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Soot Reduction Strategy: A Review

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Abstract: The aim of study is soot reduction strategy by using microwave radiation. As a first step toward the aim is a review. This study provides a review of soot particles generation and their influence on air pollution and illustrates the diesel particulate matter emission regulation, composition, and characterization including size, shape, process, and oxidation. The paper also discusses the effect of temperature, pressure, air/fuel ratio, exhaust flow rate, fuel type on soot process and heat release from soot combustion. The use of filter regeneration strategies for reducing soot emission from vehicles, stationary plants and factories due to the burning of diesel fuel have been presented with advantage and disadvantage of each method. The various strategies include catalyst, fuel injection, electric heating, engine operation condition control, non-thermal plasma, and absorbed microwave energy. The practical feasibility for soot oxidation by microwave heating technique has been explained depending on soot material property. The proposed new technique utilizing the reflected and absorbed energies of microwave for soot reduction strategy is presented to develop a new emission after-treatment system. The new system will be modeled and simulated using appropriate software. The simulation results will be validated experimentally.

Key words: Soot, exhaust after-treatment, microwave technology

INTRODUCTION

Soot is a product of incomplete combustion, produced from vehicles, industrial works, fires and household burning of coal and fuel. The soot particles are a penalty for the entire world where they contribute to climate warming, ice melting (Godfrey *et al*., 2003) and health hazard (Chen *et al*., 2009). Figure 1 shows an example of soot exhausting from a diesel vehicle. The U.S. Environmental Protection Agency defines soot or Diesel Particulate Matter (DPM) as the mass collected on a fiber filter from exhaust that has been diluted and cooled to 52°C or below (Stratakis and Stamatelos, 2003). Diesel particulate matter emissions regulations have become more stringent in recent years. The regulation of DPM emission on highway heavy-duty engines sold in the US for the 2007 model year is less than 0.01 g bhp^{-1} h^{-1} (Fig. 2) (Henrichsen and Popuri, 2001). To understand soot reduction strategy a review is provided on soot characteristics and formation and effects of operation conditions on soot process.

Fig. 1: Soot Exhaust From diesel vehicle

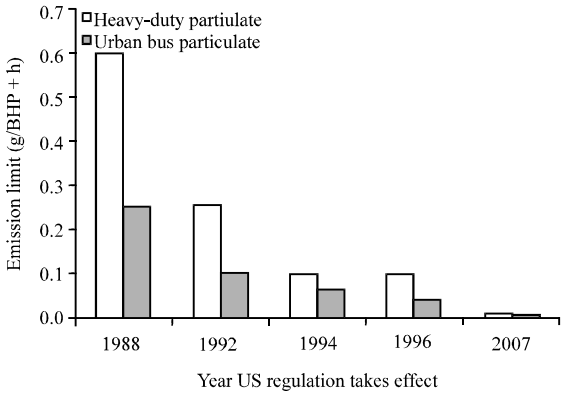

Fig. 2: US particulate emissions standards (Henrichsen and Popuri, 2001)

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The proposed techniques employing microwave technology to reduce soot will be described in detail.

SOOT CHARACTERISTICS

There are many types of soot based on the sources of the soot such as from a turbo diesel engine operated under various loads, biodiesel fuel, spark discharge generator and soot generated with a standard propane burner in a diffusion flame at various carbon to oxygen (C/O) ratios.

Saathoff *et al.* (2000) characterized various soot from different sources using Thermo Graphic Analysis (TGA). Their soot contents are divided into Elemental Carbon (EC) and Organic Carbon (OC) content. The carbon volatilized from a heated soot sample is classifies in three fractions: (350°C in helium), OC2 (650°C in helium), EC (650°C in OC1 oxygen). Figure 3 shows the ratio of EC and OC1 and OC2 to Total Carbon (TC).

