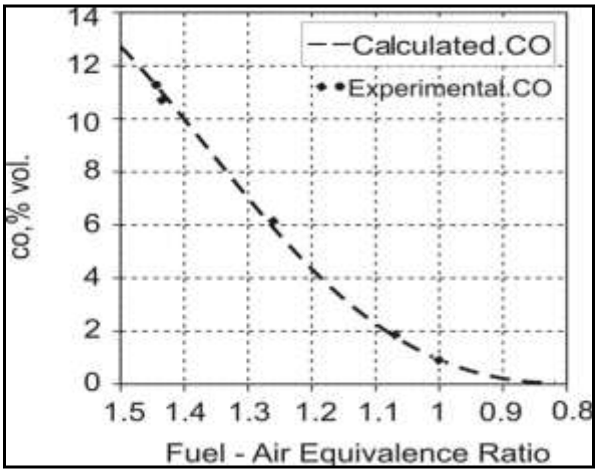


Figure 3. Comparision of measured and predicted CO concentration under steady condition as a function of fuel air equivalence ratio [3]

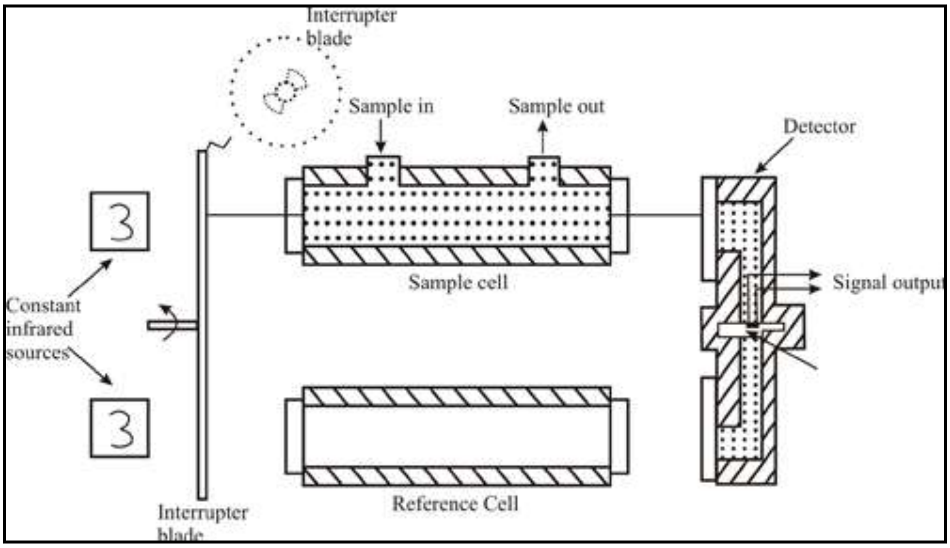
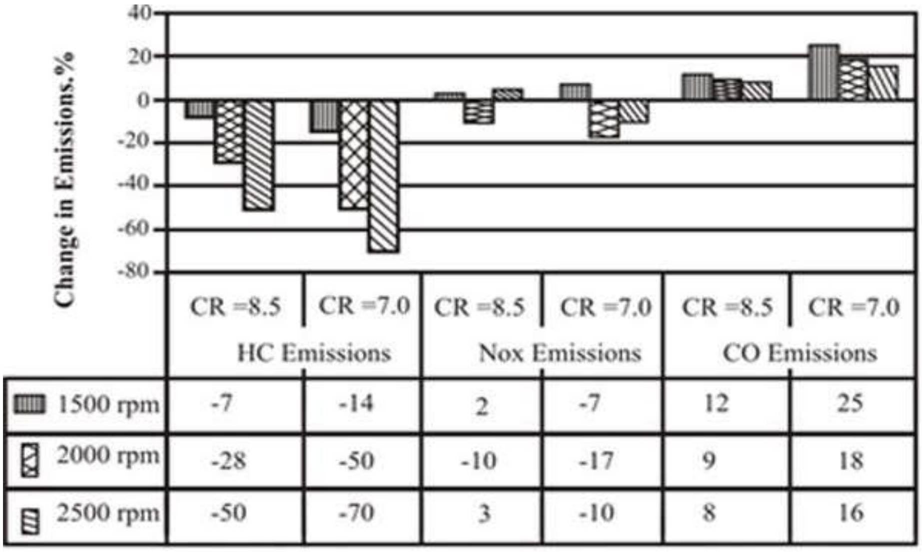


Figure 4. Schematic of an NDIR analyzer for measurement of CO and CO_2 concentration [4]



	CR =8.5	CR =7.0	CR =8.5	CR =7.0	CR =8.5	CR =7.0
	HC Emissions		Nox Emissions		CO Emissions	
▥ 1500 rpm	-7	-14	2	-7	12	25
▨ 2000 rpm	-28	-50	-10	-17	9	18
▤ 2500 rpm	-50	-70	3	-10	8	16

Figure 5. Effect of reduction in compression ratio from 10:1 to 8.5 and 7.0:1 on SI engine emissions, Single cylinder engine, 73*77 mm, bore and stroke [4]

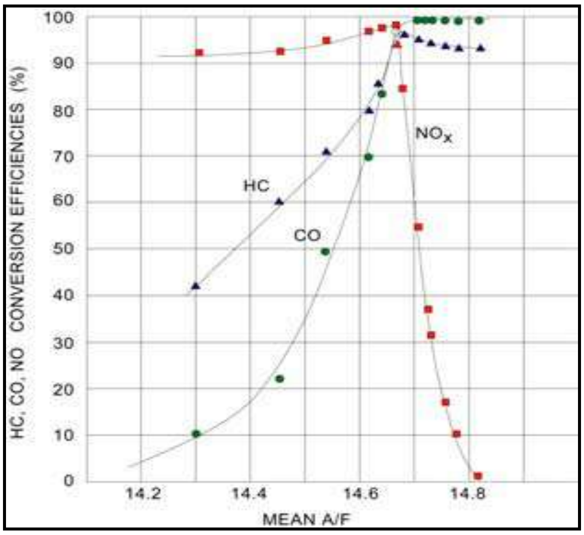


Figure 6. Variation of CO concentration in the exhaust of conventional spark- ignition engine with fuel/air equivalence ratio [5]

Measured CO for leaner than stochiometric mixtures are higher than the partial equilibriums calculation results. Partial oxidation of HC from the crevices that are entrained into burnt gases during expansion is one of the factors that have significant contribution to CO at lean mixture conditions. In multicylinder engines, maldistribution of fuel air mixture causes cylinder to cylinder variation in air fuel ratio. This is especially prominent in the carbureted or single point throttle body injected (TBI) engines. This leads to significant increase in average CO emissions because CO increases sharply as the mixture becomes richer than stoichiometric mixture. In some cylinders while no significant reduction in CO results due to leaning of mixture beyond stochiometric air fuel ratio in other cylinders. Hence, uniform mixture distribution between cylinders is very important. In multicylinder, carbureted engines cylinder to cylinder variation in air fuel ratio up to 1-2 units could result. Use of multipoint port fuel injection (PFI) systems has resulted in a largely uniform mixture distribution between cylinders. The fuel air mixture even inside the cylinder may not be entirely homogeneous due to poor fuel vaporization and non uniform mixing of residual burned gas fraction in the cylinder charge. It will also contribute to increase in CO emissions. Overall, the air fuel ratio is the most important engine parameter affecting the CO emissions, and the other factors influence CO only indirectly.

Emission Measurement: Instrumentation and Methods

NDIR Analyzer

Beer-Lambert's Law is used for operation of NDIR analyzers by measuring the degree of absorption of infrared radiations (IR) when they pass through a column of gas. The fraction of incident radiations absorbed is given by,

$$\frac{I}{I_o} = \left(1 - e^{-k.c.d}\right)$$

Where,
I = Radiation energy absorbed
I_o = Incident radiation energy
k = Characteristic absorption constant for the gas, m^2/gmol
c = Concentration of the gas, gmol/m^3
d = Length of the gas column, m

- As the absorption of IR radiations is measured only in a narrow range of wavelengths (not the entire range of wavelength of IR radiations) which has specifically a high absorbance for a particular gas, the technique is called as ‘Non-dispersive infra-red’. For example carbon monoxide has a strong absorbance in the wavelength band of 4.5-5 µm.