Kerminen *et al.* (1997) collected particles with a Berner Low-Pressure Impactor (BLPI) over the size range of approximately 0.034-16 μm diameter. Particles are impacted on greased surfaces made of aluminum foil. Diesel emission particle samples were analyzed first by heating in the argon stream to release organic compounds and detecting them as carbon dioxide after passing them through the platinum catalyst oven. After that the samples were cooled, they were heated again in the oxygen stream to determine elemental carbon. Organic carbon was released and could be detected at temperatures below 350°C, whereas elemental carbon was seen at temperatures above 500°C. The diameters of combustion-generated particles are around 15-40 nm.

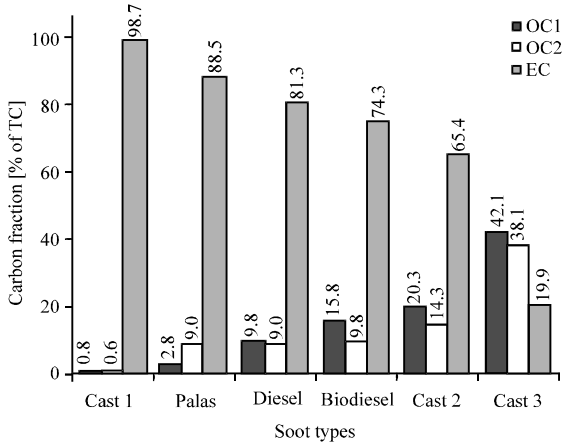

Fig. 3: Comparison of the composition of soot from various sources (Saathoff *et al.*, 2000)

After that, coagulation occurred due to the relative movement of the spherules and their small clusters where the sizes of over 80% of the number of particles were estimated around 0.1 μm and less than 0.1% had the sizes 0.7-1.0 μm. The mass of sizes 0.7-1.0 μm is about 10% of the total particulate.

Lee *et al.* (2001) used a sampling probe and optical scattering technique, reported primary particles from 20-50 nm with an average diameter of about 30 nm.

Bruce *et al.* (1991) reported the range for the primary particle diameter of 30-70 nm by using in-cylinder light-scattering measurements.

Suro *et al.* (2000) collected the samples of diesel soot by a portable impactor developed by the Delta Group at UC Davis. They analyzed the particulate matter using laser desorption ionization time of flight mass spectrometer. The structure of the soot particles emitted from a diesel engine was described as s chain composed of several tens to hundreds of primary spherical particles consist of an inner core surrounded by an outer shell. The inner cores compose of 10 nm non-planar molecules. The surrounding outer shell is a composition of micro crystallites several polycyclic aromatic hydrocarbon layers.

Stratakis and Stamatelos (2003) collected particulate samples ranging from 3.5-5.5 mg directly from the filter. Hydrocarbon and soot oxidation behavior and energy activity were analyzed based on the thermogravimetric and differential scanning calorimetry (TGA-DSC). They noted when add catalytic to diesel fuel, the TGA ignition temperature of emitted soot from modern passenger car is decreased from 630-490°C. This is because the faster catalytic oxidation of adsorbed hydrocarbon while it does not affect the oxidation behavior of soot emitted by the engine run without fuel additive.

Ullman (1989) and Lee *et al.* (1998) defined the composition of particulates is the combination of soot and other liquid-or solid-phase materials that are collected when product gases (exhaust) pass through a filter. Particulate is often separated into a soluble and an insoluble or dry fraction. The dry or soot fraction is valued by finding insoluble portion of the particulate. The soot fraction exhaust from diesel engine is not constant, typically more than 50% of the particulate. Other particulate matter constituents include unburned or partially burned fuel/lubricant oil, bound water, wear metals and fuel-derived sulfate.