- The analyzer measures differential in absorption of energy from two columns of gas; (i) the gas to be analyzed in the 'sample cell' and (ii) a gas of fixed composition like N_2 contained in the reference cell which is free of the gas of interest and relatively non-absorbing in the infrared region.
- The infrared beam from a single source is usually split into two beams of the same intensity, one each for the sample and reference cells.
- The detector is divided in two compartments separated by a flexible diaphragm; one section receives transmitted IR energy from the sample cell and the other from the reference cell.
- The detector is filled with the gas of interest, so that the energy transmitted to the detector is fully absorbed.
- The flexible diaphragm of the detector senses the differential pressure between the two sections of the detector caused by the difference in the amount of transmitted IR energy absorbed. The deflection in the diaphragm is used to generate an electrical signal that determines the concentration of the gaseous species of interest.

A rotating interrupter in the path of IR beam is put to generate AC signal output that can be amplified.

NDIR analyzers [4] enable accurate measurements of CO and CO_2 in the exhaust gases. NDIR instruments are seldom used for measurement of hydrocarbons except in the garage type analyzers, as the IR absorbance to different hydrocarbons varies substantially. The unsaturated hydrocarbons are primarily responsible for photochemical smog but they do not have an adequate absorption in the IR wavelength range that is specific to the saturated hydrocarbons and vice versa. Sensitivity and response of NDIR to the exhaust HC is typically only half of the probable true value. NO absorbs only weekly in the infrared region. Moreover, CO, CO_2 and water vapours interfere seriously; hence NDIR analyzers are also not used for NO measurement.

SI Engine Variables and Emissions

Any engine variable that affects oxygen availability during combustion would influence CO emissions. Principle design and operating variables affecting engine emissions are:

Design Variables:

- Compression Ratio
- Combustion chamber surface to volume ratio
- Ignition timing
- Valve timings and valve overlap
- Air motion, swirl tumble etc

Charge stratification:

- Operating Variables
- Air-fuel Ratio
- Charge dilution and exhaust gas recirculation (EGR)
- Speed
- Load
- Coolant temperature
- Transient engine operation: acceleration, deceleration etc.

The effect of some variables discussed below is typical in nature and variations in the trends with specific engine design change are observed.

Compression Ratio:

The compression ratio of spark ignition gasoline engines has been lowered from over 10:1 for high performance engines in earlier days to 8.5- 9.0:1 at present. Reduction in engine compression ratio was necessary for engine operation on unleaded gasoline that had a lowered octane number than the leaded gasoline. Unleaded gasoline is essential for the vehicles equipped with catalytic converters and petroleum refinery economics demanded that unleaded gasoline of a lowered octane number is produced. Reduction in engine compression ratio results in higher exhaust gas temperatures which promote oxidation of CO.

The result shows a significant increase in CO that could have resulted from partial oxidation of some of the HC to CO in the exhaust manifold. The disadvantage of lower compression ratio is reduction in engine efficiency and increase in CO_2 emissions.

Ignition Timing:

As the ignition timing is retarded, more burning takes place during expansion stroke resulting in lower peak combustion pressures and a lower of mass of charge is pushed into crevice volume. Also, at the retarded ignition timings exhaust gas temperature increases as the engine thermal efficiency is reduced. In the hotter exhaust gas with the retarded ignition timing higher oxidation rates of the HC and CO in the exhaust system are obtained. The disadvantage of the retarded ignition timing is lower engine efficiency, lower power and a poorer fuel economy.

Air –Fuel Ratio:

- Carbon monoxide results due to deficiency of oxygen during combustion and is reduced as the mixture is leaned. CO emissions are reduced to very low values as the mixture is leaned to $\phi=0.90 – 0.95$ i.e. air-fuel ratio is increased above the stoichiometric value by 5 to 10%. Further leaning of mixture shows very little additional reduction in the CO emissions.
- With increase in air fuel ratio, the initial concentration of hydrocarbons in the mixture is reduced and more oxygen is available for oxidation. Hydrocarbon emissions therefore, decrease with increase in air-fuel ratio until mixture becomes too lean when partial or complete engine misfire results which cause a sharp increase in HC emissions. For $\phi< 0.8$ engine may misfire more frequently thereby increasing HC emissions sharply.
- The highest burned gas temperatures are obtained for mixtures that are slightly (5 to 10 percent) richer than stoichiometric.