SOOT PROCESS

The soot formation start from liquid-phase or vapor-phase hydrocarbons to solid particles and possibly

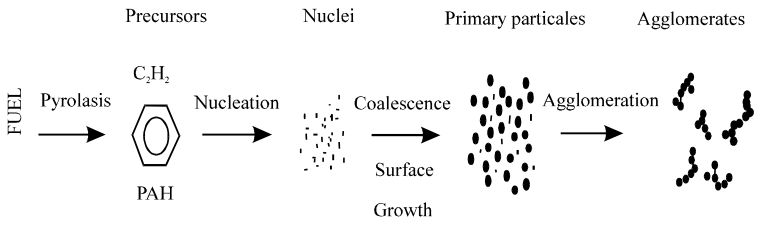

Fig. 4: Schematic diagram of soot formation from gas phase to solid agglomerated particles (Treea and Svensson, 2007)

back to gas-phase passes six processes. The processes are: pyrolysis, nucleation, coalescence, surface growth, agglomeration and oxidation. The first five processes; form the soot as shown in Fig. 4. The last process (oxidation); converts hydrocarbons to CO or CO_2, and H_2O (Treea and Svensson, 2007).

Pyrolysis is an endothermic process alters the molecular structure of organic compounds, such as fuels, when exposed to high temperature without significant oxidation even though oxygen species is available. Fuel pyrolysis produce some species which are building blocks for soot (Smith, 1981).

Nucleation is the formation of particles from gas-phase reactants. The size of smallest solid particles in luminous flames are 1.5-2 nm in diameters (Treea and Svensson, 2007).

Surface Growth is the process of adding mass to the surface of a nucleated soot particle (Treea and Svensson, 2007).

Coagulation and agglomeration:coagulation (also called coalescence) occurs when particles collide and coalesce; therefore, the numbers of particles are decreased, holding the combined mass. Agglomeration occurs when individual or primary particles stick together to form large groups of primary particles. Shape of soot particles exhaust from diesel engines are spherical for primary particles and agglomerate to form long chain-like structures (Treea and Svensson, 2007). Coagulation may be induced by the particle Brownian movement and by motions due to turbulence and to various inter-particle forces. The soot particles agglomerate further after the end of combustion, to form chain-like with 100 to 2 m diameter (Ladommatos and Zhao, 1994).

Oxidation is the conversion of carbon or hydrocarbons to combustion products. When carbon is partially oxidized to CO, the hydrocarbon will not develop to a soot particle even if entering a fuel-rich zone. Oxidation occurs at any time during the soot processes from pyrolysis through agglomeration. The most active oxidation species depends on the process, state of the mixture and the time. Oxidation of small particles is considered a two stage process: First, chemical attachment of oxygen to the surface (absorption), and second, desorption of the oxygen with the attached fuel component from the surface as a product Lee *et al*. (1998). Bartok and Sarofim (1991) stated that OH is most likely to form soot oxidation under fuel-rich and stoichiometric conditions while soot is oxidized by both OH and O_2 under lean conditions. Haynes and Wagner (1981) explained that "about 10-20% of all OH collisions with soot are effective at gasifying carbon atoms".

OPERATION CONDITIONS EFFECTS

Temperature has the greatest effect on the soot process by increasing all of the reaction rates involved in soot formation and oxidation. In a well stirred reactor, where formation is occurring, peak formation rates occur in the temperature range of 1500-1700 K (Sato *et al*., 1990). For oxidation, water evaporates at 150°C. After most hydrocarbon (called Volatile Organic Fraction (VOF)) vaporization at temperature 380-420°C, only dry soot remains which are accumulated on the filter. The ignition temperature at which the soot oxidation rate is maximized at 630°C below (Stratakis and Stamatelos, 2003) but according to Glassman (1996) soot particle oxidation occurs when the temperature is more than 1300 K. The ignition temperature of diesel soot is lower than black carbon, due to carbon content (Ma *et al*., 1997).

Pressure increasing cause more carbon collisions and higher reaction rates and soot formation. Pressure increasing more than 0.8 MPa, decreases the soot formation with increase residence time. It was suggested that this might result from higher radiation losses leading to lower temperature and lower soot formation rates (Treea and Svensson, 2007).

Stoichiometry or Equivalence Ratio of the fuel side of compression ignition flames is typically in the range between 2 and 10 where most fuels produce the maximum

Table 1: Soot characterization under different values of air/fuel ratio Palma *et al.* (2007)

Air/ fuel ratio	Bulk density (g cm^{-3})	Specific surface area (m^2 g^{-1})	Ignition temperature (K)	Oxidation max. temperature (K)	Particle size (nm)
18	1.54×10^{-2}	112	680	891	33-58
28	2.67×10^{-2}	228	650	878	24-54

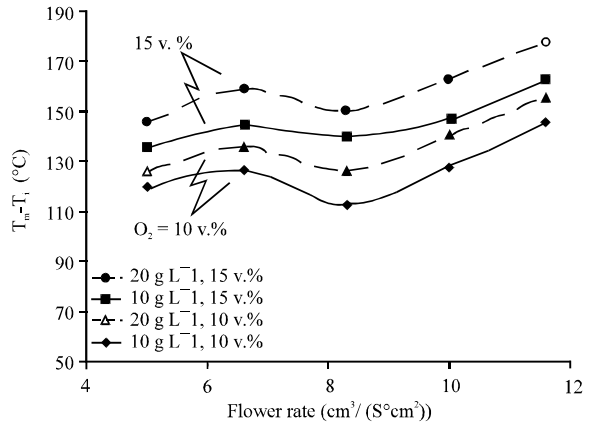

Fig. 5: Impact of filter wall flow rate, O_2 concentrations and soot loading (10 and 20 g L^{-1}) on maximum front temperature (T_m). Feed temperature (T_i) is 620°C (Chen *et al.*, 2009)

amount of soot (Treea and Svensson, 2007). If the mixture is made lean, then more oxides of nitrogen (NOx) are produced and decreases the engine efficiency (Godfrey *et al.*, 2003). Palma *et al.* (2007) reported the reactivity of air in soot collected at the exhaust of the gas-oil burner operating at different values of air/fuel ratio as in Table 1.

Exhaust Flow Rate change when change in vehicle speed and hence, change the flow rate per unit surface area of the DPF. The increased in flow rates change the value of maximum oxidation temperature between local minimum and local maximum as shown in Fig. 5 (Chen *et al.*, 2009).

Fuel Composition and Structure play an important role in soot formation in all flames. The majority of soot obtained from low soot oxygenated fuels. Increasing the number of carbon-carbon bonds increases the tendency of the fuel to soot (Treea and Svensson, 2007). The presence of catalytic converter reduces the mass of particulate by 10-30% (Kerminen *et al.*, 1997).

IN-CYLINDER SOOT FORMATION

Soot survives the combustion process through two possible pathways: (1) The exhaust valve opens and there is insufficient time to complete the combustion. (2) Portions of the flames or reaction around the perimeter become extinguished (Treea and Svensson, 2007). The ratio of soot concentration in the exhaust to that measured in peak regions of the flame can be orders of magnitude, indicating that soot deposition on walls is not a major source of exhaust particulate (Tree and Foster, 1994). Larger soot particles at the leading edge of the fuel combustion plumes than those in the center and upstream portions of the plumes (Treea and Svensson, 2007). The published values of activation energy of soot oxidation was found around 140-150 kJ mol^{-1} by Ciambelli *et al.* (1996) and Otto *et al.* (1981), 170 kJ mol^{-1} by Neeft *et al.* (1997). Stratakis and Stamatelos (2003) valued the heat release at 190 kJ mol^{-1}. Stanmore *et al.* (1999) reported a higher value of 210 kJ mol^{-1}.