Exhaust After Treatment:

Improvements in engine design and adjustment of engine parameters carried to control engine emissions were inadequate to meet the first set of stringent emission regulations introduced in the USA from 1975. Devices were developed to treat exhaust gas for conversion of engine emissions to harmless gases. Conversion of pollutants present in the exhaust gas is termed as exhaust after treatment. Two basic types of exhaust treatment systems were considered;

- Thermal reactors
- Catalytic reactors or converters

Initially, thermal reactors for oxidation of HC and CO to CO_2 and H_2O were developed. However, as high conversion rates of pollutants could not be obtained in the thermal reactors these did not find widespread application and very soon the catalytic converters became a standard exhaust after treatment device for the spark ignited engine vehicles.

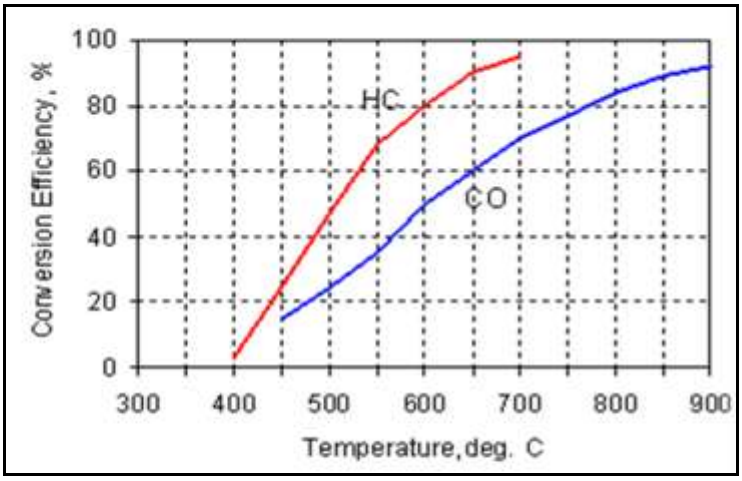


Figure 7. Conversion efficiency versus temperature for thermal oxidation of HC and CO [5]

Thermal Reactors:

If high exhaust gas temperatures are maintained and sufficient free oxygen is present in the exhaust gases, CO and HC can be oxidized in the engine exhaust system. Thermal conversion efficiency for HC and CO as a function of temperature is presented in fig.7.

- For 50% oxidation of CO and HC temperatures in excess of 500 and 600 °C, respectively are required.
- For conversion of 80 percent, temperatures required are about 600 and 750 ºC for HC and CO, respectively.
- Residence time in reactor is another important variable. At 750 ºC, conversion of HC up to 90 percent may be obtained in 100 ms while at 850 ºC only 50 ms are required. Similarly, for 90 percent oxidation of CO 250 ms and 70 ms would be necessary at 750 ºC and 850 ºC, respectively.

The thermal reactors are to be located in the exhaust manifold. In the conventional spark ignition engines, the gas temperature near the exhaust port may vary from about 300 ºC at idle to 900 ºC at full load. Exhaust gas temperature are higher for rich mixtures and increase with retard of spark timing. When the exhaust gas flows down the exhaust pipe, it cools at a rate of about 50 ºC to 100 ºC/meter. The design of the thermal reactors has the following features to ensure adequately high gas temperatures;

- The thermal reactor has to be located as close to the exhaust port as possible. In fact, the exhaust manifold is modified to act as thermal reactor.
- Exhaust manifold is made as a two wall design with an air gap in between to reduce cooling of the exhaust gases. Volume of the manifold is increased to increase residence time for the gases to undergo oxidation reactions.
- Engine is operated rich to obtain high exhaust gas temperature and secondary air amounting to 10 to 20% of air flow to the engine is injected at the port by an engine driven or electrical air pump.
- The rate of secondary air injection is controlled so that excessive cooling of the exhaust gas does not result which otherwise may chill out the oxidation reactions.

Oxidation of 1.0 percent CO in the gases increases gas temperature by about 145 ºC. Hence, with rich engine operation and secondary air injection sustained oxidation reactions of HC and CO occur giving higher conversion compared to lean engine operation when the starting temperatures are lower by about 100 °C. Rich mixture operation and use of retarded ignition timing result in deterioration of fuel economy. Overall, the thermal reactors provide rather low conversion rates as during city driving conditions engine operates at part load and exhaust gas temperature are quite low. The thermal reactors have no effect on NO_x emissions.

Catalytic Reactors:

Catalytic converters are now a standard fitment to SI engine powered vehicles. The exhaust gas flows over a bed of catalyst where pollutants are converted to harmless gases. The catalyst lowers the reaction temperature and hence high conversion rates are obtained compared to thermal reactors. A catalytic converter consists of the following main elements besides housing;

(i) Catalyst
(ii) Catalyst substrate or support, and
(iii) Intermediate coat or washcoat

Catalyst

The active catalyst material is required to possess the following main characteristics

- High specific reaction activity for pollutants
- High resistance to thermal degradation
- Good cold start performance, and
- Low deactivation caused by fuel contaminants and sulphur other desirable requirements are low cost.

The oxides of base metals such as copper, chromium, nickel, cobalt etc. have been studied. The base metal oxides are effective only at higher temperatures. In addition, they sinter and deactivate when subjected to high exhaust gas temperatures experienced at high engine loads. Their conversion efficiency is severely reduced by sulphur dioxide produced by sulphur in fuel. The noble metals platinum (Pt), palladium (Pd) and rhodium (Rh) were found to meet the above mentioned performance requirements. In practice, only the noble metals are used although these are expensive[1].

Mixtures of noble metals are used to provide higher reactivity and selectivity of conversion. Following are typical formulations;

Pt : Pd in 2:1 ratio for oxidation catalysts

(Pt + Pd): Rh in ratio of 5:1 to 10: 1 for simultaneous oxidation and reduction such as in 3-way catalysts

Palladium has higher specific activity than Pt for oxidation of CO, olefins and methane. For the oxidation of paraffin hydrocarbons Pt is more active than Pd. Platinum has a higher thermal resistance to deactivation. Rhodium is used as a NO_x reduction catalyst when simultaneous conversion of CO, HC and NO_x is desired as in the 3-way catalytic converters. The amount of noble metal used typically varies from about 0.8 to 1.8 g/l (25 to 50 g/ft^3) of catalytic converter volume. For a passenger car the total amount of noble metal in the converter is typically in the range 0.8 g to 2 g. The active metal is in a highly dispersed state when impregnated on the surface of the catalyst support. The size of the noble metal particles on the fresh converter is about 50 nm. However, when used the noble metal particles sinter and may grow to a size of around 100 nm.

Catalyst Substrate:

The active catalyst material is impregnated on the surface of catalyst substrate or support. The function of catalyst substrate is to provide maximum possible contact of catalyst with reactants.

Following are the main requirements of catalyst substrate:

- High surface area per unit volume to keep a small size of the converter.
- Support should be compatible with coating of a suitable material (washcoat) to provide high surface area and right size of pores on its surface for good dispersion and high activity of the catalyst.
- Low thermal capacity and efficient heat transfer properties for quick heat-up to working temperatures.
- Ability to withstand high operating temperatures up to around to 1000 ºC.
- High resistance to thermal shocks that could be caused by sudden heat release when HC from engine misfire get oxidized in the converter.
- Low pressure drop.
- Ability to withstand mechanical shocks and vibrations at the operating temperatures under road conditions for long life and durability of 160,000 km and longer.

The following types of catalysts supports are used;

- Pellets
- Monolithic supports
- Ceramic monoliths
- Metal monoliths

Oxidation and 3-Way Catalytic Converters:

Since 1975, in the production of gasoline vehicles, two main types of catalytic converters have been used.

- Oxidation catalytic converters
- 3-Way catalytic converters

The catalytic converters for the first time were used to reduce only HC and CO emissions from the US gasoline passenger cars in 1975. As these converters reduced HC and CO by oxidation, they were called as ‘oxidation’ catalytic converters.