REDUCTION STRATEGY

Diesel Particulate Filter (DPF) is a wall-flow type trap oxidizer system mechanically filters and collects the particulate matter from the exhaust gas (Stratakis and Stamatelos, 2003). The recollected soot or PM on the DPF leads to closing the pores in the wall of filter and resisting the exhaust flow resulting in increasing filter backpressure. Backpressure increase is undesirable since it increases fuel consumption, reduces available torque and engine operating efficiency (Stamatelos, 1997). Thus, it is necessary to clean the trap periodically by burning (oxidizing) the collected particulate (filter regeneration). Self-regeneration is not possible because the ignition temperature of diesel soot typically 500-650°C and the engine exhaust temperature is lower than 400°C Ma *et al.* (1997). Several techniques have been employed to regenerate DPF.

Catalyst deposited over the trap is used to oxide the soot indirectly as illustrated in Fig. 6 by the aid of available NOx in the exhaust gases and lower the soot ignition temperature (Hacfort *et al.*, 2000). The disadvantages are (Fino and Specchia, 2008):

- Low effect on soot oxidation because the soot temperature cannot rise to ignition point
- Low effect on trap pressure drop (backpressure)
- Not enough to reduce the energy required to soot combustion

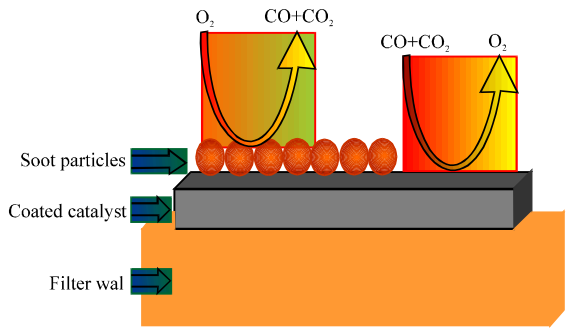

Fig. 6: Catalyst deposited over the walls of Trap (Van der Zande *et al*., 2002)

Fuel post-injection with catalyst (Bensaid *et al*., 2010) or electric heating (Chen *et al*., 2009) are used to regenerate DPF by rising the temperature to soot ignition degree. The drawbacks:

- Non uniform distribution of soot lead to multi spot ignition and this lead to excessive temperature at the end of DPF lead to damage the DPF (Chen *et al*., 2009)
- Lost heat by walls (Martirosyan *et al*., 2010)
- Slow regeneration (Ma *et al*., 1997)
- Consumed energy (Ma *et al*., 1997)
- Reduced filter service life (Ma *et al*., 1997)
- Risk due to moving parts or electrical connection in the exhaust stream (Henrichsen and Popuri, 2001)

Engine operation condition control: Cummins Power Generation work to control oxide of nitrogen and PM (Herzog, 2002) by varying parameters such as exhaust gas flow rate, air/fuel ratio, fuel type, injection typing and timing, electronic sensing and control, and engine modification. These control strategies were targeted to optimize the emission control during in-cylinder combustion (Treea and Svensson, 2007).

Non thermal plasma: There are several plasma discharge devices types which could be used for engine exhaust treatment (Hhoard *et al*., 1999). The chemical reactions that took place are a result due to electron excitation (Lepperhoff *et al*., 1999). The advantage of this method is the ability to work in cold-start and reduction of the exhaust backpressure (Lepperhoff *et al*., 2000). The disadvantages:

- Low electrical strength/efficiency and energy distribution (Ighigeanu *et al*., 2006; Whealton *et al*., 1995)
- The electrode discharge are polluting specially with corrosive gases (Ighigeanu *et al*., 2006)
- Microwave heating: Several researchers have used microwave-assisted catalytic to Palma *et al*. (2004)
- Reduce the temperature of the chemical reaction and regenerate the PM trap
- Increase the temperature of soot with microwave power

While others have used microwave, non-thermal plasma and catalyst to Ighigeanu *et al*. (2006):

- Sustain microwave plasma over a very wide pressure range
- Remove a source of contamination by the absence of internal electrodes
- Ensure fast and easy control of the process