Oxidation Catalytic Converters:

The oxidation catalyst converts “CO and HC to CO_2 and H_2O at substantially low temperatures and at higher conversion efficiency than the thermal reactors”. The required oxygen for oxidation reactions is made available either by operating engine lean or by injecting secondary air ahead of the catalytic converter when engine is operated rich.

The conversion efficiency of a catalytic converter is defined as:

$$\eta_{out} = \frac{m_{CO,in}}{m_{CO,in}} - \frac{m_{CO,out}}{m_{CO,in}} = 1 - \frac{m_{CO,out}}{m_{CO,in}}$$

Light-Off Temperature:

The catalytic conversion efficiency[6] depends upon the exhaust temperature and its relationship with temperature is shown on fig. 8. The temperature at which 50% CO or HC conversion is obtained is defined as the light off temperature. The catalyst light off temperature is desired to be as low as possible for good emission reduction during actual vehicle operation. . The light off temperature for a new catalyst varies from about 220 °C for CO to 260 – 270 °C for HC. With ageing, the catalyst light off temperature increases. For the first generation catalytic converters, the temperature of operation was in the range of 250 – 600 ºC and the typical gas space velocity during vehicle operation varied from 3 to 30 s^{-1}.

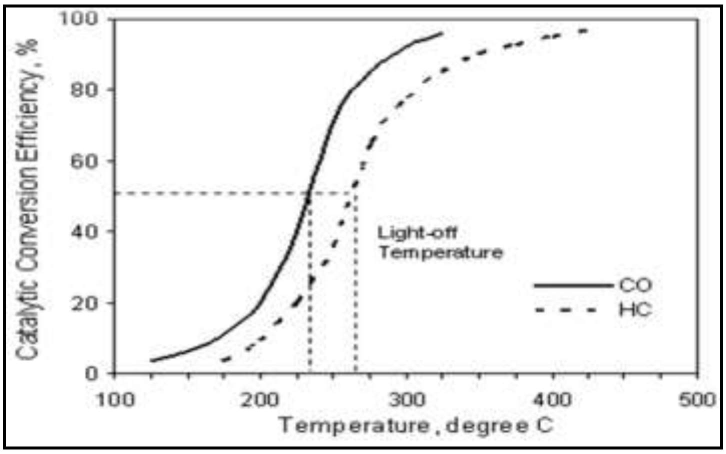


Figure 8. Conversion efficiency dependence on temperature for an oxidation catalyst [6]

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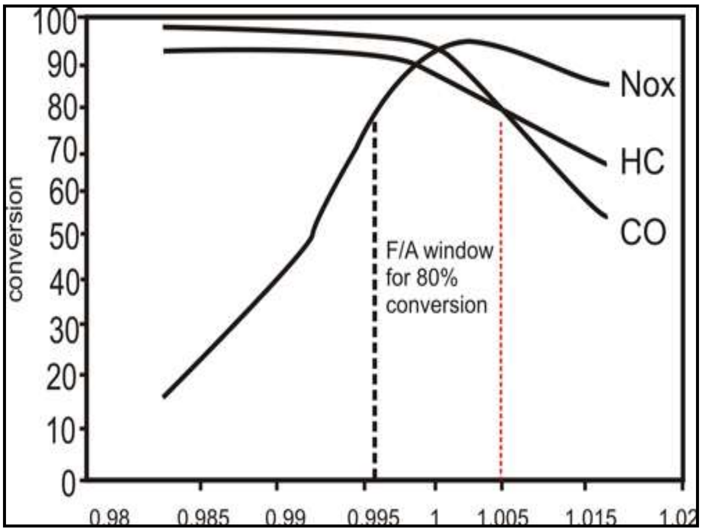

Figure 9. Conversion efficiency of a 3-way catalytic converter as a function of air-fuel ratio [7]

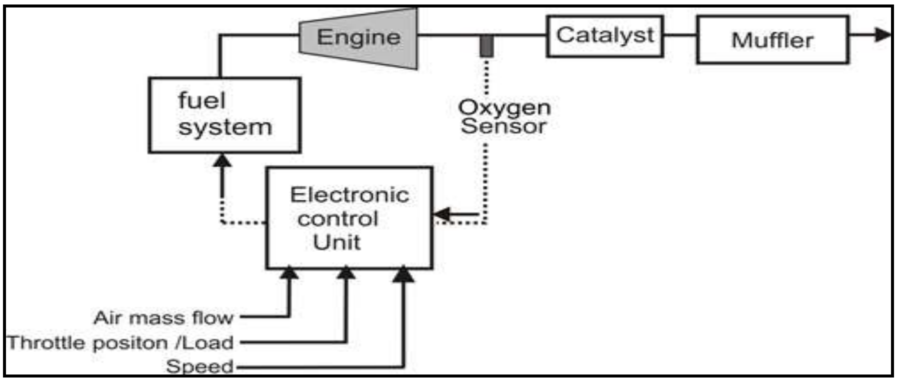

Figure 10. simple closed-loop feedback control system for air/fuel ratio control [7]

- ✓ A closed loop feedback controlled fuel management system is used for precise control of air-fuel ratio. A simple closed-loop feedback engine fuel management system is shown schematically in fig. 10.
- ✓ An oxygen sensor installed in the exhaust system detects presence of free oxygen in the exhaust gas which determines whether the fuel-air mixture is leaner or richer than stoichiometric.
- ✓ The signal from the oxygen sensor is fed to a microprocessor controlled fuel management system to adjust fuel injection rate so that the engine operates in a narrow window around the stoichiometric set point.
- ✓ Fuel-air ratio oscillates around the set point at a frequency of 0.5 to 1 Hz as the fuel flow is varied.

Signals of air mass flow rate, engine load, speed, spark timing temperatures and several other parameters are also fed to the engine electronic control unit for management of engine operation. Carburetors were found to be incompatible with such control systems although initially electronically controlled carburetors were developed and used. Now, multipoint port fuel injection system is a standard feature of engines using 3-way catalytic converters.

Conclusion

The outcomes of this research paper in the perception of study on CO emissions are listed below.

- Generally with decrease in air-fuel ratio below stoichiometric value, the formation of CO increases sharply.
- Carbon monoxide emission reduces with increase in the air-fuel ratio as more oxygen available in combustion.
- Reaction-2 paves a way for the conversion of CO to CO_2 at high temperatures.

- In most of the cases for lean mixtures, CO concentration is very small.
- For the stochiometric and lean mixtures however, partial equilibrium CO predictions have a better agreement with the measured values.
- Reduction in engine compression ratio results in higher exhaust gas temperatures which promote oxidation of CO.
- Oxidation of 1 percent CO in the gases increases gas temperature by about 145 °C.
- High conversion rates of pollutants could not be obtained in the thermal reactors.
- The thermal reactors have no effect on NO_x emissions.
- By Introducing catalyst, Palladium has higher specific activity than Pt for oxidation of CO, olefins and methane components.
- In overall under the Stoichiometric engine operating condition oxygen is enough to oxidize CO and HC and helps for the complete combustion.

Abbreviations and Acronyms

- CO – Carbon Monoxide
- HC – Hydrocarbon
- HCCI – Homogenous Charge Compression Ignition
- PM – Particulate Matter
- NO_x – Nitrogen Oxide
- ppm – Parts per million
- EGR – Engine Gas Recirculation
- TBI – Throttle Body Injection
- PFI – Port Fuel Injection
- Pt – Platinum

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Profile of Author

Dr. S. KARTHIKAYAN, FoC, National Institute of Technology Puducherry, Karaikal, completed his Research in IC Engines. He has 17 years of experience in industry and teaching.

He has published 29 papers in various reputed journals and conferences in international and national level and a text book for Anna University first year engineering students. He received DR&DO grant twice. He has received “Best Faculty Advisor Award” for the year of 2010 – 2011, from SAE India. He led the team for Supra, BAJA and effi-cycle.

He is a management committee member of SAE India Southern Section, Chennai. He is also an active member of Combustion Institute (Indian Chapter), ISTE & NIQR.

Contact Details: sskksmit@gmail.com, +91-9894457440

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