Advantage of using microwave in DPF regeneration:

- Volumetric heating means that materials can absorb microwave energy directly and internally and convert it to heat rapidly (Wu, 2002)
- Rapid heating reduce thermal gradients and stresses (McDonald *et al*., 2004)
- Efficient and fast energy transfer decrees processing time (Palma *et al*., 2004)
- Carbon has low ignition temperature by microwave heating than electric heating (Palma *et al*., 2002)
- Controlling the maximum temperature by controlling source power (Ma *et al*., 1997)
- Carbon is a strong absorber of microwave (Ma *et al*., 1997)
- Selective heating of materials through differential penetration (Ma *et al*., 1997)
- Self-limiting reaction (Ma *et al*., 1997)
- Reversed temperature gradient (heating from the inside out) fast dray moisture (McConnell, 1999)

The disadvantage are the non-homogeneous electric field and energy distribution in the mono-mode cavity, gives non-uniform temperature rise gradients (Zhang-Steenwinkel *et al*., 2005) and increases power consumption (Van der Zande *et al*., 2002). In addition, the design of a multi-mode cavity to obtain a uniform filter heating is difficult (Zhang-Steenwinkel *et al*., 2005). Supplying high microwave power increases filter regeneration efficiency but also increases thermal stresses inside the filter (Palma *et al*., 2007).

SOOT OXIDATION BY MICROWAVE

Electromagnetic waves, can exhibit reflection, refraction, polarization, and constructive and destructive interference. Low frequency electromagnetic waves with long wavelengths are called "radio waves" and are used primarily for broadcasting and communication systems within the atmosphere. Microwaves are a subset of radio

waves and fall into the frequency range from 0.3-300 GHz and wavelength from 1000-1 mm (Godfrey *et al*., 2003). The magnetron is a complete device, operates by external circuit, amplifying the power by the interrelation between the acceleration - deceleration of electrons and a time-varying electromagnetic field to generate microwave energy (Theury, 1992). The Radio-Frequency (RF) technology was employed for on-line trapped diesel particulate measurement and filter regeneration control (Zhi *et al*., 2000). Microwave energy is not the high-energy penetrating radiation as the gamma radiation that emitted at frequencies much higher than those used in microwave devices. This type of radiation is dangerous because it is "ionizing" radiation that is capable of changing molecular configurations and causing cellular damage. However, microwaves can cause physical damage if proper safety is not observed in the presence of strong microwave fields which are not contained in a shielded device. This damage would most likely be in the form of burns or hyperthermia instead of molecular damage, and low levels are not harmful due to the low penetration ability of weak fields (McConnell, 1999). The carbon is a good energy absorber where the dielectric constant (ϵ'') = 8.6, dielectric loss function (ϵ'') = 7.4 and dielectric loss tangent (tanδ) = 0.86 (Henrichsen and Popuri, 2001). The absorbed electromagnetic energy (dP_{abs}) is dissipated to thermal energy within the material in terms of source frequency (f), free space permittivity (ϵ_0) = 8.854 x 10-9 (F/m), electric field $|E|$ and volume (dV) (McConnell, 1999):

$$d_{pabs} = 2\pi f \varepsilon_0 \varepsilon' |E|^2 \, dv \quad (1)$$

CONCLUSION

Key advantages of microwave heating over other active regeneration systems include volumetric, reversed, rapid and selective heating, penetration of microwaves, and less moving parts or electrical connections in the exhaust stream. The current study proposed a new technique utilizing the characterization of reflected and absorbed microwave energies for soot reduction strategy by heating. To ascertain this promised technique the effects of soot loading, exhaust flow rate, ignition temperature, and microwave power and heating duration will be modeled and simulated to develop the new exhaust after-treatment regeneration system. The use of a supplemental air source will be determined. The simulation results will be validated experimentally.

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